

Spent Nuclear Fuel Cask and Storage Monitoring with ^4He Scintillation Fast Neutron Detectors

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1. Introduction

South Korea currently has twenty-three operating nuclear power plants. To date, more than 10,000 metric tons of uranium (MTU) have been stored in on-site temporary spent fuel storage, and about 700 tons of spent nuclear fuel are added to this inventory each year. At this rate, with a total design capacity of about 14,000 MTU, the nation's entire temporary spent fuel storage system will be saturated by 2021 [1].

With this increasing quantity of spent nuclear fuel being stored at nuclear plants across S. Korea, the demand exists for building a long-term disposal facility. However, the Korean government first requires a detailed plan for the monitoring and certification of spent fuel.

Several techniques have been developed and applied for the purpose of spent fuel monitoring, including the digital Cerenkov viewing device (DCVD), spent fuel attribute tester (SFAT), and FORK detector [2]. Conventional gamma measurement methods, however, suffer from a lack of nuclear data and interfering background radiation.

To date, the primary method of neutron detection for spent fuel monitoring has been through the use of thermal neutron detectors such as ^3He and BF_3 proportional counters. These thermal neutron detectors typically have high counting efficiencies, but are hampered by the loss of neutron energy information. Bonner spheres – thermal neutron detectors, each surrounded by a different thickness of moderating material – are often proposed as a solution to this problem [3]. The energy resolution of the network, however, is directly related to the number of detectors used, and is therefore limited. Unfolding the neutron spectrum becomes extremely complicated.

In an attempt to overcome these difficulties, a new fast neutron measurement system is currently being developed at the University of Florida. This system is based on the ^4He scintillation detector invented by Arktis Radiation Detectors Ltd. These detectors are a relatively new technological development and take advantage of the high ^4He cross-section for elastic scattering at fast neutron energies, particularly the resonance around 1 MeV.

This novel ^4He scintillation neutron detector is characterized by its low electron density, leading to excellent gamma rejection. This detector also has a fast

response time on the order of nanoseconds and most importantly, preserves some neutron energy information since no moderator is required [4]. Additionally, these detectors rely on naturally abundant ^4He as the fill gas.

This study proposes a new technique using the neutron spectroscopy features of ^4He scintillation detectors to maintain accountability of spent fuel in storage. This research will support spent fuel safeguards and the detection of fissile material, in order to minimize the risk of nuclear proliferation and terrorism.

2. ^4He Scintillation Neutron Detectors

A ^4He scintillation detector has recently been developed by Arktis Radiation Detectors. The detector consists of a cylindrical high-pressure steel vessel, filled with 150 bar of ^4He gas, and two photomultiplier tubes (PMTs) at either end [4].

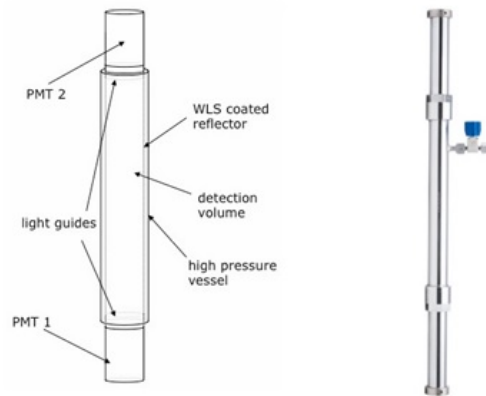


Fig. 1. ^4He scintillation detector schematic. The detection volume has a length of 20 cm and an inner radius of 2.2 cm, giving a total active volume of 304 cm^3 .

2.1 Neutron Interactions

A fast neutron that enters the detector volume and undergoes an elastic scattering interaction will transfer a portion of its kinetic energy to the ^4He nucleus. The neutron is not absorbed in this process, but rather continues to travel through the medium in a different direction and at a lower speed.

The transfer of energy to the ^4He nucleus results in the stripping of electrons, converting the atom into an alpha particle that is accelerated through the gas. This

recoil nucleus interacts with other helium atoms by excitation or ionization.

These excitations and ionizations will result in the production of excimer states [5]. These excimers decay to the ground state by the emission of photons. These photons are counted by the PMTs at either end of the detector's active volume, and are proportional to the energy of the neutron. An example of the resulting signal trace is shown in Figure 2.

2.2 Gamma Interactions

Unlike liquid scintillators, ^4He detectors are designed to be insensitive to gamma rays, and this is a significant advantage in many applications. This insensitivity is the result of several factors.

Contrary to neutrons, which interact with the nucleus of the atom, gamma rays interact with the atom's orbiting electrons. Therefore, helium's low atomic number ($Z=2$) means a very low electron density, which reduces the probability of gamma interaction.

Furthermore, if a gamma does interact, the resulting recoil electron will travel through the gas, losing energy by bremsstrahlung interactions with the surrounding nuclei, thereby slowing down and emitting this lost energy in the form of photons. Due to the low density of the gas even at the high tube pressures, the rate of energy deposition will be low, so the electron will travel farther while slowing down. It is more likely to hit a detector wall before depositing all of its energy.

The rejection of gamma events in the detector is further improved by pulse shape discrimination (PSD) algorithms. As illustrated by Figure 2, the slow component of a neutron event is significantly larger than that of a gamma event. The PSD algorithm analyzes the slow component of each event, and rejects those events below a certain threshold.

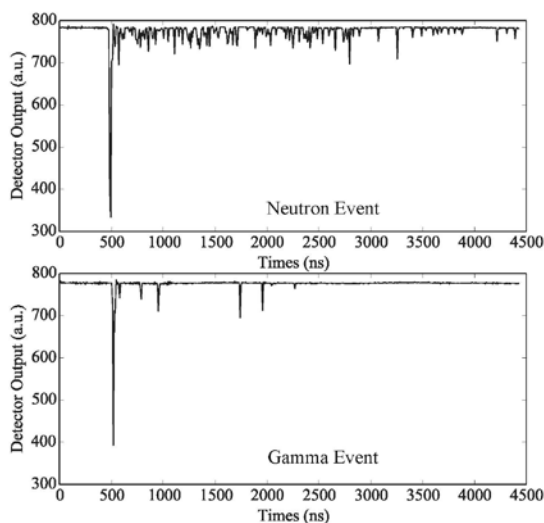


Fig. 2. Scintillation signals from (above) neutron and (below) gamma events. The fast components are comparable, but the neutron slow component is much stronger than the gamma slow component, which forms the basis for the pulse shape discrimination algorithm.

3. Monitoring System using ^4He Scintillation Neutron Detectors

3.1 Overall Review of Proposed Method

This proposed method for spent nuclear fuel cask and storage pool monitoring will mainly be carried out through the methodology described by Figure 3.

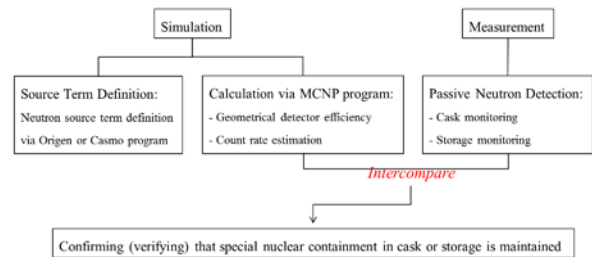


Fig. 3. Logical pathway of the proposed methods using ^4He scintillation detectors.

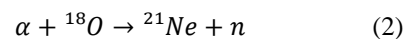
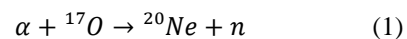
The operating data for initial fuel loading, discharge after burn-up, and storage time in the spent fuel pool or cask is obviously required and recorded. Based on the information, the current neutron source activity can be estimated via several simulation programs such as Origen or Casmo. This neutron source term will be used to predict the estimated count rates from a cask or storage pool at certain time and position via MCNP simulation.

These simulation results will be intercompared with the actual counts obtained through ^4He scintillation detectors placed at the same simulated positions.

3.2 Methodology of the Cask Monitoring System

There are two primary processes responsible for the production of neutrons in spent fuel; neutrons from a fission event and neutrons from an (α, n) reaction with oxygen. The first is from the spontaneous fission of ^{242}Cm and ^{244}Cm , which have half-lives of 162.8 days and 18.10 years, respectively.

The other principal source of neutron production in spent fuel is from (α, n) reactions with ^{17}O and ^{18}O , as shown in Equations 1 and 2, respectively.



The incident α particle in these reactions comes from the decay of uranium or plutonium, and will be born with an energy between 4 and 6 MeV. Because alpha particles will lose energy very quickly as they travel through the fuel medium, they will not travel far before losing all of their energy. Because of this short range, they will not normally reach materials eligible for a (α, n) reaction. But if oxygen is mixed with the fuel, as

in UO_2 and PuO_2 fuels, the alpha can reach these materials before losing all of its energy, and the reactions above become much more probable.

The (α, n) reaction with oxygen produces a continuous spectrum of neutron energies, with a distinct peak at 2.3 MeV that becomes more pronounced as burnup increases [6]. At energies greater than 3 MeV, the spontaneous fission neutron spectrum is dominant [7].

Although these are the primary mechanisms, other options for neutron production in spent fuel exist, including (γ, n) and $(n, 2n)$ reactions, but these are insignificant compared to the frequency of the reactions above [8].

To distinguish only the fission neutron signal, the energy cut-off method was employed. The background neutrons from the (α, n) reaction with oxygen are less than 3 MeV and the energy distribution of prompt fission neutrons is according to the Watt fission spectrum, typically distributed from 0.1 MeV to 10 MeV [Figure 4]. In the previous study, it was found that the energy of an incident neutron can partially transfer up to 64% of its energy to ^4He nucleus by elastic scattering, so the maximum energy deposited by a 3 MeV background neutron will be 1.9 MeV. Therefore, any energy deposition greater than 1.9 MeV in the detector must be from a fission neutron rather than a background neutron [9].

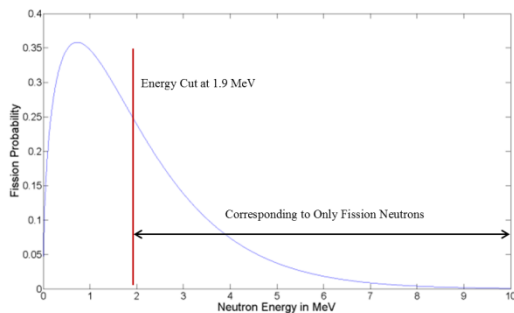


Fig. 4. Applied energy cut-off method to distinguish the fission neutron signal from the background neutron signal produced by the (α, n) reaction with oxygen.

This method was verified by a proof of concept experiment, whereby a sample of fissionable material (natural uranium) was interrogated by the 2.45 MeV neutron generator [9]. The results, as shown in Figure 5, show that the application of a simple cut technique would be able to differentiate neutrons from different sources.

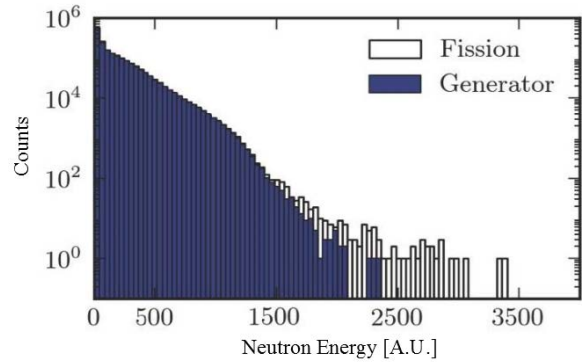


Fig. 5. Proof of concept experiment using ^4He scintillation detectors and the D-D neutron generator. The blue columns are the background response from the neutron generator, and the white columns are the signal from the neutron generator and a natural uranium slug.

3.3 Proposed System Design

A new experimental apparatus, employing some ^4He scintillation detectors, was designed and applied. ^4He scintillation detectors are vertically daisy-chained and the output signal can be analyzed individually from each detector or collectively from these daisy-chained detectors working as a single detector as shown in Figure 6.

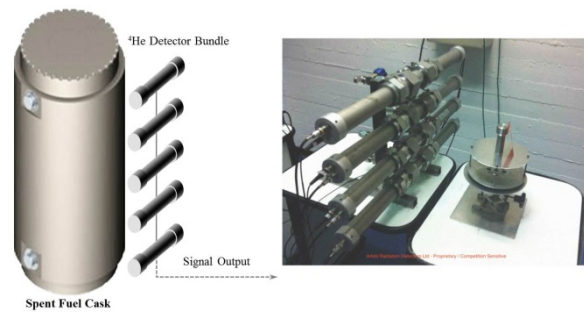


Fig. 6. The design concept (left) and actually chained ^4He scintillation detector bundle (right) for monitoring a spent nuclear fuel cask.

In previous work, the feasibility test of passive neutron interrogation using a ^4He scintillation detector was performed with MOX nuclear fuel [10]. The experimental results demonstrated the potential for this proposed application, and further optimization is being conducted with different geometries of fuel assemblies.

The spent nuclear fuel monitoring system can be described in the same manner as the previous cask monitoring system. The goal of this technical approach is to verify neutron spectra in the spent nuclear fuel storage without any transference of spent fuel assemblies. Detector performance and expected outputs are almost same as the previous approach for the cask monitoring but only three ^4He scintillation detectors are horizontally daisy-chained and placed in a pipe in order

to prevent direct contact with the pool's coolant [Figure 7].

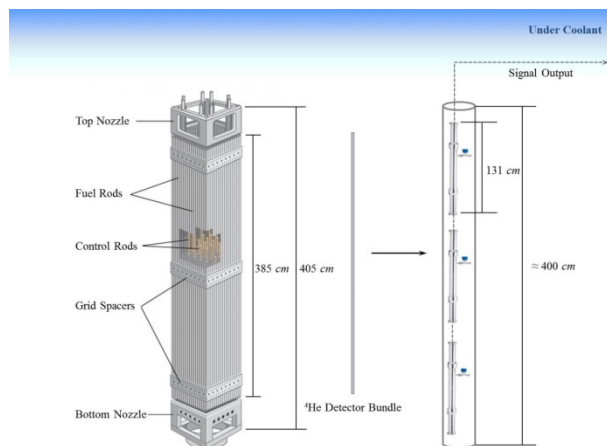


Fig. 7. The conceptual design for spent nuclear fuel storage pool monitoring.

Based on these proposed methods, the University of Florida has recently designed and built a prototype system, employing a D-D neutron generator and using natural uranium slugs embedded in a cylindrical block of polyethylene moderator. Based on the initial design and by applying the energy cut-off method, the feasibility of the proposed methods was assessed and parametric studies are being performed to find the optimal combination of experimental settings.

The next step will be to apply these methods to a real cask model and storage pool at a nuclear power plant. The experimental outputs will be compared with the simulated outputs, and an assessment of the algorithm will be performed.

3. Conclusions

This study conducted a review and conceptual design of a new system for the monitoring of spent nuclear fuel pool and cask storage. Previous experimental results using ^4He scintillator detectors have demonstrated the potential for these new detectors to be implemented in a spent fuel monitoring role. Their ability to preserve neutron energy information, while at the same time reject interfering gamma radiation, provides a new method of signal analysis. By applying energy cuts as proposed above, the spent fuel's fission signal can be isolated, and an algorithm for assembly identification developed.

With this capability verified, future work will be experimental measurements with the system of spent fuel in both wet and dry storage. Similarly, an algorithm will be developed that uses energy discrimination to isolate the fission signal, and use this detected signal to assess the contents of a fuel assembly.

The final goal of this study is to produce a ^4He -based detection and analysis system that is capable of verifying the internal integrity of the fuel assemblies,

and can certify a dry storage cask for extended transportation to a permanent disposal facility.

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