A Gamma-Compensated Helium-4 Ion Chamber for Spent Nuclear Fuel Monitoring

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Oskar Searfus[1], Jeremy Osborn[2], and Igor Jovanovic[1]

[1] *Department of Nuclear Engineering and Radiological Sciences, University of Michigan, Ann Arbor MI 48109 USA*
[2] *Department of Proliferation and Weaponization Detection, Sandia National Laboratories, Albuquerque NM 87123 USA*

Abstract

Management of the spent nuclear fuel (SNF) is an intriguing problem from a safeguards instrumentation perspective: for safety reasons, it must be packed with a large amount of shielding to prevent radiation exposure to facility workers and the public, and with neutron-absorbing materials to prevent criticality. SNF is typically also at high temperature in dry storage and during transportation, precluding the use of many types of radiation sensors internal to a fuel cask. The radiation environment of SNF casks includes intense gamma radiation originating mostly from fission fragments, and a smaller but still sizable amount of fast neutron radiation, both from spontaneous fission of transuranic isotopes and (alpha,n) reactions with oxygen in the fuel matrix. Fast neutrons emitted by SNF are of particular interest for safeguards, as plutonium is the most significant contributor to the neutron source term. A gamma-"compensated" ^{4}He ion chamber would in some ways be an ideal instrument for use in SNF safeguards: ^{4}He has a high cross-section for fast neutron elastic scattering, it does not contain any materials that are consumed or transmuted, and it is a simple gas-based detector that is resilient to high temperature and radiation damage. The detector must be primarily sensitive to fast neutrons, as the thermal neutron population in a cask is depressed by neutron-absorbing materials. The gamma-ray contribution to the chamber current can be compensated by sheathing two identical tubes in different thicknesses of tungsten, a strong attenuator of gamma rays. In this way, the fractional difference in current between the two tubes is directly proportional to the fractional contribution of gamma rays to the signal in the thinner tube, which can then be subtracted to yield the fast neutron-only signal. In this work, Monte Carlo simulations are used to predict the performance of a ^{4}He-based detector system deployed at a realistic SNF cask. The sensitivity to a fuel diversion scenario is evaluated, and the gamma-compensation relationship is validated across the lifetime of a typical SNF cask.

1 Introduction

A top-priority R&D need for the IAEA is to "develop safeguards equipment to establish and maintain knowledge of spent fuel in shielding/storage/transport containers at all points in their life cycle." Fast neutrons emitted by spent nuclear fuel (SNF) are of particular interest for safeguards, as plutonium is the most significant contributor to the neutron source term. Potential detectors for SNF safeguards must be primarily sensitive to fast neutrons, as thermal neutron absorbing materials are added to SNF casks to prevent criticality. Furthermore, it is desirable for a potential detector to be as close to the neutron source as possible without compromising the safety features of the cask: an optimal location is on top of the inner steel canister, but inside the neutron-shielding concrete overpack.

^{4}He has a small but significant niche as a fast neutron detector, relying exclusively on neutron elastic scatter [1]. While ^{4}He has a zero neutron absorption cross-section, it has a significant neutron scattering cross-section in the fission neutron spectrum energy range. Up to 64% of the incident neutron energy can be transferred to the ^{4}He nucleus in a single scatter. In common detector designs, the ^{4}He is either used as a proportional gas at low pressure or as a gas scintillator at high pressure, the former having better energy resolution and lower efficiency, the latter having poorer energy resolution and higher efficiency [2]. In pulse-mode operation, these detectors exhibit excellent γ-ray discrimination, as γ-induced fast electrons produce low-amplitude pulses in the low-density gas when compared to the short-ranged helium recoil nuclei [1].

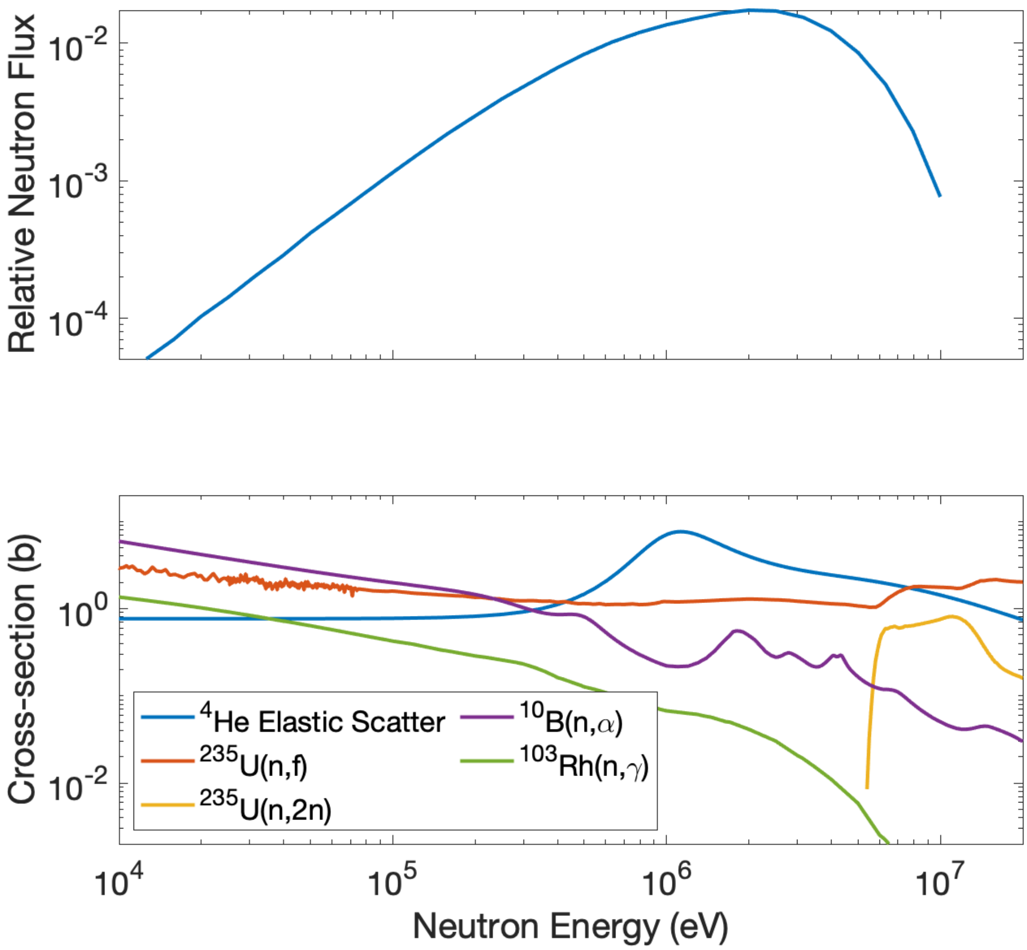


Figure 1: Typical fission neutron energy spectrum (above). Cross-sections of several reactions of interest for fast neutron detection (below). [3]

In comparison to fission chambers and boron-lined tubes, two other detector types of interest for high radiation field environments, ^{4}He-based detectors hold several significant advantages. In ^{4}He detectors, the fill gas is also the neutron sensitive component, making their fabrication much less complex than the lined counterparts. Boron and uranium linings are vulnerable to physical and electrical degradation in high temperature and radiation environments over long periods of time [4], so the lack of any neutron-converting lining is a major benefit of ^{4}He detectors in this application. Since the ^{4}He detectors are based on neutron elastic scattering rather than absorption, the detector material is not depleted as in boron and uranium-lined tubes. Consequently, a ^{4}He detector should have a constant sensitivity to neutrons regardless of the irradiation time and, as discussed previously, gamma-ray contributions are a small fraction of the total signal.

The operating principle of the proposed ^{4}He compensated ion chamber neutron detector is as follows: two identical cylindrical ^{4}He ion chambers are sheathed in tungsten tubes of different thickness. Tungsten, which strongly attenuates gamma rays, decreases the contribution of gamma rays to the ionization current in the detector by a fraction approximately determined by the tungsten thickness. In this way, the fractional difference in current between the two tubes is directly proportional to the fractional contribution of gamma rays to the signal in the thicker tube, which can then be subtracted to give the fast neutron-only signal. The constituent materials necessary to fabricate the detectors are widely available and inexpensive, allowing for widespread application at SNF storage sites worldwide. This detector would be challenging to spoof in the case of SNF diversion, as the SNF would have to be replaced by a fast neutron source that produces neutron flux of the same order as SNF at the location of detector. Such detectors could significantly improve the technical safeguards capabilities for SNF: unlike the technologies proposed to date, it could be deployed indefinitely in the interior of a SNF cask, addressing the need to monitor the growing quantity of SNF in storage worldwide.

The procedure for determining the contribution of gamma rays and neutrons to the tube signal is straightforward. If the response of each tube to pure sources of gamma rays and neutrons is known, as in simulation, the difference in tube current at these boundary conditions can be linearly interpolated to produce a function which relates fractional current difference in the two tubes to the gamma-ray contribution to the signal in either tube:

$$I_\gamma = \frac{\Delta I_\gamma - \Delta I_n}{\Delta I}, \tag{1}$$

where I_γ is the fractional contribution of gamma rays to the thin tube current, and

$$\Delta I = \frac{I_{thin} - I_{thick}}{I_{thin}}, \tag{2}$$

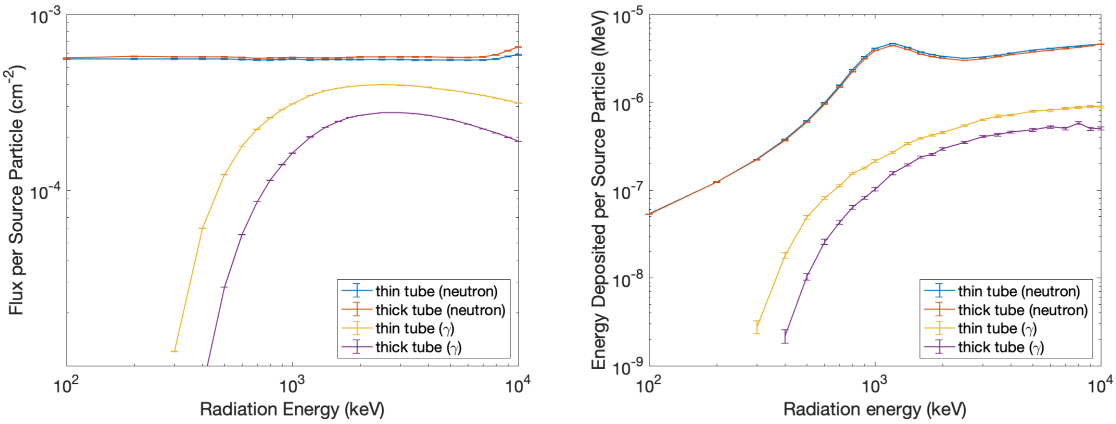
where I_{thin} and I_{thick} are the currents in the thin and thick tubes, respectively,and ΔI_γ and ΔI_n are the values of ΔI when the tubes are exposed to pure gamma and neutron field, respectively. This linear interpolation is possible, as the neutron and gamma-ray generation of signal is independent, *i.e.* the generation of signal by a gamma ray is independent of the generation of signal by a neutron. Consequently, the signal contribution of gamma rays and neutrons are in linear superposition. The contribution of gamma rays to the signal in the thick tube can be calculated in a similar way.

2 Detector Prototype Design

The proposed detector prototype is a system comprised of two identical stainless steel tubes, each with a length of 12" and 1" ID. These tubes are filled with 1 atm ^{4}He and operated as ion chambers. The "thin" tube is encased with a 90% tungsten-copper alloy (WCu) tube of 1/4" wall thickness, while the "thick" tube is encased with a WCu tube of 1/2" wall thickness. WCu is used in place of pure W, as it is more machinable and economical, while sacrificing little of tungsten's gamma-ray shielding properties. The two tubes are operated with a center-to-center distance of 20 cm, and must be used only in applications where both tubes are exposed to the same radiation flux at their surfaces.

3 Simulation Results

The described detector system was simulated using MCNP6.2 [5]. The response of the detector system to gamma rays and neutrons in the 100 keV to 10 MeV spectral region was generated, as shown in Figure 2. In this simulation, the center-to-center distance of the detectors was 20 cm, with the simulated source directly between the detector centers.



(a) (b)

Figure 2: (a) Simulated neutron and gamma-ray flux inside each tube as a function of source radiation energy. (b) Simulated neutron and gamma-ray energy deposition in the active volume as a function of source radiation energy.

The simulated detector system response shows several key findings. First, both tubes are clearly much more sensitive to neutrons than to gamma rays of equal energy at every simulated energy. Furthermore, there is little difference in the detector response to neutrons when comparing the thick and thin tubes, while there is a significant difference in detector response to gamma rays when making the same comparison, as desired. This confirms the hypothesis that any difference in current between the two tubes is solely due to the prevalance of gamma rays in the signal.

A SNF dry cask model was obtained and edited for simulating the detector response using MCNP6.2 [5]. The MCNP model was developed by Khudoleeva [6] and based on a 32-assembly HOLTEC HI-STORM 100S Version B dry cask [7]. The gamma and neutron source terms, also developed in the same work [6], were used here as the radiation source terms. Khudoleeva calculated the source terms by using ORIGEN-ARP to simulate fuel with an initial enrichment of 3.9% ^{235}U to a burnup of 45 GWd/MTU followed by 3 years decay time. This resulted in an 18-group gamma spectrum and 44-group neutron spectrum as well as a total source term per fuel assembly of 4.23×10^8 n/s and 2.04×10^{16} γ/s.

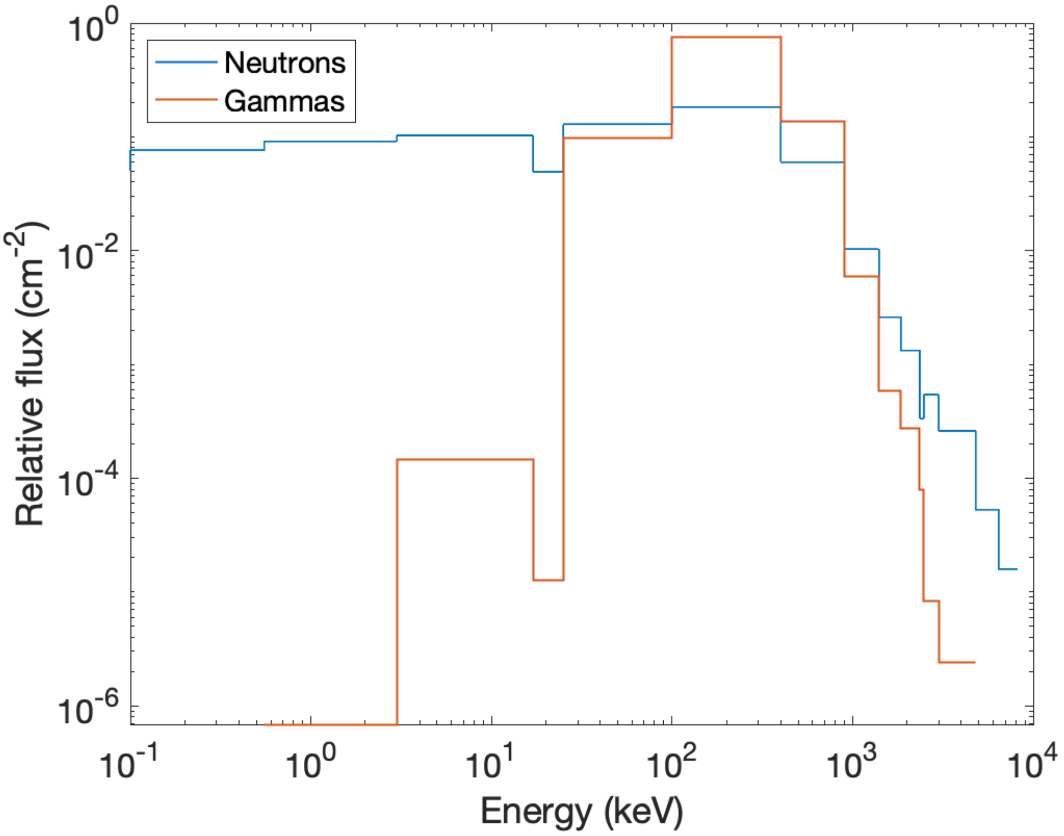
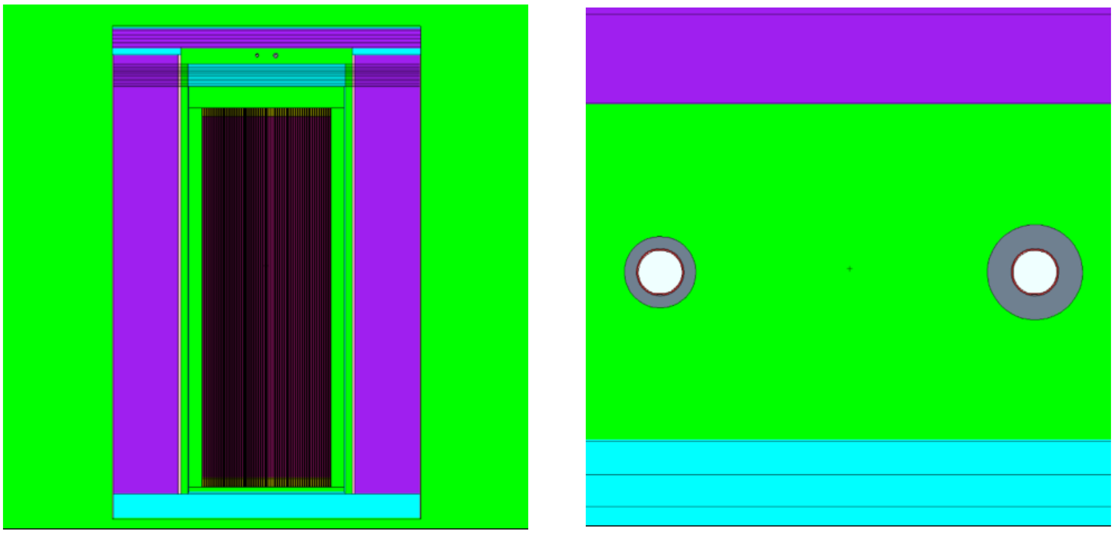


Figure 3: The simulated relative neutron and gamma-ray flux in the air gap of the SNF cask

For this work, the model was modified by populating the SNF material definition with spent fuel isotopic concentrations calculated by an MCNP6.2 simulation of a PWR 3-cycle irradiation, resulting in burnup of 45 GWd/MTU. Updating the isotopics of the SNF material improved the realism of the neutron transport simulations of the SNF cask, as many fission fragments depress the thermal neutron population, further depressing secondary fission neutrons. As a result of the updated fuel material, the criticality of the populated cask was calculated to be $k_{eff} = 0.24880 \pm 0.00032$. The 32-assembly cask was modeled with a homogeneous loading pattern. The ^{4}He detector prototype design, as seen in Figure 4, was modeled within the cask in the air region above the multi-purpose canister (MPC) and below the concrete cask lid. Figure 3 displays the neutron and gamma-energy spectra at the detector location.



(a) (b)

Figure 4: (a) MCNP-generated image of the SNF cask with the ^{4}He detector prototype. (b) Magnified view on the ^{4}He detector prototype modeled in the air gap between the MPC and concrete cask lid.

Table 1 presents the simulated response of the detector system to the SNF neutron and gamma-ray source. This simulation shows a fractional difference in current between the two tubes of 40.8%. The fractional difference of current due to neutrons alone is 10.1%, and that due to gammas alone is 45.6%. As a result, using Equation 1, the contribution of gamma rays to the current in the thin tube is 87%.

Table 1: Gamma and neutron flux and energy deposition in each tube's active volume for the simulated SNF cask.

Particle Flux (cm^{-2}s^{-1})	**Thin Tube**	**Thick Tube**
Neutrons	$5.67\times10^2 \pm 1.02$	$5.34\times10^2 \pm 0.96$
Gammas	$2.73\times10^5 \pm 9.0 \times 10^2$	$1.28\times10^5 \pm 5.8 \times 10^2$
Energy Deposition (MeVs^{-1})		
Neutrons	$2.86\times10^1 \pm 8.6 \times 10^{-2}$	$2.57\times10^1 \pm 7.96 \times 10^{-2}$
Gammas	$1.95\times10^2 \pm 9.0$	$1.06\times10^2 \pm 9.4$
Total	$2.23\times10^2 \pm 9$	$1.32\times10^2 \pm 9$

4 Conclusions and Further Work

This work represents the first step in assessing the feasibility of a ^{4}He-based ion chamber for SNF monitoring. Based on simulations, it appears that monitoring the fast neutron radiation levels inside a SNF cask with this proposed detector is feasible, as the detector has a characteristic fractional current difference between the two tubes for the simulated cask. Despite gamma rays being eight orders of magnitude more abundant than neutrons in this simulated SNF cask, fast neutrons accounted for 13% of the total signal in the detector system, a fraction which can be numerically determined by the difference in current between the two tubes.

The next step in simulation is to use Geant4 to more accurately model the transport of electrons generated by gamma rays in the active volume of the gas as well as the inner wall of the detector. Furthermore, realistic diversion and spoofing scenarios will be modeled to determine important detectability limits of this system.

Fabrication of a detector system prototype is in the planning stage. When the prototype is fabricated, it should be exposed to SNF neutron and gamma-ray sources or realistic surrogates to validate and adjust relationships predicted by simulation.

5 Acknowledgements

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