Boron Carbide Contents Variation Effect on Shielding Properties of Styrene Butadiene Rubber Composites

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Abstract

The main objective of this experimental approach is to find out effect of boron carbide contents on shielding properties of composite material. The Formulation of Styrene Butadiene Rubber composites have been prepared by using Boron Carbide contents, ranging from 5% to 25%. These composites have been subjected to various tests and experiments have been conducted to study thermal neutron shielding properties of these composites by neutron irradiation. Moreover, linear attenuation coefficient has been calculated from this data. Monte Carlo N-Particle transport code simulations have been used to compare the simulated results to experimental results. Mechanical properties such as hardness, swelling uptake studies, and thermal gravimetric analysis have been carried out, and it is observed that hardness increased 5% and swelling index values, specifically lower for water, decreased by increasing boron carbide contents. For thermal neutron shielding properties, graphs show the slope of composite with higher concentration of boron carbide is steeper than the one with lower amount of boron. Furthermore, both the experimentally calculated and MCNP estimated values are in good agreement. These results suggested that shielding capability of composites increases with the boron carbide amount. More the boron carbide content, less thickness of composite will serve the purpose of shielding.

Keywords: *Styrene Butadiene Rubber, Radiation Shielding, Polymer Composites, Boron Carbide, Neutron Shielding.*

INTRODUCTION

Development of nuclear technologies render its use in almost every field of life including industry, medicine, research, and others, resulting in emission of neutrons from various devices such as medical vaults and doors, hot cells, nuclear storage and transport containers, nuclear waste management sites, particle accelerators, and nuclear detection system [1]. Neutrons are produced during fission reactions as a result of cosmic ray collision with nitrogen and oxygen atom at upper atmosphere and interaction of high energy particles with matter. However, excessive exposure to these radiations may cause harmful effects on humans and environment. The only way to protect the humans and environment is to shield these radiations in control and containment [2].

Radiation protection always remains a serious concern among the scientists. Different modeling approaches are used which focus on the shielding of neutrons produced in different environment [3]. Nuclear radiation shielding applications require such material which could provide safety and protection for environment and people, and these materials must exhibit high hydrogen content and less weight at acceptable cost [2]. Hydrogen is effective for fast neutrons while boron can eliminate the thermal neutrons more effectively because of high neutron absorption cross section [1]. Protection against nuclear radiation is sometimes achieved by thickly shielded lead plates, and cast reinforced concrete between the source and human or environment. However, the problem of making or moving the thick protective layers, and it is difficult to repair soon once it is destroyed. That's why it cannot meet the development needs of the nuclear power industry. Therefore, the composites filled with ^{10}B have the advantage of convenience and safety in the nuclear environment which has become the focus of the nuclear protection research [4]. There have been several studies of polymeric composition containing materials with high neutron cross section to determine their thermal neutron shielding properties and mechanical properties [5-8]. Materials made of polyethylene or epoxy resin mixed with boron compounds are widely used as neutron shielding materials, but they usually exhibit sizable weight and volume which results in low-flexibility performance. The Proper selection of materials and thickness are some of the requirements for appropriate and effective shielding against neutrons. It involves optimally analyzing the weight, volume and cost considerations. Generally, it requires lighter material that has higher ability to attenuate the neutrons. Nurazila Mat Zali et al studied the effect of B_4C contents on properties of thermoplastic natural rubber composites as neutron shielding material [9]. Their work proved that Polyethylene is effective and the most popular resin for neutron shielding. However, its heatproof temperature is fairly low. Concrete can endure high temperatures, but it is not suitable for additional shielding in narrow and restricted spaces such as the tokamak fusion device [3].Water, lead, graphite, iron, boron, concrete, and polyethylene are some of the commonly used shielding materials with their own advantages and disadvantages [7]. Polymer composites are the materials that can be designed to effectively attenuate neutrons by virtue of properties like efficiency, light-weight, cost-effectiveness and flexibility. Polymer composites filled with ^{10}B have the advantage of convenience and safety, greater durability, withstanding ability of wider range of temperatures, and these can be molded into desired shapes and less space is occupied by them [4]. In present research, SBR/B_4C composites were prepared. The effects of different concentrations of B_4C loaded to SBR and its mechanical properties as well as attenuating properties of the neutrons were studied.

MATERIALS AND METHODS

The main raw materials were selected from simple elements or compound materials, taking into consideration cost, radiation shielding performance, physical and thermal properties. Styrene butadiene rubber composites with varying contents of boron carbide were prepared by using two-roll mill. All the ingredients were mixed in two-roll mill according to the recipe of the composition. Mixing time was about 25 minutes. Other chemicals were added in rubber to change its softness, rupture energy, fatigue resistance and abrasion resistance. Mass fractions of each composite of the SBR were determined for MCNP stimulations, and are mentioned in table-1:

Table 1: Mass fractions of elements in each SBR composite

Sample Type	Material Composition	Density (g/cm^3)	Element Contents (Mass Fraction)						
			Nitrogen	Sulfur	Zinc	Oxygen	Hydrogen	Boron	Carbon
0	**SBR_0**	1.04	0.00187	0.019	0.028	0.0089	0.086	0.00	0.855
1	**SBR_5**	1.06	0.0018	0.0180	0.026	0.0084	0.081	0.049	0.815
2	**SBR_{10}**	1.10	0.0016	0.0168	0.024	0.0078	0.076	0.0997	0.77
3	**SBR_{15}**	1.15	0.00152	0.016	0.022	0.0073	0.070	0.149	0.73
4	**SBR_{20}**	1.19	0.0014	0.014	0.02	0.0067	0.065	0.1996	0.69
5	**SBR_{25}**	1.22	0.00128	0.013	0.019	0.0061	0.059	0.248	0.65

Sheets were cut into pieces then compression molded by an electrically-heated hydraulic press at 150ºC and 15MPa for a suitable duration of each sample. To study neutron attenuation, Two different establishments were approached for neutron irradiation namely R-Block and PARR-I. R-Block irradiated the individual sheets of composites with the thermal neutron beam using polyethylene moderator and neutron source with neutron strength of 10^6 n/s. PARR-I utilized thermal neutron irradiation technique by using diffractometer to study the neutron shielding properties of SBR/ B_4C composites. The thermal neutrons were obtained from a 10 MW with a core flux of $1 \times 10^{14} n/cm^2/s$ and were converted to mono-energetic neutron beam through a mono-chromator (Cu_{220} crystal). Neutrons beam of wavelength of 1.2 A^0 was bombarded on various thicknesses of the samples for equal interval of time. The transmitted beam was collected with a BF_3 detector (Reuter Stokes, USA) with Ortec counter timer counting system.

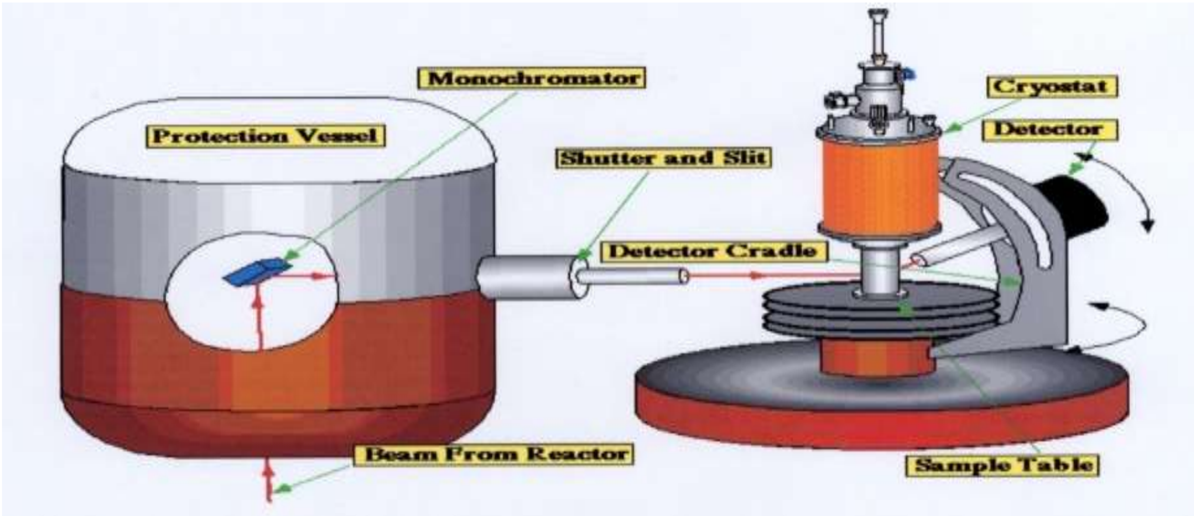

Fig. 1: Diffractometer for Neutron Attenuation Experiment

Linear Attenuation Coefficient, Relaxation Length and Half Value Thickness was measured using these equations

$$\ln I_\circ / I = \mu x \quad \text{..................1}$$

$$\lambda = 1/\mu \quad \text{...................2}$$

$$\chi_{1/2} = \ln 2/\mu \quad \text{..................3}$$

The mass attenuation coefficient was calculated by dividing the linear attenuation coefficient of each composite with its density. The mechanical behavior was also tested in which important mechanical properties are strength, hardness, ductility, and stiffness [10]. Hardness of composites was measured using Kori Durometer which measured Shore A hardness of composites.

To observe mechanical properties of materials factors considered the nature of the applied load and its duration, as well as the environmental conditions. Mechanical properties were determined by using a universal tensile testing machine. Three specimens of each composite were tested. Tensile strength, elasticity and elongation at break of these composites were measured [1]. Polymer stability in different media is of a great practical importance [11]. Swelling measurements were taken by using the following procedure. Tiny pieces of sample of approximately uniform size and weight were accurately weighed (W_1) by using an electric balance of sensitivity 10^{-4}g and immersed in 50 ml of respective solvent at room temperature for 24 h, 48 h and 72 h. After that the sample was taken out and placed between two pieces of filter paper. Then the sample was placed in weighing bottle and reweighed (W_2), the degree of swelling (SI %) was calculated by using the following equation [12].

$$\text{Swelling Index (SI \%)} = (W_2\text{-}W_1/W_1) \times 100$$

The swelling of the prepared composites was measured in various solvents. Density measurement of composites has also been done experimentally. Thermo gravimetric analysis or thermal gravimetric analysis (TGA) was performed for sample as it is heated, from typically 25^0C to 1000^0C. **M**onte **C**arlo **N**-**P**article (MCNP) transport code based techniques and widely used for solution of particle transport problems is also utilized for theoretical results.

RESULTS

In this project a comprehensive research has been carried out to prepare and characterized the SBR/B_4C composites. All the properties of these composites are compared briefly.

Mechanical Properties of Composites

The results of tensile testing and hardness testing are as follows: All of the six formulations were mechanically tested according to the parameters described in Material and methods section.

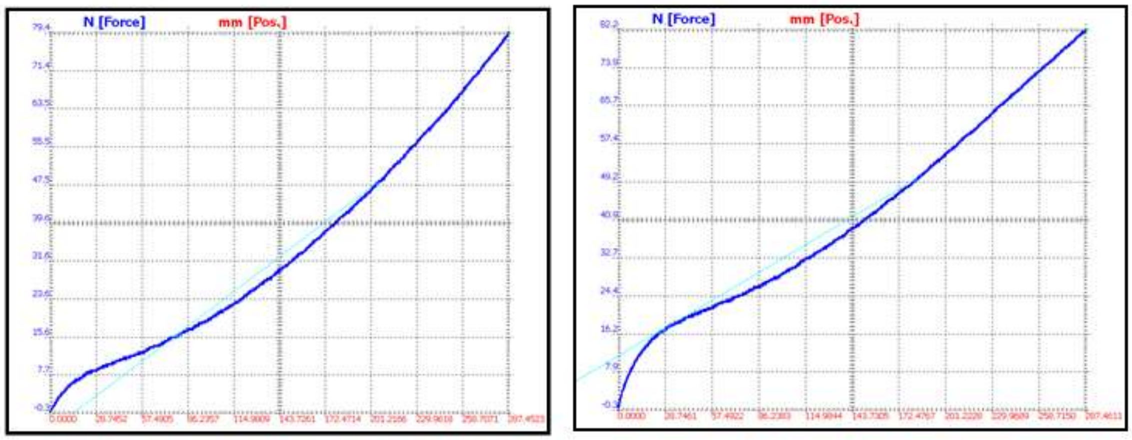

Fig. 2: Comparison of Stress-Strain curve for (a) SBR_0 and (b) SBR_{25}

Figure No.2 shows comparison of SBR_0 and SBR_{25} as a reference. As SBR_0 curve has less initial linear portion so its elasticity is less than SBR_{25}. Hence point of permanent deformation reaches earlier in SBR_0 curve than SBR_{25} curve. It means boron carbide content imparts desirable characteristics.

Tensile strength increased with increase in boron carbide content while % elongation at break decreases. This is due to the fact that more the tensile strength of composite, less it will be elongated and hence its % elongation at break decreases.

Study of Hardness

Shore A hardness of all six composites was measured and represented graphically. It can be seen hardness of the composites increases as boron carbide contents increased.

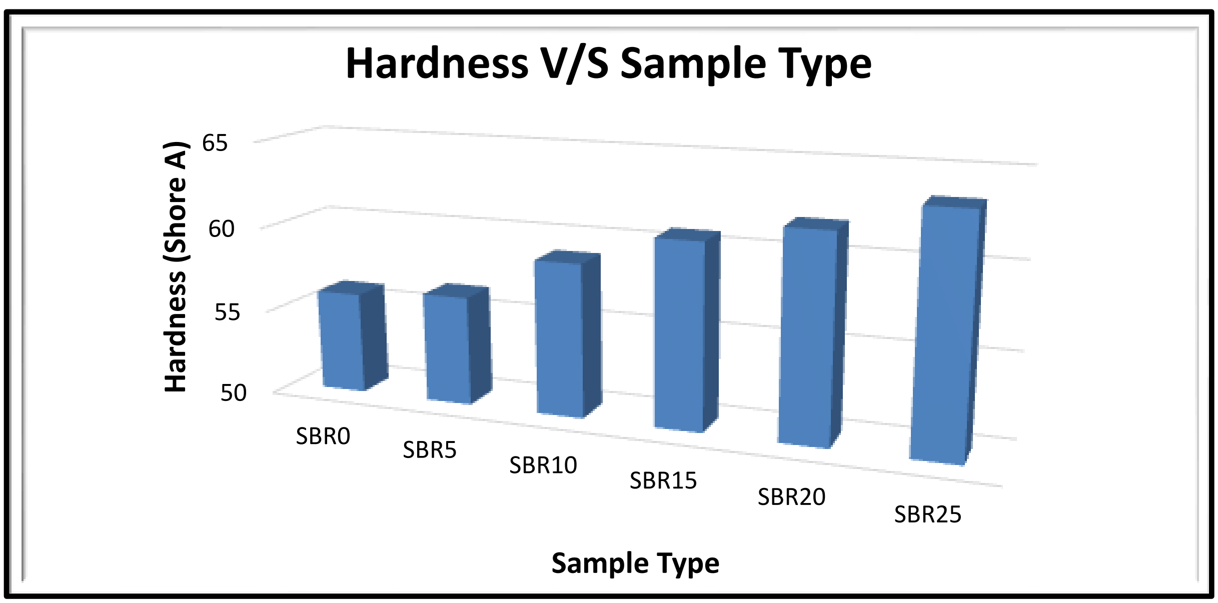

Fig. 3: Variation of hardness with boron carbide content

Moreover, it has also observed that hardness has increased 5% in SBR_0 to SBR_{25} by increasing boron carbide contents.

Solvent Uptake Properties of Composites

Swelling measurement describes the interaction of the solvent with particular material, and swelling index provides the measure of absorbed solvent. Swelling Index experiment was performed on composites to check the behavior of composites for each concentration in different materials. Three different time frames were selected to evaluate the swelling index. Data of swelling measurement after 24, 48 and 72 hours for composites is represented by three graphs.

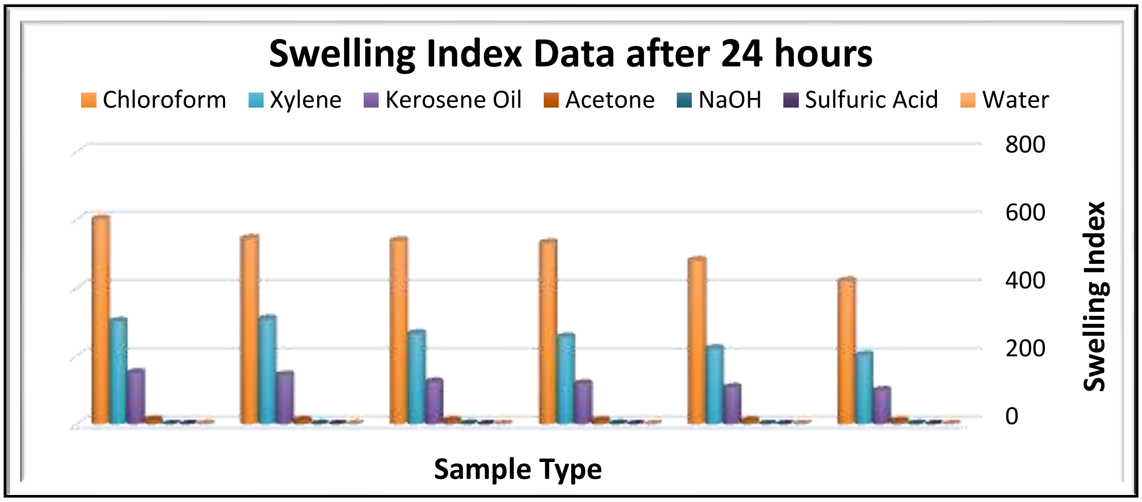

Fig. 4: Swelling index of solvents after 24 hours

After 24 hours reasonable solvent uptake occurs particularly in nonpolar solvents which illustrate the nonpolar nature of composites as "like dissolves like". While the swelling index values for all the composites are specifically lower for water. It's a desirable trait as water is the most commonly encountered solvent.

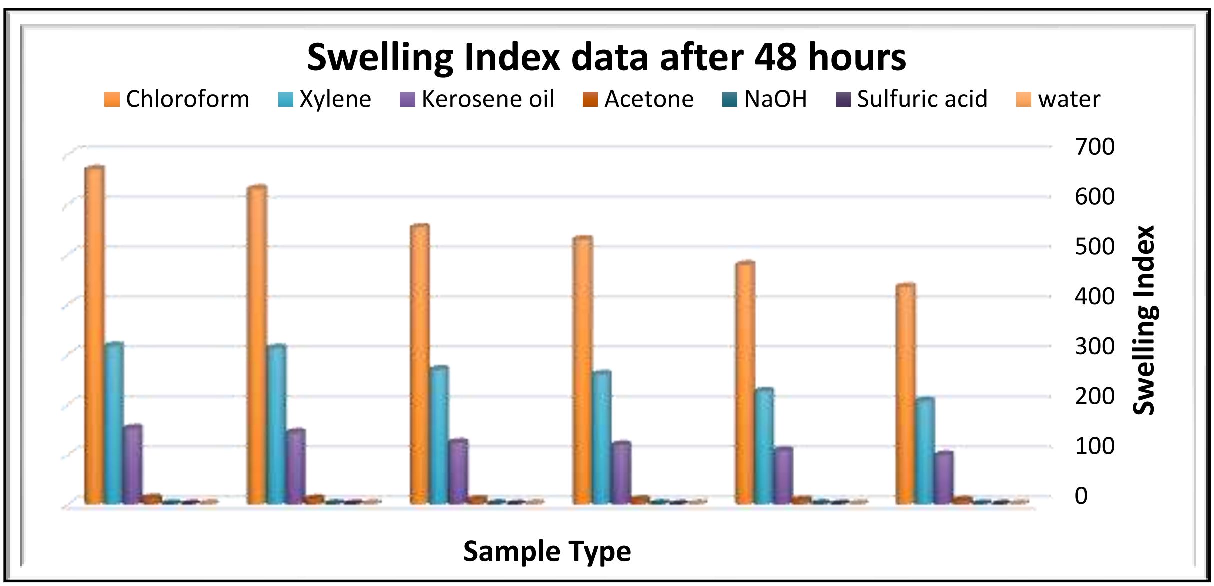

Fig. 5: Swelling Index of composites after 48 hours.

In fig. 5, it can be seen that after 48 hours slight changes occur in swelling index values. This suggests that composites are tending to be in equilibrium with the environment.

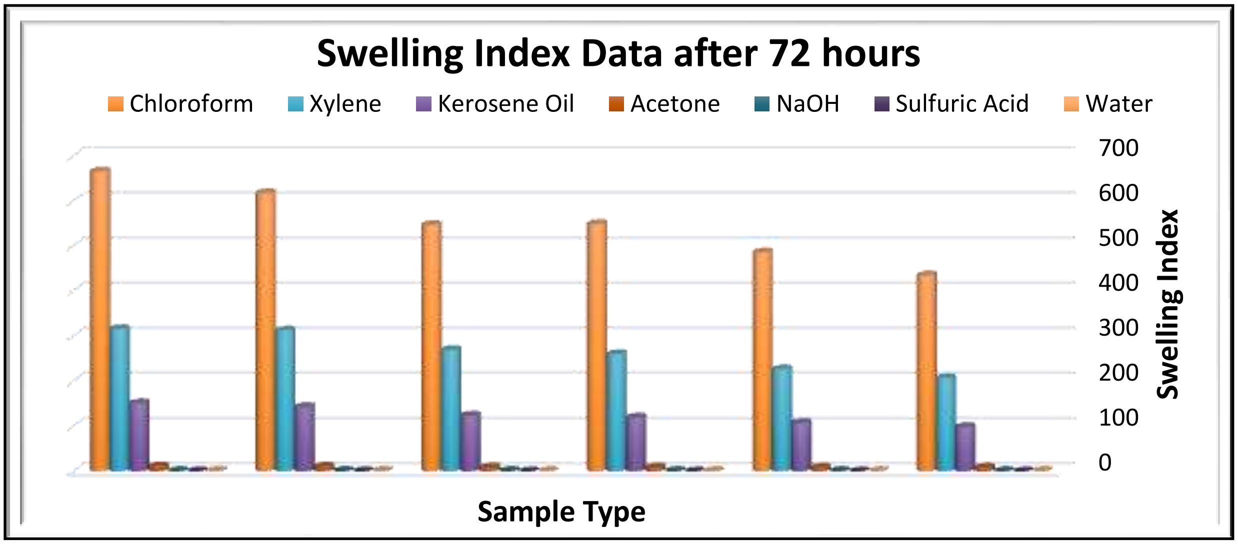

Fig. 6: Swelling Index data for composites after 72 hours

Figure no. 6 shows a complete swelling equilibrium was established after 72hours: there is no remarkable change in swelling index after certain period of time. Additionally, inconsiderable difference is observed in swelling for 2nd and 3rd intervals.

Moreover, with increase in boron carbide content in composites, swelling index decreases no matter solvent is polar or nonpolar. This can be attributed to the fact that addition of more filler in composites left the less room for solvent molecules to get absorbed.

Thermo-gravimetric properties of composites:

Thermo-gravimetric properties are very important in analyzing the thermal stability of the polymers under consideration. Four TGA curves were generated using Lab scale TGA machine as shown in figure no. 7

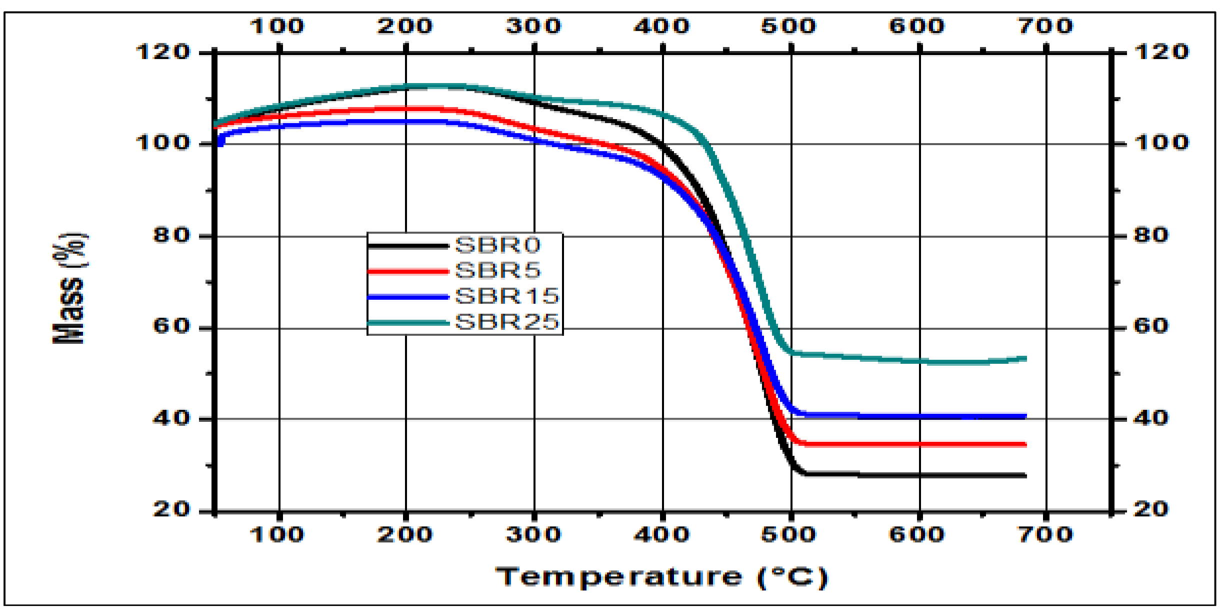

Fig. 7: Curves of thermal stability of composites

TGA curves illustrates, all the composites are stable against temperature up to 250^{0}C while further increase in temperature results in minor mass loss up to 350^{0}C. Furthermore, major mass loss occurs within the temperature range of 400^{0}C to 500^{0}C that can be seen in figure no. 7.

Nuclear properties of composites

Neutron irradiation techniques have been employed and Variation in counts is observed with change in boron carbide content. The indication of decrease in count rate with increase in boron carbide contents in the composites suggests that boron carbide increases shielding capability of composites. A desirable feature obtained without distorting mechanical and other properties of materials. Furthermore, decrease in count rate is also observed when thickness of sheet was doubled irrespective of type of composite as predicted by Beer-lambert's law [13]. The facts are represented graphically in figure no. 8 to understand the effect of boron carbide contents on neutron shielding capability.

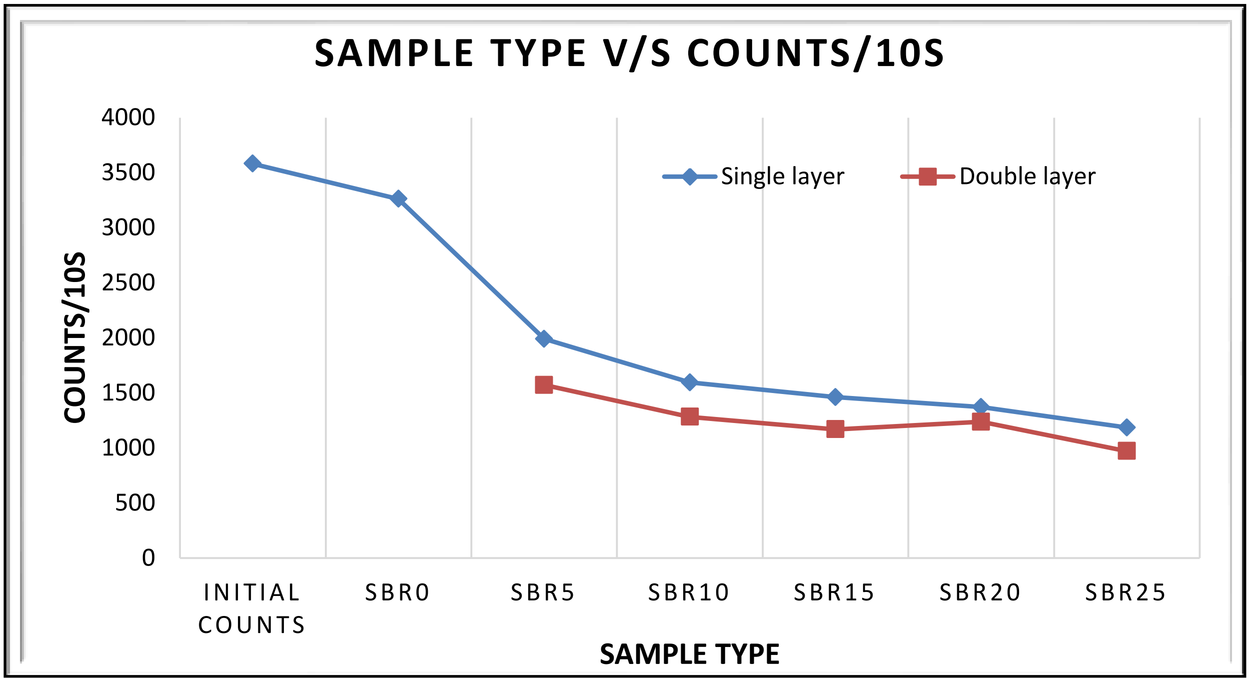

Figure No. 8: Curves of neutron count rate with Boron Carbide content

Another neutron irradiation experiment was performed using diffracto-meter arrangement that provides the variation of neutron count rates as a function of thicknesses where the Initial counts were 75000 counts/sec. Decrease in counts with increase in thickness is observed as predicted by Beer-lambert's law [13]. Furthermore it can be seen that with increased boron carbide contents less thickness of any material can be used for shielding and boron carbide addition is economical as well. Variation of neutron counts with various thicknesses of composites are plotted and shown in figure no. 9.

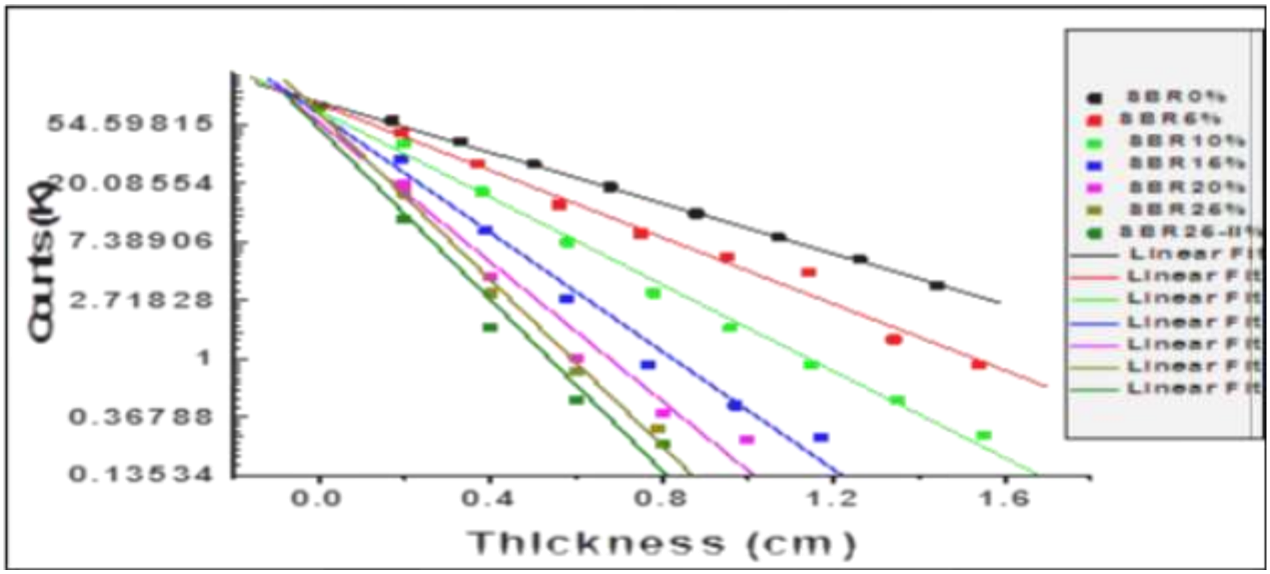

Figure No.9: Variation of neutron counts with thickness of sample

Negatively sloped lines indicating the fact that increase in thickness of composites decrease the count rate. Besides this, the slope of composite with higher concentration of boron carbide is steeper than those having low amount of boron carbide. This behavior reflects the boron carbide's neutron shielding capability to composites.

Linear Attenuation Coefficient of composites has been determined experimentally. It can be seen that with increase in boron carbide contents, linear attenuation coefficient is increased showing more neutron shielding capability in the composites.

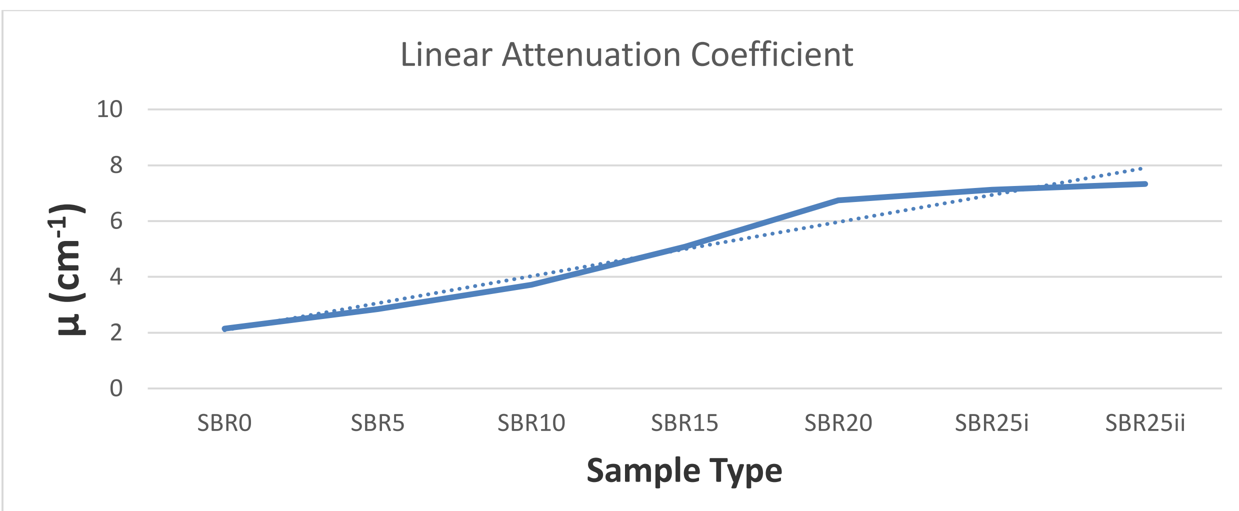

Fig. 1: Curve of linear attenuation coefficient with boron carbide content

Figure no. 10 illustrates the effect of boron carbide contents on the linear attenuation coefficient of the composites. An increasing trend in linear attenuation coefficient with boron carbide content is observed

As like neutron shielding capability of composites Densities, and Mass attenuation coefficients were also calculated. The increasing trend in both was observed with increasing boron carbide contents.

Table 2: Nuclear Properties of Composites

Formulation	Density (g/cc)	Linear Attenuation coefficient (cm^{-1})	Mass attenuation coefficient (cm^2/g)	Relaxation Length (cm)	Half Value Thickness (cm)
SBR_0	1.04	2.15	2.07	0.46	0.32
SBR_5	1.06	2.85	2.69	0.35	0.24
SBR_{10}	1.10	3.72	3.38	0.27	0.19
SBR_{15}	1.15	5.08	4.42	0.20	0.14
SBR_{20}	1.19	6.75	5.67	0.15	0.10
SBR_{25i}	1.22	7.12	5.84	0.140	0.10
SBR_{25ii}	1.25	7.33	5.86	0.136	0.09

Relaxation length is the inverse of linear attenuation coefficient. Relaxation length values shows decreasing trend with boron carbide content. This fact suggests that lower thickness of material will be enough to shield the neutrons if boron carbide content is kept high. Same trend is illustrated by the half value thicknesses of composites. Nuclear properties of composites are mentioned in the table no. 2.

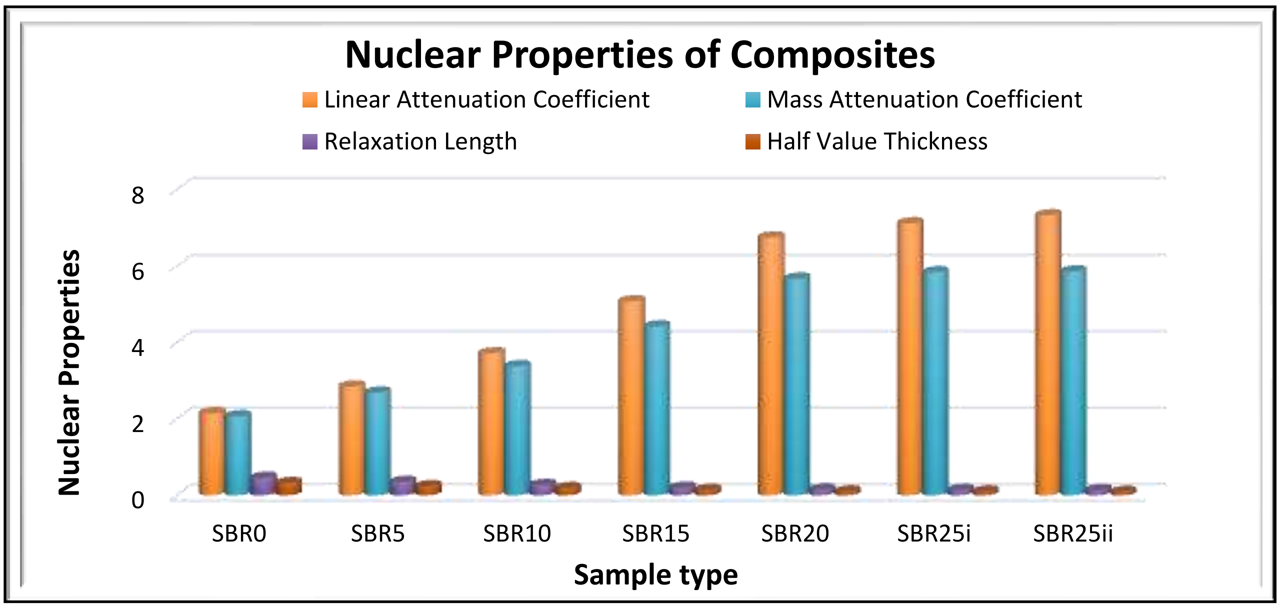

Fig. 2: Nuclear Properties of Composites

Comparison of Experimental and Simulated Results

Experimentally determined results of neutron shielding were verified by using **M**onte **C**arlo **N**-**P**article transport code. MCNP model is developed for the problem geometry that contains a source emitting a neutron beam of 1.2 A^0 wavelengths. It is the same wavelength that was used physically on target sheets of composites. This beam is irradiated at the sheet and transmitted beam is collected by detector.

Counts reaching the detector, with and without placement of shield are determined by MCNP are compared with experimental results as shown in figure no. 12. Values from both the sources are in good agreement as can been seen by small acceptable values of relative errors at each step.

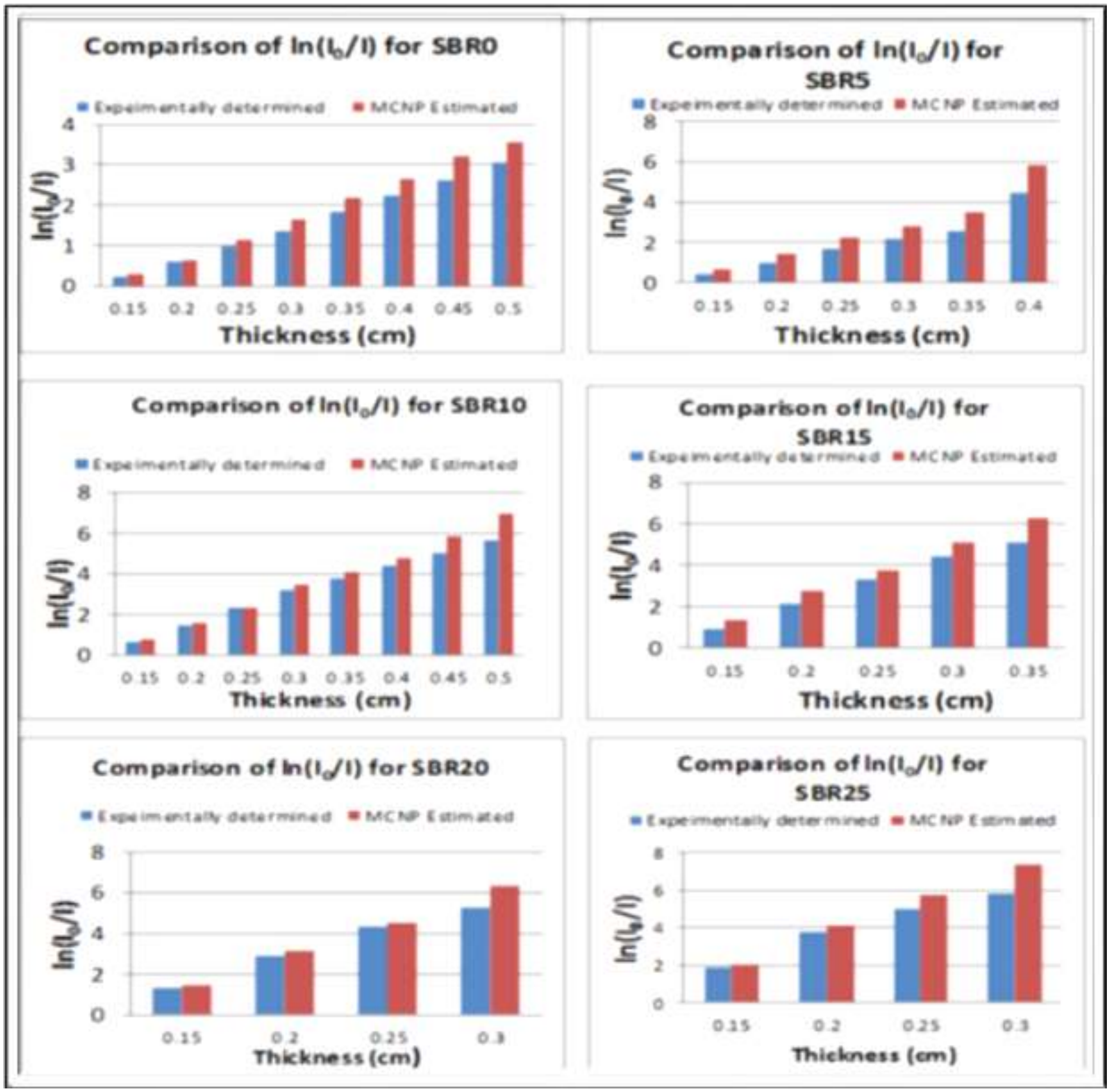

Fig. 32: Comparison of Experimentally determined and MCNP Estimated ln (I_0/I)

Comparison of linear attenuation coefficient (μ)

Linear attenuation coefficient of composites has also been calculated both experimentally, and by MCNP: given in figure no. 13. There comes out to be very slight difference as mentioned follows:

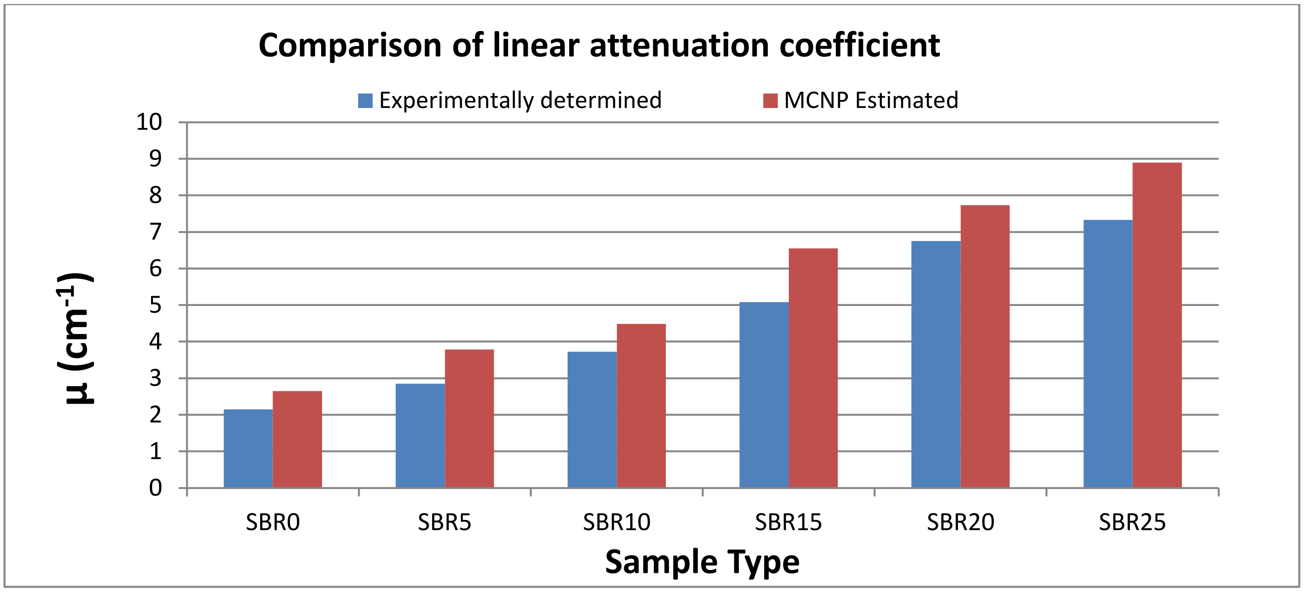

Fig. 43: Values of experimental and MCNP linear attenuation coefficient

Fig.13 shows the difference between the experimental and MCNP estimated values of linear attenuation coefficient. Moreover there is no difference in trend for all composites.

Discussion

Styrene butadiene composites have been formed by keeping amount of all constituents same except for the boron carbide in order to evaluate the effect of boron carbide concentration on the properties of composites. These composites have been subjected to various tests based on multiple techniques. Linear attenuation coefficient, mass attenuation coefficient, relaxation lengths and half value thicknesses of these composites have also been calculated. Mechanical properties have been improved by addition of boron carbide. Hardness also increases with boron carbide amount as SBR_0 has hardness 55.95 which increases to 63.45 in case of SBR_{25}. Solvent uptake studies data reveals that all of these composites interacts more with the non-polar solvents than the polar ones. While comparing the swelling index values of these composites, it comes out that no matter whether the solvent is polar or non-polar increased amount of boron carbide resulted in less interaction with solvent: SBR_{25} interacts much less than the SBR_0 in all kinds of solvents. Thermal stability of these composites has also been determined by TGA. These results show two stage mass losses starting from 250^0C. These results provide us with the temperature range in which these composites can be used safely otherwise composites would start degradation. Nuclear properties of these composites include Neutron shielding properties have been measured using neutron irradiation techniques. Counts measurement has been taken by the R-Block with and without the placement of samples between the sources (Am-Be source) and detector. Neutrons from Am-Be source were thermalized with the help of polyethylene moderator. These results showed decrease in neutron counts by increase in boron carbide content in the composites. Results of various thicknesses of samples in a neutron irradiation experiment at PARR-1 suggested that decrease in counts with increase in thickness is observed as predicted by Beer-lambert's law. It can further be seen that less thickness of any type of material is required if boron carbide content is increased and boron carbide addition is economical as well.

Experimentally determined results of neutron shielding were verified by using MCNP (**M**onte **C**arlo **N**-**P**article transport code). MCNP developed the problem geometry and also calculated the counts and linear attenuation coefficients. Both the experimentally calculated and MCNP estimated values are in good agreement.

Conclusion

Styrene butadiene composites of six SBR formulations (SBR_0, SBR_5, SBR_{10}, SBR_{15}, SBR_{20}, SBR_{25}) have been prepared with boron carbide contents ranging from 5% to 25%. After the formation of composites, these have been subjected to various tests based on multiple techniques to figure out the effect of variation of boron carbide amount on the properties of composites. From the outcomes of various tests, certain conclusion can be drawn. More the boron carbide content, less thickness of composite will serve the purpose of shielding with best physical properties.

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