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Research Article

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Assessment of shielding performance of nitrile butadiene rubber through simulation and experiments at MNSR beam line

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HIGHLIGHTS

- NBR is used as one of the most powerful flexible neutron shields.
- Research reactors could use flexible neutron shield around non-uniform surfaces.
- Neutron dose rate decreases about to its half value using the NBR with suitable thickness.
- Measurements help to estimate the NBR behavior before its installation.

ABSTRACT

New nitrile butadiene rubber (NBR) materials are being considered to use for neutron shielding especially for the positions where need a flexible neutron shield. Such light, low-cost, and suitable material could be used for sealing of the gaps or even for shielding of low radiation environments. In the present work, experimental investigation of NBR shielding performance of neutrons and gamma rays was performed using the beam line of the Isfahan Miniature Neutron Source Reactor. MCNPX code was used to simulate the 30 kW research reactor beam line. Six NBR sheets with 2 cm thickness were used at the outlet of the beam line. The sheets were added top of each other respectively to measure the shielding performance of the material for neutron as well as gamma rays emerged from the outlet. The experiment situations were modeled using the computational code. The obtained results showed the flexible and cheap material could be used as a good neutron shield while it acts as a very weak shield for gamma rays. Also there is good conformity between simulation and experimental data with maximum 37% relative discrepancy.

KEYWORDS

NBR
Neutron dose rate
Gamma dose rate
Shielding power
MNSR

HISTORY

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1 Introduction

Nitrile rubber, also known as nitrile butadiene rubber, NBR, is a synthetic rubber derived from acrylonitrile (ACN) and butadiene. NBR is the polymerization of Acrylonitrile (CH_2=CHCN) and Butadiene (CH_2CH-CH=CH_2) into one large multiple-unit chains. NBR is used in the automotive and aeronautical industry to make fuel and oil handling hoses, seals, grommets, and self-sealing fuel tanks. It is used in the nuclear industry to make protective gloves. NBR's stability at high temperatures from 40 to 108 °C makes it an ideal material for aeronautical applications. NBR is also used to produce moulded goods, footwear, adhesives, sealants, sponges, expanded foams, and floor mats (Threadingham et al., 2000; Ceresana, 2018).

Setiawan et al. (2020) reported that a rubber neutron shielding, could be made flexible, in accordance with the shape of the space that must be protected from neutron radiation. They explained although Polyethylene resin is effective and is the most popular for neutron shielding, but has poor heat resistant while concrete is more effective for neutron rays and gamma rays and can withstand high temperatures, but is not suitable for additional protection in tight and confined spaces. They investigated attenuation factor of NBR, without and with fillers, Gd (Gadolinium), and B_4C (Boron Carbida) each composition has 5% by weight. They calculated attenuation coefficient (μ) using a comparison of the flux before and after the neutron beam through the sample using gold plates. Their results showed the neutron flux decreases 2.032, 3.772, 4.359 times for NBR, NBR-Gd, and NBR-

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B_4C respectively. The results obtained from the attenuation coefficient of NBR rubber samples without filler, with Gd filler and B_4C filler found that the greatest attenuation coefficient on NBR rubber with B_4C filler. Their work also shows no significant reduction in neutron intensity for NBR with a thickness of more than 1.4 cm (Sumirat et al., 2020).

Hegazi (Hegazi, 2018) investigated NBR nanosilica-added as shielding materials for gamma radiations. In this work, Rubber silica nanocomposites were prepared by mixing nitrile rubber with surface modified and unmodified nanosilica. The Linear attenuation coefficient was measured experimentally and calculated for NBR rubber with different concentration of nano silica. This report shows that the addition of nanosilica particles improves the mechanical and nuclear properties of NBR rubber. The increase of modified nanosilica particles increases the total γ-ray attenuation coefficient for all energies. This study shows that NBR composite with surface modified with nanosilica particles, has higher tensile strength and elongation at break (Hegazi, 2018). Jumpee et al. (Jumpee and Wongsawaeng, 2015) reported that the most effective neutron shielding material can be obtained by appropriately mixing high hydrogen-content materials, heavy elements and thermal neutron absorbers. High hydrogen-content materials can undergo elastic scattering with fast and intermediate-energy neutrons. Hence, they computationally designed an optimized flexible and lightweight neutron shielding material. Their results from the MCNP transport code showed that the 10-mm thick sample #2 (NR +SBR +B_2O_3) and 100 mm thick sample #10 (4 alternating layers of NR+Fe_2O_3/NR+SBR+B_2O_3) exhibited excellent neutron and secondary gamma ray shielding (Jumpee and Wongsawaeng, 2015).

Aziz et al. (Aziz et al., 2023) investigated the production of composite materials through the combination of natural rubber and acrylonitrile butadiene rubber, along with nano-silica-loaded bismuth (III) oxide, in varying concentrations ranging from 0 to 45 parts per hundred parts of rubber (phr). The gamma attenuation properties of the composites at different concentrations of B_2O_3 were measured in their work. Additionally, the mechanical properties of the resulting composites, including hardness, tensile strength, and elongation, were tested by them. Their obtained results indicated that the prepared composites could be used for several radiation-protection applications. Overall, they explained that the addition of bismuth to the rubber blend can improve the mechanical properties and make it more suitable for radiation-shielding applications (Aziz et al., 2023). Other rubbers are widely investigated and used for construction of proper neutron and gamma shields.

Güngör et al. (Güngör et al., 2018) investigated the effect of bismuth (III) oxide addition on the mechanical, thermal, rheological and gamma ray attenuation capability of Ethylene Propylene Diene monomer (EPDM) rubber. In their work, EPDM samples including 10%, 20% and 30% (w/w) bismuth oxide were irradiated with doses of 25.6, 39 and 57 kGy up to irradiation period of 133 days using a gamma ray source with an initial dose rate of 18.3 Gy.h^{-1}. Their results showed that attenuation ratio from 68.6 to 94% were obtained for samples with different EPDM composite material thickness values (Güngör et al., 2018). Some of these rubbers are prepared to be both neutron and gamma shield.

Onjun et al. (Onjun et al., 2014) indicated that different amounts of boron compounds (i.e. boric acid and borax) are mixed with natural rubber to form a thermal neutron radiation shielding block. With proper treatment on natural rubber, uniform distribution of a boron compound can be achieved, which results in an enhancement of thermal neutron shielding capability. It is found that the neutron shield capability of those shielding blocks depends sensitively on both borax and boric acids concentration, and the thickness of the shielding block. Even though the boron content in a natural rubber shielding block mixed with boric acid is higher than that with borax at the same amount, the shielding blocks with borax can shield thermal neutron better due to the loss of boric acid by a bubble formation during the process. Their results showed that an inclusion of lead oxide (PbO_2) in the shielding block can slightly increase its thermal neutron shielding capability and results in good gamma ray shielding capability, in which a minimum thickness of shielding blocks is required to be effective (Onjun et al., 2014).

Simulation evaluation using the developed nuclear codes helps to better clarification of the shield material behaviour against the neutron spectra available at any section of a nuclear reactor. Košt'ál et al. (Košt'ál et al., 2021) investigated different neutron filter materials using MCNPX6 code while they experimentally studied 3 different filters B_4C, Cd, and In, on the case of the ^{55}Mn(n,γ) reaction in LR-0 reactor for nuclear data validation and verification. Measured values of that cross section in the given filtered reference spectra were reported by them (Košt'ál et al., 2021).

Flexible neutron and gamma shields such as Benzophenone (BP) are used as a filler to improve the radiation resistance performance of composites. This neutron and -ray shielding material demonstrate superior flexible performance, excellent thermal stability, good radiation resistance, and wide potential applications. Also flexible, and heat-resistant neutron and gamma-ray shielding materials are developed to act as sealing material around gaps or ports for example in a tokamak fusion device to prevent radiation streaming (Sukegawa and Anayama, 2014).

Hence, in the present work, shilelding performance of 2, 4, 6, 8, 10 and 12 cm NBR layer was theoritically and experimentally invesigated using MCNPX2.6 computational code as well as the Isfahan Miniature Neutron Source Reactor (MNSR) vertical beam line. In this study, some 6×6 cm^2 NBR sheets with 2 cm thickness were used so that they were added top of the beam line one by one to make 12 cm thickness finally.

This manuscript tests experimentally and theoretically, the NBR as flexible possible neutron shield material for the first time. Also, a benchmark study of MCNPX computational code for such calculations is done via the code output comparison with the experimental data.

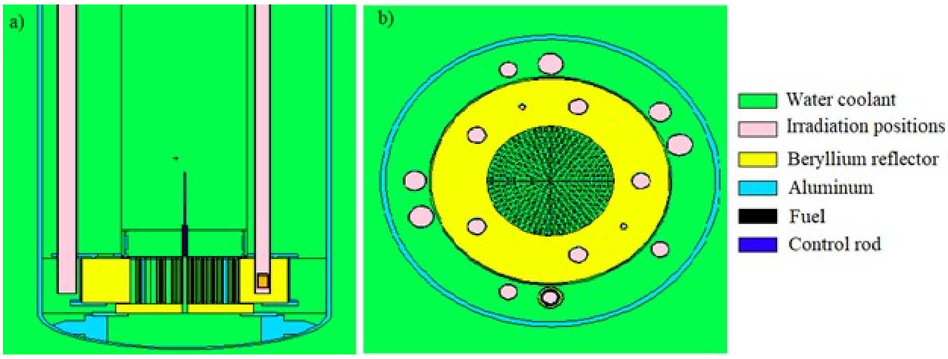

Figure 1: Cross-sectional view of the Iranian MNSR a) $x-z$ plane b) $x-y$ plane.

2 Experimental

2.1 Preparation of the materials

The Isfahan Miniature Neutron Source Reactor (MNSR) is a low temperature and tank-in-pool type reactor. It is a small and safe nuclear research reactor and the core design embodies highly enriched uranium (90.2 w/o U-235) as fuel, light water as moderator, coolant and biological shielding, and metal beryllium as reflector. It is designed for the use in universities, hospital and research institutes mainly for neutron activation analysis, production of short-lived radioisotopes, education and training. There are five inner and five outer irradiation sites in and around the annular beryllium reflector. MNSRs use only one control rod for reactivity control and for shutting down the reactor during normal and accidental conditions. The core, cooled by natural convection, is located inside and near the bottom of water-filled cylindrical container which is suspended in a large pool of water (Chengzhan and Yongchun, 1994).

Technical specifications of the MNSR are as shown in Table 1 and a cross sectional view of the reactor is shown in Fig. 1. This reactor is equipped with several vertical beam lines to perform out-of-core irradiation experiments such as neutron radiography and prompt gamma neutron activation analysis (Dastjerdi et al., 2019; Mokhtari and Dastjerdi, 2023; Dastjerdi et al., 2023a; Ghasemi et al., 2023; Dastjerdi et al., 2023b; Rahmati et al., 2023).

MCNPX code was used to simulate the MNSR core and vessel. The code is a general radiation transport code with the Monte Carlo method to trace different types of particles within a vast spectrum of energies. First, the geometry of the system is simulated using this code. The output production process is done with the help of tallies. Using tallies and useful lateral cards, some quantities such as flux, flow, dose, current, the angular distribution of particles, the spatial distribution of particles, energy spectrum, etc. can be calculated (Pelowitz, 2008).

Neutron and gamma sources of MNSR at the 4 cm beam line exit were written in MCNPX input and the gamma and neutron dose rates top of the modeled NBR layer were calculated using different thicknesses of the NBR layer. DE/DF tally cards and ANSI/ANS-6.1.1-1977 flux to dose conversion factors were used to calculate the gamma dose rates. Flux to dose conversion factor of NCRP-38, ANSI/ANS-6.1.1-1977 was used to calculate the neutron dose rates (Pelowitz, 2008).

Table 2 showes thermal ($E_n < 0.4$ eV), epithermal ($E_n < 1$ keV) and fast neutron ($E_n > 1$ MeV) flux measured top of the MNSR 4.1 cm diameter beam line (Vatani et al., 2023).

Table 1: Main specifications of Iranian MNSR (Rahmati et al., 2023).

Parameters	Description
Reactor type	Tank-in-pool
Nominal power	30 kW
Fuel	UAl_4 dispersed in Al
U-235 enrichment	90.2%
Fuel density (g.cm^{-3})	3.456
Number of fuel elements	343
Core diameter	23 cm
Core height	23 cm
Moderator and coolant	Light water

Table 2: Neutron flux top of Iranian MNSR vertical beam line with 4.1 cm diameter (Vatani et al., 2023).

Parameter	Value
Thermal neutron flux (n.cm^{-2}.s^{-1})	$1.69E+06$
Epi-thermal neutron flux (n.cm^{-2}.s^{-1})	$1.60E+05$
Fast neutron flux (n.cm^{-2}.s^{-1})	$1.56E+05$
Total neutron flux (n.cm^{-2}.s^{-1})	$2.01E+06$

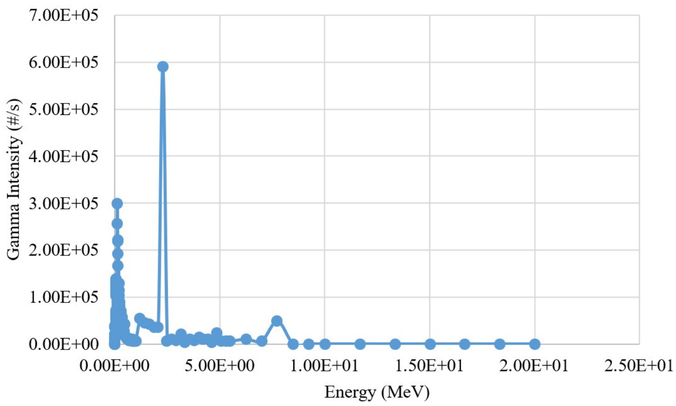

Figure 2: The calculated amma spectra at outlet of the vertical beam line.

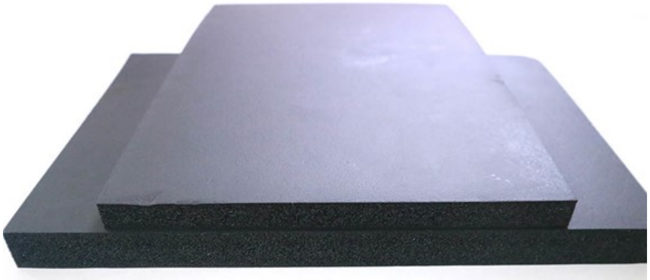

Figure 3: The original prepared NBR sheet view.

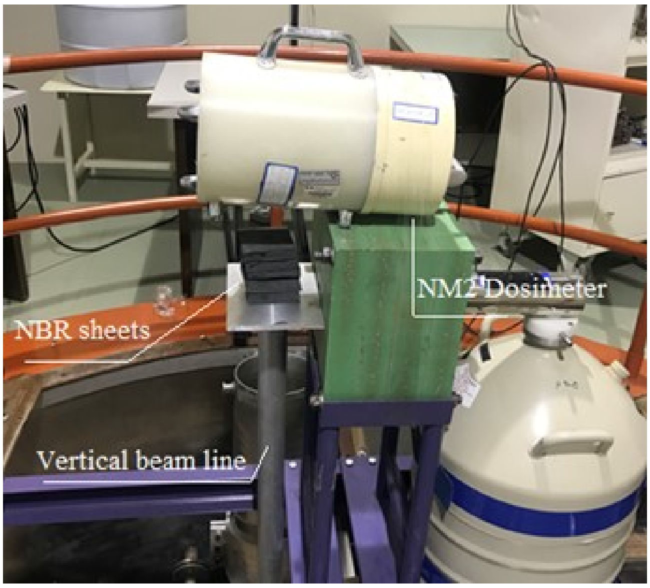

Figure 4: The 6×6 cm^2 cutted NBR sheets and NM2 neutron dosimeter top of MNSR vertival beam line.

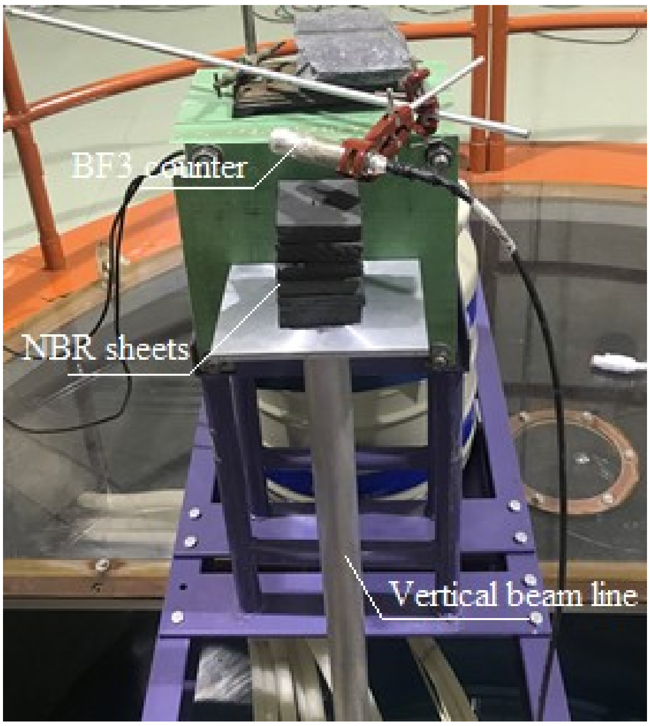

Figure 5: The 6×6 cm^2 cutted NBR sheets and BF_3 counter top of MNSR vertival beam line.

Beam diameter is 4.1 cm. Neutron beamline is completely parallel (L/D is over 145 or $\theta = 0.39°$; θ is the divergent angle of beamline). The neutron flux inhomogeneity from center to edge is less than 2%. However the beam divergence was modeled inside the MCNPX input.

The calculated gamma spectra on the vertical beam line was used to calculate gamma dose rates using MCNPX code at top of the beam line outlet (Fig. 2).

Home-made NBR sheets with 2 cm thickness and 0.14 g.cm^{-3} density was prepared according to Fig. 3. The sheet was cutted to 6×6 cm^2 pieces so that cover the MNSR beam line outlet area. The pieces were placed top of MNSR vertical beam line according to Fig. 4. As is seen in Fig. 4 a neutron dosimeter type NM2B made by Nuclear Technlogy LTD, UK was used to measure neutron dose rate reduction by adding the 2 cm NBR layers.

Also to measure count rate reduction of thermal neutrons, a BF_3 counter was used to determine the NBR shielding performance against the thermal neutrons (Fig. 5)

In overal, 1) The pieces of NBR were exposed to the PGNAA neutron beam line and a BF_3 detector recorded the neutrons passing through them (to investigate the attenuation of thermal neutrons). 2) The pieces of NBR were placed against the PGNAA neutron beam and the neutron dose passing through them was measured with the NM2 neutron dosimeter (to check the weakening of the total dose of neutrons). 3) A cadmium layer with a thickness of 0.5 mm was placed in front of the PGNAA neutron beam according to Fig. 6 and NBR pieces were placed in front of the neutron beam passing through the cadmium and the neutron dose passing through them was measured with the neutron dosimeter (to investigate the weakening of epithermal and fast neutron doses). Measurements were made at 10 sec intervals. Each measurement was repeated 12 times and the average of the responses was considered as the final response.

The irradiation condition was modeled using MCNPX code according to Fig. 7 to calculate the neutron and gamma dose rate reduction of NBR sheets theoritically. Also, the NBR sheet shielding abilityes for Co-60 gamma rays was investigated experimentally using 10 μCi Co-60 source according to Fig. 8. Also the experiment condition was modeled using MCNPX code to calculate the NBR shielding abiltiy using different thickness of the felexible sheet placed in front of the 10 μCi Co-60 source.

3 Results and discussion

Total neutron count was measured top of PGNAA beam line of MNSR when 2 cm NBR sheets were added respectively. The measurements using BF_3 counter showed 2 cm NBR reduced the thermal neutron count up to 69.31% and 4 cm sheet reduced the value up to 91.5%. Clearly light elements of nitrogen, carbon and hydrogen inside the NBR sheet causes noticably shielding of thermal neutrons. Adding more thicknesses up to 12 cm did not reduce the count rate noticably. which shows if thermal neutrons are to be shielded it seems 4 cm NBR flexible sheet is adequate.

Table 3: NBR sheet shielding ability for thermal, epithermal+fast and total neutrons of MNSR vertical beam line.

Parameter	Background	2	4	6	8	10	12
Count by BF_3	80184	24611	7175	2234	671	235	85
BF_3 count reduction factor	-	3.25	11.17	35.89	119.49	341.20	943.34
Total neutron dose rate by NM2 (μSv.h^{-1})	4253	3624	3281	2888	2624	2407	2238
Reduction value of total neutron dose rate (%)	-	14.79	22.85	32.09	38.30	43.40	47.38
Fast and Epithermal neutron dose rate by NM2 (μSv.h^{-1})	3269	2983	2726	2501	2336	2169	2039
Reduction value of Fast and Epithermal neutron dose rate (%)	-	8.75	16.61	23.49	28.54	33.65	37.63

Figure 6: The 6×6 cm^2 cutted NBR sheets and cadmium sheet top of MNSR vertival beam line.

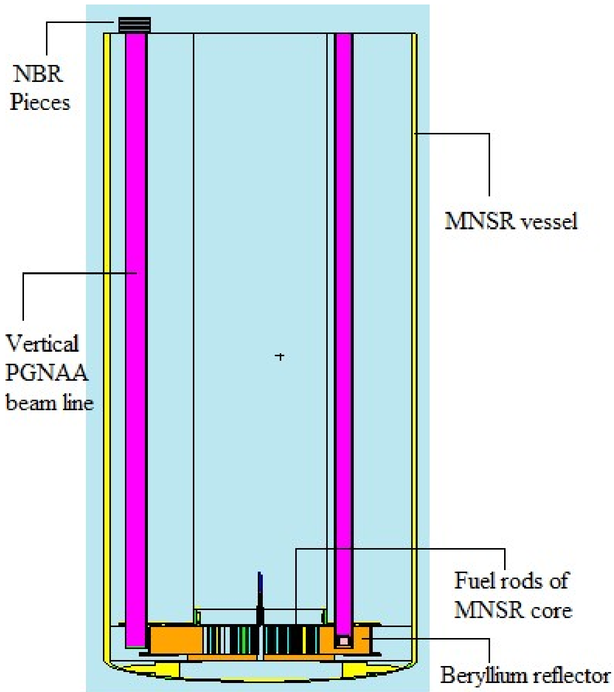

Figure 7: Simulation of the 6×6 cm^2 cutted NBR sheets top of MNSR vertival beam line.

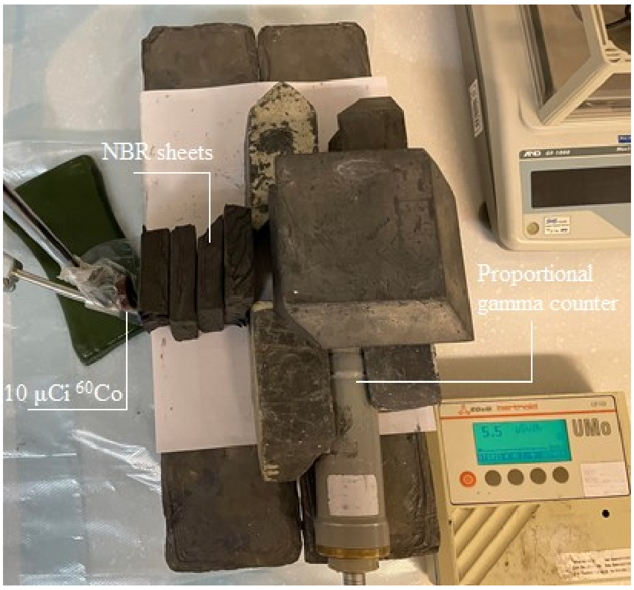

Figure 8: Test of the 6×6 cm^2 cutted NBR sheets in front of Co-60 source.

Total neutron dose rate measurements using NM2 dosimeter showed that using 12 cm NBR sheet caused the value decreases about 47.88%. Then if the sheet is going to be used against a neutron spectra involved fast and epithermal neutrons too, 12 cm of it remove only half of total neutron dose rate. Application of a cadmium sheet before NBR pieces showed 12 cm of it could shield fast and epithermal neutrons about 37.63% as a result of neutron slowing down during their propagation inside the NBR sheet (Table 3).

MCNPX simulations showed if 2 to 12 cm NBR sheet is used in front of neutron spectra emerged from PGNAA beam line of MNSR, total neutron dose rate would be shielded from 15% to 63% respectively. There is good agrement between the experimental measurements and the theoritically calculated neutron dose rates with 12% to 37% relative discrepancies. The noticable discrepancy between the valuses obtained using 12 cm NBR sheet may be arisen of this fact that the NBR sheets modeled inside the computational code have good integrity while in practical, there is gap between the sheets placed top of each other (Fig. 9).

Whereas the NBR sheet involves carbon element, prompt gamma of the sheet which receives neutron exposure was calculated using MCNPX code. The obtained calculations showed by increasing the NBR sheet thickness, secondary gamma dose rates (gammas from the sheet activation) rises from 0.07 μSv.h^{-1} to 4.71 μSv.h^{-1} which is not significat in comparison with first gamma rays' dose rates (MNSR spent fuel gammas emarged from the nuclear core) (Fig. 10). Whereas these secondary gamma dose rates were noticeably less than the backgrounds of the experimental, no experimental measurement was possible at this stage.

Also fast+epithermal neutron dose rates were calculated using different thicknesses of the NBR sheet placed top of the vertical PGNAA beam line of MNSR. The carried out simlations showed there is 2% to 31% relative discrepancies between the theoritical and experimental values of 2 and 12 cm NBR neutron dose rates (Fig. 11). The same reason mentioned above is valid for the observed noticable discrepancies between the experimental and simulation fast+epithermal neutron dose rates of 12 cm NBR.

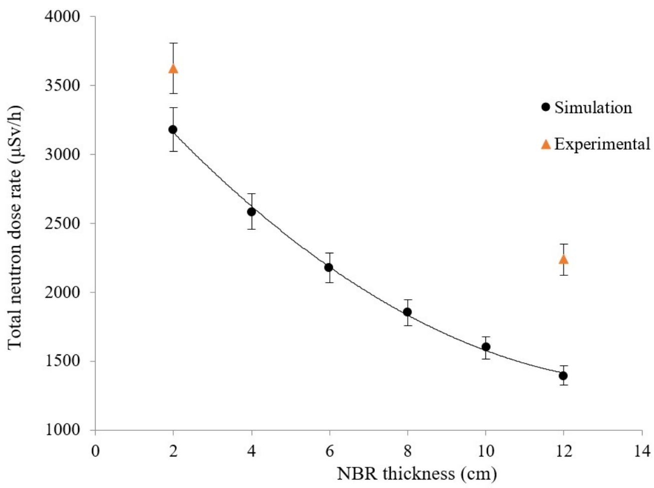

Figure 9: Simulation of total neutron dose rate variation on NBR sheet thickness added top of MNSR neutron beam line, NM2 was used as the neutron dosimeter.

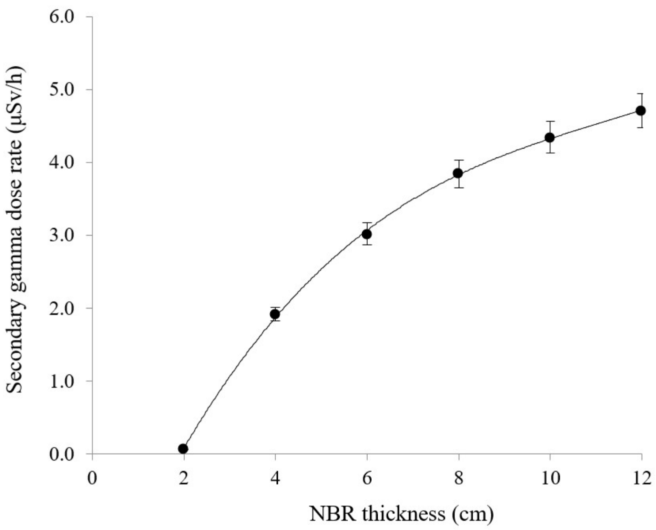

Figure 10: Simulation of secondary gamma dose rate variation on NBR sheet thickness added top of MNSR neutron beam line, LB1236 was used as the gamma dosimeter.

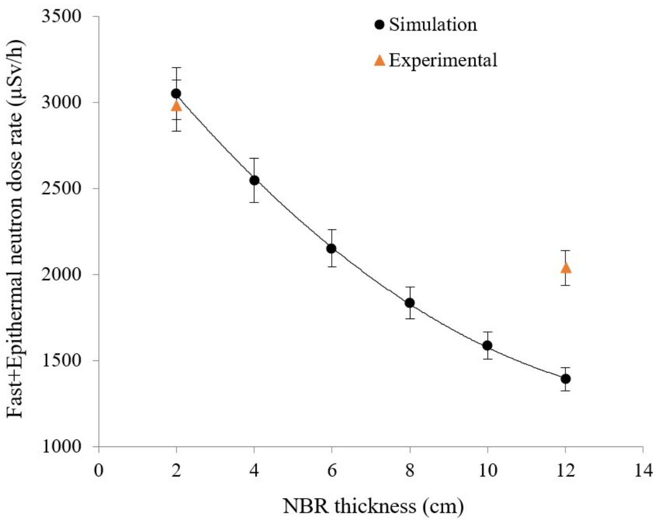

Figure 11: Simulation of Fast+Epithermal neutron dose rate variation on NBR sheet thickness added top of MNSR neutron beam line, NM2 was used as the neutron dosimeter, cadmium sheet was used to remove thermal section of the neutron specta.

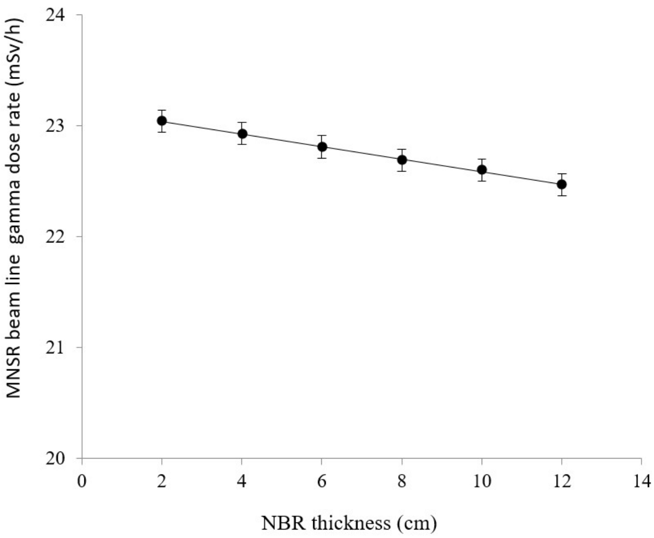

Figure 12: Simulation of first gamma dose rate variation on NBR sheet thickness added top of MNSR beam line, LB1236 was used as the gamma ray dosimeter.

Obviously it is not expected NBR sheet acts as a good shield for gamma rays. Anyway, its shielding ability against the gamma spectra emerged from MNSR core was investigated theoritically. Figure 12 showes there is about 2.4% reduction in gamma dose rate according to the simulation data. The measured experimental dose rate of MNSR PGNAA beam line is 23.2 mSv.h^{-1}. It should be taken in attention whereas the beam line involves low-energy gamma, the NBR material tested as gamma shield for this beam line could reduce some of the low-energy gammas while has not noticable effect agains Co-60 high-energy gammas as is seen in Fig. 13.

The gamma shielding ability of NBR material for hard gamma rays was theoritically and experimentally investigated using 60Co source. Carried out calculations showed by adding the NBR sheets from 2 up 8 cm the gamma dose rate would change less than 0.08 μSv.h^{-1} which was in tolerance range of the used gamma dosimeter. Simulation of the test condition showed the same behaviour so that the gamma dose rate decreases about 1.4% (Fig.13).

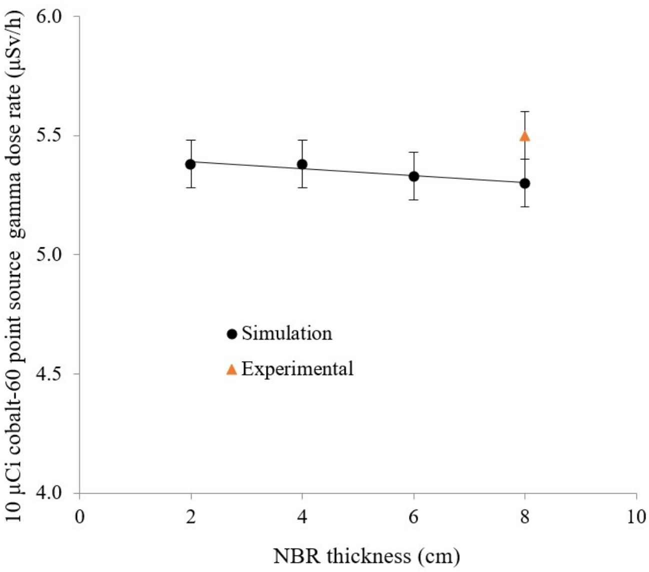

Figure 13: Simulation of first gamma ray dose rate variation on NBR sheet thickness added top of MNSR neutron beam line, LB1236 was used as the gamma ray dosimeter.

4 Conclusions

Natural rubber, which is abundant in hydrogen, can lower the energy of a neutron. For sealing the gaps between the main shields and the neutron sources, flexible materials are suggested. Many of these materials with different combinations, which ensures more neutron shielding are being investigated and tested. The present work experimentally and theoritically investigated the NBR sheet as a felexible neutron shield. Test of NBR sheet using vertical beam line of MNSR showed that the material with thichness of 12 cm decreases total neutron dose rate about 47% according to experimental reported data. Also the material plays good role in reduction of fast and epithermal neutron dose rate because of its composion (carbon, hydrogen and nitrogen) which is good moderator of neutrons. Application of 12 cm of NBR decreases fast and epithermal neutron dose rate about 37.63%. Computational investigation always helps that different amterials to be evaluated when experimental tests are expensive or difficult. In the present work, the simulation data were in good conformity with the experimental ones with maximum 37% relative discrepancy. In overall, NBR material could be suggested as a cheap and flexible neutron shield material especially when a sealing is needed or disordered surfaces are to be shielded. The sheet has minor effect on gamma rays on which 12 cm of it could decreases the gamma ray dose rate about 2.4% in the case of MNSR beam outlet.

Conflict of Interest

The authors declare no potential conflict of interest regarding the publication of this work.

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