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Radiological Characterization of the Philippine Research Reactor-1 Legacy Neutron Sources via Monte Carlo Simulation

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Abstract

Three legacy neutron sources (LNS), one ^{239}PuBe and two ^{252}Cf, are currently stored in the Philippine Research Reactor-1 (PRR-1) facility. These LNS have been kept in the storage tank for the irradiated fuel in the PRR-1 facility for three decades. However, there is limited information available on these sources. Thus, in this paper, the dose rates associated with the irradiated fuel and the three LNS were calculated using Monte Carlo simulations. The dose distribution for the three LNS in a 200-L concrete-filled stainless steel container was also calculated. Results showed that the storage tank has provided adequate shielding against neutrons from the three LNS and the neutron doses are negligible compared with the gamma dose from the irradiated fuel. Simulation results demonstrated that the container has given sufficient radiation shielding for handling LNS. The results of this study will serve as a basis for developing procedures for safe handling, storage, transfer, disposal and use of LNS upon commissioning of the PRR-1 Subcritical Assembly for Training, Education, and Research (SATER). Moreover, this offers a reference for updating and improving the radiation protection measures in the PRR-1 facility.

***Keywords**:* dose rate, gamma dose, legacy neutron sources, Monte Carlo simulation, neutron dose

1. Introduction

Although isotopic neutron sources (INS) have low neutron emission rates, they have important advantages compared with neutron generators and nuclear reactors because of their small size, portability and relatively low cost. INS are highly practical for various applications because these sources are easy to handle, operate and maintain as it requires no high-voltage source and typically, no fissile materials. These neutron sources are widely used in radiation monitoring, instrument calibration, neutron activation, nuclear training and education and other industrial and field applications. They are also utilized in designing shielding and radiation protection protocols for the safe handling of the source (Hassan, 2001; Vega-Carrillo and Martinez-Ovalle, 2015).

There are four main types of isotopic neutron sources: spontaneous fission sources, radioisotope (*α, n)* sources, photoneutron sources and reactions from accelerated charged particles. The most common example of a spontaneous fission source is ^{252}Cf. This isotope is a transuranic heavy nuclide, which is unstable and decays into other elements. In each fission event, several fast neutrons are promptly emitted; hence, ^{252}Cf is a reasonably convenient isotopic neutron source (Hassan, 2001). It is an intense neutron emitter, which decays by *α*-emission with a 96.91% probability and with a 3.09% probability by spontaneous fission. It has a half-life of 2.645 years (Vega-Carrillo and Martinez-Ovalle, 2016; Hila *et al.*, 2021). ^{252}Cf is used for prompt gamma neutron activation analysis (PGNAA) of coal, cement, minerals and detection of explosives. Other uses include neutron radiography, reactor start-up sources, calibration standards, and cancer therapy, namely treatment for cervical cancer and malignant tumors (Martin *et al.*, 2000; Zhang *et al.*, 2012).

Aside from the ^{252}Cf spontaneous fission source, other commonly used neutron sources are radioisotope (*α, n*) sources such as ^{239}PuBe and ^{241}AmBe. These sources both consist of heavy nuclides that are *α*-emitters (^{239}Pu and ^{241}Am) combined homogeneously with a light element (e.g., ^{9}Be) to induce the (*α, n*) reaction. ^{239}PuBe source is one of the most commonly used among the (*α, n*) isotopic neutron sources (Hassan, 2001). Its half-life is 2.41 x 10^4 years (Vega-Carrillo *et al.*, 2012). The ^{239}PuBe source has varying isotopic compositions depending on the source origin (Bagi *et al.*, 2013). The total Pu content and its isotopes can be determined from the combination of alpha activities of Pu isotopes and its isotopic composition, which can be measured using various methods (Bagi *et al.*, 2016; Abdessamad *et al.*, 2017). However, the neutron source strength of ^{239}PuBe tends to vary with time due to the buildup of ^{241}Am from the decay of ^{241}Pu impurities in the source (Vega-Carrillo *et al.*, 2012). Meanwhile, the ^{241}AmBe neutron source has a highly stable neutron flux, providing many advantages, especially for neutron detector and dosimeter calibration.

Although neutron sources are generally beneficial, there is a need for radiation safety protocols in the use of these sources. Neutron shielding is an essential aspect of radiation safety for neutron sources, which should account for the dose that can be derived from neutron radiation as well as primary gamma radiation from the source. There are also secondary neutrons that can arise from the interaction of source neutrons with the shielding material. Hence, shielding design for neutron sources has to consider three types of radiation: the primary neutron and gamma radiation, as well as the secondary gamma. Monte Carlo methods are typically used to optimize the design of shielding for different neutron sources (Vega-Carrillo *et al.*, 2018).

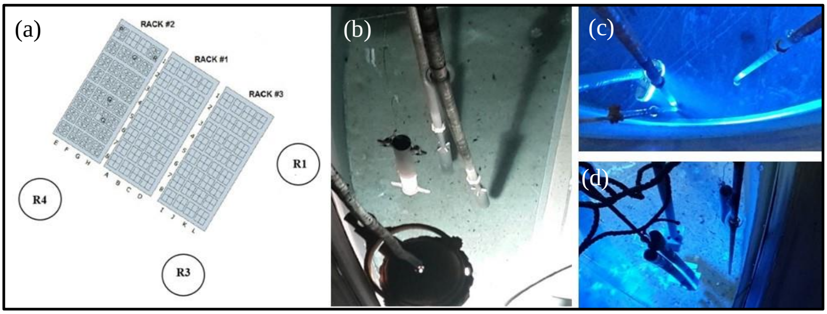
Legacy neutron sources (LNS) are especially important to characterize due to the lack of information available on these sources. LNS are sources that pre-date effective regulatory requirements and which may not have been disposed of, either at all or in an appropriate manner (International Atomic Energy Agency [IAEA], 2011). Radiological characterization aids in addressing the potentially harmful effects of neutron radiation from the LNS by providing a basis for the appropriate shielding. This characterization should include the following information about the LNS: initial activity, half-life, current activity and emission rate. Hence, a survey of existing data, calculations, measurements, sampling and analyses are necessary (IAEA, 1998).

The Philippine Research Reactor-1 (PRR-1) facility is currently in possession of three LNS that have been stored together with slightly irradiated TRIGA fuel in a storage tank for almost 35 years. These LNS include one ^{239}PuBe and two ^{252}Cf, which are legacy sources from the former operation of PRR-1.

This study aimed to characterize the radiation hazard associated with the three legacy neutron sources and to determine the dose distributions from the slightly irradiated Training, Research, Isotopes, General Atomics (TRIGA) fuel rods. This will serve as a basis for preparing the procedures for the safe handling, storage, transfer, disposal and use of isotopic neutron sources upon commissioning of the PRR-1 Subcritical Assembly for Training, Education, and Research (SATER).

2. Methodology

Figure 1 shows the PRR-1 storage tank, which contains the locations of the legacy neutron sources relative to the fuel rods. Although there is information on neutron sources provided in the PRR-1 authorization, the actual identities of these LNS have not been confirmed initially. Previous work was done to confirm these LNS through gamma spectrometry and radiation dose survey in preparation for the reoperation of the PRR-1 facility (Gatchalian *et al.*, 2019).



R1, and R3 and R4 correspond to ^{239}PuBe and ^{252}Cf sources, respectively. Note that the labeling convention follows internal documentation.

Figure 1. Top view of the PRR-1 storage tank with the location of the neutron sources relative to the fuel rods (a) and photographs of the actual locations of neutron sources (b)-(d)

Table 1 provides the details of the PRR-1 LNS, while Figure 2 presents the corresponding neutron spectra. Monte Carlo N-Particle (MCNP) simulation was performed to characterize the radiological hazard associated with each source. The F4 mesh tally in MCNP 5 v.1.6 (X-5 Monte Carlo Team, 2003) was utilized to map the neutron and gamma doses. The flux tally was modified to dose using flux-to-dose conversion factors (DE and DF cards) in MCNP. The ICRP-74 conversion factors were used for neutron fluence to convert to ambient dose equivalent, while the ANSI/ANS-6.1.1 factors were used for gamma (International Committee on Radiological Protection, 1996; American Nuclear Society, 1977). Between the two conversion factors, the ICRP-74 consisted of more recent data. However, the ANSI/ANS-6.1.1 was applied for gamma dose calculation to be conservative as it tends to overestimate gamma dose compared with ICRP-74 (International Committee on Radiological Protection, 1996).

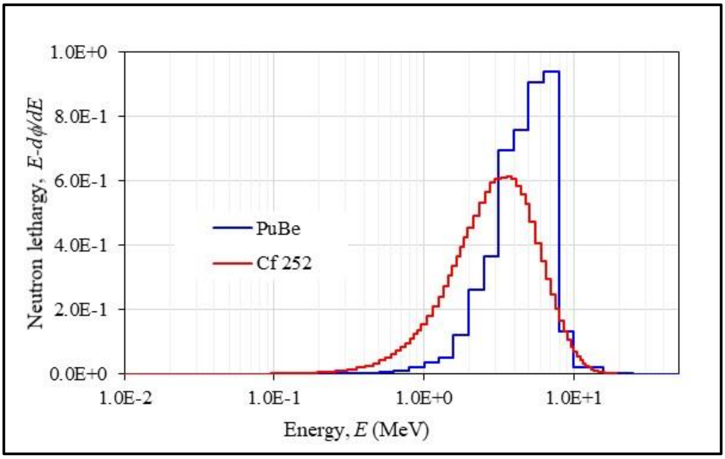


Figure 2. Lethargy neutron spectra of ^{239}PuBe (IAEA, 2001) and ^{252}Cf isotopic neutron sources (Chadwick *et al*., 2006)

Neutron dose and gamma dose from secondary photons produced from neutron interactions were calculated by performing fixed source calculations with MCNP. Volumetric sources were defined with neutron spectra as presented in Figure 2. Calculation results were scaled based on the neutron emission rates as listed in Table 1.

Table 1. Isotopic neutron sources in the PRR-1 facility

Neutron source	Mass (g)	Initial activity (Bq)	Reference date	Half-life (days)	Current activity* (Bq)	Emission rate (n/s)	Plan
^{239}PuBe	32	7.40×10^{10}	Unknown	8.80×10^{6}	Unknown	$3.60 \times 10^{6**}$	To be repurposed
^{252}Cf	2.05×10^{-4}	4.28×10^{9}	4-Apr-84	9.68×10^{2}	3.20×10^{5}	3.59×10^{4}	To be disposed
^{252}Cf	2.16×10^{-4}	4.07×10^{9}	4-Apr-84	9.68×10^{2}	3.04×10^{5}	3.74×10^{4}	To be disposed

*as of July 31, 2020; **based on initial activity

MCNP models were prepared, which included volumetric neutron sources that are incorporated in a detailed model of the PRR-1 storage tank. The PRR-1 storage tank contains the TRIGA fuel rods that are clustered in an assembly containing four rods. Every cluster occupies a single hole in the storage rack. The storage rack is an aluminum structure that measures an 8 x 4 rectangular array of holes where 30 TRIGA fuel clusters were accommodated. A detailed description of the computational modeling performed for the storage tank is provided in previous work (Gatchalian *et al.*, 2021). For the calculation of the dose distribution, a rectangular mesh centered at the tank with mesh element dimensions of 1 x 1 x 50.8 cm was used to calculate the x-y plane distribution of the dose rate. A 1 x 1 cm mesh size along the x and y axes was selected to obtain a good resolution, while the 50.8 cm mesh size along the z-axis was chosen to coincide with the active length of the fuel. Although the actual location of the neutron sources in the tank varies in terms of vertical positioning, they were declared in the MCNP model at a vertical position that coincides with the midpoint of the fuel length. This is to estimate the neutron doses from the legacy LNS relative to the gamma dose from the slightly irradiated PRR-1 TRIGA fuel.

3. Results and Discussion

3.1 Neutron Dose from LNS in the Storage Tank

The dose distributions from photons emitted by the slightly irradiated TRIGA fuel rods and neutrons from the three LNS are presented in Figure 3. While the calculated gamma dose rates were scaled based on measured dose rates around the tank, the calculated neutron dose rates were scaled using the corresponding emission rates as listed in Table 1. For relative comparison of the doses, the combined plots are also presented in a three-dimensional (3D) plot in Figure 4, where the dose values for R3 and R4 sources have been multiplied 100 x to be visible. The plots demonstrate that, in the current storage condition, the storage tank has provided enough shielding against neutrons from the LNS such that the neutron doses are insignificant compared with the gamma dose from the storage tank.

For underwater neutron sources, the dose rate decreased to a fraction of a µSv/h at a distance of 50 cm or more from the source. Considering that the water level in the storage tank was 3.5 m, it is expected that the only contributors in the dose rates at the storage tank platform are photons from the slightly irradiated fuel rods. The vertical photon dose distribution from the irradiated fuel rods is presented in Figure 5 to provide an estimate of the gamma doses in the storage platform, where most activities involving the storage tank are performed. Table 2 provides the measured and MC-calculated dose rates at selected locations in the vicinity of the storage tank. The MC-calculated dose rate was higher than the measured dose rate at the lateral surface, which demonstrates that the calculation result provided a conservative estimate of measured doses in the area. However, at the storage platform level, the measured dose rate was higher than the calculated value. This is because the measured dose rate was linked to the background level radiation in the platform. Meanwhile, the calculated dose rate indicated that, under normal conditions, the radiation exposure from the photons emitted by the fuel has a negligible contribution in the storage tank platform location.

Table 2. Maximum dose rates at the vicinity of the storage tank (μSv/h)

Location	MC-calculated dose rates	Measured dose rates
Lateral surface	39.66±4.03	~10.5
Just above the water level	$1.8 \times 10^{-2} \pm 5.8 \times 10^{-4}$	*
Just above the tank	$1.1 \times 10^{-2} \pm 4.7 \times 10^{-4}$	*
Storage platform level	$2.7 \times 10^{-3} \pm 2.0 \times 10^{-4}$	3.02×10^{-1}

* means no measured values due to lack of access

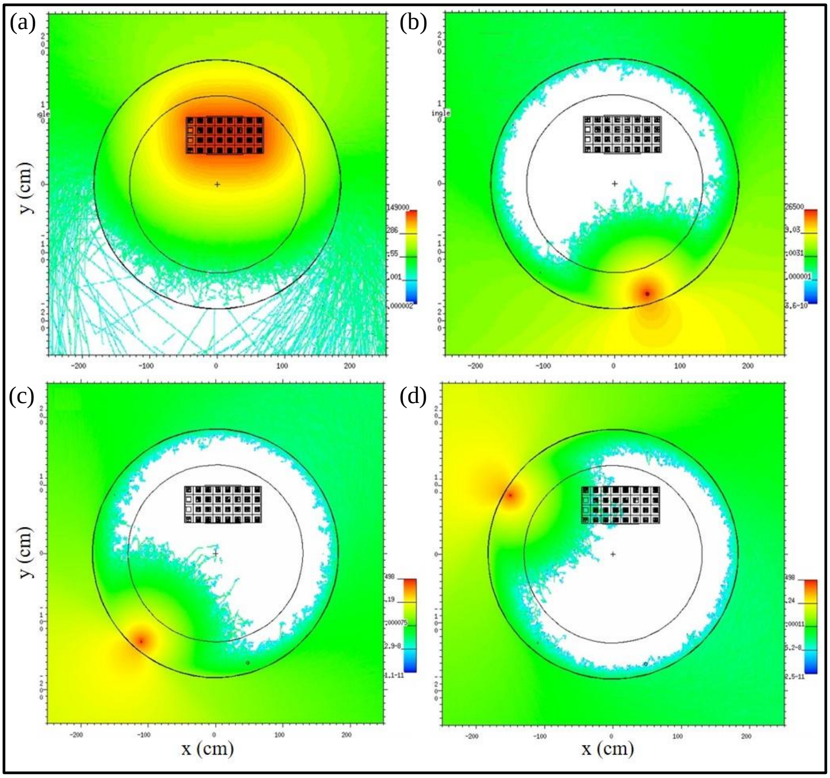


Figure 3. Horizontal photon dose distribution from the slightly irradiated fuel rods (a); horizontal neutron dose distribution from R1: ^{239}PuBe LNS (b), R3: ^{252}Cf LNS (c) and R4: ^{252}Cf LNS (d); dose rates are in μSv/h.

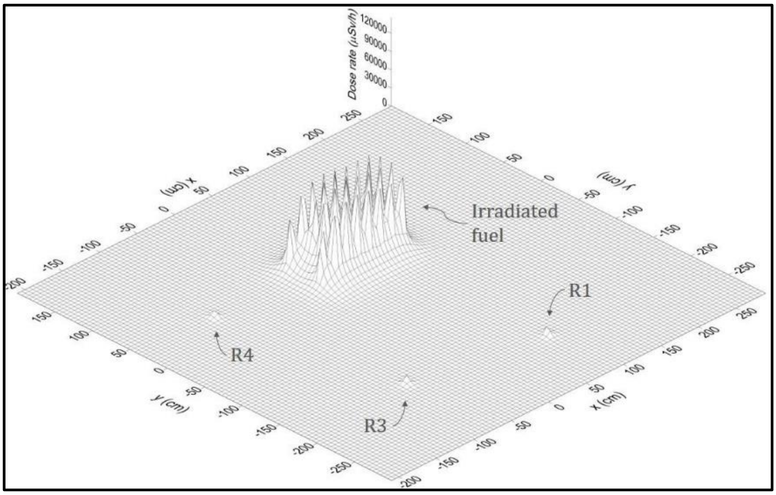


Figure 4. Relative horizontal distribution of photon dose from irradiated fuel and neutron doses from R1, R3, and R4 LNS; note that the values of doses from R3 and R4 were multiplied 100 x for visibility.

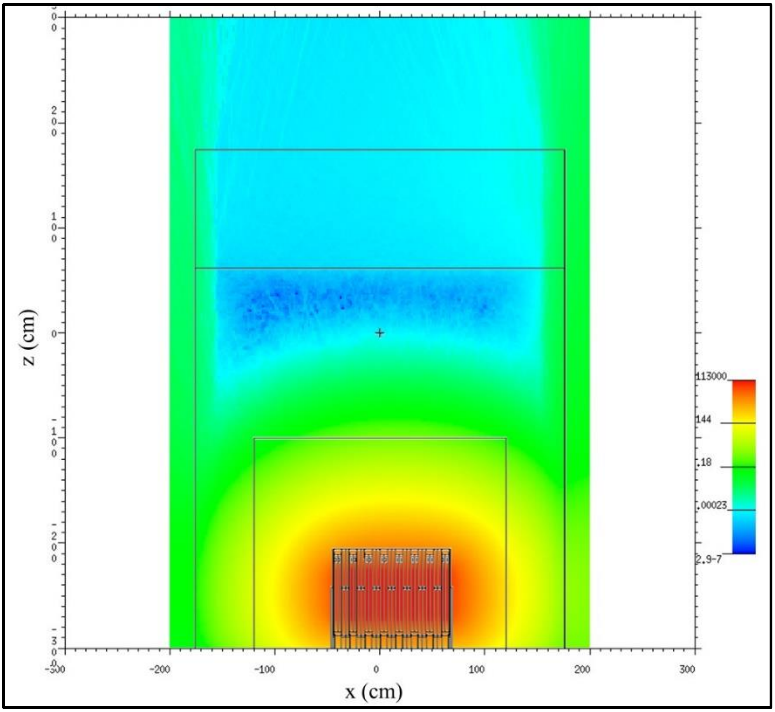


Figure 5. Vertical photon dose distribution from the slightly irradiated fuel rods

3.2 Dose Distribution for LNS in Standard 200-L Stainless-Steel Drum

Although the current storage location of the LNS has given sufficient shielding against neutrons emitted by the LNS, the facility operators have planned to gradually remove the LNS from the storage tank to further improve the safety conditions in the PRR-1 facility. As indicated in Table 1, there is a plan to repurpose the ^{239}PuBe source, while the ^{252}Cf sources will be disposed and transferred to the PNRI Radioactive Waste Management Facility (RWMF). The radiological hazards associated with the movement of the three LNS were assessed by modeling the sources contained in a standard 200-L stainless-steel drum with a 4-cm central cylindrical opening that will be fitted with a polyethylene plug.

The vertical and horizontal neutron and secondary photon dose distributions for the ^{239}PuBe and two ^{252}Cf LNS are presented in Figures 6 and 7, respectively.

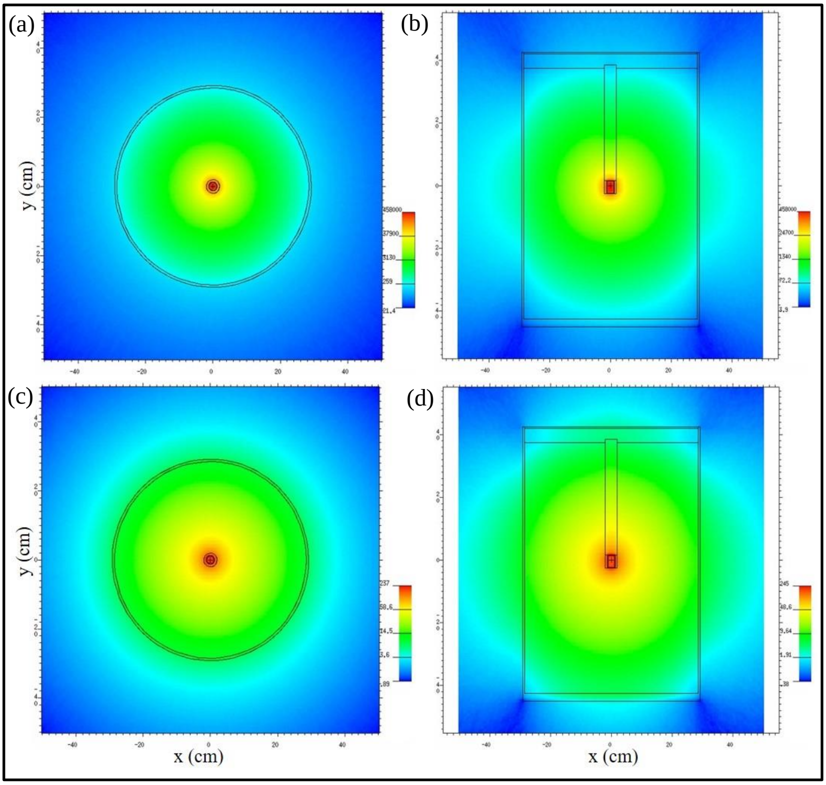


Figure 6. Horizontal (a) and vertical (b) neutron dose distributions from a ^{239}PuBe contained in a 200-L concrete-filled stainless-steel drum; horizontal (c) and vertical (d) dose distributions from secondary photons; dose rates are in μSv/h.

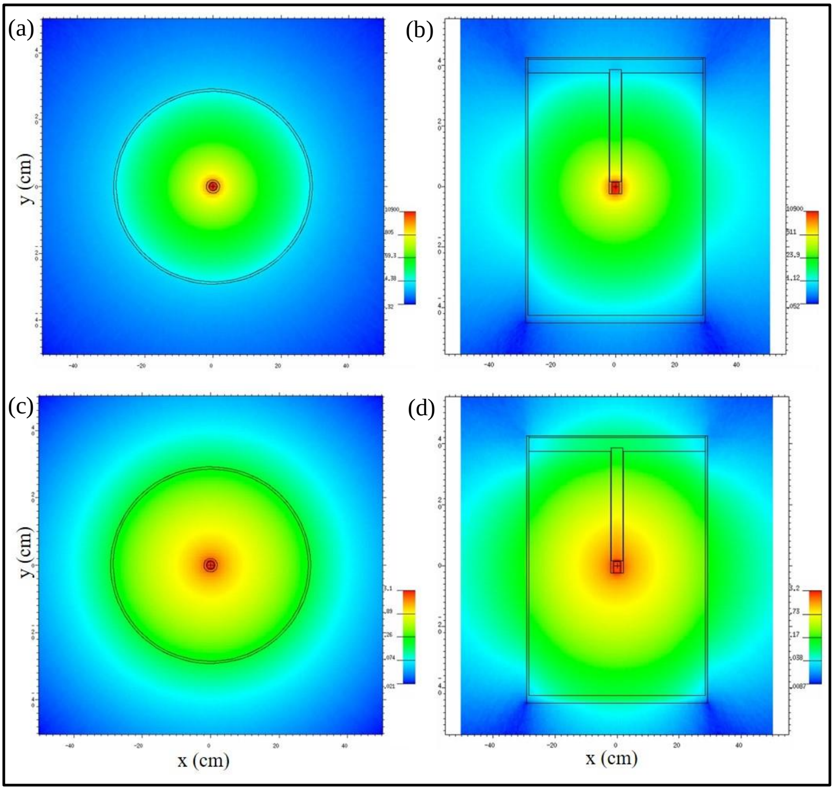


Figure 7. Horizontal (a) and vertical (b) neutron dose distributions from two ^{252}Cf sources contained in a 200-L concrete-filled stainless-steel drum; horizontal (c) and vertical (d) dose distributions from secondary photons; dose rates are in μSv/h.

The dose distributions were nearly symmetrical, and it was evident in the contour plots that a 200-L concrete-filled stainless-steel container will provide sufficient shielding against the combined gamma and secondary photon dose distributions from the LNS. A summary of calculated maximum dose rates at the surface of the containers is presented in Tables 3 and 4.

Table 3. Calculated dose rates at the lateral surface of the LNS container (μSv/h)

Neutron source	Container	Lateral surface		
		Neutron	Photon	Total
^{239}PuBe	200-L concrete-filled stainless-steel drum	198.89±1.77	9.18±0.08	208.07±1.85
2 ^{252}Cf		3.18±0.03	0.222±0.002	3.402±0.032

Table 4. Calculated dose rates at the top and bottom surfaces of the LNS container (μSv/h)

Neutron source	Container	Top and bottom surfaces		
		Neutron	Photon	Total
^{239}PuBe	200-L concrete-filled stainless-steel drum	37.74±1.28	4.39±0.08	42.13±1.36
2 ^{252}Cf		0.475±0.012	0.113±0.001	0.588±0.013

4. Conclusion and Recommendation

In this paper, an MCNP simulation was performed to characterize the radiation hazard associated with the irradiated fuel and three LNS. Relevant quantities such as photon and neutron dose distributions of these materials were determined. Based on the results of calculations, the storage tank in the PRR-1 has afforded adequate shielding against neutrons from the three LNS – neutron doses are negligible compared with the gamma dose from the storage tank. The dose distribution for the three LNS in a 200-L concrete-filled stainless steel container also showed that this container is also sufficient in shielding against the combined neutron and secondary photon dose from the LNS. It was also found that the water in the storage tank has given adequate radiation shielding with an MCNP-calculated dose rate of 39.66±4.03 μSv/h at the surface of the tank during normal conditions of the facility and $2.7 \times 10^{-3} \pm 2.0 \times 10^{-4}$ μSv/h at the storage platform level. The results obtained in this study will serve as a basis for the procedures for safe handling, storage, transfer, disposal and use of isotopic neutron sources upon commissioning of the PRR-1 SATER. Furthermore, these will provide a reference for an updated dose map of the facility that includes neutron radiation, which will be incorporated in the existing radiation area monitoring procedure of the facility.

5. References

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