Monte Carlo Model of a Low-Energy Neutron Interrogation System for Detecting Fissile Material

Submitted to the Department of Nuclear Science and Engineering on May 12, 2006 in Partial Fulfillment of the Requirements for the Degrees of Master of Science in Nuclear Science and Engineering and Bachelor of Science in Nuclear Science and Engineering

Abstract

The undeniable threat of nuclear terrorism presents an opportunity for innovation in developing active interrogation technology. The proposed system aims to detect the smuggling of special nuclear material (SNM) in maritime containers. Identifying the importation of SNM will be instrumental in protecting the American public from a nuclear terrorist attack made possible by the construction of a weapon with fissile material from abroad.

The proposed system uses a directionally-biased beam of low-energy neutrons (60 – 100 keV) generated from a $^7\text{Li}(p,n)^7\text{Be}$ reaction run near threshold. These neutrons are directed towards a cargo container of unknown composition. If SNM is present in the container and the neutrons can reach it, high-energy fission neutrons will be detectable outside the cargo container.

MCNP models indicate that even low-energy neutrons will be able to penetrate through reasonable amounts of material likely to be encountered in cargo environments. The only major exception is hydrogenous material, which could alter the radiation signature. The presence of shielding material may further alter these results. Small amounts of shielding that is hydrogenous will thermalize incident neutrons and raise the likelihood of generating fissions. An abundance of shielding material could mask the presence of fissile material but will also result in changes in the induced gamma energy spectrum and greatly increase the flux of thermal neutrons even outside the cargo container. Still, this material would not be resistant to other radiological techniques and the presence of an abundance of hydrogen will be evident and potentially raise suspicion in and of itself.

Further MCNP simulations of the neutron source impinging on cargo containers suggest that this technique can respond, as expected, qualitatively differently to containers containing SNM from containers that do not. Containers that contain small amounts of fissile isotopes as in the case of a few grams of uranium-235 in a kilogram of depleted uranium will also respond to this method but much more weakly.

The system as proposed is viable and further simulation and experimental work will elucidate the behavior of this system under a wide range of cargo environments.

Thesis Supervisor: Richard C. Lanza
Title: Senior Research Scientist in Nuclear Science and Engineering
Acknowledgements

I would first like to thank Dr. Richard Lanza for his advice and encouragement. I also thank him for his efforts in generating the backing for this project among those interested in supporting the development of non-proliferation technology without which I would not have the privileged opportunity to pursue graduate studies.

Professor Jacquelyn Yanch has been immensely patient in providing her generous insight into the development of models of radiation transport phenomena, methods essential to the completion of this thesis.

Dr. Brandon Blackburn and his expertise in nuclear applications of non-proliferation technology have been instrumental in shaping the direction of this project.

Also, Eduardo Padilla’s additional insight and suggestions have further enriched the depth of this thesis.
# Table of Contents

ABSTRACT ....................................................................................................................... 3  
ACKNOWLEDGEMENTS ................................................................................................. 4  
TABLE OF FIGURES ...................................................................................................... 6  
LIST OF TABLES ........................................................................................................... 7  

1.0 INTRODUCTION ..................................................................................................... 8  
  1.1 Motivation for the Development of SNM Detection Systems ......................... 8  
  1.2 Prior and Current Approaches to Detecting Fissile Material .......................... 9  
  1.3 Thesis Objectives ............................................................................................... 11  

2.0 LOW-ENERGY NEUTRON PENETRATION ......................................................... 14  
  2.1 Selecting a Neutron Energy ............................................................................... 14  
  2.2 Neutron Penetration at Varying Energies ....................................................... 16  
  2.3 Neutron Penetration in Specific Media ............................................................. 18  
    2.3.1 MCNP Problem Geometry ....................................................................... 18  
    2.3.2 Tallying Approach .................................................................................... 19  
    2.3.3 Tallying Method Details ........................................................................... 21  
    2.3.4 Low-Z Hydrogenous Materials ................................................................. 25  
    2.3.5 Low-Z Non-hydrogenous Materials .......................................................... 28  
    2.3.6 High-Z Materials ....................................................................................... 32  
  2.4 Summary ............................................................................................................. 35  

3.0 NEUTRON INTERROGATION IN CARGO ENVIRONMENTS ......................... 38  
  3.1 Cargo Model Geometry ..................................................................................... 38  
  3.2 Basal Signal Modeling ....................................................................................... 40  
    3.2.1 Basal Neutron Flux .................................................................................... 40  
    3.2.2 Basal Gamma Flux .................................................................................... 42  
  3.3 Neutron Interrogation of Cargo Model ............................................................. 44  
    3.3.1 Neutron Source Definition ....................................................................... 44  
    3.3.2 Neutron Interrogation in Cargo With versus Without Fissile Material... 46  
  3.4 Shielding of the Incident Neutron Beam ........................................................... 51  
    3.4.1 Moderation of Incident Neutrons with Little Shielding ......................... 52  
    3.4.2 Absorption of Incident Neutrons with Moderate Shielding .................. 53  
  3.5 Discriminating Fissile Isotopes from Fissionable Isotopes ............................. 54  
  3.6 Improving Selectivity ......................................................................................... 56  
    3.6.1 Increase in Thermal Neutron Flux ............................................................... 57  
    3.6.2 Changes in Gamma Energy Spectrum ....................................................... 59  
  3.7 Summary ............................................................................................................. 61  

4.0 FUTURE WORK AND CONCLUSIONS ............................................................... 62  
  4.1 Special Nuclear Material Decay ........................................................................ 62  
  4.2 Limitations of System and Proposed Solutions .............................................. 63  
  4.3 Conclusions ....................................................................................................... 64  

REFERENCES ............................................................................................................... 66  

APPENDIX A MONTE CARLO METHODS IN MCNP ................................................. 68  
APPENDIX B REPRESENTATIVE MCNP INPUT DECKS ....................................... 70  
APPENDIX C SPECIAL NUCLEAR MATERIAL AGING MODEL ............................ 81
Table of Figures

| Figure 2-1: Total neutron interaction cross section for boron-11 [Cullen, 2003] | 17 |
| Figure 2-2: Penetration MCNP Problem Geometry | 19 |
| Figure 2-3: Total neutron flux through steel with 14 MeV incident neutrons | 24 |
| Figure 2-4: Total neutron flux through HDPE with 60 keV incident neutrons | 26 |
| Figure 2-5: Total neutron flux through HDPE with 14 MeV incident neutrons | 26 |
| Figure 2-6: Total neutron flux through concrete for 60 keV incident neutrons | 28 |
| Figure 2-7: Neutron Radiative Capture Cross Section for Fe-56 [Cullen, 2003] | 29 |
| Figure 2-8: Thermal (E< 1 eV) neutron flux in concrete for 60 keV incident neutrons | 29 |
| Figure 2-9: Thermal neutron flux in steel for 60 keV incident neutrons | 30 |
| Figure 2-10: Neutron Total Cross Section for Al-27 [Cullen, 2003] | 31 |
| Figure 2-11: Total neutron flux through Al for 60 keV incident neutrons | 31 |
| Figure 2-12: Total neutron flux through Al for 100 keV incident neutrons | 32 |
| Figure 2-13: Total neutron flux through W for 100 keV incident neutrons | 33 |
| Figure 2-14: Total neutron flux through W for 14 MeV incident neutrons | 33 |
| Figure 2-15: High (E< 1 MeV) neutron flux in depleted uranium for 100 keV incident neutrons | 34 |
| Figure 3-1: Interrogation in Cargo MCNP Problem Geometry | 39 |
| Figure 3-2: Energy-angle distribution of 7Li(p, n)7Be source [Kerr et al., 2005] | 45 |
| Figure 3-3: Neutron distribution from neutron source in air with no cargo | 45 |
| Figure 3-4: Neutron flux at all energies with HEU in cargo container | 47 |
| Figure 3-5: Neutron flux at all energies with Pb in cargo container | 47 |
| Figure 3-6: Fast neutron flux (E > 1 MeV) with HEU in cargo container | 49 |
| Figure 3-7: Fast neutron flux with Pb in cargo container | 49 |
| Figure 3-8: Fast neutron flux with Pu in cargo container | 50 |
| Figure 3-9: Fast neutron flux with HEU and 5 cm concrete | 52 |
| Figure 3-10: Fast neutron flux with HEU and 25 cm concrete | 53 |
| Figure 3-11: Fast neutron flux for DU in cargo container | 55 |
| Figure 3-12: Fast neutron flux with DU and 25 cm concrete | 56 |
| Figure 3-13: Thermal flux with HEU and 25 cm concrete | 57 |
| Figure 3-14: Thermal flux with DU and 5 cm concrete | 58 |
| Figure 3-15: Gamma energy spectrum for fissile and fissionable material with light or heavy shielding | 59 |
List of Tables

Table 2-1: Uranium-235 Fission Cross Sections at Selected Energies [Cullen, 2003] ..... 15
Table 2-2: Material Compositions [MatWeb, 2006], [ICRU, 1989].......................... 36
Table 2-3: Neutron Penetration in Materials from MCNP models .............. 36
Table 3-1: HEU spontaneous fission neutron emission rate [Cullen, 2003], [Kerr et al.,
2005], [Gallagher, 2005]................................................................................... 41
Table 3-2: Pu spontaneous fission neutron emission rate [Cullen, 2003], [Kerr et al.,
2005], [Rudisill and Crowder, 2000]................................................................. 42
Table 3-3: Gamma spectrum for 7 kg HEU given in flux (photons/cm²/sec) .......... 44
Table 3-4: Gamma spectrum for 1 kg Pu given in flux (photons/cm²/sec)............ 44
1.0 Introduction

The threat of nuclear terrorism has captured the consciousness of both the American public and the American government. While nuclear terrorism as an issue is rather broadly defined, the particular threat of the importation of nuclear material with the intention of creating a weapon to use in the country is perceived as one of the more significant vulnerabilities. Special nuclear material (SNM) in some nuclear-capable nations is known to be insecure: inventory controls at these sites are ineffective and rogue organizations exist locally who would be interested in acquiring this material for potential distribution to terrorist organizations globally [National Research Council, 2002].

1.1 Motivation for the Development of SNM Detection Systems

Particularly worrisome is a scenario in which a terrorist acquires SNM abroad and imports it piecewise into the United States via shipping ports [Helfand et al., 2002]. In this case, both the material acquisition and importation is feasible though difficult. The material is potentially available at poorly guarded sites abroad though there is no indication that any terrorist organization has ever managed to take advantage of this situation [Cameron, 2000]. Additionally, limited means exist today to detect SNM entering the United States. Unlike highly radioactive material, which is desirable for a dirty bomb, SNM does not always emit sufficient radiation for a passive detection system and thus is difficult to detect directly. Highly enriched uranium is particularly unsuited to detection via passive measurement of gamma rays and spontaneous fission neutrons because of its minute emissions during natural decay. This fact is especially true if natural uranium fuel stock is used for enrichment, in which case radioactive impurities, as seen in spent fuel sources, will not be present [National Research Council, 2002].

The development of a system for detecting SNM at shipping ports would be immensely helpful to establishing security. With several of the most active seaports
in the world, protecting the United States will require widely deployable or at least the appearance of widely deployable detection technology [National Research Council, 2002]. In addition, because most SNM does not produce easily detectable radiation without being stimulated, an active probing system will likely be necessary as a basis for detecting nuclear weapons [Slaughter et al., 2003].

1.2 Prior and Current Approaches to Detecting Fissile Material

Several methods for SNM detection do exist and many of these approaches have been implemented in various forms over the past decade. All focus on finding a characteristic emission or set of emissions that is known to only come from SNM. Either this signal may be radiation that is induced via an active probing of the material or instead simply radiation emitted as a result of the natural decay occurring in weapons material; the latter case would require only a passive detector.

The expected composition of material of a hypothetical rogue weapon will determine the effectiveness of active versus passive detection methods. Nuclear weapons being imported clandestinely into the country will almost certainly be based on either uranium-235 or plutonium-239 owing to the greater availability of these fissile isotopes relative to others. These devices will of course not be entirely free of other isotopes as enrichment of any isotope becomes increasingly costly for minimal gains in weapon yield. Additionally, they could have been procured from reactor refuse, intercepted reprocessed fuel, or weapons stockpiles among other possible sources [Fetter et al., 1990]. In these cases, other isotopes such as uranium-238, and uranium-234, uranium-232 or plutonium-241, and plutonium-240 will appear. Decay chain daughters and fission products will also be present for material that has aged appreciably. These additional isotopes may have important effects on the emitted radiation under varying circumstances. While neither plutonium-239 nor uranium-235 emits significant spontaneous neutron or
gamma radiation [Turner, 1995], these other isotopes (plutonium-240 and uranium-232, in particular) could prove to be very useful for revealing the presence of fissile material. The degree to which these latter isotopes are present in the material will affect the efficacy of passive detection methods since these approaches depend upon the natural emissions of SNM [Fetter et al., 1990].

Methods under current research span a wide range but focus exclusively on active techniques. Researchers presently avoid the investigation of passive approaches mainly because most natural radiation emitted by SNM is easily absorbed with a reasonable amount of shielding [Slaughter et al., 2003]. Additionally, not all material suitable for creating a weapon would contain the isotopes most easily detected with a passive system [Moss et al., 2003]. For these reasons, current research efforts concentrate on the development of active systems.

One set of approaches attempts to identify SNM by applying high-energy, high-intensity photon fluxes on a target in order to establish a density image, which will give some qualitative idea of the atomic number \(Z\) of the material inside [Slaughter et al., 2003]. With atomic numbers of 92 and 94 for uranium and plutonium, respectively, this method can be sensitive to, though not selective for the presence of SNM. Unfortunately, many innocuous materials also have high \(Z\) and will not be distinguishable from SNM with this technique. Alternatively, at very high energies (on the order of several MeV), one can look for gamma rays and neutrons resulting from photoneutron and photofission events. The impinging photons must exceed a threshold energy that depends on the particular target nuclide in order for these reactions to occur [Slaughter et al., 2003]. One group using 6-7 MeV gamma rays from a \(^{19}\)F(p, \(\gamma\)\(\alpha\))\(^{16}\)O source generates neutrons selectively in nuclear materials [Micklich and Smith, 2005]. These materials have a photoneutron emission threshold at least 1.5 MeV lower than benign materials and so the approach is selective for SNM under photon bombardment. However,
this technique is limited by the radiation dose rate that the target will tolerate [Micklich and Smith, 2005].

There has also been work in detecting the characteristic signal of certain identifiable fission products. Neutron irradiation will lead to fission of fissile material, and the products of this process emit delayed neutrons and gamma rays with half-lives approximately sixty seconds or less [Slaughter et al., 2003]. The gamma rays emitted in this process are of such high intensity that they will not experience significant attenuation in most cargo environments [Norman et al., 2004]. In addition, some groups [Moss et al., 2004] have used deuterium-tritium (DT) generators to produce 14 MeV neutrons for a neutron interrogation probe. This approach has had some success particularly because neutrons of this energy can penetrate a reasonable amount of shielding [Moss et al., 2004]. However, uranium-238, a non-fissile isotope, will also fast fission under this type of irradiation producing neutrons and gamma rays that will be detected by the system. Thus, a positive signal may not truly indicate the presence of material for a weapon [Dietrich et al., 2005].

### 1.3 Thesis Objectives

The approach studied in this thesis uses low-energy neutrons (60-100 keV) from a $^7$Li(p,n)$^7$Be source and detects fission neutrons produced from fissile isotopes. Neutrons seen in this fashion unambiguously indicate the presence of fissile material. Non-fissile isotopes such as uranium-238 only fast fission above a threshold of 1 MeV, which is well above the energy range of the chosen neutron source [Dietrich et al., 2005]. In addition to specificity, this technique also has the advantage of producing a directed flux of neutrons [Dietrich et al., 2005] as opposed to the isotropic distribution produced in DT generators. One possible caveat is that lower energy neutrons will be less able to penetrate shielding in the cargo. However, since the number of scattering events for a given change in
energy decreases logarithmically with decreasing energy, only five scatters will be sufficient to moderate a 14 MeV neutron to 100 keV energies [Dietrich et al., 2005].

In order to direct future experimental work, the objective of this thesis is to establish numerical models that will provide useful predictions of the behavior of the proposed system. Using the Monte Carlo N-Particle (MCNP) radiation transport code, the planned setup will be examined in simulations aimed at producing better-informed future experimental work.

The first goal will be to establish that the chosen neutron source and consequent spectrum will be sufficiently versatile for this application in a variety of cargo environments and interfering materials. Using MCNP models of bare solid materials likely to appear in cargo environments and varying neutron source energy spectra, the penetration ability of possible chosen neutron energies will be compared with one another for likely interacting media.

Once low neutron energies are known to be effective, the effects of incident neutrons from a model source will be analyzed for several cargo environments. These cargo models will vary by internal target material composition and intervening extraneous cargo that behaves as shielding. This analysis will provide a preliminary evaluation of the ability of this system to discriminate innocuous from dangerous material.

In a cargo environment, the basal signal generated from radioactive decay of material will have to be determined so experimental work can be better informed and future users of a non-prototype implementation of this system can know the expected changes in emitted radiation for a dangerous cargo container under interrogation.
Given these results, future experimental studies may be implemented based on the output of these simulations.
2.0 Low-Energy Neutron Penetration

More than one school of thought exists as to the best choice of energy for SNM detection applications. Any choice of neutron energy must be a compromise between penetration ability of the impinging neutrons and SNM selectivity. As discussed in Section 1.3, neutrons with an energy below several hundred keV will fission fissile isotopes such as uranium-235 but not non-fissile fissionable isotopes such as uranium-238. While these low-energy neutrons then interact with fissile material selectively, they may not penetrate as far through the material because they have less energy to lose in scattering events.

Conversely, high-energy neutrons will reach further into material; however, owing to high fast fission cross sections, these neutrons will generate fission neutrons in materials that cannot be used for constructing weapons. This will lead to false positives in a system relying on high-energy neutrons. However, a system based on low-energy neutrons could be prone to false negatives when neutrons undergo capture in cargo material before reaching fissile material. All these factors must be considered and weighed in order to develop an effective SNM detection system.

2.1 Selecting a Neutron Energy

Thermal neutrons (~0.0253 eV) can be quickly eliminated from consideration because most materials comprise isotopes with high thermal neutron interaction cross sections. It is true that thermal neutrons will most readily interact with fissile nuclei; the fission cross section of uranium-235 is more than ten times higher at thermal energies than at 60 keV and 280 times than at 14 MeV (See Table 2-1). Still, with relatively high total cross sections for most cargo materials, thermal neutrons will be quickly absorbed. After low- and high-energy neutrons traverse several mean free paths in a material, these neutrons will have lost much of their energy to scattering events and will have both penetrated into cargo and thermalized. Any benefit that could be derived from thermal neutrons will be
gained once neutrons of a higher than thermal energy have been moderated to thermal energies with the advantage of increased penetration into cargo (see Section 2.2 for descriptions of neutron moderation in media). The same is not true for high-energy neutrons (14 MeV) being moderated to low energy (100 keV); if a high-energy neutron encounters fissile material, it could generate fast fission in non-fissile nuclei, an undesirable result for selectivity.

**Table 2-1: Uranium-235 Fission Cross Sections at Selected Energies** [Cullen, 2003]

<table>
<thead>
<tr>
<th>Energy (keV)</th>
<th>2.53x10^{-5}</th>
<th>60</th>
<th>100</th>
<th>14x10^{-3}</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fission Cross Section (barns)</td>
<td>584.4</td>
<td>52.5</td>
<td>7.4</td>
<td>2.06</td>
</tr>
</tbody>
</table>

The remaining spectrum choices are high-energy neutrons (~14 MeV) and low-energy neutrons (~60-100 keV). High-energy neutrons can be readily produced with deuterium-tritium generators [Knoll, 2000]. A disadvantage of this type of source though is its isotropic distribution of neutron emission [Blackburn, 2005]. For a system for scanning cargo, in order to improve energy efficiency and minimize surrounding dose rate, it is most desirable to choose a system that produces a directed neutron beam naturally. This can be readily accomplished with a source that already produces a bias in the velocity distribution of the emitted neutrons.

Low-energy neutrons for this application can be produced with a high-energy proton source (i.e. an accelerator) driven near threshold for the $^7\text{Li}(p,n)^7\text{Be}$ reaction. The kinematics of this reaction lead to a forward-biased distribution of low-energy neutrons that can be directed towards a target such as a cargo container [Blackburn, 2005].
2.2 Neutron Penetration at Varying Energies

Two factors determine how well a neutron beam will penetrate an intervening material: the rate of energy loss in the medium and the mean free path of the neutrons at the chosen energy.

It can be argued that the difference in penetration ability of a neutron at 14 MeV should not be significantly greater than that at 100 keV because the expected number of elastic scattering events \( n \) it takes to moderate a neutron depends logarithmically on the ratio of the final energy \( E' \) and initial energy \( E \) is [Turner, 1995]:

\[
    n \propto \ln \left( \frac{E}{E'} \right) 
\]

Therefore, for a chosen nucleus, one can expect it to take five times fewer scattering events for a neutron to slow from 14 MeV to 100 keV than from 100 keV to thermal energies (~0.0253 eV). Consequently, one could estimate that the penetration will not be greatly increased even after significantly increasing interrogating neutron energy [Lanza, 2005].

Because momentum must be conserved in the interaction, the amount of momentum, and consequently energy, that can be transferred from the neutron to a target nucleus depends on the mass of the target nucleus. The expected amount of energy transferred in a given scattering collision will be much less for a heavier nucleus than for a lighter one. After an elastic collision with a nucleus with mass number \( A \), the ratio \( \alpha \) of the minimum final energy of a neutron to its initial energy is:

\[
    \alpha = \frac{(A - 1)^2}{(A + 1)^2} 
\]

A neutron whose final energy \( \alpha \) times its initial energy will have undergone the maximal energy transfer for that elastic scattering event. A hydrogen nucleus
weighs roughly the same as a neutron. In a scattering event with hydrogen \((A=1)\), as much as the entire energy can be transferred to the nucleus, but for a uranium-238 nucleus, at most only 1.67 percent of the initial neutron energy can be given to the uranium nucleus. Consequently, one will expect more scatters and, assuming comparable atomic density, greater penetration of a neutron in a material composed of isotopes with high mass number. Between the effects of reduced energy transfer in heavy nuclei and limited decrease in number of scattering events relative to 14 MeV neutrons, one can expect significant penetration of neutrons in heavy materials even at energies around 100 keV.

![MT=1: (n,total) Cross section for B11 from ENDFB 6.8 from NEA](image)

**Figure 2-1: Total neutron interaction cross section for boron-11** [Cullen, 2003]

However, an additional factor must be accounted for in estimating the penetration ability of neutrons of a particular energy: the mean free path of the neutron and, particularly, how the mean free path changes with decreasing energy. A neutron may only require a few scatters to reach 100 keV from 14 MeV but if the mean free path of the neutron near 14 MeV is significantly greater than around 100 keV, the 14 MeV may demonstrate significantly greater penetration ability.
The mean free path is inversely related to the macroscopic cross section, and cross sections typically decrease with increasing energies (notable exceptions being resonance regions in the neutron spectrum) [Turner, 1995]. For example, in boron-11, one can expect an energy transfer (from Equation 2-2) no greater than 31 percent. But, as can be seen in Figure 2-1, the cross section drops a factor of three between 60 keV and 14 MeV. So, while a 14 MeV neutron may only have a few scatters to be moderated from 14 MeV to 60 keV, it will be able to cover three times the distance between each scattering event.

Due to these competing factors and the complicated true variations in cross section, numerical computations with MCNP are necessary to effectively characterize the differences in penetration ability of neutrons at varying energies in media (see Appendix A for an overview of MCNP methods).

2.3 Neutron Penetration in Specific Media

Simulations of neutron penetration in several media were performed using the MCNPX code. An especially useful feature of this implementation of MCNP is the mesh tally, which can overlay the defined problem geometry with a grid of cells at specified intervals. Details of how the tally is utilized for studying neutron penetration in media is given in Section 2.3.2. For the mesh tally, as particles are followed as normal through the problem geometry and mesh grid, all particle tracks are tallied individually for each cell. After all the data are acquired for each initial particle run, the tallies associated with each cell can be combined into a readily visualizable data set representing the entire problem [Waters, 2002].

2.3.1 MCNP Problem Geometry

The penetration simulations comprise two components: a neutron source and a medium. The neutron source as defined emits monoenergetic neutrons of a chosen energy directed in a single direction towards the center of one face of a 500 cm x
500 cm x 500 cm slab of a specified material as depicted in Figure 2-2. A sphere of air with a radius of 500 cm centered on the slab uniformly surrounds the slab.

![Diagram of slab and sphere](image)

**Figure 2-2: Penetration MCNP Problem Geometry**

### 2.3.2 Tallying Approach

It should be noted that with the neutron source specified as the origin for the geometry so long as the radiation transport problem is examined from points within 250 cm of the center of a given face, the problem is cylindrically symmetric about the neutron source with the direction of the neutrons chosen as the axis untransformed from Cartesian coordinates.

In order to preserve the cylindrical symmetry, one might expect the slab to be made semi-infinite in the direction away from the particle source. However, in a real experiment, neutrons would also be reflected out of the material and scattered from the air back into the medium. As will be seen below, most saliently for hydrogenous media, more than a few tens of centimeters of radial distance away from the neutron source, the incoming neutron flux penetrates uniformly.
Thus, an appropriate tallying method will be to use a cylindrical mesh with the origin \((r=0)\) for the mesh being the point on the slab surface nearest to the neutron source. So long as the mesh does not extend past half the width of the slab (in this case 250 cm), the MCNP problem geometry will be cylindrically symmetric and no significant information will be lost when using a single angular bin in the cylindrical mesh. In these models, 200 cm is the maximal extent of the mesh because for most tested materials, obtaining statistically significant flux tallies is generally prohibitively computationally taxing beyond 200 cm. Because of the cylindrical symmetry of the problem for the mesh’s extent, a single bin in the \(\theta\) direction can be used to cover the regions of space for a given angle. The symmetry allows for the radiation transport to be uniform in a given angle; any deviations from this ideal situation are purely consequences of the stochastic nature of the Monte Carlo simulation process. By summing over all angles, the variance can be appreciably reduced from what could be obtained with a rectangular mesh grid divided in all three dimensions. Also, radial and axial bin divisions will be delimited every 10 cm to provide sufficient detail to catch spatial features of the neutron distribution. The MCNP paradigm and algorithm are elucidated in Appendix A.

Additionally, the mesh tally can be split up into several energy bins so the neutron spectrum can be measured throughout the material volume. The important regions to monitor are the ones that we will arbitrarily label for convenience the thermal region (less than 1 eV), the low energy region (between 1 eV and 1 MeV), and the high energy region (between 1 MeV and 14 MeV). The thermal region is significant because this region has the highest interaction cross sections and will contribute most significantly to inducing fissions in any potentially present SNM. The low-energy region is where one would expect to see neutrons when using either a high- or low-energy source. When using low-energy neutrons, you would not expect to see high-energy neutrons unless the impinging neutrons were
inducing fission events. Examining how both the high- and low-energy neutrons migrate can shed light on how both interrogating particles and particles born in fissile material responses will be transported through expected cargo material.

An example input deck used to make the measurements depicted throughout the rest of this chapter is available in Appendix B.

2.3.3 Tallying Method Details

Prior penetration studies for active neutron interrogation applications have used the distance reached by one percent of incident neutrons as a metric for penetration ability [Dietrich et al., 2005]. This approach is sensible because any neutrons that make it through the material will be able to induce the emission of detectable fission neutrons.

In this paper, however, the neutron flux at a given point has been chosen to be the measured quantity in comparing penetration in various materials. This metric is more apt for determining how effectively an active interrogation system will perform because the fission rate, and consequently the induced flux of fission neutrons, depends directly on the flux and not the neutron concentration. The volumetric reaction rate $R$ for a given flux $\Phi$, atomic number density $N$, and microscopic cross section $\sigma$ is [Knief, 1992]:

$$R = \Phi \sigma N \quad \text{(2-3)},$$

and the flux $\Phi$ in turn depends on the neutron density $n$ (used as a metric in [Dietrich et al., 2005]) as a velocity distribution and the neutron velocities $v$:

$$\Phi = \int n(v) \nu dv \quad \text{(2-4)}$$

The number of fission neutrons generated, then, will not depend immediately on how many neutrons reach a given point but instead on the flux of those neutrons.
Since neutrons will become moderated when traversing a material, their velocity will be reduced, and as seen in Equation 2-4, the flux $\Phi$ will also decrease. Thus, flux has been chosen as the appropriate quantity to be measured with the simulations discussed below. Because the flux decreases as the neutrons spread out and slow down, the penetration estimates given here will be underestimates relative to those given by Dietrich, et al [Dietrich et al., 2005].

Since neutron attenuation is an exponential function with distance [Turner, 1995] and the mesh size of 200 cm is many mean free paths of the chosen materials (See Table 2-3), the flux throughout the material will vary over several orders of magnitude. The mesh tally feature of MCNPX allows for simulated measurements of particle flux, so the plotted quantity $P$ in the figures below will depend logarithmically on the flux $\Phi$ as:

$$P = -\log_{10}(\Phi + 10^{-30})$$  \hspace{1cm} (2-5)

MCNPX gives flux estimates as expected fluxes per source particle so the flux values extracted directly from the simulation output will generally be less than one. The calculated quantity on the right side of Equation 2-5 is negative in order to ensure that $P$ will be a positive number. In addition, the $10^{-30}$ additive factor in Equation 2-5 prevents the logarithm from approaching infinity when MCNP fails to count a particle track in a particular cell. When this happens, the simulation reports a flux of zero, the logarithm of which is not defined. This adjustment will keep the value of $P$ for all cells finite while not introducing artifacts of imposing an artificial upper bound on $P$.

Furthermore, the fluxes given below are averages over a particular cell, which is a volumetric region in MCNP. To make these flux estimates, MCNP uses what is known as a track-length estimate. The effectiveness of this method depends on the assumption that throughout the cell, the flux is almost uniform. This will be true in
regions where the neutrons do not interact and the reduction in flux magnitude is due to the larger area occupied by the neutron beam; however, in regions where the mean free path is much less than the cell depth of 10 cm, the track-length estimate will fail to resolve these changes in flux behavior. Consequently, estimates for specific distances with particular features have been rounded to the nearest five centimeters.

Additionally, there is an error associated with each flux value generated by MCNP based on the expected deviation in sampling of the neutron interaction probabilities. The data in the simulations below are expressed logarithmically and each contour represents 0.5 of units in the quantity \( P \) from Equation 2-5. Thus, the tolerance in the plot is equivalent to half an order of magnitude. Any figure with a relative error that exceeds this quantity (a factor of \( \sqrt{10} \approx 0.31 \)) will result in a value whose confidence is too small for the simulation result to be useful in that mesh. Any elements with this large an error have been eliminated from the graph and the range displayed in the plot has been reduced to reflect this uncertainty.

To illustrate, a sample figure has been given in Figure 2-3. This represents the penetration of 14 MeV neutrons in steel. Each square in the plot represents an annular region where the vertical axis represents the distance along the z-axis (the direction of incident neutron flux) and the horizontal axis represents the radial distance from the neutron source. The flux is summed over all angles as explained above.

The values given in the legend are the values for \( P \) calculated from the flux using Equation 2-5. Since the relative error is greater than 0.31 when \( P \) exceeds 10, contours representing values after this point have been stricken from the figure. The magnitude of the flux has been delimited by contours each one representing 0.5 in \( P \) or half an order of magnitude. For increasing \( P \), the flux is decreasing.
As can be inferred from the graph, the neutrons are both penetrating into the material and spreading out from scattering events. In different materials, both these attributes will vary in a way that cannot be captured easily in a single number; they are better expressed as a two-dimensional plot for each material at a particular energy.

Figure 2-3: Total neutron flux through steel with 14 MeV incident neutrons

The neutron behavior in several materials has been examined with incident neutron energies of 60 keV, 100 keV, and 14 MeV. Neutron scattering energy transfer depends on the mass of the nucleus and several categories of materials have been chosen for emphasizing relevant aspects of neutron interactions in media. These materials include low-Z (low atomic number) hydrogenous materials, low-Z non-hydrogenous materials, and high-Z materials. A summary of chosen material compositions and neutron behavior features in those materials are
available in Table 2-2 and Table 2-3, respectively. Granted, the scattering cross sections themselves do not depend on Z but rather on properties of each isotope’s nuclear structure so the individual choices of nuclei to represent each of the above categories will also affect the observed neutron penetration beyond the consequences of simply the atomic number of these materials.

2.3.4 Low-Z Hydrogenous Materials

*High-density polyethylene (HDPE)*

High-density polyethylene (HDPE) has significant hydrogen content. With near the density of water and greater hydrogen relative contribution, HDPE will be one of the most effective materials for stopping neutrons. Hydrogen has a mass number A of 1 and as discussed in Section 2.2, as much as the entire kinetic energy of a neutron can be transferred to a hydrogen nucleus in a single scattering event. This observation makes hydrogen an element that is effective for shielding. High-density polyethylene also contains carbon nuclei, which also are effective at scattering neutrons, but with mass number A=12, not as much energy will be transferred in a collision as can be seen with Equation 2-2.

Figure 2-4 demonstrates that penetration of low-energy neutrons through this material is relatively weak. The situation is somewhat improved with 14 MeV neutrons, however the increased depth is not greater by more than a factor of two or so for any given flux (see Figure 2-5 and Table 2-3). Due to the high hydrogen content of HDPE, this material represents a “worst-case scenario” for the loss of neutron penetration ability between a 14-MeV- and a 100-keV interrogation system. MCNP simulations indicate the neutrons thermalize (meaning the flux at energies below 1 eV is greater to or equal than the flux at all other energies) after roughly 10 cm for thermal flux in HDPE at 60 keV.
Figure 2-4: Total neutron flux through HDPE with 60 keV incident neutrons

Figure 2-5: Total neutron flux through HDPE with 14 MeV incident neutrons
For 60 keV incident neutrons, the neutron flux reflected into the material scattered from the air back into the material becomes significant after a radial distance of 40 cm; this is visible in the figures where the contours become perpendicular to the source direction. Also, the 14 MeV neutrons are more penetrating in the direct line of the neutron source, but the neutrons do not spread radially much more than in the 60 keV scenario. This “tear drop” shape of the neutron distribution is characteristic of a heavily moderated variable-energy neutron distribution penetrating through media.

**Concrete**

Concrete is also a hydrogenous medium with a somewhat higher density than HDPE (2.3 g/cc vs. 0.96 g/cc) but the hydrogen atomic fraction is significantly reduced (30% vs. 60%) [ICRU, 1989]. Consequently, more of the scattering events in concrete result in less energy loss than for HDPE in spite of concrete’s higher density. Due to the hydrogenous content of the medium, the penetration ability of 14 MeV neutrons does differ from that of 100 keV neutrons similarly to the differences observed in HDPE. In these simulations, scattering from the air was significant in concrete; it is clear that these neutrons have undergone appreciable scatter because the spectrum in the regions where the contours have flattened out is much softer than nearer the neutron source.

It is important to note the significant boost in penetration for the concrete over HDPE with a reduction of hydrogen atomic number density by only half. While concrete is still a hydrogenous medium, for a given flux, the penetration has increased in concrete (compare Figure 2-4 and Figure 2-6).
2.3.5 Low-Z Non-hydrogenous Materials

Steam

Steel is an alloy of several relatively low-Z elements (most prominently iron with Z=26) so appreciable energy transfer during an individual neutron scattering event is possible. Lower Z impurities will serve to increase the moderation capability of steel. Steel at 60 keV expresses a surprisingly similar penetration profile over all energies to concrete at the same energy, but steel has no hydrogen content. It turns out this resemblance in the overall flux disappears on examining the energy spectrum. The thermal spectrum at any given depth and radius is approximately three orders of magnitude smaller in steel than in concrete for 60 keV neutrons (compare Figure 2-8 and Figure 2-9). This implies that Fe is absorbing many of the neutrons before they reach thermal energies and this can be confirmed by examining the Fe-56 cross section as a function of energy in Figure 2-7.
Figure 2-7: Neutron Radiative Capture Cross Section for Fe-56 [Cullen, 2003]

Figure 2-8: Thermal (E< 1 eV) neutron flux in concrete for 60 keV incident neutrons
Figure 2-9: Thermal neutron flux in steel for 60 keV incident neutrons

Aluminum

Aluminum is also a low-Z element (Z=13) so energy transfer will also be considerable as in the case of iron in steel. Still, an incident neutron beam at 60 keV penetrates quite deeply into the material. Aluminum-27, the most abundant isotope of aluminum, contains several low energy resonances in the total neutron cross section, one of which lies between 60 and 100 keV as shown in Figure 2-10. These counterintuitively allow for greater penetration by 60 keV neutrons than 100 keV neutrons in aluminum (Compare Figure 2-11 and Figure 2-12). In fact, the total neutron flux of 14 MeV neutrons penetrating aluminum does not differ markedly from the 60 keV neutron flux.

Also, the incident neutron flux does not thermalize significantly until it has reached 125 cm into the material for 100 keV neutrons and 115 cm for 60 keV neutrons,
allowing for significant penetration. The effect of these low-energy resonances may be important in optimizing the chosen neutron interrogation energy as a number of isotopes of other elements likely to be present in cargo environments also have resonances in this range.

![MT=1: (a,total) Cross section for Al27 from ENDFB 6.8 from NEA](image)

**Figure 2-10: Neutron Total Cross Section for Al-27** [Cullen, 2003]

![Figure 2-11: Total neutron flux through Al for 60 keV incident neutrons](image)
2.3.6 High-Z Materials

*Tungsten*

Tungsten is a high Z (Z=74) metal with a significant neutron capture cross section. Owing to the high mass of tungsten nuclei, little energy will be transferred during scattering collisions. Most of the flux attenuation will be due to absorption especially the contribution from W-186 (having an isotopic abundance of 28.426 percent), which has a resonance-average radiative capture cross section of 346.8 barns; consequently, very little thermal flux is observed in penetration simulations.

Also, because the capture cross section is still relatively high even at several hundred keV, low-energy neutrons penetrate roughly as far into tungsten as neutrons with initial energies of 14 MeV. The difference between the thicknesses at which the same flux is reached in the two materials differs by at most 20 cm
within 100 cm of material, which is as deep as statistically significant simulation results are available (See Figure 2-13 and Figure 2-14). For most of the extent of the initial beam, this difference is closer to 10 cm.

Figure 2-13: Total neutron flux through W for 100 keV incident neutrons

Figure 2-14: Total neutron flux through W for 14 MeV incident neutrons
Depleted Uranium

Depleted uranium, the refuse of uranium enrichment processes, comprises mostly uranium-238 and a small fraction (0.25 percent for simulations in this study) of uranium-235. If only U-238 were present, these models would exhibit some absorption of neutrons at most energies and little fission; generally there will be none except those generated by incident neutrons with energies above the fast fission threshold for U-238. However, U-235 has an appreciable fission cross section at all energies so some high-energy neutrons will be generated although depleted uranium cannot be used directly for weapons.

Figure 2-15: High (E < 1 MeV) neutron flux in depleted uranium for 100 keV incident neutrons

The implication of this result is that enriched uranium will not be strictly distinguishable from natural or depleted uranium because of the presence of uranium-235 in both materials suggesting the possibility of false positives for active
interrogation of depleted uranium. Depleted uranium will still produce high-energy neutrons in response to exposure to low-energy neutrons (See Figure 2-15).

Additionally, because of the high mass of uranium nuclei, little energy will be transferred in scattering collisions. In this model, no neutrons were able to pass resonance absorptions and reach thermal energies.

2.4 Summary

From following the changes in penetration ability of neutrons at various energies, several effects of materials and their implications for SNM detection systems emerge. A summary of the material compositions used in these simulations is available in Table 2-2. To quantify the penetration ability of neutrons through all these materials, two metrics derived from the results of MCNP simulations above are compared in Table 2-3. The first (Penetration) is a direct measure of the penetration distance of the neutrons; this is taken as the point when the flux falls to $10^{-4}$ neutrons/cm$^2$/source particle. The second (Penetration $\Delta$) is a comparative measure of how far it takes for the flux to be reduced from $10^{-3}$ neutrons/cm$^2$/source particle to $10^{-4}$. The second metric is necessary because the first may be confounded by the initial scattering of neutrons both laterally into the material and backwards into the air surrounding the medium. Because of this effect, a neutron beam that actually may penetrate far into a material may appear to reach a certain flux sooner even though this particular flux may only be a small reduction from its initial intensity.

It is clear that one of the dominant factors in determining the penetration ability of various neutrons is the absorption and scattering cross sections for these nuclei, which can be inferred from their mean free path and material density. Of secondary importance is the mass of the target nucleus; some heavier nuclei are also more able to absorb neutrons.
Table 2-2: Material Compositions [MatWeb, 2006], [ICRU, 1989]

<table>
<thead>
<tr>
<th>Material</th>
<th>Composition (atom fraction); these are given in natural isotopic abundances unless the isotope is specified</th>
</tr>
</thead>
<tbody>
<tr>
<td>HPDE</td>
<td>H-1 (.666), H-2 (.0001), C-12 (.3296), C-13 (.0037)</td>
</tr>
<tr>
<td>Concrete</td>
<td>H-1 (.3053), C (.0029), O-16 (.5005), Na-23 (.0092), Mg (.0007), Al-27 (.0103), Si (.1510), K (.00358), Ca (.0149), Fe (.0016)</td>
</tr>
<tr>
<td>Steel (Type 314)</td>
<td>Fe (.6083), C (.0115), Mn-55 (.0201), P-31 (.0008), S (.0005), Si (.0196), Cr (.1907), Ni (.1314), Mo (.0172)</td>
</tr>
<tr>
<td>Aluminum</td>
<td>Al-27 (1.0)</td>
</tr>
<tr>
<td>Tungsten</td>
<td>W (1.0)</td>
</tr>
<tr>
<td>Depleted U</td>
<td>U-238 (.9975), U-235 (.0025)</td>
</tr>
</tbody>
</table>

Table 2-3: Neutron Penetration in Materials from MCNP models

<table>
<thead>
<tr>
<th>Material</th>
<th>Density (g/cc)</th>
<th>Energy (keV)</th>
<th>Average Mean Free Path (cm)</th>
<th>Penetration (cm)</th>
<th>Penetration Δ (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HDPE</td>
<td>0.96</td>
<td>60</td>
<td>0.45</td>
<td>20</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>1.00</td>
<td>0.46</td>
<td>2</td>
<td>55</td>
<td>25</td>
</tr>
<tr>
<td>Concrete</td>
<td>2.3</td>
<td>60</td>
<td>1.05</td>
<td>30</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>1.00</td>
<td>1.05</td>
<td>30</td>
<td>15</td>
<td></td>
</tr>
<tr>
<td></td>
<td>14000</td>
<td>3.03</td>
<td>45</td>
<td>25</td>
<td></td>
</tr>
<tr>
<td>Steel</td>
<td>7.85</td>
<td>60</td>
<td>1.80</td>
<td>35</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>1.00</td>
<td>2.02</td>
<td>35</td>
<td>20</td>
<td></td>
</tr>
<tr>
<td></td>
<td>14000</td>
<td>3.07</td>
<td>60</td>
<td>30</td>
<td></td>
</tr>
<tr>
<td>Aluminum</td>
<td>2.7</td>
<td>60</td>
<td>13.8</td>
<td>70</td>
<td>45</td>
</tr>
<tr>
<td></td>
<td>1.00</td>
<td>13.1</td>
<td>50</td>
<td>30</td>
<td></td>
</tr>
<tr>
<td></td>
<td>14000</td>
<td>16.2</td>
<td>75</td>
<td>45</td>
<td></td>
</tr>
<tr>
<td>Tungsten</td>
<td>19.25</td>
<td>60</td>
<td>1.38</td>
<td>25</td>
<td>15</td>
</tr>
<tr>
<td></td>
<td>1.00</td>
<td>1.55</td>
<td>25</td>
<td>10</td>
<td></td>
</tr>
<tr>
<td></td>
<td>14000</td>
<td>1.92</td>
<td>45</td>
<td>15</td>
<td></td>
</tr>
<tr>
<td>Depleted U</td>
<td>19.1</td>
<td>60</td>
<td>1.82</td>
<td>25</td>
<td>10</td>
</tr>
<tr>
<td></td>
<td>1.00</td>
<td>1.82</td>
<td>25</td>
<td>10</td>
<td></td>
</tr>
<tr>
<td></td>
<td>14000</td>
<td>2.21</td>
<td>55</td>
<td>20</td>
<td></td>
</tr>
</tbody>
</table>

As expected, hydrogenous materials are the most effective for shielding cargo from neutron interrogation most consistently over several energies leading to the temptation in their use for obscuring SNM from a neutron interrogation system. However, the presence of an abundance of hydrogenous material completely
surrounding a large space may be detectable and could be an indicator of hazardous material in and of itself. One possibly abundant hydrogenous material that may not be so unusual in shipping in large quantities is crude oil. It is worth noting that as of 2006, for those who would be interested in the clandestine importation of nuclear material, the use of crude oil as a neutron shield would be geopolitically convenient. The following chapter will explore the behavior of the proposed system in simulated cargo environments including the effects of hydrogenous shielding on SNM detection.
3.0 Neutron Interrogation in Cargo Environments

Several features of cargo environments are necessary to consider in predicting the behavior of an interrogation system. First, the background signal must be established for each isotope in a possible target material. Then, it must be decided what types of materials may be placed in between SNM and where fluxes are tallied in order to simulate a cargo environment. It is clear that cargo environments encountered in the real world will be highly variable so either an average cargo environment must be assumed or several variations on a basic cargo environment can be studied [Gallagher, 2005]. Using a neutron source model based on the proposed target reaction, the system can be simulated containing SNM components.

3.1 Cargo Model Geometry

In order to reduce the variance in particle tallies, the cylindrical geometry approach from the simulations discussed in Chapter 2.0 is maintained for the following studies of interrogated cargo. The motivation for the associated mesh tallying approach for estimating flux throughout the problem is identical to that outlined in Section 2.3.2; the only difference is that instead of a single material, the problem consists of several concentric cylindrical layers of intervening cargo materials with simulated SNM.

Figure 3-1 illustrates a schematic of the simulation geometry. This method follows the method of Kerr et al who used a spherical mock cargo container and overall geometry instead of a cylindrical one [Kerr et al., 2005]. A 300 cm wide, 300 cm tall cylindrical shell composed of steel 0.5 cm thick approximates a basic cargo container, though real cargo containers are much larger. Inside the container shell are several layers that may or may not be present. Most of the container is filled with air at standard pressure. In the center of the container is a target material of varying mass; this target consists either of innocuous materials like lead and
depleted uranium or it comprises SNM including either highly enriched uranium (HEU) or plutonium. Surrounding the target material may be shielding material consisting of both hydrogenous and non-hydrogenous compounds of varying thickness likely to appear in cargo environments. While there may be numerous other objects in the path of a real smuggled weapon, varying the shielding thickness and composition will allow for the systematic study of these scenarios. Additional configurations for SNM can be constructed by varying the shape of the target and other materials while maintaining the cylindrical symmetry of the problem.

Figure 3-1: Interrogation in Cargo MCNP Problem Geometry

The neutron source is a modified version of that used by Kerr et al in their studies of active interrogation via the same method examined in this thesis [Kerr et al., 2005]. This source is placed 50 cm outside the cargo container’s axial surface and the particles are directed along the concentric axis of the materials in this
geometry. The neutrons effectively have 200 cm total distance to travel to reach the target SNM.

Because this neutron source is intended to represent the emissions of a $^7\text{Li(p,n)}^7\text{Be}$ reaction, most but not all of the neutrons will be directed forward, hence the lighter arrows at shallower angles.

Examples of the input decks used for the simulation of both radioactive decay emissions by fissile material and responses to neutron interrogation are available in Appendix B.

3.2 Basal Signal Modeling

The fissile material models will contain unstable isotopes. Highly enriched uranium, as well as natural uranium, emits primarily alpha particles and gamma rays. The high LET of the alpha particles prevents nearly all of them from exiting the uranium, and those which do escape the uranium are certainly unable to reach the exterior of a cargo container. Emitted gamma rays on the other hand will be able to penetrate the uranium they are produced in and most media they will encounter on their way to a detector. This characteristic gamma flux will provide a basal signal with which any analysis of neutron-induced photons must compare. A basal flux that exceeds the induced signal will result in difficulty for the use of changes in gamma intensity under neutron interrogation for inferring the presence of fissile material.

The composition of the special nuclear material used in generating these basal data is available in Table 3-1 and Table 3-2.

3.2.1 Basal Neutron Flux

Neutron emissions from spontaneous fission events for uranium isotopes can be neglected. As can be seen in Table 3-1, even after accounting for all isotopes in the
chosen composition, any reasonable amount of uranium (on the order of a few kilograms) will spontaneously emit only a few neutrons/sec/kg. The simulations used to measure the basal radiation fluxes contained a 7 kg target of highly enriched uranium and 1 kg of plutonium. The rate of spontaneous fission neutron emissions from any unstable isotope results approximately from Equation 3-1

\[
\text{neutron\_emission\_rate} = \frac{\text{frac}}{\text{MM}} \cdot N_a \cdot \frac{\ln(2)}{t_{1/2}} \cdot SFF \cdot \nu \cdot \Phi \quad (3-1)
\]

Equation 3-1 assumes that all isotopes have approximately the same molar mass so that the difference between the atom and mass fractions of each isotope will not depend significantly on the difference in masses of those isotopes. This assumption holds well for heavy elements such as uranium but would not for lighter ones such as hydrogen or lithium.

**Table 3-1: HEU spontaneous fission neutron emission rate** [Cullen, 2003], [Kerr et al., 2005], [Gallagher, 2005]

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Atom %</th>
<th>Half-life (years)</th>
<th>Spontaneous fission fraction</th>
<th>Fission neutrons/sec/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-232</td>
<td>10^8</td>
<td>68.9</td>
<td>(9.0 \times 10^{13})</td>
<td>(1.8 \times 10^{-5})</td>
</tr>
<tr>
<td>U-234</td>
<td>1.0</td>
<td>(2.455 \times 10^5)</td>
<td>(1.7 \times 10^{11})</td>
<td>(0.097)</td>
</tr>
<tr>
<td>U-235</td>
<td>93.5</td>
<td>(7.038 \times 10^6)</td>
<td>(7.0 \times 10^{11})</td>
<td>(0.013)</td>
</tr>
<tr>
<td>U-238</td>
<td>5.5</td>
<td>(4.468 \times 10^9)</td>
<td>(5.0 \times 10^7)</td>
<td>(0.087)</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td></td>
<td></td>
<td></td>
<td><strong>0.98</strong></td>
</tr>
</tbody>
</table>

As can be seen by comparing the total in Table 3-1 with the neutron output from interrogated cargo described in Section 3.3.2, these neutrons contribute negligibly to the total neutron flux produced during active stimulation of uranium.
Table 3-2: Pu spontaneous fission neutron emission rate [Cullen, 2003], [Kerr et al., 2005], [Rudisill and Crowder, 2000]

<table>
<thead>
<tr>
<th>Isotope</th>
<th>Atom %</th>
<th>Half-life (years)</th>
<th>Spontaneous fission fraction</th>
<th>Fission neutrons/sec/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu-239</td>
<td>91.95</td>
<td>24110</td>
<td>3×10^{-12}</td>
<td>15.8</td>
</tr>
<tr>
<td>Pu-240</td>
<td>4.34</td>
<td>6564</td>
<td>5.7×10^{-8}</td>
<td>5.21×10^{4}</td>
</tr>
<tr>
<td>Pu-241</td>
<td>0.29</td>
<td>14.35</td>
<td>&lt;2.0×10^{-14}</td>
<td>&lt; 0.56</td>
</tr>
</tbody>
</table>

In plutonium, however, a significant neutron flux is produced by the Pu-240 content of plutonium-derived fissile material. These neutrons are assumed to be born in the simulation uniformly throughout the plutonium. For Pu-240, MCNP simulations with 1 kg of plutonium as a target material predict a flux of 5.495×10^{-6} ± 1.1×10^{-9} neutrons/cm² per source particle just beyond the exterior radial surface of the steel shell. Normalizing this figure by the neutron emission rate obtained in Table 3-2 implies a real flux of 0.28629 ± 5.7×10^{-5} neutrons/cm²/sec due to the radioactive decay of the plutonium.

Since these estimates were derived when no neutron shielding was present in the model, any shielding that is introduced in later models will reduce the actual basal neutron flux. Thus, the above figure represents a maximal basal rate of neutron flux. So long as the induced neutron signal under active interrogation is significantly higher than the predicted basal signal, the basal signal can be neglected from the latter analysis. It is important to note that the detection of these fission neutrons due to decay will not interfere with the active interrogation approach. Fission neutrons would still be indicative of the presence of fissile material because few innocuous materials exist which undergo significant spontaneous fission and produce high-energy neutrons.

3.2.2 Basal Gamma Flux

The gamma emissions, however, do remain significant in both HEU and plutonium, and the transport of these background photons through the cargo
geometry must be modeled in MCNP. The basal estimates below use the same problem geometry as given in Figure 3-1. The shielding in the cargo models in Section 3.4 will focus on hydrogenous and other low Z materials, through which high-energy photons can penetrate easily so shielding from media surrounding the fissile material has been neglected in the following basal gamma emission simulations. As assumed for the neutrons above, the gammas are born uniformly throughout the fissile material volume.

For each isotope, the basal emission has been modeled based on the known gamma decay spectrum [Cullen, 2003]. The MCNP simulation delivers a flux estimate per source particle. For these surface tallies, the flux is estimated by counting each particle and weighing it by the cosine of the angle between the tallying surface and the particle direction. Normalizing this figure with the known activity using Equation 3-2 results in the actual flux expected to be observed just outside the cargo container. For a given flux from MCNP, where \( M \) is the target fissile material mass and with common variables defined as in Equation 3-1, the number of photons produced per unit time is:

\[
\text{photons} \text{ produced} = \frac{\frac{N \ln(2) \cdot \Phi}{t_{1/2}}}{MM} \cdot \frac{NM}{MM} \tag{3-2}
\]

Divided up coarsely by energy, the mass-weighted gamma outputs for each isotope are given in Table 3-3 for highly enriched uranium and Table 3-4 for plutonium. The results in both these tables assume the same composition of fissile material used to calculate the results in Table 3-1 and Table 3-2, respectively. Like the neutron analysis, these fluxes are those observed just outside the radial surface of the steel cargo container shell.

When compared with those photon fluxes obtained during simulations of neutron interrogation explained in Section 3.6.2, these basal gamma signal contributions are clearly small but still non-negligible, especially for plutonium isotopes.
Table 3-3: Gamma spectrum for 7 kg HEU given in flux (photons/cm²/sec)

<table>
<thead>
<tr>
<th>Isotope</th>
<th>E &lt; 50 keV</th>
<th>E &lt; 100 keV</th>
<th>E &lt; 1 MeV</th>
<th>E &lt; 14 MeV</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>U-232</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>U-234</td>
<td>≈ 0</td>
<td>≈ 0</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>U-235</td>
<td>≈ 0</td>
<td>0.054</td>
<td>0.074</td>
<td>0</td>
<td>0.128</td>
</tr>
<tr>
<td>U-238</td>
<td>0</td>
<td>0.0011</td>
<td>0.0031</td>
<td>0</td>
<td>0.0042</td>
</tr>
</tbody>
</table>

Table 3-4: Gamma spectrum for 1 kg Pu given in flux (photons/cm²/sec)

<table>
<thead>
<tr>
<th>Isotope</th>
<th>E &lt; 50 keV</th>
<th>E &lt; 100 keV</th>
<th>E &lt; 1 MeV</th>
<th>E &lt; 14 MeV</th>
<th>Total</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pu-239</td>
<td>0.0068</td>
<td>2.379</td>
<td>48.5</td>
<td>8.02×10⁻⁴</td>
<td>50.9</td>
</tr>
<tr>
<td>Pu-240</td>
<td>2.08×10⁻⁴</td>
<td>0.169</td>
<td>0.362</td>
<td>0</td>
<td>0.530</td>
</tr>
<tr>
<td>Pu-241</td>
<td>0.0673</td>
<td>34.3</td>
<td>104</td>
<td>0</td>
<td>138</td>
</tr>
</tbody>
</table>

3.3 Neutron Interrogation of Cargo Model

As described in Section 3.1, these cargo containers are bombarded from a nearby neutron source. This source model will approximate the flux produced in a \(^7\text{Li}(p,n)^7\text{Be}\) reaction run near threshold, so unlike sources based on deuterium-tritium targets, this source will be directed in the forward direction.

3.3.1 Neutron Source Definition

A calculated MCNP definition of the \(^7\text{Li}(p,n)^7\text{Be}\) type source is available from Kerr et al. and prior studies indicate that the beam will be reasonably well-focused in a uniform direction [Kerr et al., 2005]. The energy-angle distribution for this source is depicted in Figure 3-2.

It can be seen that in this calculation, the most likely energy of an outgoing neutron is approximately 60 keV and the neutron energy does depend to some degree on the neutron’s angle. The angular distribution is markedly forward-peaked, which is quite convenient for a neutron-based interaction. Examining this neutron source alone without any intervening cargo container but only a sphere of
air for neutrons to interact with reveals a behavior (see Figure 3-3) consistent with earlier studies leading to prior results such as Figure 3-2.

Figure 3-2: Energy-angle distribution of $^7\text{Li}(p, n)^7\text{Be}$ source [Kerr et al., 2005]

Figure 3-3: Neutron distribution from neutron source in air with no cargo
The coordinates in Figure 3-3 and similar plots throughout this chapter have a similar definition to those in Chapter 2.0 (explained in detail in Section 2.3.3). However, in these plots the geometry has been rotated ninety degrees and reflected so that now the untransformed z-axis lies horizontally and the radial direction is vertical. In these cases, the cylindrical axis of the cargo container runs parallel to the untransformed z-axis above, equivalent to the direction of the neutron source. Additionally, since the neutron beam is forward directed, less attenuation occurs with distance so the difference in flux intensity between contours has been reduced from half an order of magnitude to only a quarter (See Section 2.3.3 and Equation 2-5). This is made possible by the fact that since the space in the cargo contains material of mostly lower density than the space in the penetration problems of Chapter 2.0, statistics are significantly improved even with a similar number of particle runs.

In Figure 3-3, the neutron distribution matches exactly what would be expected in light of Figure 3-2. The majority of the neutrons are emitted in a narrow cone of not more than thirty degrees or so from the direction of maximum intensity. This intensity falls slowly as the neutron beam widens and travels through the space where the cargo container will be placed in further simulations.

3.3.2 Neutron Interrogation in Cargo With versus Without Fissile Material

By introducing the cargo container model depicted in Figure 3-1, the response of the cargo materials to the neutron source of the previous section can be examined. Two models can be compared: one with fissile material and the other without. Here, the fissile material is HEU with the isotopic composition from Table 3-1 and the chosen innocuous material is pure lead. The same volume of Pb and HEU have been used to maintain the geometry so while 7 kg of HEU is present in this simulation, roughly 4.5 kg of Pb is used in the following one. The response for
neutrons at all energies to HEU and Pb is given below in Figure 3-4 and Figure 3-5, respectively.

**Figure 3-4:** Neutron flux at all energies with HEU in cargo container

**Figure 3-5:** Neutron flux at all energies with Pb in cargo container
In both these figures, the effects of the cargo container and its contents on the neutron distribution are readily visible. The small drop in neutron flux at 0 cm along the untransformed axis is due to the steel layer making up the base of the cargo container. There is a similar reduction at a distance of 300 cm where the top of the cargo container is. At a radial distance of 150 cm, the radial wall of the container limits the amount of neutrons that pass through it. Incidentally, it is the neutron and photon fluxes on the exterior of this wall that are crucial in modeling the system because this is equivalent to where a detector will sit in the real-world application of this technique. The particle distribution inside the container is not immediately relevant to the detection of fissile material when the detector lies outside the container. It is important instead for understanding how cargo composition and geometry affect the neutron interrogation system when the incident neutrons interact with the subject cargo container.

There is little in the above figures that distinguishes the alarm HEU case from the harmless Pb case because the low energy neutrons from the source dominate the neutron distribution. The only lingering difference is a small escalation in neutron flux where the HEU is present owing to the dominant contribution of fast neutrons at the location, which are not present when the target material comprises only Pb. In order to observe the distinction in neutron response, a detector must isolate fission neutrons with energies greater than 1 MeV. These responses are given in Figure 3-6 and Figure 3-7 for HEU and Pb respectively.
Figure 3-6: Fast neutron flux \( (E > 1 \text{ MeV}) \) with HEU in cargo container

Figure 3-7: Fast neutron flux with Pb in cargo container
The categorical difference between these two responses coincides with what might be predicted by the physics involved and does not pose any immediate barriers to the implementation of the system described in this thesis. The HEU fissions as expected and produces a flux that is statistically significant even outside the container. For a pulse of $10^9$ neutrons, one can expect $180 \pm 2$ neutrons to be detectable outside the cargo container (assuming a $1000 \text{ cm}^2$ detector with 10 percent efficiency). The Pb, of course, does not fission at all. The difference between the two materials is unambiguous when detecting neutrons at high energies. Even only 1 kg of plutonium will give a similar strong and unambiguous response outside the cargo container, however in the model this signal is reduced in intensity by a factor of 0.3 relative to the 7 kg of HEU (See Figure 3-8).

This signal magnitude issue becomes somewhat more complicated, though not insoluble, when introducing intervening material that can shield the fissile material from the incident neutron beam.
3.4 Shielding of the Incident Neutron Beam

In the simulations for the following portion of the study, fissile material or innocuous material was surrounded with a cylindrical uniform layer of a hydrogenous shielding material and the effect of this on the measured neutron intensity at key locations in the geometry are compared. Otherwise, the geometry is identical to the simulations conducted in Section 3.3.2. These studies will focus on the effect of this additional moderating material. Hydrogenous material was chosen for shielding analyses exclusively because hydrogen will be the most effective in stopping neutrons from reaching fissile material due to its small nuclear mass and thus could make an effective shield.

A single set of processes for neutron slowing occurs when moderating or shielding material (especially hydrogenous material) intervenes between the interrogating beam and target material but two contrary outcomes are possible depending on the amount of material present. If the amount of shielding is appreciable but relatively small, incident low-energy neutrons will become moderated as they pass through the material and scatter off nuclei transferring their kinetic energy. The neutrons continue to lose energy to the medium until they reach thermal energies at which point, on average, the neutrons receive as much energy in a collision as they lose.

If a significant portion of neutrons have thermalized in this way but have not yet been absorbed, many of the important interaction cross sections will be higher in the material. In particular, their absorption cross section and thus reaction rates in material beyond the shielding will be greatly increased. If this material is fissile, an increase in fissions will result, and many more fast neutrons will be visible outside the cargo container. If there is enough intervening shielding between the neutron source and target material, not only will the neutrons be thermalized but also many will be absorbed and few will reach the target material. If the target is fissile
material, a *reduction* in observed fast neutrons outside the cargo container will occur and it is possible no significant signal will be generated. Whichever outcome occurs for a given geometry, it will be determined by the amount of shielding present, its composition, and the competition of the scattering and absorption cross sections at the corresponding energies.

Example cases of the above two possibilities for neutron interrogation shielding are discussed in the sections below.

3.4.1 Moderation of Incident Neutrons with Little Shielding

The simulations from Section 3.3.2 were repeated with highly enriched uranium and plutonium as target materials while introducing a uniform 5 cm thick concrete layer surrounding the fissile material. This concrete layer serves as a hydrogenous shield and will interact with the incident neutrons as described in the above section either simply slowing them down or blocking their passage to the fissile material.

![Figure 3-9: Fast neutron flux with HEU and 5 cm concrete](image)

Figure 3-9 shows the resulting high energy ($E > 1$ MeV) neutron flux for 7 kg HEU surrounded by 5 cm of concrete. By comparing this figure with Figure 3-6, it
can be seen that rather than shielding the fissile material from the neutron beam, the thin concrete layer effectively moderates the neutrons. The neutrons that pass through the concrete exhibit a softer energy spectrum and the slower neutrons have larger fission cross sections in uranium-235. Thus, the fission rate increases and more fast neutrons are produced. The contour corresponding to a $P$ (see Section 2.3.3 for definition of the quantity $P$) of 8.5, reaches a maximum radial distance of 125 cm in the unshielded simulation (see Figure 3-6). However, adding a thin concrete layer extends this reach to 145 cm (see Figure 3-9).

### 3.4.2 Absorption of Incident Neutrons with Moderate Shielding

Further increases in shielding thickness may moderate the neutrons more but eventually will result in their absorption as the absorption cross section in many materials increases with decreasing incident neutron energy [Cullen, 2003]. As can be seen in Figure 3-10, when a layer of 25 cm of concrete is added to the fissile material model, it will be much more difficult to gain statistically significant measurements of the fast neutron output.

![Figure 3-10: Fast neutron flux with HEU and 25 cm concrete](image)

The simulation depicted in Figure 3-10 followed $10^9$ source particles initiated in the neutron source outside the cargo container but only $9800 \pm 300$ neutrons
would be expected to be produced and reach the outside of the cargo container. A reasonably sized (1000 cm²) detector outside the steel wall of the cargo container could only expect to see a flux of 3 neutrons for each pulse of the neutron source (assuming $10^9$ neutrons/pulse and a 10 percent efficiency).

Still, to hide fissile material by surrounding it with a foot of concrete would neither be realistic nor subtle. The concrete also would not be resistant to conventional radiological techniques, e.g. x-ray analysis, rendering the high Z of the obscured nuclear material detectable. Other materials may exist such as crude oil that may be more effective in concealing nuclear material without drawing suspicion. Crude oil is more hydrogenous than concrete and can be expected to appear in large volumes during shipping. Still, like concrete and other hydrogenous materials, crude oil is relatively transparent to x-ray inspections and other imaging approaches. If significant hydrogenous material were found to be present, further examination of the cargo with these techniques would reveal the presence of high Z material.

### 3.5 Discriminating Fissile Isotopes from Fissionable Isotopes

One original purpose of choosing a low-energy ($E < 1$ MeV) spectrum for neutron interrogation was to have a radiation source that would generate fissions in fissile nuclei but would not risk false alarms from fast fissions in fissionable nuclei. For this system then to effectively satisfy that criterion, it should respond much more weakly, or preferably not at all, to depleted uranium than to an equivalent mass of highly enriched uranium. For the simulation with results given in Figure 3-11, the 7 kg of HEU from the simulations above has been substituted with depleted uranium, containing 0.25 atom percent uranium-235. It is clear from this image that while fast neutrons are produced by limited fission in the depleted uranium, the number is not nearly enough to reach outside. The system's response is
markedly different for depleted uranium than for a real threat of highly enriched uranium.

**Figure 3-11: Fast neutron flux for DU in cargo container**

However, introducing a moderating material will increase the fission rate in depleted uranium as it did for fissile material in Section 3.4.1. Adding a 5 cm thick layer of concrete around the mass of depleted uranium results in a marked increase in fission neutrons (see Figure 3-12). The strong boost makes dubious the suggestion that the low uranium-235 amount is what is preventing a significant fission neutron signal from appearing. It is more likely that the thermalization of neutrons has increased the amount that can be absorbed. The output in Figure 3-11 is so small not because the probability of a fission induced by a particular neutron is so low but rather because the resonance absorption probability of a neutron is so high. Figure 3-12 shows a much larger fission rate because the thermalized neutrons cannot get lost in resonance absorptions. They already have fallen to thermal energies and are much more likely to cause fissions in the small uranium-235 component that is present in the depleted uranium.
Still, given the resemblance of the neutron distribution of Figure 3-10 to Figure 3-12, it is clear that there may be a difficulty in distinguishing heavily shielded HEU from lightly shielded depleted uranium through a low-energy neutron interrogation approach.

The upshot of this analysis is that such a small amount as 18 g of uranium-235 is in principle detectable given the right conditions of material composition and cargo geometry. Granted, the time to reach a given confidence threshold may be large and will depend on the ability to reduce background radiation and dark current in the chosen detector.

### 3.6 Improving Selectivity

Since the observable high-energy neutron fluxes may be very similar for shielded fissile material and unshielded innocuous cargo, the selectivity of this detection system can be improved by implementing a means of detecting neutron interaction with any cargo that does serve to shield against neutrons. When neutrons slow or stop in media, and especially in hydrogenous material, two changes occur in the nearby radiation distribution: thermal neutron flux increases and the gamma...
energy distribution shifts. If either of these quantities can be easily measured, it will be much easier to assess how much shielding is present in an unknown cargo container and what effect this shielding would have on the detectable signal from SNM. The details of these changes in radiation signals are discussed in the sections below.

3.6.1 Increase in Thermal Neutron Flux

When a non-thermal neutron beam passes through any material, the neutrons may scatter elastically off nuclei in the medium and transfer energy. Collisions with low mass nuclei result in greater energy transfer in each collision. Consequently, the thermal neutron distribution surrounding any hydrogenous material will be somewhat moderated because of its low mass number.

![Figure 3-13: Thermal flux with HEU and 25 cm concrete](image)

Using the same simulations examined in Sections 3.4.2 and 3.5, the thermal neutron fluxes are compared for 25 cm of concrete shielding HEU and 5 cm of concrete shielding depleted uranium in Figure 3-13 and Figure 3-14, respectively. The heavily shielded HEU has a greater content of hydrogenous material and so
the thermal flux even outside the cargo container, where it is detectable, is much higher.

The thermal neutron flux just outside the cargo container’s radial surface is almost an order of magnitude higher when heavier shielding is present inside the container. The flux distribution is, of course, still dominated by the low-energy neutrons whose energy has not changed significantly since being emitted from the $^7\text{Li}(p,n)^7\text{Be}$ reaction. If thermal neutrons can be reliably detected and discriminated from the many other neutrons and gamma rays generated in this process, the amount of shielding can be readily estimated. The geometry of any SNM and shielding and other cargo will greatly affect how all these components interact and appear outside the cargo container. Because this interrogation method is not intended to image a cargo container but only decide whether or not SNM may be present, an exact determination of the effect of the shielding will not be possible without a significant revision of the approach. For typical contexts where the composition of a cargo container is at least roughly known, an estimate of shielding between the neutron source and any fissile isotopes will be sufficient for detecting SNM.

![Figure 3-14: Thermal flux with DU and 5 cm concrete](image)

58
3.6.2 Changes in Gamma Energy Spectrum

With the introduction of shielding, the neutrons will interact with a greater diversity of material within the cargo container and the reactions undergone will differ. Consequently, the energy spectrum of the photons produced in these processes can be expected to change in a measurable way. These alterations in the observable photon spectrum can also be used to characterize how much shielding is present inside a cargo container and then to draw an improved inference over whether SNM may be present inside a container.

![Gamma energy spectrum](image)

**Figure 3-15: Gamma energy spectrum for fissile and fissionable material with light or heavy shielding**

In Figure 3-15, the gamma fluxes from the two simulations considered in Section 3.5 are compared with one another: one case where HEU is surrounded by a heavy amount of concrete and a second where depleted uranium is surrounded by
only enough concrete to moderate the incident neutrons. The fractional difference between the two scenarios is also included on a separate axis. The marked deviations are derived from the variances of the fluxes as computed from the relevant physical distributions with MCNP.

Significant and potentially measurable changes between the two cargo model scenarios are visible for the 50 keV to 100 keV and the 100 keV to 1 MeV energy bins. Here, a twenty to forty percent increase can signify a large amount of shielding that may be concealing the fast fission spectrum from fissile material inside.

These photons are produced when neutrons are either captured or slowed inside the hydrogenous shielding. However, the shielding serves to prevent the neutrons from reaching the fissile material and generating fission photons. This is the main explanation for why the flux does not increase uniformly. Lower-energy photons are produced in the shielding while high-energy fission photons are suppressed when fewer neutrons penetrate the shielding and reach the fissile material. But counteracting this feature is the increased thermalization of the neutrons which do penetrate the shielding. These neutrons will generate more fissions and produce more fission photons. The competition of these two effects will determine how the photon spectrum shifts.

A material with a denser hydrogen content will both thermalize and capture the neutrons further than one with less hydrogen. When the depleted uranium in these simulations is surrounded with HDPE instead of concrete, the fractional increase in photon output falls by half. HDPE contains about three times as many hydrogen nuclei per unit volume than concrete does.
3.7 Summary

The above studies show that for a generalized cargo container, it will be possible to distinguish one that contains SNM from one that does not using the output fast neutron spectrum. It turns out that in the fast spectrum, fissile material responds in a qualitatively different way from other materials and simulations suggest this distinction should be observable experimentally.

Hydrogenous material that may be in cargo containers interferes with this process by slowing or stopping probing neutrons. Fortunately, shielding will exert an effect on other aspects of the radiation even outside the cargo container.
4.0 Future Work and Conclusions

A number of additional avenues of simulation work are possible for further analysis of the behavior of this system. For example, the composition of fissile material is not constant with time since SNM comprises unstable isotopes. Additionally, many more potential realistic scenarios that cannot be tested experimentally may be presented and it will be useful to also model how this system will behave under those circumstances. The potential of these opportunities and their relevance to the results of this thesis are discussed below.

4.1 Special Nuclear Material Decay

It is very probable that an individual or group who chose to import SNM into the United States would obtain their SNM from stockpiles comprising either old weapons or used fuel [Fetter et al., 1990]. It may have been a few decades since this material was manufactured, and since the SNM comprises unstable isotopes, it will have decayed appreciably. New isotopes will be generated in the decay process that may significantly affect the behavior of fissile material under neutron interrogation.

In order to model the changes in the material composition of fissile material, a script for solving the Bateman equations, which can be used to calculate the transmutation of isotopes, has been implemented in the mathematics computation package Matlab [The MathWorks, 2004]. The detailed script is available in Appendix C.

Given an initial mass of each isotope, the script will determine the number of nuclei from each isotope in the decay chains for the initial isotopes. For each isotope, there is a function that calculates the relative change in the number of nuclei for each isotope. These time steps are integrated numerically by means of Matlab’s ode45 function, an implementation of the Runge-Kutta numerical
integration algorithm. Isotopes with a half-life of less than $10^4$ seconds have been eliminated from the decay chain models in order to prevent the magnitude of changes in isotope’s composition between time stops to exceed the current value of that isotope’s concentration. Exceptions to this stipulation are those isotopes near the beginning of the decay chain; because of their small half-life, they quickly come into a non-negligible transient equilibrium with the uranium or plutonium in the model.

The final isotopic composition of the decayed SNM is written to a material input deck for use in MCNP. Because a number of the isotopes produced in the decay of uranium and plutonium do not have publicly available MCNP cross section libraries as of the time of the writing of this thesis, the final material definition for MCNP cannot include all the isotopes present. Once these cross section libraries or substitutes for them are available, the simulations described in Chapter 4.0 can be repeated for aged SNM models.

Additionally, the Matlab script includes a computation of helium production after significant radioactive decay in the material. The amount of helium is computed from the difference in the sum of mass numbers. Dividing this quantity by four for the mass number of an alpha particle will give the number of helium atoms produced. This number can later be coupled with the anticipated geometry, composition and storage environment of the material to estimate the amount of helium that remains in the material and how this gas production has affected the volume and integrity of the SNM.

4.2 Limitations of System and Proposed Solutions

As expected, the measurable response of the proposed system will be substantially reduced by the presence of abundant low Z material. As demonstrated above, the ability to distinguish shielded dangerous material from unshielded material is weak when observing only the fast spectrum. This weakness can be improved by
expanding the types of radiation detected in the system to gamma rays and sensitivity at energies other than fast, particularly the thermal region of the neutron energy spectrum. Further simulations and experimental work will shed light on this aspect of the system's behavior.

Variations in geometry of cargo, SNM, and shielding may also affect the signal. Comparisons of system performance with diverse cargo environments where SNM may be separated into many smaller pieces interspersed with neutron absorbing or scattering material may further lessen the effectiveness of this system. These scenarios remain as a future area to explore in further simulation studies.

4.3 Conclusions

The above preliminary simulations indicate that the proposed detection system will be able to meet its intended goal of detecting a dangerous amount (e.g. ~1 kg of Pu-239) of hidden fissile material in cargo containers or potentially even less. The low-energy approach is not a severe handicap on the penetration of the neutrons in the cargo containers under the circumstances anticipated to be present during the prospective use of this system.

The minimum threshold of material of detectable material will depend on the energy distribution of the neutrons when they reach the material. If the beam has been well thermalized and still reaches the SNM, the response will be strong. For better or worse, this approach can be made sensitive to even the very small amounts of uranium-235 present in depleted uranium.

Because this detection ability of the system depends so greatly on the internal composition of a cargo container, the observed experimental performance of the system will vary significantly with the chosen experimental test geometry. Further experimental work in studying the use of this system in real-world-like scenarios
will confirm or refute the validity of these simulated results and offer new opportunities for exploration of the proposed system.
References

Blackburn, Brandon W. 2005. Personal communication.


Yip, Sidney. 2006. 22.106 neutrons interactions and applications lectures. Massachusetts institute of technology, Cambridge, MA.
Appendix A  Monte Carlo Methods in MCNP

Monte Carlo techniques are a means of sampling a known distribution stochastically in order to approximate the behavior of a large system that may even be computationally intractable in simulating directly [Yip, 2006].

Monte Carlo techniques use random numbers and cumulative probability distributions to create a set of elements that will represent random sampling of the real physical system itself. In the case of MCNP, each particle is generated in a source distribution and tracked one at a time. The distributions for the source position and velocity are defined by the user in developing an input deck, a series of lines describing the radiation transport problem given as input to the MCNP executable.

The input deck defines the problem geometry with a series of surfaces combined to single out volumetric regions identified as cells. The input deck also provides the material compositions and associated cross section libraries for particle interactions within those cells. The particle tracks depend on the mean free paths of the particles in those materials. The likelihood of a particle interacting after a certain distance in a given material with known cross sections is known, and from this, one can derive the cumulative probability distribution for its interaction over a certain distance. Choosing several random numbers between 0 and 1 and selecting the corresponding lengths from the values for the cumulative probability distribution will yield a representative sampling of the track lengths between collisions for these particles in a real system.

An analogous analysis can be done for all the ways a particle’s trajectory and interactions may vary. Tallies are also defined in the input deck which will count particles passing either through surfaces or cells and these are normalized appropriately typically to make estimates of the particle flux in those areas. These
tallies can be divided into bins representing specific ranges of energies, angles, or time coordinates.

The MCNP algorithm repeats the above processes for a number of particles specified in the input deck. The results are summed, averaged and an estimated error in the results is generated based on the deviations in the sampled distributions.
Appendix B  Representative MCNP Input Decks

Below is an example of an input deck for the penetration studies. The included deck is for measuring neutron penetration in aluminum for 100 keV photons. By adjusting the chosen material, cell density, and source energy, other problems can be computed using the same code.

```
C          Cells
C
1  3 -2.7 1 -2 3 -4 5 -6 imp:n=1 imp:p=1  $ Al
2  0 7 imp:n=0 imp:p=0
3  2 -1.293e-3 #1 #2 imp:n=1 imp:p=1  $ air
C          Surfaces
C
1  pz 0
2  pz 500
3  py -250
4  py 250
5  px -250
6  px 250
7  sz 250 500

mode n p
sdef par=1 vec 0 0 0 pos 0 0 -.01 dir=1 erg=.1
phys:p 100 1 0 0 1 $(upper energy limit no brem no coherent photnuclear doppler)
print 40 110 130 140
c          tallies
tmesh
cmsh1l:n flux
ergsh1l 0 1e-6 1 14
coral1 0 19i 200
corb1l 0 19i 200
corc1l 360
cmsh2l:n flux
ergsh2l 1e-6 1
cora2l 0 19i 200
corb2l 0 19i 200
corc2l 360
cmsh3l:n flux
ergsh3l 1 14
cora3l 0 19i 200
corb3l 0 19i 200
corc3l 360
cmsh4l:n flux
ergsh4l 0 14
cora4l 0 19i 200
corb4l 0 19i 200
corc4l 360
endmd
c          Material cards to follow
```
Below is a sample input deck for modeling basal neutron emissions from a cargo container as detailed in Section 3.2.1. Other isotopes can be modeled by adjusting the spectrum parameters and materials.

```
C Cells
1 5 -15.61 4 -5 -9 imp:n=1 imp:p=1 $ Pu target
2 2 -0.96 9 -10 4 -5 imp:n=1 imp:p=1 $ hydrogenous shell
3 2 -0.96 -10 5 -6 imp:n=1 imp:p=1 $ hydrogenous upper cap
4 2 -0.96 -10 3 -4 imp:n=1 imp:p=1 $ hydrogenous lower cap
5 3 -1.3e-3 10 -11 -6 3 imp:n=1 imp:p=1 $ air shell
6 3 -1.3e-3 -11 6 -7 imp:n=1 imp:p=1 $ air upper cap
7 3 -1.3e-3 -11 2 -3 imp:n=1 imp:p=1 $ air lower cap
8 4 -7.85 11 -13 2 -7 imp:n=1 imp:p=1 $ steel shell
9 4 -7.85 -13 7 -8 imp:n=1 imp:p=1 $ steel upper cap
10 4 -7.85 -13 1 -2 imp:n=1 imp:p=1 $ steel lower cap
11 3 -1.3e-3 -12 #1 #2 #3 #4 #5 #6 #7 #8 #9 #10 imp:n=1 imp:p=1 $ outs air
12 0 12 imp:n=0 imp:p=0 $ void

C Surfaces
1 pz 0
2 pz 0.5
3 pz 143.8
4 pz 147.8
5 pz 152.2
6 pz 156.2
7 pz 300
8 pz 300.5
9 cz 2.2 $ -1 kg == 1.044
10 cz 6.2
11 cz 150
12 so 400
13 cz 150.5

mode n p
sdef cel=1 par=1 pos = 0 0 150 axs = 0 0 1 erg=d1 rad=d2 ext=d3
spl1 -3 .799 4.903
si2 0 4
si3 -4 +4
phys:p 100 1 0 0 1 $(upper energy limit no brem no coherent photnuclear doppler)
print 40 110 130 140
```
Below is a sample input deck for modeling basal gamma emissions from a cargo container as detailed in Section 3.2.2. Other isotopes can be modeled by adjusting the spectrum intensities and materials.

```
c tallies
tmesh
cmesh1l:p flux
ergsh1 .1

coram 0  9i 20

corb1 130 14i 160

corcl3 360

cmesh2l:p flux
ergsh2 .1 1

coram 0  9i 20

corb21 130 14i 160

corcl3 360

cmesh3l:p flux
ergsh3 .1 4

coram 0  9i 20

corb3 130 14i 160

corcl3 360

demd
f2:n 13 $ photons passing through steel wall
fs2 -1 -8
sd2 10 284158.626 5
e2 1e-6 .1 1

t2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.0 1.0+37

c
m1 92238.66c 0.9975 92235.66c 0.0025 $ DU
m2 1001.66c .6665667 1002.66c .0001 6012.42c .3296333 6013.42c .0037 $ HDPE
m3 8016.62c -.231781 7014.62c -.755267 6000.60c -.00124 18000.59c -.012827 $ air
m4 26000.55c -.6168 6000.66c -.0025 25055.66c -.02 &
15031.66c -.045 16000.66c -.0003 14000.66c -.01 &
24000.50c -.180 28000.50d -.1400 42000.66c -.03
m5 94239 91.95 94241.50 0.29 94240.50 4.34 31000.50 3.41

$c m5 is d-Phase plutonium with 3.41% natural Gallium
nps 100000000
```

Below is a sample input deck for modeling basal gamma emissions from a cargo container as detailed in Section 3.2.2. Other isotopes can be modeled by adjusting the spectrum intensities and materials.
12  0 12 imp:n=0 imp:p=0 $ void

c Surfaces
c
1  pz 0
2  pz 0.5
3  pz 143.8
4  pz 147.8
5  pz 152.2
6  pz 156.2
7  pz 300
8  pz 300.5
9  cz 2.2 $ -1 kg == 1.044
10 cz 6.2
11 cz 150
12 so 400
13 cz 150.5

mode n p
sdef cel=1 par=2 pos = 0 0 150 axs = 0 0 1 erg=d1 rad=d2 ext=d3
sil L 0.030037 0.038661 0.04041 0.042088 0.046204 0.046625 0.04756 &
0.051624 0.05403 0.056825 0.067673 0.068699 0.06873 &
0.07496 0.077598 0.078422 0.091618 0.099878 0.103032 &
0.11537 0.119685 0.12235 0.123226 0.123626 0.124501 &
0.125181 0.129297 0.141655 0.144201 0.146095 0.158347 &
0.16019 0.16145 0.16781 0.171388 0.173715 0.179212 &
0.18455 0.1881 0.19349 0.195683 0.19687 0.203545 0.218 &
0.225417 0.237774 0.242085 0.243383 0.24493 0.248882 0.25537 &
0.263914 0.265724 0.281141 0.285334 0.29745 0.302909 0.307807 &
0.311729 0.31644 0.319802 0.320865 0.323853 0.332842 0.336113 &
0.34151 0.345008 0.3508 0.354031 0.361841 0.367072 0.368557 &
0.375045 0.380173 0.382737 0.39256 0.393136 0.399535 0.40688 &
0.41102 0.412436 0.413707 0.422598 0.42667 0.428098 0.430187 &
0.44574 0.44682 0.451483 0.45765 0.46126 0.46371 0.4743 0.48152 &
0.487015 0.493145 0.497 0.52639 0.5388 0.55053 0.5573 0.5794 &
0.582797 0.586092 0.59583 0.59798 0.5996 0.6069 0.6089 0.612834 &
0.6171 0.618334 0.61928 0.624754 0.63309 0.637795 0.63997 &
0.645896 &
0.64931 0.650529 0.652053 0.65481 0.65886 0.664536 0.6682 0.6708 &
0.674 0.68597 0.6881 0.690715 0.6932 0.697769 0.6996 0.70101 &
0.703678 0.71296 0.714563 0.71776 0.7203 0.727806 0.735906 0.7427 &
0.747974 0.75622 0.76361 0.766467 0.76919 0.7771 0.779431 0.7869 &
0.7885 0.792608 0.7969 0.8032 0.805648 0.80821 0.813504 0.816 &
0.8213 &
0.8268 0.8289 0.8325 0.8373 0.84025 0.843787 0.8792 0.891 0.8954 &
0.8981 0.9055 0.9117 0.9187 0.9319 0.9403 0.9556 0.9576 0.96837 &
0.9797 0.9827 0.9869 0.9927 1.0057 1.0094 1.0573
spl 2.17E-06 1.05E-04 1.62E-06 1.65E-06 7.37E-06 5.80E-07 5.60E-07 &
2.71E-04 1.97E-06 1.13E-05 4.56E-07 1.64E-06 3.00E-06 1.10E-06 &
3.80E-07 4.10E-06 1.41E-06 3.00E-07 2.23E-07 1.22E-05 2.30E-06 &
4.62E-06 5.97E-06 3.20E-07 3.00E-08 1.60E-11 1.97E-07 6.13E-07 &
7.11E-07 6.31E-05 3.20E-07 1.74E-07 2.86E-06 1.24E-06 1.00E-08 &
6.20E-08 1.23E-06 2.90E-08 1.10E-06 3.00E-11 3.10E-08 6.60E-07 &
2.10E-08 1.09E-07 8.30E-07 8.90E-08 1.07E-06 3.70E-08 5.69E-06 &
<table>
<thead>
<tr>
<th>s1</th>
<th>s2</th>
<th>s3</th>
<th>s4</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.20E-08</td>
<td>1.51E-07</td>
<td>1.44E-07</td>
<td>7.30E-08</td>
</tr>
<tr>
<td>8.00E-07</td>
<td>2.65E-07</td>
<td>1.60E-08</td>
<td>2.10E-08</td>
</tr>
<tr>
<td>5.50E-08</td>
<td>2.58E-07</td>
<td>1.32E-07</td>
<td>4.80E-08</td>
</tr>
<tr>
<td>1.12E-06</td>
<td>6.62E-07</td>
<td>5.56E-06</td>
<td>1.80E-08</td>
</tr>
<tr>
<td>8.80E-07</td>
<td>1.55E-05</td>
<td>3.05E-06</td>
<td>2.59E-06</td>
</tr>
<tr>
<td>2.50E-08</td>
<td>6.80E-08</td>
<td>1.80E-10</td>
<td>1.47E-05</td>
</tr>
<tr>
<td>4.30E-08</td>
<td>8.80E-08</td>
<td>8.40E-09</td>
<td>1.89E-09</td>
</tr>
<tr>
<td>5.40E-10</td>
<td>4.60E-08</td>
<td>2.65E-09</td>
<td>8.70E-09</td>
</tr>
<tr>
<td>4.20E-09</td>
<td>3.80E-10</td>
<td>8.60E-10</td>
<td>6.15E-09</td>
</tr>
<tr>
<td>2.00E-09</td>
<td>1.20E-09</td>
<td>1.16E-09</td>
<td>9.50E-09</td>
</tr>
<tr>
<td>4.57E-09</td>
<td>2.53E-08</td>
<td>2.56E-08</td>
<td>8.70E-08</td>
</tr>
<tr>
<td>6.60E-08</td>
<td>2.25E-08</td>
<td>9.70E-08</td>
<td>1.66E-08</td>
</tr>
<tr>
<td>8.70E-09</td>
<td>1.11E-09</td>
<td>9.00E-09</td>
<td>3.00E-10</td>
</tr>
<tr>
<td>3.95E-08</td>
<td>5.20E-10</td>
<td>7.90E-10</td>
<td>2.80E-08</td>
</tr>
<tr>
<td>3.80E-10</td>
<td>8.10E-10</td>
<td>3.47E-08</td>
<td>3.20E-10</td>
</tr>
<tr>
<td>1.36E-09</td>
<td>8.60E-10</td>
<td>3.50E-10</td>
<td>2.00E-10</td>
</tr>
<tr>
<td>1.21E-09</td>
<td>4.50E-10</td>
<td>2.40E-10</td>
<td>5.00E-10</td>
</tr>
<tr>
<td>1.90E-10</td>
<td>4.80E-10</td>
<td>1.34E-09</td>
<td>3.60E-10</td>
</tr>
<tr>
<td>7.50E-11</td>
<td>1.40E-10</td>
<td>8.40E-11</td>
<td>1.30E-10</td>
</tr>
<tr>
<td>2.80E-10</td>
<td>2.80E-10</td>
<td>1.10E-10</td>
<td>2.10E-10</td>
</tr>
</tbody>
</table>

**si2:** 0.4  
**si3:** -4 +4  
**phys:** p 10 0 0 1  

$(upper energy limit no brem no coherent photnuclear doppler)$

print 40 110 130 140  

c tallies  

tmesh  
cmesh1l:p flux  
ergsh1l 0 .1  
coral1 0 9i 20  
corb1l 130 14i 160  
corc1l 360  
cmesh2l:p flux  
ergsh2l 1.1  
cora2l 0 9i 20  
corb2l 130 14i 160  
corc2l 360  
cmesh3l:p flux  
ergsh3l 0 14  
cora3l 0 9i 20  
corb3l 130 14i 160  
corc3l 360  
endmd  
f2:p 13 $ photons passing through steel wall  
fs2 -1 -8  
sd2 10 284158.626 5  
e2 .05 .1 1 14  
t2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1.0 1.0+37  
c  
m1 92238.66c 0.9975 92235.66c 0.0025 $ DU  
m2 1001.66c .6665667 1002.66c .0001 6012.42c .3296333 6013.42c .0037 $ HDPE  
m3 8016.62c -.231781 7014.62c -.755267 6000.60c -.000124 18000.59c -.012827 $ air  
m4 26000.55c -.6168 6000.66c -.0025 25055.66c -.02 $ 15031.66c -.045 16000.66c -.0003 14000.60c -.01 $ 24000.50c -.180 28000.50d -.140 42000.66c -.03
The following input deck can be used to simulate the neutron flux in a cargo container that may or may not contain fissile material responding to the interrogating source. By altering the composition of the target and the surrounding shielding or by adjusting the geometry, further scenarios for neutron interrogation of cargo can also be simulated. The neutron source has been calculated in previous literature for a $^7$Li(p,$n$)$^7$Be based target with a neutron beam where 60 keV is the most likely output energy.

C Cells
1 5 -18.74 4 -5 -9 imp:n=1 imp:p=1 $ \text{U target}$
2 3 -1.3e-3 9 -10 4 -5 imp:n=1 imp:p=1 $ \text{hydrogenous shell}$
3 3 -1.3e-3 -10 5 -6 imp:n=1 imp:p=1 $ \text{hydrogenous upper cap}$
4 3 -1.3e-3 -10 3 -4 imp:n=1 imp:p=1 $ \text{hydrogenous lower cap}$
5 3 -1.3e-3 10 -11 -6 3 imp:n=1 imp:p=1 $ \text{air shell}$
6 3 -1.3e-3 -11 6 -7 imp:n=1 imp:p=1 $ \text{air upper cap}$
7 3 -1.3e-3 -11 2 -3 imp:n=1 imp:p=1 $ \text{air lower cap}$
8 4 -7.85 11 -13 2 -7 imp:n=1 imp:p=1 $ \text{steel shell}$
9 4 -7.85 -13 7 -8 imp:n=1 imp:p=1 $ \text{steel upper cap}$
10 4 -7.85 -13 1 -2 imp:n=1 imp:p=1 $ \text{steel lower cap}$
11 3 -1.3e-3 -12 #1 #2 #3 #4 #5 #6 #7 #8 #9 #10 imp:n=1 imp:p=1 $ \text{outs}$
air
12 0 12 imp:n=0 imp:p=0 $ \text{void}$

C Surfaces
1 pz 0
2 pz 0.5
3 pz 142
4 pz 146
5 pz 154
6 pz 158
7 pz 300
8 pz 300.5
9 cz 4 $-$5 kg == 5.12
10 cz 8
11 cz 150
12 so 400
13 cz 150.5

mode n p
sdef par=1 vec 0 0 1 pos 0 0 -50 dir=d1 erg=fdir d2
sil -1.00 -0.90 -0.80 -0.70 -0.60 -0.50 -0.40 -0.30 -0.20 -0.10 -0.00
& 0.10 0.20 0.30 0.40 0.50 0.60 0.61 0.62 0.63 0.64 0.65
&
0.66 0.67 0.68 0.69 0.70 0.71 0.72 0.73 0.74 0.75 0.76
& 0.77 0.78 0.79 0.80 0.81 0.82 0.83 0.84 0.85 0.86 0.87
& 0.88 0.89 0.90 0.91 0.92 0.93 0.94 0.95 0.96 0.97 0.98
& 0.99 1.00
sp1 0 4.885E-05 6.476E-05 8.926E-05 1.273E-04 1.878E-04 2.891E-04
4.667E-04 & 7.906E-04 1.400E-03 2.564E-03 4.754E-03 8.718E-03 1.551E-02
1.018E-02 & 1.064E-02 1.106E-02 1.151E-02 1.202E-02 1.248E-02 1.302E-02
1.351E-02 & 1.411E-02 1.468E-02 1.526E-02 1.587E-02 1.651E-02 1.717E-02
1.785E-02 & 1.858E-02 1.932E-02 2.003E-02 2.087E-02 2.162E-02 2.251E-02
2.334E-02 & 2.419E-02 2.512E-02 2.609E-02 2.706E-02 2.806E-02 2.915E-02
3.022E-02 & 3.133E-02 3.247E-02 3.382E-02 3.506E-02 3.667E-02 3.830E-02
4.018E-02
ds2 s 11 12 13 14 15 16 17 18 19 20 21 22 23 24 25 26 27 28 29 30 &
31 32 33 34 35 36 37 38 39 40 41 42 43 44 45 46 47 48 49 50 &
51 52 53 54 55 56 57 58 59 60 61 62 63 64 65 66
sill 0.00 0.01
sp11 0 1.000E+00
sp12 0 1.000E+00
sp13 0 1.000E+00
sp14 0 1.000E+00
sp15 0 1.000E+00
sp16 0 1.000E+00
sp17 0 1.000E+00
sp18 0 1.000E+00
sp19 0 1.000E+00
sp20 0 1.000E+00
sp21 0 1.000E+00
sp22 0 1.000E+00
sp23 0 1.000E+00
sp24 0 1.000E+00
sp25 0 1.000E+00
sp26 0 1.000E+00
sp27 0 1.000E+00
sp28 0 1.000E+00
sp29 0 1.000E+00
sp30 0 1.000E+00
sp31 0 1.000E+00
sp32 0 1.000E+00
sp33 0 1.000E+00
sp34 0 1.000E+00
sp35 0 1.000E+00
sp36 0 1.000E+00
sp37 0 1.000E+00
sp38 0 1.000E+00
sp39 0 1.000E+00
sp40 0 1.000E+00
sp41 0 1.000E+00
sp42 0 1.000E+00
sp43 0 1.000E+00
sp44 0 1.000E+00
sp45 0 1.000E+00
sp46 0 1.000E+00
sp47 0 1.000E+00
sp48 0 1.000E+00
sp49 0 1.000E+00
sp50 0 1.000E+00
sp51 0 1.000E+00
sp52 0 1.000E+00
sp53 0 1.000E+00
sp54 0 1.000E+00
sp55 0 1.000E+00
sp56 0 1.000E+00
sp57 0 1.000E+00
sp58 0 1.000E+00
sp59 0 1.000E+00
sp60 0 1.000E+00
sp61 0 1.000E+00
sp62 0 1.000E+00
sp63 0 1.000E+00
sp64 0 1.000E+00
sp65 0 1.000E+00
sp66 0 1.000E+00
<p>| | | | | | | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0.00</td>
<td>0.01</td>
<td>0.02</td>
<td>0.03</td>
<td>0.04</td>
<td>0.05</td>
</tr>
<tr>
<td>sp27</td>
<td>8.274E-02</td>
<td>1.511E-01</td>
<td>1.847E-01</td>
<td>2.010E-01</td>
<td>2.080E-01</td>
<td>1.724E-01</td>
</tr>
<tr>
<td>sp28</td>
<td>7.914E-02</td>
<td>1.425E-01</td>
<td>1.738E-01</td>
<td>1.890E-01</td>
<td>1.958E-01</td>
<td>1.970E-01</td>
</tr>
<tr>
<td>sp29</td>
<td>7.694E-02</td>
<td>1.425E-01</td>
<td>1.738E-01</td>
<td>1.890E-01</td>
<td>1.958E-01</td>
<td>1.970E-01</td>
</tr>
<tr>
<td>sp30</td>
<td>7.396E-02</td>
<td>1.377E-01</td>
<td>1.688E-01</td>
<td>1.837E-01</td>
<td>1.903E-01</td>
<td>1.916E-01</td>
</tr>
<tr>
<td>sp31</td>
<td>7.151E-02</td>
<td>1.332E-01</td>
<td>1.639E-01</td>
<td>1.786E-01</td>
<td>1.849E-01</td>
<td>1.863E-01</td>
</tr>
<tr>
<td>sp32</td>
<td>6.887E-02</td>
<td>1.314E-01</td>
<td>1.590E-01</td>
<td>1.731E-01</td>
<td>1.805E-01</td>
<td>1.805E-01</td>
</tr>
<tr>
<td>sp33</td>
<td>6.648E-02</td>
<td>1.262E-01</td>
<td>1.547E-01</td>
<td>1.731E-01</td>
<td>1.805E-01</td>
<td>1.805E-01</td>
</tr>
<tr>
<td>sp34</td>
<td>6.454E-02</td>
<td>1.207E-01</td>
<td>1.510E-01</td>
<td>1.701E-01</td>
<td>1.715E-01</td>
<td>1.715E-01</td>
</tr>
<tr>
<td>sp35</td>
<td>6.202E-02</td>
<td>1.140E-01</td>
<td>1.443E-01</td>
<td>1.644E-01</td>
<td>1.701E-01</td>
<td>1.715E-01</td>
</tr>
<tr>
<td>sp36</td>
<td>6.021E-02</td>
<td>1.129E-01</td>
<td>1.387E-01</td>
<td>1.561E-01</td>
<td>1.613E-01</td>
<td>1.624E-01</td>
</tr>
<tr>
<td>sp37</td>
<td>5.805E-02</td>
<td>1.100E-01</td>
<td>1.337E-01</td>
<td>1.519E-01</td>
<td>1.568E-01</td>
<td>1.577E-01</td>
</tr>
<tr>
<td>sp38</td>
<td>5.626E-02</td>
<td>1.067E-01</td>
<td>1.337E-01</td>
<td>1.519E-01</td>
<td>1.568E-01</td>
<td>1.577E-01</td>
</tr>
<tr>
<td>sp39</td>
<td>5.434E-02</td>
<td>1.032E-01</td>
<td>1.318E-01</td>
<td>1.446E-01</td>
<td>1.493E-01</td>
<td>1.499E-01</td>
</tr>
<tr>
<td>sp40</td>
<td>5.248E-02</td>
<td>1.002E-01</td>
<td>1.286E-01</td>
<td>1.412E-01</td>
<td>1.459E-01</td>
<td>1.464E-01</td>
</tr>
<tr>
<td>sp41</td>
<td>5.088E-02</td>
<td>9.997E-02</td>
<td>1.255E-01</td>
<td>1.380E-01</td>
<td>1.426E-01</td>
<td>1.429E-01</td>
</tr>
<tr>
<td>sp42</td>
<td>4.923E-02</td>
<td>9.726E-02</td>
<td>1.224E-01</td>
<td>1.350E-01</td>
<td>1.394E-01</td>
<td>1.396E-01</td>
</tr>
</tbody>
</table>

77
<table>
<thead>
<tr>
<th>SP</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>sp43</td>
<td>0.476E-02 9.431E-02 1.197E-01 1.319E-01 1.364E-01 1.365E-01</td>
</tr>
<tr>
<td>si44</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09</td>
</tr>
<tr>
<td>sp44</td>
<td>0.618E-02 9.186E-02 1.165E-01 1.290E-01 1.336E-01 1.335E-01</td>
</tr>
<tr>
<td>si45</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09</td>
</tr>
<tr>
<td>sp45</td>
<td>0.466E-02 8.858E-02 1.144E-01 1.264E-01 1.308E-01 1.307E-01</td>
</tr>
<tr>
<td>si46</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09</td>
</tr>
<tr>
<td>sp46</td>
<td>0.435E-02 8.731E-02 1.135E-01 1.254E-01 1.297E-01</td>
</tr>
<tr>
<td>si47</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10</td>
</tr>
<tr>
<td>sp47</td>
<td>0.394E-02 8.485E-02 1.107E-01 1.228E-01</td>
</tr>
<tr>
<td>si48</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09</td>
</tr>
<tr>
<td>sp48</td>
<td>0.378E-02 8.269E-02 1.103E-01 1.224E-01</td>
</tr>
<tr>
<td>si49</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10</td>
</tr>
<tr>
<td>sp49</td>
<td>0.358E-02 7.984E-02 1.081E-01 1.212E-01</td>
</tr>
<tr>
<td>si50</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09</td>
</tr>
<tr>
<td>sp50</td>
<td>0.332E-02 7.696E-02 9.565E-02 1.093E-01</td>
</tr>
<tr>
<td>si51</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11</td>
</tr>
<tr>
<td>sp51</td>
<td>0.316E-02 7.462E-02 9.345E-02 1.074E-01</td>
</tr>
<tr>
<td>si52</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11</td>
</tr>
<tr>
<td>sp52</td>
<td>0.296E-02 7.234E-02 9.145E-02</td>
</tr>
<tr>
<td>si53</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11</td>
</tr>
<tr>
<td>sp53</td>
<td>0.276E-02 6.995E-02</td>
</tr>
<tr>
<td>si54</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11</td>
</tr>
<tr>
<td>sp54</td>
<td>0.255E-02 6.786E-02</td>
</tr>
<tr>
<td>si55</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11 0.12</td>
</tr>
<tr>
<td>sp55</td>
<td>0.234E-02 6.576E-02</td>
</tr>
<tr>
<td>si56</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11 0.12</td>
</tr>
<tr>
<td>sp56</td>
<td>0.214E-02 6.367E-02</td>
</tr>
<tr>
<td>si57</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11 0.12</td>
</tr>
<tr>
<td>sp57</td>
<td>0.194E-02 6.158E-02</td>
</tr>
<tr>
<td>si58</td>
<td>0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11 0.12</td>
</tr>
<tr>
<td>sp58</td>
<td>0.174E-02 5.949E-02</td>
</tr>
</tbody>
</table>

78
1.000E-01 9.547E-02 9.131E-02 8.718E-02 3.283E-02
si58 0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11 0.12
sp58 0 2.996E-02 6.234E-02 8.381E-02 9.547E-02 1.024E-01 1.052E-01
1.028E-01 &
9.861E-02 9.394E-02 8.966E-02 8.549E-02 5.028E-02
si59 0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11 0.12
sp59 0 2.906E-02 6.089E-02 8.163E-02 9.355E-02 1.005E-01 1.036E-01
1.016E-01 &
9.735E-02 9.262E-02 8.815E-02 8.395E-02 6.714E-02
si60 0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11 0.12
sp60 0 2.824E-02 5.923E-02 7.917E-02 9.176E-02 9.864E-02 1.018E-01
1.006E-01 &
9.524E-02 9.031E-02 8.556E-02 8.131E-02 7.172E-02 2.198E-02
si61 0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11 0.12
sp61 0 2.738E-02 5.789E-02 7.652E-02 9.037E-02 9.694E-02 9.995E-02
9.944E-02 &
9.524E-02 9.031E-02 8.556E-02 8.131E-02 7.172E-02 2.198E-02
si62 0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11 0.12
sp62 0 2.647E-02 5.616E-02 7.811E-02 8.867E-02 9.485E-02 9.772E-02
9.778E-02 &
si63 0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 0.08 0.09 0.10 0.11 0.12
sp63 0 2.569E-02 5.490E-02 7.460E-02 8.808E-02 9.342E-02 9.609E-02
9.648E-02 &
9.303E-02 8.800E-02 8.295E-02 7.844E-02 7.432E-02 5.398E-02
si64 0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 &
0.08 0.09 0.10 0.11 0.12 0.13 0.14
sp64 0 2.471E-02 5.332E-02 7.788E-02 8.819E-02 9.145E-02 9.373E-02
9.410E-02 &
1.962E-03
si65 0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 &
0.08 0.09 0.10 0.11 0.12 0.13 0.14
sp65 0 2.381E-02 5.197E-02 7.768E-02 8.977E-02 9.040E-02 9.163E-02
9.189E-02 &
8.984E-02 8.487E-02 7.969E-02 7.500E-02 7.086E-02 6.710E-02
1.547E-02
si66 0.00 0.01 0.02 0.03 0.04 0.05 0.06 0.07 &
0.08 0.09 0.10 0.11 0.12 0.13 0.14
sp66 0 2.288E-02 5.095E-02 7.786E-02 9.228E-02 9.068E-02 8.938E-02
8.942E-02 &
8.792E-02 8.320E-02 7.794E-02 7.314E-02 6.900E-02 6.531E-02
3.004E-02
phys:p 100 1 0 0 1 $(upper energy limit no brem no coherent
photnuclear doppler)
print 40 110 130 140
c tallies
tmesh
cmesh1:n flux
ergsh1 0 14
coral 0 16i 170
corb1 -50 39i 350
corc1 360
cmesh21:n flux
ergsh21 0 1e-6
cora21 0 16i 170
corb21 -50 39i 350
corc21 360
cmesh31: n flux
  ergsh31 1e-6 1
cora31 0 16i 170
corb31 -50 39i 350
corc31 360
cmesh41: n flux
  ergsh41 1.14
cora41 0 16i 170
corb41 -50 39i 350
corc41 360
endmd
f2:p 13 $ photons passing through steel wall
fs2 -1 -8
sd2 10 284158.626 5
e2 .05 .1 1.14
t2 100 200 300 400 500 600 700 800 900 1.0e+37
f12:n 13 $ neutrons passing through steel wall
fs12 -1 -8
sd12 10 284158.626 5
e12 1e-6 1.14
t12 5 10 40 50 60 70 80 90 100 110 120 130 140 150 160 170 180 190 200 210 220 & 230 240 250 260 270 280 290 300 310 320 330 340 350 360 370 380 390 400 410 & 420 430 440 450 460 470 480 490 500 510 520 530 540 550 560 570 580 590 600 & 700 800 900 1000 1100 1200 1300 1400 1500 1600 1700 1800 1900 2000 1.0e+37
ml 92238.66c 0.9975 92235.66c 0.0025 $ DU
m2 1001.66c .6665667 1002.66c .0001 6012.42c .3296333 6013.42c .0037 $ HDPE
m3 8016.62c -.231781 7014.62c -.755267 6000.60c -.00124 18000.59c -.012827 $ air
m4 26000.55c -.6168 6000.66c -.0025 25055.66c -.02 & 15031.66c -.045 16000.66c -.0003 14000.60c -.01 & 24000.50c -.180 28000.50d -.1400 42000.66c -.03
m5 92234.66c 0.01 92235.66c 0.935 92238.66c 0.055 $ HEU
nps 100000000
Appendix C  Special Nuclear Material Aging Model

The following script includes the age of the weapons material and the mass fraction of uranium and/or plutonium isotopes. It uses the functions below to calculate the amount of each fissile isotope produced and outputs the composition to an external material input deck.

```
totalComp.m

% Calculates total composition given age and initial composition of
% U-232, -234, -235, -238, Pu-239, -240, -241

Na = 6.02214199e23; % Avogadro's number

time = 50; % years
(time = time * 365*24*60*60; % seconds

numIsotopes = 12;
numInitIsotopes = 7;

% Uranium + Plutonium Starting Composition
TotalMass = 10; % kg

UPuInitCompFrac = zeros(numInitIsotopes,1);
UPuInitCompMassNumber = zeros(numInitIsotopes,1);

% Fractions below are mass; fractions in simulation are atom
UPuInitCompFrac(1) = .1; % U-232
UPuInitCompMassNumber(1) = 232;
UPuInitCompFrac(2) = .1; % U-234
UPuInitCompMassNumber(2) = 234;
UPuInitCompFrac(3) = .5; % U-235
UPuInitCompMassNumber(3) = 235;
UPuInitCompFrac(4) = .2; % U-238
UPuInitCompMassNumber(4) = 238;

UPuInitCompFrac(5) = .02; % Pu-239
UPuInitCompMassNumber(5) = 239;
UPuInitCompFrac(6) = .03; % Pu-240
UPuInitCompMassNumber(6) = 240;
UPuInitCompFrac(7) = .05; % Pu-241
UPuInitCompMassNumber(7) = 241;

MatCardFileName = 'output';

% Calculate number of nuclei (i.e. convert mass fraction to atom fraction)
% if given in mass fraction
MassIsotopes = TotalMass .* UPuInitCompFrac;
AtomsIsotopes = 1000 * MassIsotopes./UPuInitCompMassNumber.*Na;
TotalAtoms = sum(AtomsIsotopes);
UPuInitComp = AtomsIsotopes/TotalAtoms; % Atom fraction
```
TotalInitMassNumber = sum(UPuInitComp .* TotalAtoms .* UPuInitCompMassNumber);
HeliumProduced = zeros(numInitIsotopes,1);

%UPuInitComp = UPuInitCompFrac; % if given in atom fraction

% Run simulations
if UPuInitComp(1)
    fprintf('Running U-232 decay...
');
    U232DecayIsotopes = U232run(time)*UPuInitComp(1); % Fraction of material that consists of each isotope
    U232NumIsotopes = size(U232DecayIsotopes, 2); % Number of isotopes in decay chain
    U232DecayMassNum = zeros(U232NumIsotopes, 1); % Mass number of each isotope in chain
    U232DecayMassNum(1) = 232;
    U232DecayMassNum(2) = 228;
    U232DecayMassNum(3) = 224;
    U232DecayMassNum(4) = 212;
    U232DecayMassNum(5) = 208;
    U232MassNumberPerIsotope = U232DecayIsotopes' .* TotalAtoms .* U232DecayMassNum;

    % Mass of helium produced
    HeliumProduced(1) = ((UPuInitComp(1) * TotalAtoms * UPuInitCompMassNumber(1)) - sum(U232MassNumberPerIsotope))/Na;
end

if UPuInitComp(2)
    fprintf('Running U-234 decay...
');
    U234DecayIsotopes = U234run(time)*UPuInitComp(2);
    U234NumIsotopes = size(U234DecayIsotopes, 2);
    U234DecayMassNum = zeros(U234NumIsotopes,1);
    U234DecayMassNum(1) = 234;
    U234DecayMassNum(2) = 230;
    U234DecayMassNum(3) = 226;
    U234DecayMassNum(4) = 222;
    U234DecayMassNum(5) = 220;
    U234DecayMassNum(6) = 210;
    U234DecayMassNum(7) = 209;
    U234DecayMassNum(8) = 206;
    U234MassNumberPerIsotope = U234DecayIsotopes' .* TotalAtoms .* U234DecayMassNum;

    % Mass of helium produced
    HeliumProduced(2) = ((UPuInitComp(2) * TotalAtoms * UPuInitCompMassNumber(2)) - sum(U234MassNumberPerIsotope))/Na;
end

if UPuInitComp(3)
    fprintf('Running U-235 decay...
');
    U235DecayIsotopes = U235run(time)*UPuInitComp(3);
    U235NumIsotopes = size(U235DecayIsotopes, 2);
    U235DecayMassNum = zeros(U235NumIsotopes,1);
    U235DecayMassNum(1) = 235;
    U235DecayMassNum(2) = 231;
    U235DecayMassNum(3) = 231;
U235DecayMassNum(4) = 227;
U235DecayMassNum(5) = 227;
U235DecayMassNum(6) = 223;
U235DecayMassNum(7) = 207;
U235MassNumberPerIsotope = U235DecayIsotopes' .* TotalAtoms .* U235DecayMassNum;

HeliumProduced(3) = ((UPuInitComp(3) * TotalAtoms * UPuInitCompMassNumber(3)) - sum(U235MassNumberPerIsotope))/Na;
end

if UPuInitComp(4)
  fprintf('Running U-238 decay...
');
  U238DecayIsotopes = U238run(time)*UPuInitComp(4);
  U238NumIsotopes = size(U238DecayIsotopes, 2);
  U238DecayMassNum = zeros(U238NumIsotopes, 1);
  U238DecayMassNum(1) = 238;
  U238DecayMassNum(2) = 234;
  U238DecayMassNum(3) = 234;
  U238DecayMassNum(4) = 238;
  U238DecayMassNum(5) = 230;
  U238DecayMassNum(6) = 226;
  U238DecayMassNum(7) = 222;
  U238DecayMassNum(8) = 210;
  U238DecayMassNum(9) = 210;
  U238DecayMassNum(10) = 210;
  U238DecayMassNum(11) = 209;
  U238DecayMassNum(12) = 206;
  U238MassNumberPerIsotope = U238DecayIsotopes' .* TotalAtoms .* U238DecayMassNum;

  HeliumProduced(4) = ((UPuInitComp(4) * TotalAtoms * UPuInitCompMassNumber(4)) - sum(U238MassNumberPerIsotope))/Na;
end

if UPuInitComp(5)
  fprintf('Running Pu-239 decay...
');
  Pu239DecayIsotopes = Pu239run(time)*UPuInitComp(5);
  Pu239NumIsotopes = size(Pu239DecayIsotopes, 2);
  Pu239DecayMassNum = zeros(Pu239NumIsotopes, 1);
  Pu239DecayMassNum(1) = 235;
  Pu239DecayMassNum(2) = 231;
  Pu239DecayMassNum(3) = 231;
  Pu239DecayMassNum(4) = 227;
  Pu239DecayMassNum(5) = 227;
  Pu239DecayMassNum(6) = 223;
  Pu239DecayMassNum(7) = 207;
  Pu239DecayMassNum(8) = 239;
  Pu239MassNumberPerIsotope = Pu239DecayIsotopes' .* TotalAtoms .* Pu239DecayMassNum;

  HeliumProduced(5) = ((UPuInitComp(5) * TotalAtoms * UPuInitCompMassNumber(5)) - sum(Pu239MassNumberPerIsotope))/Na;
end

if UPuInitComp(6)
  fprintf('Running Pu-240 decay...
');
Pu240DecayIsotopes = Pu240run(time)*UPuInitComp(6);
Pu240NumIsotopes = size(Pu240DecayIsotopes, 2);
Pu240DecayMassNum = zeros(Pu240NumIsotopes,1);
Pu240DecayMassNum(1) = 240;
Pu240DecayMassNum(2) = 236;
Pu240DecayMassNum(3) = 232;
Pu240DecayMassNum(4) = 228;
Pu240DecayMassNum(5) = 228;
Pu240DecayMassNum(6) = 224;
Pu240DecayMassNum(7) = 208;
Pu240MassNumberPerIsotope = Pu240DecayIsotopes' .* TotalAtoms .*
Pu240DecayMassNum;

HeliumProduced(6) = ((UPuInitComp(6) * TotalAtoms * 
UPuInitCompMassNumber(6)) - sum(Pu240MassNumberPerIsotope))/Na;
end
if UPuInitComp(7)
fprintf('Running Pu-241 decay...\n');
Pu241DecayIsotopes = Pu241run(time)*UPuInitComp(7);
Pu241NumIsotopes = size(Pu241DecayIsotopes, 2);
Pu241DecayMassNum = zeros(Pu241NumIsotopes,1);
Pu241DecayMassNum(1) = 241;
Pu241DecayMassNum(2) = 241;
Pu241DecayMassNum(3) = 237;
Pu241DecayMassNum(4) = 233;
Pu241DecayMassNum(5) = 233;
Pu241DecayMassNum(6) = 229;
Pu241DecayMassNum(7) = 225;
Pu241DecayMassNum(8) = 225;
Pu241DecayMassNum(9) = 209;
Pu241MassNumberPerIsotope = Pu241DecayIsotopes' .* TotalAtoms .*
Pu241DecayMassNum;

HeliumProduced(7) = ((UPuInitComp(7) * TotalAtoms * 
UPuInitCompMassNumber(7)) - sum(Pu241MassNumberPerIsotope))/Na;
end

% Calculate helium produced
TotalHeliumProduced = sum(HeliumProduced);

% Output to screen
fprintf('
\nFractional (by atom) isotopic composition at %.3e 
seconds\n', time);
fprintf('=================================\n');
fprintf('Americium: \n');
if UPuInitComp(7) fprintf('\t241: %.3e \n', Pu241DecayIsotopes(2)); end
fprintf('Plutonium: \n');
if UPuInitComp(5) fprintf('\t239: %.3e \n', Pu239DecayIsotopes(8)); end
if UPuInitComp(6) fprintf('\t240: %.3e \n', Pu240DecayIsotopes(8)); end
if UPuInitComp(7) fprintf('\t241: %.3e \n', Pu241DecayIsotopes(1)); end
fprintf('Neptunium: \n');
if UPuInitComp(7) fprintf('\t237: %.3e \n', Pu241DecayIsotopes(3)); end
fprintf('Uranium: \n');
if UPuInitComp(1) fprintf('\t232: %.3e \n', U232DecayIsotopes(1)); end

84
if UPuInitComp(7) fprintf('	233: %.3e 
', Pu241DecayIsotopes(5)); end
if UPuInitComp(2) || UPuInitComp(4) fprintf('	234: %.3e 
',
U234DecayIsotopes(1) + U238DecayIsotopes(4)); end
if UPuInitComp(3) || UPuInitComp(5) fprintf('	235: %.3e 
',
U235DecayIsotopes(1) + Pu239DecayIsotopes(1)); end
if UPuInitComp(6) fprintf('	236: %.3e 
', Pu240DecayIsotopes(2)); end
if UPuInitComp(4) fprintf('	238: %.3e 
', U238DecayIsotopes(1)); end
fprintf('Protactinium:
');
if UPuInitComp(3) || UPuInitComp(5) fprintf('	231: %.3e 
',
U235DecayIsotopes(3) + Pu239DecayIsotopes(3)); end
if UPuInitComp(2) || UPuInitComp(4) fprintf('	234: %.3e 
', U238DecayIsotopes(3)); end
fprintf('Thorium:
');
if UPuInitComp(3) || UPuInitComp(5) fprintf('	227: %.3e 
',
U235DecayIsotopes(5) + Pu239DecayIsotopes(5)); end
if UPuInitComp(1) || UPuInitComp(6) fprintf('	228: %.3e 
',
U232DecayIsotopes(2) + Pu240DecayIsotopes(5)); end
if UPuInitComp(7) fprintf('	229: %.3e 
', Pu241DecayIsotopes(6)); end
if UPuInitComp(2) || UPuInitComp(4) fprintf('	230: %.3e 
',
U234DecayIsotopes(2) + U238DecayIsotopes(6)); end
if UPuInitComp(3) || UPuInitComp(5) fprintf('	231: %.3e 
',
U235DecayIsotopes(2) + Pu239DecayIsotopes(2)); end
if UPuInitComp(6) fprintf('	232: %.3e 
', Pu240DecayIsotopes(3)); end
if UPuInitComp(4) fprintf('	234: %.3e 
', U238DecayIsotopes(2)); end
fprintf('Actinium:
');
if UPuInitComp(7) fprintf('	225: %.3e 
', Pu241DecayIsotopes(8)); end
if UPuInitComp(3) || UPuInitComp(5) fprintf('	227: %.3e 
',
U235DecayIsotopes(4) + Pu239DecayIsotopes(4)); end
if UPuInitComp(6) fprintf('	228: %.3e 
', Pu240DecayIsotopes(4)); end
fprintf('Radium:
');
if UPuInitComp(3) || UPuInitComp(4) fprintf('	222: %.3e 
',
U234DecayIsotopes(6) + Pu239DecayIsotopes(6)); end
if UPuInitComp(1) || UPuInitComp(6) fprintf('	224: %.3e 
',
U232DecayIsotopes(3) + Pu240DecayIsotopes(6)); end
if UPuInitComp(7) fprintf('	225: %.3e 
', Pu241DecayIsotopes(7)); end
if UPuInitComp(2) || UPuInitComp(4) fprintf('	226: %.3e 
',
U234DecayIsotopes(3) + U238DecayIsotopes(7)); end
if UPuInitComp(3) || UPuInitComp(5) fprintf('	207: %.3e 
',
Pu239DecayIsotopes(7)); end
if UPuInitComp(6) fprintf('	208: %.3e 
', Pu240DecayIsotopes(7)); end
if UPuInitComp(4) fprintf('	210: %.3e 
', U238DecayIsotopes(8)); end
fprintf('Radon:
');
if UPuInitComp(2) || UPuInitComp(4) fprintf('	222: %.3e 
',
U234DecayIsotopes(8) + U238DecayIsotopes(12)); end
if UPuInitComp(4) fprintf('	210: %.3e 
', U238DecayIsotopes(8) + U234DecayIsotopes(9)); end
if UPuInitComp(2) || UPuInitComp(4) fprintf('	206: %.3e 
',
U234DecayIsotopes(9) + U238DecayIsotopes(12)); end
if UPuInitComp(3) || UPuInitComp(5) fprintf('	207: %.3e 
',
U235DecayIsotopes(7) + Pu239DecayIsotopes(7)); end
if UPuInitComp(1) || UPuInitComp(6) fprintf('	208: %.3e 
',
U232DecayIsotopes(5) + Pu240DecayIsotopes(7)); end
if UPuInitComp(2) || UPuInitComp(4) fprintf('	210: %.3e 
',
U234DecayIsotopes(5) + U238DecayIsotopes(8)); end
if UPuInitComp(1) fprintf('
\texttt{12}: %.3e 
', U232DecayIsotopes(4)); end
fprintf('Helium produced: %.3e g
', TotalHeliumProduced);
%
% Output to material card

    cardID = fopen(MatCardFileName, 'w');
    fprintf(cardID, 'm*
');
    fprintf('Americium
');
    if UPuInitComp(7) fprintf(cardID, '95241.66c %.5E 
', Pu241DecayIsotopes(2)); end
    fprintf('Plutonium
');
    if UPuInitComp(5) fprintf(cardID, '94239.66c %.5E 
', Pu239DecayIsotopes(8)); end
    if UPuInitComp(6) fprintf(cardID, '94240.66c %.5E &
', Pu240DecayIsotopes(1)); end
    if UPuInitComp(7) fprintf(cardID, '94241.66c %.5E ', Pu241DecayIsotopes(1)); end
    fprintf('Neptunium
');
    if UPuInitComp(7) fprintf(cardID, '93237.66c %.5E ', Pu241DecayIsotopes(3)); end
    fprintf('Uranium
');
    if UPuInitComp(l) fprintf(cardID, '92232.66c %.5E &
', Pu241DecayIsotopes(5)); end
    if UPuInitComp(2) || UPuInitComp(4) fprintf(cardID, '92234.66c %.5E ', Pu234DecayIsotopes(1) + Pu238DecayIsotopes(4)); end
    if UPuInitComp(3) || UPuInitComp(5) fprintf(cardID, '92235.66c %.5E &
', U235DecayIsotopes(1) + Pu239DecayIsotopes(1)); end
    if UPuInitComp(6) fprintf(cardID, '92236.66c %.5E ', Pu240DecayIsotopes(2)); end
    if UPuInitComp(4) fprintf(cardID, '92238.66c %.5E ', Pu238DecayIsotopes(1)); end
    fprintf('Protactinium
');
    if UPuInitComp(3) || UPuInitComp(5) fprintf(cardID, '91231.66c %.5E &
', U235DecayIsotopes(3) + Pu239DecayIsotopes(3)); end
    if UPuInitComp(7) fprintf(cardID, '91233.66c %.5E ', Pu241DecayIsotopes(4)); end
    if UPuInitComp(4) fprintf(cardID, '91234.66c %.5E &
', U235DecayIsotopes(2) + Pu239DecayIsotopes(2)); end
    if UPuInitComp(6) fprintf(cardID, '92235.66c %.5E &
', Pu240DecayIsotopes(3)); end
    if UPuInitComp(4) fprintf(cardID, '92236.66c %.5E &
', Pu240DecayIsotopes(2)); end
    if UPuInitComp(3) || UPuInitComp(5) fprintf(cardID, '92237.66c %.5E &
', U235DecayIsotopes(5) + Pu239DecayIsotopes(5)); end
    if UPuInitComp(1) || UPuInitComp(6) fprintf(cardID, '90227.66c %.5E &
', U238DecayIsotopes(5) + Pu240DecayIsotopes(5)); end
    if UPuInitComp(7) fprintf(cardID, '90229.66c %.5E &
', Pu241DecayIsotopes(6)); end
    if UPuInitComp(2) || UPuInitComp(4) fprintf(cardID, '90230.66c %.5E &
', U234DecayIsotopes(2) + Pu238DecayIsotopes(5)); end
    if UPuInitComp(3) || UPuInitComp(5) fprintf(cardID, '90231.42c %.5E ', U235DecayIsotopes(2) + Pu239DecayIsotopes(2)); end
    if UPuInitComp(6) fprintf(cardID, '90232.66c %.5E ', Pu240DecayIsotopes(3)); end
    if UPuInitComp(4) fprintf(cardID, '90234.66c %.5E &
', U238DecayIsotopes(2)); end
    fprintf('Actinium
');
    if UPuInitComp(7) fprintf('
\texttt{225}: %.3e 
', Pu241DecayIsotopes(8)); end
%if UPuInitComp(3) || UPuInitComp(5) fprintf('\t227: %.3e \n', U235DecayIsotopes(4) + Pu239DecayIsotopes(4)); end
%fprintf('Radium:\n');
%if UPuInitComp(3) || UPuInitComp(4) fprintf('\t223: %.3e \n', U235DecayIsotopes(6) + Pu239DecayIsotopes(6)); end
%if UPuInitComp(1) || UPuInitComp(6) fprintf('\t224: %.3e \n', U232DecayIsotopes(3) + Pu240DecayIsotopes(6)); end
%if UPuInitComp(7) fprintf('\t225: %.3e \n', Pu241DecayIsotopes(7)); end
%if UPuInitComp(2) || UPuInitComp(4) fprintf('\t226: %.3e \n', U234DecayIsotopes(3) + U238DecayIsotopes(6)); end
%if UPuInitComp(6) fprintf('\t228: %.3e \n', Pu240DecayIsotopes(4)); end
%fprintf('Radon:\n');
%if UPuInitComp(2) || UPuInitComp(4) fprintf('\t222: %.3e \n', U234DecayIsotopes(4) + U238DecayIsotopes(7)); end
%fprintf('Polonium:\n');
%if UPuInitComp(2) || UPuInitComp(4) fprintf('\t210: %.3e \n', U234DecayIsotopes(7) + U238DecayIsotopes(10)); end
%fprintf('Bismuth:\n');
%if UPuInitComp(2) || UPuInitComp(4) fprintf(cardID, '83209.66c %.5E &n', U234DecayIsotopes(8) + U238DecayIsotopes(11) + Pu241DecayIsotopes(9)); end
%if UPuInitComp(2) || UPuInitComp(4) fprintf('\t210: %.3e \n', U234DecayIsotopes(6) + U238DecayIsotopes(9)); end
%fprintf('Lead:\n');
%if UPuInitComp(2) || UPuInitComp(4) fprintf(cardID, '82206.66c %.5E ', U234DecayIsotopes(9) + U238DecayIsotopes(12)); end
%if UPuInitComp(3) || UPuInitComp(5) fprintf(cardID, '82207.66c %.5E ', U235DecayIsotopes(7) + Pu239DecayIsotopes(7)); end
%if UPuInitComp(1) || UPuInitComp(6) fprintf(cardID, '82208.66c %.5E \n', U232DecayIsotopes(5) + Pu240DecayIsotopes(7)); end
%if UPuInitComp(2) || UPuInitComp(4) fprintf('\t210: %.3e \n', U234DecayIsotopes(5) + U238DecayIsotopes(8)); end
%if UPuInitComp(1) fprintf('\t212: %.3e \n', U232DecayIsotopes(4)); end
fclose(cardID);

The following script is called by totalComp.m and computes the output composition of the decay of uranium-232. It uses U232Decay.m to calculate the change in composition for each time step.

function fN=U232run(t);

% Initialize initial conditions vector
numNuclides = 10;
N0=zeros(numNuclides, 1);

% Set initial amount of material N(i) (number of nuclei)
N0(1) = 10^23;

decayTime = t; %50*365*24*60*60; % seconds
%decayTime = decayTime/lelO; % Introduce shift
numTimePointsRecorded = 50;
The following script calculates the change in each time step for each isotope in the decay chain of uranium-232.

```matlab
%results = ode45(@U235Decay, [0:decayTime/numTimePointsRecorded:decayTime], N0);
% options = odeset('AbsTol', 10^-24);
[T,N] = ode45(@U232Decay, [0,decayTime], N0);
finalNumIsotopes = N(size(N,1),:);
fNtemp = finalNumIsotopes./N0(1);
fN(1)=fNtemp(1); % U-232
fN(2)=fNtemp(2); % Th-228
fN(3)=fNtemp(3); % Ra-224
fN(4)=fNtemp(6); % Pb-212
fN(5)=fNtemp(10); % Pb-208

% T = T*1e10;
% subplot(2,3,1), plot(T,N(:,1)), title('U-232');
% subplot(2,3,2), plot(T,N(:,2)), title('Th-238');
% subplot(2,3,3), plot(T,N(:,3)), title('Ra-224');
% subplot(2,3,4), plot(T,N(:,6)), title('Pb-212');
% subplot(2,3,5), plot(T,N(:,10)), title('Pb-208');

%for i=1:numNuclides-1
% subplot(4, 4, i), plot(T, N(:, i+1))
%end

The following script calculates the change in each time step for each isotope in the decay chain of uranium-232.

function dNdt=U232Decay(t, N);
% U232Decay returns the differential equations for use with ode45.

% Nuclides
% 1 = U-232
% 2 = Th-228
% 3 = Ra-224
% 4 = Rn-220
% 5 = Po-216
% 6 = Pb-212
% 7 = Bi-212
% 8 = Po-212
% 9 = Tl-208
% 10 = Pb-208
numNuclides = 10;

% Nuclide Half Lives (seconds)
halfLife = zeros(numNuclides,1); % Initialize half life vector
halfLife(1) = 2.1742722e9;
halfLife(2) = 6.03242197e7;
halfLife(3) = 3.16224e5;
halfLife(4) = 55.6;
halfLife(5) = .145;
halfLife(6) = 3.83e4;
```
halfLife(7) = 3.633e3;
halfLife(8) = .299e-6;
halfLife(9) = 183.2;
halfLife(10) = 1/eps;

% Convert to years
% halfLife = halfLife .* 3.1687646e-8;

% Nuclide Decay Constants (1/second)
lambda = zeros(numNuclides,1); % Initialize
lambda = log(2)./halfLife;

% Nuclide decay equations (Remove all species with half life < 10^4 seconds)
dNdt=zeros(numNuclides,1); % Initialize differential equations vector
dNdt(1)=-lambda(1)*N(1);
dNdt(2)=lambda(1)*N(1)-lambda(2)*N(2);
dNdt(3)=lambda(2)*N(2)-lambda(3)*N(3);
dNdt(4)=0; % lambda(3)*N(3)-lambda(4)*N(4);
dNdt(5)=0; % .9862*lambda(4)*N(4)-lambda(5)*N(5);
dNdt(6)= lambda(3)*N(3)-lambda(6)*N(6); % .0138*lambda(4)*N(4)-lambda(6)*N(6);
dNdt(7)= 0; % (1-6e-5)*.0138)*lambda(4)*N(4)+lambda(5)*N(5)-lambda(7)*N(7);
dNdt(8)= 0; % 6e-5*lambda(6)*N(6)-lambda(8)*N(8);
dNdt(9)= 0; % lambda(7)*N(7)+.03*lambda(8)*N(8)-lambda(9)*N(9);
dNdt(10)=lambda(6)*N(6); % lambda(6)*N(6)+lambda(15)*N(15)+lambda(16)*N(16);

% Set N to zero if dNdt < -N
for i=1:numNuclides,
    if -dNdt(i) > N(i)
        dNdt(i) = -N(i);
    end
end

% Introduce time
%dNdt = dNdt*lel0;

The following script is called by totalComp.m and computes the output composition of the decay of uranium-234. It uses U238Decay.m to calculate the change in composition for each time step.

function fN=U234run(t);

% Initialize initial conditions vector
numNuclides = 13;
N0=zeros(numNuclides, 1);

% Set initial amount of material N(i) (number of nuclei)
N0(4) = 10^23;
decayTime = t;%*365*24*60*60; % seconds
%decayTime = decayTime/lel0; % Introduce shift
numTimePointsRecorded = 50;
%results = ode45(@U235Decay, [0:decayTime/numTimePointsRecorded:decayTime], NO);
% options = odeset('AbsTol', 10^24);
[T N] = ode45(@U238Decay, [0, decayTime], NO);
finalNumIsotopes = N(size(N,1),:);
fNtemp = finalNumIsotopes./NO(4);
fN(1)=fNtemp(4); % U-234
fN(2)=fNtemp(5); % Th-230
fN(3)=fNtemp(6); % Ra-226
fN(4)=fNtemp(7); % Rn-222
fN(5)=fNtemp(8); % Pb-210
fN(6)=fNtemp(9); % Bi-210
fN(7)=fNtemp(10); % Po-210
fN(8)=fNtemp(12); % Bi-209
fN(9)=fNtemp(13); % Pb-206

T = T*1e10;
% subplot(3,4,1), plot(T,N(:,2)), title('Th-234');
% subplot(3,4,2), plot(T,N(:,3)), title('Pa-234');
% subplot(3,4,3), plot(T,N(:,4)), title('U-234');
% subplot(3,4,4), plot(T,N(:,5)), title('Th-230');
% subplot(3,4,5), plot(T,N(:,6)), title('Ra-226');
% subplot(3,4,6), plot(T,N(:,7)), title('Rn-222');
% subplot(3,4,7), plot(T,N(:,8)), title('Pb-210');
% subplot(3,4,8), plot(T,N(:,9)), title('Bi-210');
% subplot(3,4,9), plot(T,N(:,10)), title('Po-210');
% subplot(3,4,10), plot(T,N(:,11)), title('Pb-209');
% subplot(3,4,11), plot(T,N(:,12)), title('Bi-209');
% subplot(3,4,12), plot(T,N(:,13)), title('Pb-206');

for i=1:numNuclides-1
% subplot(4, 4, i), plot(T, N(:, i+1))
end

The following script is called by totalComp.m and computes the output composition of the decay of uranium-235. It uses U235Decay.m to calculate the change in composition for each time step.

function fN=U235run(t);
% returns final fraction of initial nuclei of each isotope

% Initialize initial conditions vector
numNuclides = 18;
NO=zeros(numNuclides, 1);

% Set initial amount of material N(i) (number of nuclei)
NO(1) = 10^23; % U-235
NO(18) = 0; % Pu-239
NO(1) = 1e16; % 10 kg U-235

decayTime = t; % 365*24*60*60; % 100*365*24*60*60; % seconds
The following script calculates the change in each time step for each isotope in the decay chain of uranium-235.

function dNdt=U235Decay(t, N);
% U235Decay returns the differential equations for use with ode45.
% 15 = Po-211
% 16 = Tl-207
% 17 = Pb-207

numNuclides = 18;

% Nuclide Half Lives (seconds)
halfLife = zeros(numNuclides,1); % Initialize half life vector
halfLife(18) = 7.60837485e11;
halfLife(1) = 2.221e16;
halfLife(2) = 9.187e4;
halfLife(3) = 1.034e12;
halfLife(4) = 6.871e8;
halfLife(5) = 1.617e6;
halfLife(6) = 1320;
halfLife(7) = 9.85e5;
halfLife(8) = 56;
halfLife(9) = 3.96;
halfLife(10) = 456;
halfLife(11) = 1.78e-3;
halfLife(12) = .10e-3;
halfLife(13) = 2166;
halfLife(14) = 128.4;
halfLife(15) = .516;
halfLife(16) = 286.2;
halfLife(17) = 1/eps;

% Convert to years
% halfLife = halfLife .* 3.16887646e-8;

% Nuclide Decay Constants (1/second)
lambda = zeros(numNuclides,1); % Initialize
lambda = log(2)./halfLife;

% Nuclide decay equations (Remove all species with half life < 10^4 seconds)
dNdt=zeros(numNuclides,1); % Initialize differential equations vector
dNdt(18)=-lambda(18)*N(18);
dNdt(1)=lambda(18)*N(18)-lambda(1)*N(1);
dNdt(2)=lambda(1)*N(1)-lambda(2)*N(2);
dNdt(3)=lambda(2)*N(2)-lambda(3)*N(3);
dNdt(4)=lambda(3)*N(3)-lambda(4)*N(4);
dNdt(5)=.9862*lambda(4)*N(4)-lambda(5)*N(5);
dNdt(6)= 0; % .0138*lambda(4)*N(4)-lambda(6)*N(6);
dNdt(7)= (1-6e-5)*.0138*lambda(4)*N(4)+lambda(5)*N(5)-lambda(7)*N(7);
dNdt(8)= 0; % 6e-5*lambda(6)*N(6)-lambda(8)*N(8);
dNdt(9)= 0; % lambda(7)*N(7)+.03*lambda(8)*N(8)-lambda(9)*N(9);
dNdt(10)= 0; % .97*lambda(8)*N(8)-lambda(10)*N(10);
dNdt(11)= 0; % lambda(10)*N(10)+lambda(9)*N(9)-lambda(11)*N(11);
dNdt(12)= 0; % 2.3e-6*lambda(11)*N(11)-lambda(12)*N(12);
dNdt(13)= 0; % (1-2.3e-6)*lambda(11)*N(11)-lambda(13)*N(13);
dNdt(14)= 0; % lambda(12)*N(12)-lambda(14)*N(14);
dNdt(15)= 0; % .00275*lambda(14)*N(14)-lambda(15)*N(15);
dNdt(16)= 0; % .99725*lambda(14)*N(14)-lambda(16)*N(16);
dNdt(17)=lambda(7)*N(7)+.0138*lambda(4)*N(4); lambda(15)*N(15)+lambda(16)*N(16);

% Set N to zero if dNdt < -N
for i=1:numNuclides,
    if -dNdt(i) > N(i)
        dNdt(i) = -N(i);
    end
end

% Introduce time
%dNdt = dNdt*1e10;

The following script is called by totalComp.m and computes the output composition of the decay of uranium-238. It uses U238Decay.m to calculate the change in composition for each time step.

function fN=U238run(t);

% Initialize initial conditions vector
numNuclides = 13;
N0=zeros(numNuclides, 1);

% Set initial amount of material N(i) (number of nuclei)
N0(1) = 10^23;

decayTime = t;%*365*24*60*60; % seconds
%decayTime = decayTime/le10; % Introduce shift
numTimePointsRecorded = 50;
%results = ode45(@U235Decay, [0:decayTime/numTimePointsRecorded:decayTime], N0);
% options = odeset('AbsTol', 10^24);
[T N] = ode45(@U238Decay, [0,decayTime], N0);
finalNumIsotopes = N(size(N,1),:);

fNtemp = finalNumIsotopes./N0(1);
fN(1)=fNtemp(1); % U-238
fN(2)=fNtemp(2); % Th-234
fN(3)=fNtemp(3); % Pa-234
fN(4)=fNtemp(4); % U-234
fN(5)=fNtemp(5); % Th-230
fN(6)=fNtemp(6); % Ra-226
fN(7)=fNtemp(7); % Rn-222
fN(8)=fNtemp(8); % Pb-210
fN(9)=fNtemp(9); % Bi-210
fN(10)=fNtemp(10); % Po-210
fN(11)=fNtemp(12); % Bi-209
fN(12)=fNtemp(13); % Pb-206

%T = T*1e10;
% subplot(3,4,1), plot(T,N(:,2)), title('Th-234');
% subplot(3,4,2), plot(T,N(:,3)), title('Pa-234');
% subplot(3,4,3), plot(T,N(:,4)), title('U-234');
% subplot(3,4,4), plot(T,N(:,5)), title('Th-230');
% subplot(3,4,5), plot(T,N(:,6)), title('Ra-226');
% subplot(3,4,6), plot(T,N(:,7)), title('Rn-222');
% subplot(3,4,7), plot(T,N(:,8)), title('Pb-210');
% subplot(3,4,8), plot(T,N(:,9)), title('Bi-210');
% subplot(3,4,9), plot(T,N(:,10)), title('Po-210');
The following script calculates the change in each time step for each isotope in the decay chain of uranium-238.

```matlab
function dNdt=U238Decay(t, N);
% U238Decay returns the differential equations for use with ode45.

% Nuclides
% 1 = U-238
% 2 = Th-234
% 3 = Pa-234
% 4 = U-234
% 5 = Th-230
% 6 = Ra-226
% 7 = Rn-222
% 8 = Pb-210
% 9 = Bi-210
% 10 = Po-210
% 11 = Pb-209
% 12 = Bi-209
% 13 = Pb-206

numNuclides = 13;

% Nuclide Half Lives (seconds)
halfLife = zeros(numNuclides,1); % Initialize half life vector
halfLife(1) = 1.40996345e17;
halfLife(2) = 2.082240e6;
halfLife(3) = 2.4120e4;
halfLife(4) = 7.74722333e12;
halfLife(5) = 2.37876108e12;
halfLife(6) = 5.04910816e10;
halfLife(7) = 3.303504e5;
halfLife(8) = 7.03719449e8;
halfLife(9) = 4.331232e5;
halfLife(10) = 1.19556864e7;
halfLife(11) = 11710.8;
halfLife(12) = 6.31138519e26;
halfLife(13) = 1/eps;

% Nuclide Decay Constants (1/second)
lambda = zeros(numNuclides,1); % Initialize
lambda = log(2)./halfLife;

% Nuclide decay equations (Remove all species with half life < 10^4 seconds)
dNdt=zeros(numNuclides,1); % Initialize differential equations vector
```
\[
dNdt(1) = -\lambda(1)*N(1);
dNdt(2) = \lambda(1)*N(1) - \lambda(2)*N(2);
dNdt(3) = \lambda(2)*N(2) - \lambda(3)*N(3);
dNdt(4) = \lambda(3)*N(3) - \lambda(4)*N(4);
dNdt(5) = \lambda(4)*N(4) - \lambda(5)*N(5);
dNdt(6) = \lambda(5)*N(5) - \lambda(6)*N(6);
dNdt(7) = \lambda(6)*N(6) - \lambda(7)*N(7);
dNdt(8) = (1-1.474e-8)*\lambda(7)*N(7) - \lambda(8)*N(8);
dNdt(9) = \lambda(8)*N(8) - \lambda(9)*N(9);
dNdt(10) = \lambda(9)*N(9) - \lambda(10)*N(10);
dNdt(11) = 0; \quad dNdt(11) = 1.407e-8*\lambda(7)*N(7) - \lambda(11)*N(11);
dNdt(12) = 1.407e-8*\lambda(7)*N(7);
dNdt(13) = \lambda(10)*N(10);
\]

% Set \( N \) to zero if \( dNdt < -N \)
for \( i=1:\text{numNuclides} \)
  if \( -dNdt(i) > N(i) \)
    \( dNdt(i) = -N(i); \)
  end
end

% Introduce time
\( dNdt = dNdt*1\text{e}10; \)

The following script is called by totalComp.m and computes the output composition of the decay of plutonium-239. It uses Pu239Decay.m to calculate the change in composition for each time step.

function \( fN=Pu239run(t); \)

% returns final fraction of initial nuclei of each isotope

% Initialize initial conditions vector
numNuclides = 18;
N0=zeros(numNuclides, 1);

% Set initial amount of material \( N(i) \) (number of nuclei)
N0(18) = 10^23; % Pu-239
N0(1) = 0; % U-235
N0(1) = 1e16; % 10 kg U-235

decayTime = t; %*365*24*60*60; % seconds
decayTime = decayTime/1e10; % Introduce shift
numTimePointsRecorded = 50;
results = ode45(@U235Decay,
[0:decayTime/numTimePointsRecorded:decayTime], N0);
% options = odeset('AbsTol', 10^-24);
[T N] = ode45(@U235DecayReduced, [0,decayTime], N0);
finalNumIsotopes = N(size(N,1),:);
fNtemp = finalNumIsotopes./N0(18);
fN(1)=fNtemp(1); % U-235
fN(2)=fNtemp(2); % Th-231
fN(3)=fNtemp(3); % Pa-231

95
function dNdt=Pu239Decay(t, N);

% Pu239Decay returns the differential equations for use with ode45.

% Nuclides
% 1 = U-238
% 2 = Th-234
% 3 = Pa-234
% 4 = U-234
% 5 = Th-230
% 6 = Ra-226
% 7 = Rn-222
% 8 = Pb-210
% 9 = Bi-210
% 10 = Po-210
% 11 = Pb-209
% 12 = Bi-209
% 13 = Pb-206

numNuclides = 13;

% Nuclide Half Lives (seconds)
halfLife = zeros(numNuclides,1); % Initialize half life vector
halfLife(1) = 1.40996345e17;
halfLife(2) = 2.082240e6;
halfLife(3) = 2.4120e4;
halfLife(4) = 7.74722533e12;
halfLife(5) = 2.37876108e12;
halfLife(6) = 5.04910816e10;
halfLife(7) = 3.303504e5;
halfLife(8) = 7.03719449e8;
halfLife(9) = 4.331232e5;
halfLife(10) = 1.19556864e7;
halfLife(11) = 11710.8;
halfLife(12) = 6.31138519e26;
halfLife(13) = 1/eps;

% Nuclide Decay Constants (1/second)
lambda = zeros(numNuclides,1); % Initialize
lambda = log(2)./halfLife;

% Nuclide decay equations (Remove all species with half life < 10^4
seconds
dNdt=zeros(numNuclides,1); % Initialize differential equations vector

% Introduce time
%dNdt = dNdt*lel0;

function fN=Pu240run(t);
% Initialize initial conditions vector
numNuclides = 7;
N0=zeros(numNuclides, 1);

% Set initial amount of material N(i) (number of nuclei)
N0(1) = 10^23;
decayTime = t;%*365*24*60*60; % seconds
%decayTime = decayTime/lel0; % Introduce shift
numTimePointsRecorded = 50;

The following script is called by totalComp.m and computes the output
composition of the decay of plutonium-240. It uses Pu240Decay.m to calculate the
change in composition for each time step.
The following script calculates the change in each time step for each isotope in the decay chain of plutonium-240.

```matlab
function dNdt=Pu240Decay(t, N);
% Pu240Decay returns the differential equations for use with ode45.

% Nuclides
% 1 = Pu-240
% 2 = U-236
% 3 = Th-232
% 4 = Ra-228
% 5 = Th-228
% 6 = Ra-224
% 7 = Pb-208
numNuclides = 7;

% Nuclide Half Lives (seconds)
halfLife = zeros(numNuclides,1); % Initialize half life vector
halfLife(1) = 2.07139662e11;
halfLife(2) = 7.39063206e14;
halfLife(3) = 4.4337481e17;
halfLife(4) = 181452324;
halfLife(5) = 60324219.7;
halfLife(6) = 316224;
halfLife(7) = 1/eps;

% Nuclide Decay Constants (1/second)
lambda = zeros(numNuclides,1); % Initialize
lambda = log(2)./halfLife;

% Nuclide decay equations (Remove all species with half life < 10^4 seconds
```
% Initialize differential equations vector
dNdt=zeros(numNuclides,1);

% Initialize initial conditions vector
numNuclides = 10;
N0=zeros(numNuclides, 1);

% Set initial amount of material N(i) (number of nuclei)
N0(1) = 10^23;
decayTime = t;%*365*24*60*60; % seconds
decayTime = decayTime/le10; % Introduce shift
numTimePointsRecorded = 50;
T = decayTime/numTimePointsRecorded:
results = ode45(@(U235Decay, [0:decayTime/numTimePointsRecorded:decayTime], N0);
% options = odeset('AbsTol', '10^-24);
[T N] = ode45(@(Pu241Decay, [0,decayTime], N0);
finalNumIsotopes = N(size(N,1),:);
fNtemp = finalNumIsotopes./N0(1);
fN(1)=fNtemp(1); % Pu-241
fN(2)=fNtemp(2); % Am-241
fN(3)=fNtemp(4); % Np-237
fN(4)=fNtemp(5); % Pa-233
fN(5)=fNtemp(6); % U-233
fN(6)=fNtemp(7); % Th-229
fN(7)=fNtemp(8); % Ra-225
fN(8)=fNtemp(9); % Ac-225
fN(9)=fNtemp(10); % Bi-209

function fN=Pu241run(t);

% Set initial amount of material N(i) (number of nuclei)
N0(1) = 10^23;
decayTime = t;%*365*24*60*60; % seconds
decayTime = decayTime/le10; % Introduce shift
numTimePointsRecorded = 50;
T = decayTime/numTimePointsRecorded:
results = ode45(@(U235Decay, [0:decayTime/numTimePointsRecorded:decayTime], N0);
% options = odeset('AbsTol', '10^-24);
[T N] = ode45(@(Pu241Decay, [0,decayTime], N0);
finalNumIsotopes = N(size(N,1),:);
fNtemp = finalNumIsotopes./N0(1);
fN(1)=fNtemp(1); % Pu-241
fN(2)=fNtemp(2); % Am-241
fN(3)=fNtemp(4); % Np-237
fN(4)=fNtemp(5); % Pa-233
fN(5)=fNtemp(6); % U-233
fN(6)=fNtemp(7); % Th-229
fN(7)=fNtemp(8); % Ra-225
fN(8)=fNtemp(9); % Ac-225
fN(9)=fNtemp(10); % Bi-209

%T = T*le10;
% subplot(3,4,1), plot(T,N(:,2)), title('Am-241');
% subplot(3,4,2), plot(T,N(:,3)), title('U-237');
% subplot(3,4,3), plot(T,N(:,4)), title('Np-237');
The following script calculates the change in each time step for each isotope in the decay chain of plutonium-241.

```matlab
function dNdt=Pu241Decay(t, N);

% Pu241Decay returns the differential equations for use with ode45.

% Nuclides
% 1 = Pu-241
% 2 = Am-241
% 3 = U-237
% 4 = Np-237
% 5 = Pa-233
% 6 = U-233
% 7 = Th-229
% 8 = Ra-225
% 9 = Ac-225
% 10 = Bi-209

numNuclides = 10;

% Nuclide Half Lives (seconds)
halfLife = zeros(numNuclides,1); % Initialize half life vector
halfLife(1) = 452841888.;
halfLife(2) = 1.36389034e10;
halfLife(3) = 583200.;
halfLife(4) = 6.76580493e13;
halfLife(5) = 2329948.8;
halfLife(6) = 5.02386262e12;
halfLife(7) = 2.31627837e11;
halfLife(8) = 1287360.;
halfLife(9) = 864000.;
halfLife(10) = 1/eps;

% Nuclide Decay Constants (1/second)
lambda = zeros(numNuclides,1); % Initialize
lambda = log(2)./halfLife;

% Nuclide decay equations (Remove all species with half life < 10^4 seconds)
dNdt=zeros(numNuclides,1); % Initialize differential equations vector
dNdt(1)=-lambda(1)*N(1);
dNdt(2)=0.9999761*lambda(1)*N(1)-lambda(2)*N(2);
dNdt(3)=2.39e-5*lambda(2)*N(2)-lambda(3)*N(3);
dNdt(4)=2.39e-5*lambda(1)*N(1)+lambda(2)*N(2)-lambda(4)*N(4);
dNdt(5)=lambda(4)*N(4)-lambda(5)*N(5);
dNdt(6)=lambda(5)*N(5)-lambda(6)*N(6);
dNdt(7)=lambda(6)*N(6)-lambda(7)*N(7);
```
dNdt(8) = lambda(7) * N(7) - lambda(8) * N(8);
dNdt(9) = lambda(8) * N(8) - lambda(9) * N(9);
dNdt(10) = lambda(9) * N(9);

% Set N to zero if dNdt < -N
for i = 1:numNuclides,
    if -dNdt(i) > N(i)
        dNdt(i) = -N(i);
    end
end