Demonstration of Gamma Ray Insensitivity of Boron Coated Straw-based Neutron Multiplicity Counter

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Abstract

Pebble bed reactors (PBRs) are advanced reactor designs that can potentially improve the safety, efficiency, and economics of the nuclear energy production. PBRs rely on tristructural-isotropic (TRISO) fuel for enhanced fission product retention and improved spent fuel management. Unique identification of individual fuel pebbles would allow determining the fuel transit time for validation of computational models, preventing excessive burnup accumulation or premature fuel discharge, and improving fuel accountability. One of the unique signatures for fuel identification is the ²³⁵U mass and burnup level, which can be extracted through neutron coincidence counting. However, neutron coincidence counting of partially spent fuel can be challenging due to the intense gamma-ray background emitted by fission products. Boron coated straw (BCS) detectors, featuring high neutron detection efficiency and low sensitivity to gamma rays, are suitable for this application. In this work, we have designed and tested a high-neutron-efficiency BCS-based neutron multiplicity counter (NMC, external diameter 17.5 cm, inner diameter 5 cm, length 40 cm) optimized for TRISO fuel assay. We used the NMC to measure a 8.1 Ci ¹⁹²Ir gamma-ray source and obtained an intrinsic gamma-ray efficiency of 10⁻¹² under an average gamma-ray exposure rate of 340.87 R/h. We simulated the active interrogation of used TRISO-fueled pebble with burnup ranging from 9 GWD/t to 90 GWD/t and achieved a relative uncertainty and error below 2.5% in ²³⁵U mass assay in 100 s.

Keywords: PBR, TRISO, neutron coincidence counting

1. Introduction

Pebble bed reactors (PBRs) are advanced reactor designs that can potentially improve the safety, efficiency, and economics of the nuclear energy production. PBRs rely on tristructural-isotropic (TRISO) fuel for enhanced fission product retention and improved spent fuel management. Unique identification of individual fuel pebbles would allow determining the fuel transit time for validation of computational models, preventing excessive burnup accumulation or premature fuel discharge, and improving fuel accountability. One of the unique signatures for fuel identification is the ²³⁵U mass and burnup level, which can be extracted through neutron coincidence counting. In our previous work [1], we simulated the neutron active interrogation of fresh TRISO-fueled pebbles with an external thermal neutron source to induce fission inside the pebble. We have developed a neutron multiplicity counter (NMC) based on pie-shape boron coated straw (BCS) detectors to assay ²³⁵U mass based on the correlated neutron counts from induced fission and achieved a relative uncertainty below 2% in 100 s.

PBR core typically employs the MEDUL fuel management scheme to achieve higher average fuel burnup and power output, where used spent fuel pebbles that have not reached the target burnup level are re-inserted into the reactor core [2]. In this work, we simulated the neutron active interrogation of used fuel pebbles with burnup in the 9-90 GWD/t range. Used fuel pebbles are strong passive gamma-ray emitters with gamma-ray intensity up to 10^{13} cps [3]. Therefore, a high-gamma-ray-insensitivity of the BCS-based NMC is crucial to ensure the gamma-ray background does not interfere with the neutron coincidence counting. Used fuel pebbles also emit (α , n) neutrons, delayed neutrons, and spontaneous fission neutrons through (α , n) reactions, beta decay of fission product, and spontaneous fission of fission product, respectively. In particular, spontaneous fission nuclides such as ²⁴⁰Pu, ²⁴²Cm, and ²⁴⁴Cm have a high neutron fission yield of 1.02×10^3 ,

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 2.10×10^7 , 1.08×10^7 n/s/g respectively [4], and their contributions to doubles may become non-negligible at high burnup levels.

This paper is organized as follows. In Section 2, a detail description of the BCS-based NMC is presented. In Section 3, the high gamma-ray-insensitivity of the BCS-based NMC are demonstrated by measuring a gamma-ray source with intensity comparable to used fuel pebble's. In Section 4, determination of nuclide concentration of used fuel pebble and passive neutron and gamma-ray source terms are presented. In Section 5, simulation of active interrogation of used fuel pebble is carried out. Finally, the discussion and conclusions are presented in Section 6.

2. BCS-Based Neutron Multiplicity Counter

Figure 1 shows the design of a custom BCS-based NMC for interrogation of TRISO-fueled pebbles. The NMC is a cylinder that measures 55 cm in length and 17.5 cm in diameter, as shown in Fig. 1a. It has a central cavity with a diameter of 10 cm, which serves as the sample placement area, as shown in Fig. 1b. The inner and outer surfaces of the NMC are covered with 0.508 mm-thick cadmium to prevent thermal neutrons from reentering the sample cavity. The NMC is composed of 192 straws that are organized in a hexagonal lattice, with an inter-straw distance of 0.9091 cm, as shown in Fig. 1c. Surrounding the straws is high-density polyethylene (HDPE). Each straw is 40 cm long and has a diameter of 4.7244 mm. Fig. 1d shows that the inner surface of each straw is coated with a 1.3 µm-thick layer of B₄C (96% enriched with ¹⁰B) to absorb thermal neutrons and generate ⁷Li ions and alpha particles through ¹⁰B(¹n,⁴\alpha)⁷Li reactions. Six septa are present inside each straw to increase the surface area for improving the detection efficiency and reducing the system die-away time. The straw is filled with a gas mixture of Ar/CO₂ (9:1) at 0.7 atmosphere to detect ⁷Li/alpha ions. When alpha/⁷Li particles ionize the gas, ionized electrons/ions induce electrical signals on the anode wire at the center of the straw. Based on the timestamps of acquired pulses, we calculated the neutron singles and doubles count rates using the signal-triggered shift-register algorithm [5]. The pre-delay time was set to 2 µs, the gate was set to 32 µs, and the long delay time was set to 2 ms. The uncertainty associated with the singles and doubles were calculated using [6]:

$$\sigma_{S} = \frac{S + 2Df_{1}}{T},$$

$$\sigma_{D} = \frac{D + 2S^{2}G + 4DSGf_{1} + 2D^{3}f_{1}/S^{2}}{T}$$
(1)

where S and D are the singles and doubles count rates, respectively, G is the time gate width, f_1 is the gate utilization factor, and T is the interrogation time.



Figure 1: Design of a custom BCS-based NMC for interrogation of TRISO-fueled pebbles. (a) is the external view of the counter, (b) is the internal view with the readout electronics, straws and HDPE, (c) is the cross section of pie-6 straws, and (d) shows the $1.3 \,\mu$ m-thick layer of B₄C converter.

3. Demonstration of High-Gamma-Ray-Insensitivity of BCS-Based NMC

The partially-spent TRISO-fueled pebble can emit passive gamma-rays with intensity up to 10^{13} cps [3]. Therefore, a high-gamma-ray-insensitivity of the NMC is crucial to ensure that passive gamma-rays emitted by the pebble do not interfere with neutron measurements. Gamma-rays produce low-amplitude pulses by depositing a small amount of energy, which can be effectively rejected by properly setting the discriminator threshold of the system [7]. To demonstrate

the system's high-gamma-ray insensitivity, we used our BCS-based NMC to measure a Nucletron Flexisource ¹⁹²Ir HDR (High Dose-Rate) source in a Elekta Flexitron Remote Afterloader [8], as shown in Fig. 2. The ¹⁹²Ir source has a diameter of 0.6 mm and length of 3.5 mm [9], with an activity of 8.1 Ci at the date of experiment. The source can be transported along the blue wire through a control software. We placed the source at the center of sample cavity of the NMC and measured source for 10 min. The NMC was connected to a 14-bit 500MS/s DT5730S digitizer for list mode data acquisition. The NMC was powered by a BK Precision 9110 power supply at 5 V. We calculated the intrinsic gamma-ray efficiency, defined as the ratio between the number of counts and the number of gamma-rays that reaches the NMC's inner surface. Fig. 3 shows the intrinsic gamma-ray efficiency and the intrinsic neutron efficiency obtained by measuring a ²⁵²Cf source [1] as a function of discriminator threshold. A gamma-ray efficiency of 10⁻¹² can be achieved, comparable to ³He-based systems, without sacrificing the neutron efficiency significantly.



Figure 2: Measurement of the 8.1 Ci ¹⁹²Ir HDR source. The gamma-ray source was placed at the center of sample cavity of the NMC and the measurement lasted for 10 min.

4. Burnup Calculation for HTR-10 Reactor Using SCALE 6.2.4

We used *SCALE* 6.2.4 to simulate the nuclide concentration of used fuel pebble and the passive neutron and gammaray source terms due to decay of fission products. SCALE is a widely-used modeling and simulation suite for nuclear safety analysis and design [10]. First, we developed a high-fidelity Monte Carlo model of the HTR-10 in SCALE 6.2.4. The HTR-10 reactor provided valuable benchmark experimental data for validating computational model of PBR [11]. Fig. 4 shows the vertical cross sections of the HTR-10 model. The overall radius and height of HTR-10 are 190 cm and 610 cm, respectively. The pebbles are arranged in a Hexagonal Close-Packed (HCP) lattice, as shown in Fig. 4b, and the difference between the experimental and simulated packing fraction is 0.23%. Each TRISO-fueled pebble contains approximately 8335 TRISO fuel particles arranged in a body-central-cubic lattice, as shown in Fig. 4c. The initial ²³⁵U enrichment is 17%. We used the *KENO-VI* module in SCALE to calculate the the effective multiplication factor (k_{eff}). The nuclear data library used was ENDF-VII.1 and the number of generations was 500 with 1000 neutrons per generation. An effective multiplication factor of 1.0008 ± 0.0013 was obtained, which is in good agreement with the experimental value of 1.00000 ± 0.00369 [11]. The excellent agreement of k_{eff} between the simulation and experiment suggests that we have an accurate Monte Carlo model of the HTR-10, based on which we can perform depletion calculation of the fuel and extract the isotopic composition of partially-spent fuel.

The *t6-depl* sequence in *TRITON* (Transport Rigor Implemented with Time-dependent Operation for Neutronic depletion) module of SCALE was employed to calculate the material composition at different burnup levels. The reactor power



Figure 3: Intrinsic gamma-ray and neutron efficiency as a function of discriminator threshold.



Figure 4: Full SCALE model of HTR-10. (a): Vertical cross-section of the reactor. Green spheres represent the TRISO-fueled pebble and the orange spheres represent the pure graphite pebbles. (b): Zoomed-in view of the mixed pebble region. (c): Zoomed-in view of a single TRISO-fueled pebble, with each black dot representing one TRISO fuel particle.

was set to 74.07 MW/MTU and the depletion time was 1200 days in ten time intervals. The resulting fuel burnup ranged from 8.88 GWD/t to 88.88 GWD/t in steps of 8.88 GWD/t. The fuel material was then fed to the *ORIGEN* (Oak Ridge Isotope Generation) module of SCALE to extract the nuclide concentration, gamma-ray source term, and neutron source terms, including the (α , n) neutron, delayed neutron, and SF neutron. Fig. 5 shows the concentration of ten nuclides of the highest weight fraction as a function of burnup. The ²³⁵U mass decreases and the mass of fission products increases with burnup as the fuel burns.



Figure 5: Mass of 10 nuclides of the highest weight fraction as a function of fuel burnup.

5. Simulation of Interrogation of Partially-Spent TRISO-Fueled Pebbles

Based on the nuclide concentration of used fuel pebble, we simulated the neutron active interrogation of used fuel pebble at each burnup. For each burnup, we simulated the neutrons singles and doubles contributed by the gamma-ray source, (α , n) neutron source, delayed neutron source, SF neutron source, and induced fission by the interrogation source. Fig. 6 shows the MCNP simulation setup of active interrogation of used TRISO-fueled pebbles. A 3 cm radius TRISO-fueled pebble was placed at the center of the sample cavity of the NMC. A layer of 1-cm thick of tungsten was added to the inner surface of the NMC to shield the gamma-rays. MCNP simulation shows that the tungsten shielding reduces the gamma-ray intensity by approximately 16% without affecting the neutron count rate. The interrogation source was a thermal neutron beam with a source strength of 10^6 n/s and the interrogation time was 100 s.

Table 1 and Table 2 shows the comparison of singles and doubles count rates from both induced fission and passive neutrons and gamma-rays sources. The singles count rate contributed by gamma-rays was estimated by multiplying the shielded gamma-ray intensity with the intrinsic gamma-ray efficiency of the system, which was assumed to be 10^{-12} based on the gamma-ray-insensitivity measurement in section 3. We observed that the contribution from passive gamma-rays and neutrons are negligible compared to singles/doubles from induced fission in active interrogation. Therefore, our system is feasible for assay of ²³⁵U in both fresh and used TRISO-fueled pebbles.

Table **??** shows the relative uncertainty and relative error associated with 235 U mass estimated based on the doubles count rate using the method in [1]. Both the relative uncertainty and relative error associated with the 235 U mass assay are below 2.5% with an inspection time of 100 s.



Interrogation beam

Figure 6: Active interrogation measurement setup.

Table 1: Comparison	of neutron singles rates	(unit: cps) from each	source term as a function	of burnup and ²³⁵ U mass.
1	0			

Burnup (GWD/t)	²³⁵ U mass (g)	Active interrogation	Delayed & (alpha,n)	SF
8.8884	0.79	2.111698×10 ⁴	0.37	0.00
17.7768	0.74	1.993981×10 ⁴	0.31	0.00
26.6652	0.68	1.876520×10^4	0.55	0.00
35.5536	0.63	1.755169×10^4	0.87	0.45
44.442	0.58	1.634818×10 ⁴	1.74	1.64
53.3304	0.53	1.516904×10^4	3.48	3.59
62.2188	0.48	1.396813×10 ⁴	5.61	6.96
71.1072	0.43	1.277833×10^{4}	9.95	12.02
79.9956	0.38	1.157827×10^4	16.10	19.80
88.884	0.33	1.039044×10^4	24.40	31.82

Table 2: Comparison of neutron doubles rates (unit: cps) from each source term as a function of burnup and ²³⁵U mass.

Burnup (GWD/t)	²³⁵ U mass (g)	Active interrogation	Delayed & (alpha,n)	SF
8.8884	0.79	1.744620×10^{3}	0.00	0.00
17.7768	0.74	1.644850×10^{3}	0.00	0.00
26.6652	0.68	1.553740×10^{3}	0.00	0.00
35.5536	0.63	1.447560×10^{3}	0.00	3.00×10 ⁻²
44.442	0.58	1.335340×10^{3}	0.00	0.19
53.3304	0.53	1.256900×10^{3}	0.00	0.44
62.2188	0.48	1.173780×10^{3}	2.00×10 ⁻²	0.85
71.1072	0.43	1.072920×10^{3}	1.00×10 ⁻²	1.62
79.9956	0.38	9.447600×10 ²	2.00×10 ⁻²	2.79
88.884	0.33	8.629800×10 ²	1.00×10 ⁻²	4.44

Burnup (GWD/t)	U235 mass (g)	Relative error (%)	Relative uncertainty (%)
8.8884	0.79	-0.87	1.68
17.7768	0.74	-0.58	1.67
26.6652	0.68	0.30	1.66
35.5536	0.63	0.00	1.65
44.442	0.58	-1.04	1.66
53.3304	0.53	0.88	1.68
62.2188	0.48	2.52	1.72
71.1072	0.43	2.18	1.80
79.9956	0.38	-2.24	1.94
88.884	0.33	-1.05	2.15

Table 3: Relative uncertainty and relative error associated with the ²³⁵U mass estimated based on neutron doubles.

6. Discussions and Conclusions

In this work, we have demonstrated the high-gamma-ray-insensitivity of the BCS-NMC, with a value of 10⁻¹², under a high gamma-ray exposure rate of 340.87 R/h. We have developed a high-fidelity Monte Carlo model of the HTR-10 reactor in SCALE 6.2.4 and performed fuel burnup calculation. Fuel composition, passive gamma-ray and neutron source terms were extracted for used fuel pebble with burnup between 8.88 GWD/t and 88.88 GWD/t. We simulated the neutron active interrogation of used fuel pebbles and the contributions to the singles and doubles from passive neutron and gammaray emitters were negligible compared to induced fission. Therefore, measurement of ²³⁵U mass in used TRISO-fueled pebble using our BCS-NMC is feasible. The relative uncertainty and error associated with the ²³⁵U mass assay is below 2.5% for pebbles with burnup below 90 GWD/t with an interrogation time of 100 s, which is compatible with practical reactor operation.

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References

- M. Fang, J. Lacy, A. Athanasiades, A. Di Fulvio, Boron coated straw-based neutron multiplicity counter for neutron interrogation of triso fueled pebbles, Annals of Nuclear Energy 187 (2023) 109794. doi:https://doi.org/10.1016/j.anucene.2023.109794.
 URL https://www.sciencedirect.com/science/article/pii/S0306454923001135
- [2] S. Topan, I. Dwi, et al., Study on fuel multipass effect on core performance of small pebble bed reactor (2016).
- [3] B. Su, Design and construction of a prototype advanced on-line fuel burn-up monitoring system for the modular pebble bed reactor, Tech. rep., University of Cincinnati, Cincinnati, OH (US) (2004).
- [4] D. Reilly, N. Ensslin, H. Smith Jr, S. Kreiner, Passive nondestructive assay of nuclear materials, Tech. rep., Nuclear Regulatory Commission (1991).
- [5] S. Croft, D. Henzlova, D. Hauck, Extraction of correlated count rates using various gate generation techniques: Part i theory, NIMA 691 (2012) 152 – 158. doi:10.1016/j.nima.2012.06.011.
- [6] M. Prasad, N. Snyderman, J. Verbeke, Analytical error bars and rsd for neutron multiplicity counting, Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 903 (2018) 25–31. doi:https://doi.org/10.1016/j.nima.2018.06.010.
- [7] G. F. Knoll, Radiation detection and measurement, John Wiley & Sons, 2010.

- [8] Elekta flexitron afterloader, https://www.elekta.com/products/brachytherapy/flexitron/, accessed: 2023-03-27.
- [9] A. Lopes, E. Sabondjian, A. R. Baltazar, In vivo dosimetry for superficial high dose rate brachytherapy with optically stimulated luminescence dosimeters: A comparison study with metal-oxide-semiconductor field-effect transistors, Radiation 2 (4) (2022) 338–356.
- [10] B. T. Rearden, M. A. Jessee, Scale code system, Tech. rep., Oak Ridge National Lab.(ORNL), Oak Ridge, TN (United States) (2018).
- [11] W. K. Terry, Evaluation of the initial critical configuration of the htr-10 pebble-bed reactor, Tech. rep., Idaho National Lab.(INL), Idaho Falls, ID (United States) (2005).