Neutron shielding using Li_3BO_3/Epoxy Composite

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Abstract: This work aims to perform wall tiles for neutron attenuation through radiotherapy laboratories to protect patients and employee. Different ratios of micro-particles lithium borate were finally mixed with epoxy before hardener adding. It was found that the optimal percent of lithium borate 20% that preserving high mechanical properties. Thermal behavior proved the availability of the composite through wide range of temperature. Morphological structure showed homogenous pattern of the composite. Electrical conductivity illustrated very small changes with high dose of gamma irradiation reached to 200 kilo gray that approved their stability while it increased dramatically by raising the dose to 500 KGy. The composite carry out high shielding efficiency reached to 6.3 and 6.78 cm shielding thickness to reach to half value of the original neutron efficiency even with using 5 curry source (Americium–Beryllium) for total neutrons and non thermal one respectively. Low decrease of the shielding efficiency upon irradiation up to 500 Kilo gray of the composite for non-thermal neutrons which recommended effectively usage of the composite as neutron shielding. Durability of the composite has been investigated by examine the mechanical properties with irradiation dose.
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Key wards; epoxy, lithium borate, neutron, shielding, gamma ray, wall tiles

Introduction

Nuclear equipments and instrumentation make a large use of neutron shielding materials. These are generally used for collimating beam lines, shielding detectors and, last but not least, protecting the environment where researchers are operating. The materials usually employed to this aim are characterized by a relevant abundance in atomic nuclides that present a high neutron absorption cross-section.

Significant textbooks and handbooks providing broad coverage on shielding are available [1-11]. In addition, many reports, monographs and journal articles have been published on various particular aspects of nuclear radiation shielding. Of particular significance as sources of such information are the following organizations: (in the USA) Radiation Shielding Information Center, Oak Ridge National Laboratory, Oak Ridge, Tennessee; National Council on Radiation Protection and Measurements, Beheads, Maryland; Radiation Protection and Shielding Division of the American Nuclear Society, La Grange Park, Illinois; (in Europe) OECD Nuclear Energy Agency Data Bank, Gif-Sur-Yvette, France; and the European Shielding Information Services, Ispra, Italy.

Typical elements are boron and cadmium, whose neutron absorption cross-section for thermal neutrons (λ = 1:8 ˚A) are 767 and 2520 barn, respectively. Some rare earth elements, too, are characterized by a huge neutron absorption cross-section (e.g. Promethium, Samarium, Europium, Gadolinium, and Dysprosium). However, the high-cost prevents their generalized use, leaving the utilization of these nuclides for special applications. When thermal neutrons are considered, i.e. neutrons that are slowed down by moderators kept around room temperature, cadmium is frequently used as a shielding material, thanks to the high value of its neutron absorption crosssection which, moreover, increases for decreasing values of the neutron energy, E, with the well-known law $1/\sqrt{E}$ [12]. Cadmium shielding is generally applied in the environment of instrumentation located around nuclear fission reactors. However, when the energy of neutrons increases, the absorption cross-section of cadmium is subjected to a drastic decrease, especially in the energy range of the so called epithermal neutrons [13]. Therefore, when epithermal neutrons represent the main component of the neutron flux (which is generally the case, for the pulsed neutrons produced by a spallation source) its use as a shielding material is restricted to a number of limited applications. In this case, boron becomes much more effective, due to its much higher neutron absorption cross-section, up to energy levels of several tens of eV. Just to give an example, for neutrons of 1 eV energy, the neutron absorption crosssection of

cadmium is 20 barn while the corresponding cross-section of boron is well beyond 100 barn [13]. This is a simple explanation for the massive use of boron in theshielding of instruments and beam lines located at a spallation neutron source. A further advantage of boron is represented by the fact that its nuclear transmutation following neutron absorption, produces stable elements (lithium and helium) that are not radioactive.

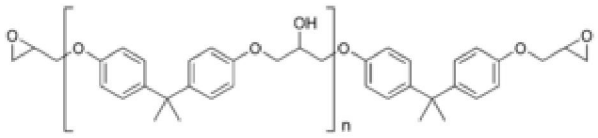
Lithium-6 (^{6}Li) has a large absorption crosssection of 940 b for thermal neutrons, and the absorption is predominantly (n,α) reaction without gamma-ray generation. Thus, ^{6}Li is normally used for thermal-neutron shielding in medical and biological applications. The natural abundance of ^{6}Li is small, 7.5%, so enriched ^{6}Li (to ~96%) is usually used.

The target of this work is synthesis of mobile polymer tiles used in neutrons' shielding. Mixing lithium borate with 20 % ratio by weight to epoxy to prepare the composite. Characterizations of the composite were carried out using some techniques such as thermal gravimetric analysis, scan electron microscopy and hardness measurement. The composites were exposed to gamma irradiation for ageing investigation . Electrical conductivities of the irradiated composites were measured with different frequencies prior measurement of neutron's attenuation.

2. Experimental work

2.1. Materials

The samples were prepared using phenyl epoxy and their solidifying agents of commercial grade were purchased from Optco, Egypt. Lithium borate was of analytical grade purchased from Ayah Co for chemical supplies (local company).



phenyl epoxy

2.2. Preparation of the sample

The epoxy formula consisted of standard bisphenol-A. with technical purity 95% and epoxide weight: as a resin polyoxypropylendiamine as a hardener while erosail was also added to prevent fillers from sedimentation. Lead oxide filler was first dispersed with equal particulate sizes in range of nano particles and loading fractions in the epoxy formula. The mixture was stirred at room temperature and heated to 80^{o}C for 30 minuets. The mixture was degassed to allow the entrapped air to be released, then poured with great care into the specimen moulds and left to cure was carried out. The specimen moulds were made of Teflon with different geometries. These moulds guarantee the dimensions of different shapes which suits different examinations. After 24 hours, the samples were extruded from moulds and 7 days later were processed either by shaping or machining to obtain the final ASTM dimensions.

2.3. Characterization of the sample

2 .3.1.Thermal properties

Thermal gravimetric analysis (TGA)

Shimadzu TGA -50, Japan was used to characterize the polymer composite thermally. Sample weight used was few milligrams, while thermal scanning was from ambient to 600 ^{o}C.

2.3.2. Morphological properties (SEM)

Investigation and magnification of the polymer composite surface was carried out by SEM, JEOL-JSM-5400, Japan.

2.3.3. Mechanical properties

Durability testing and hardness

Square shape of polymer composite of dimension not less than 20(L) x 20 (W) mm was used to examine by analogue manual instrument of hardness tester with thin pin it is termed Baxio USA, while durability was examined by measuring the attenuation coefficient, HVL, TVL and λ after gamma irradiation of shielding materials for 50, 100, 200 and 500 KGy for the calculation of change of the previous factor.

2.4. Neutron Sources

The neutron irradiation cell contains two neutron sources were used in this experiment. Both sources were manufactured by Amersham Co., UK and donated to the National Institute of Standard from Schlumberger Logelco Company in Egypt. ^{241}Am-Be cylindrical source is kept in a stainless steel pressurized vessel of with 3 cm base diameter and 7 cm height. It contains about 0.2 TBq (5 Ci) ^{241}Am. It has a neutron output of 1.291×10^7 n S^{-1}. The ^{252}Cf is a point source 50 μg of original activity 27 mCi, kept in a cylindrical seal welded double encapsulation with dimension of 3.3 cm base diameter and 10 cm height with present activity 0.39 mCi. Model No. NSR-R outer capsule is made of multi-phase MP35N alloy. ^{241}Am-Be source holder is made from Perspex with cylindrical shape and dimensions 4 cm diameter and 11 cm height, when the source admitted inside it and secured by a covered screw. The two sources are enclosed in two PVC tubes supported vertically inside the shield with diameter 4 and 5 cm, respectively, 90cm in height and 60cm imbedded in the shield. The two PVC tubes were fixed at the center of a rectangular shaped wooden box of 120 cm × 140 cm side and 120 cm height, having 20 cm distance

between them as shown in Figure (1)

2.5. Sources Mechanics

Two gearbox motors with 0.5 horsepower having 30 turns/min upper fixed at the wooden holders above the shield box are used to move the sources virtually up and down (in and out of the shield). The source is clipped from its Perspex holder as shown in the Figure (1). The source holder is hanged by a polymer wire on prespex pulley to motor to other pulley and finally to a mass equivalent to that of both sources and its holder.

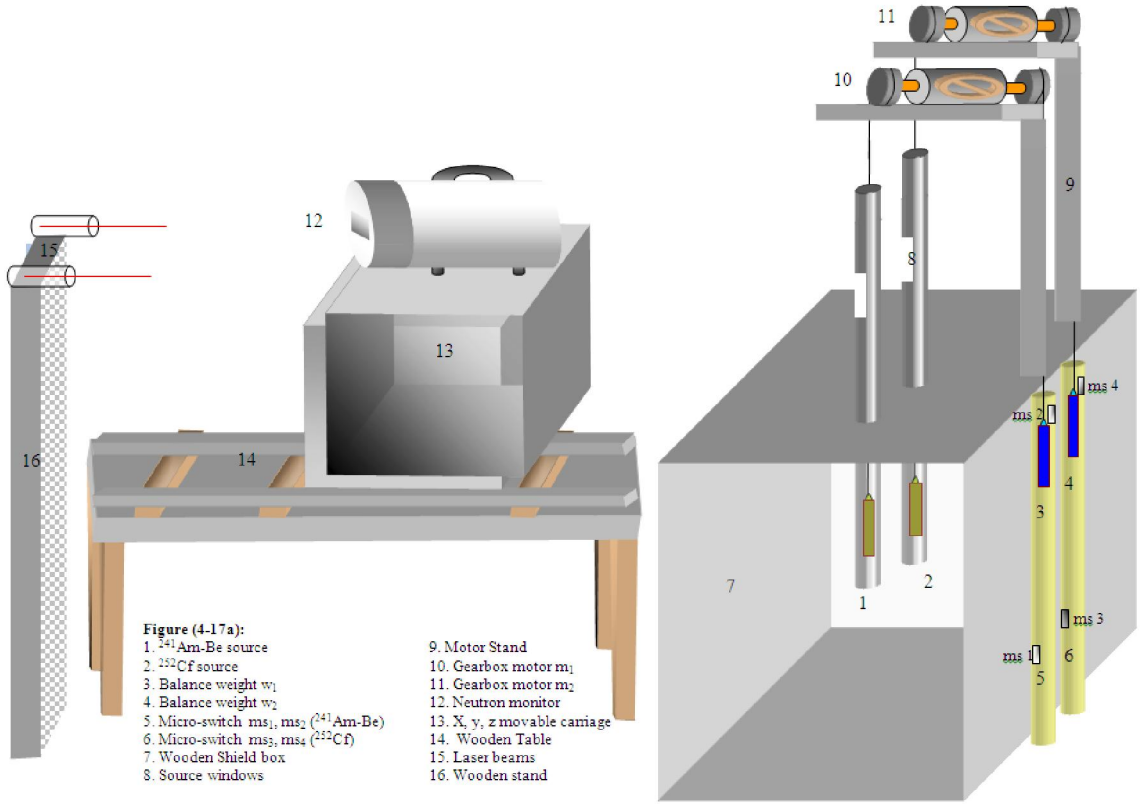


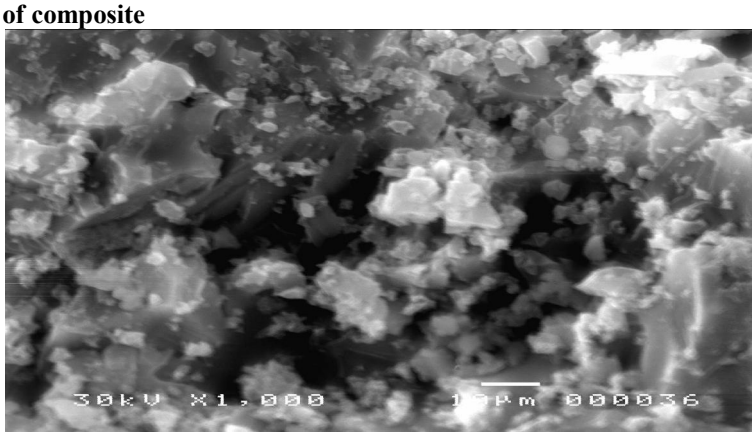
Figure (4-17a):

1. ^{241}Am-Be source
2. ^{252}Cf source
3. Balance weight w_1
4. Balance weight w_2
5. Micro-switch ms_1, ms_2 (^{241}Am-Be)
6. Micro-switch ms_3, ms_4 (^{252}Cf)
7. Wooden Shield box
8. Source windows
9. Motor Stand
10. Gearbox motor m_1
11. Gearbox motor m_2
12. Neutron monitor
13. X, y, z movable carriage
14. Wooden Table
15. Laser beams
16. Wooden stand

3. Results and Discussion

3.1. Characterization of composite

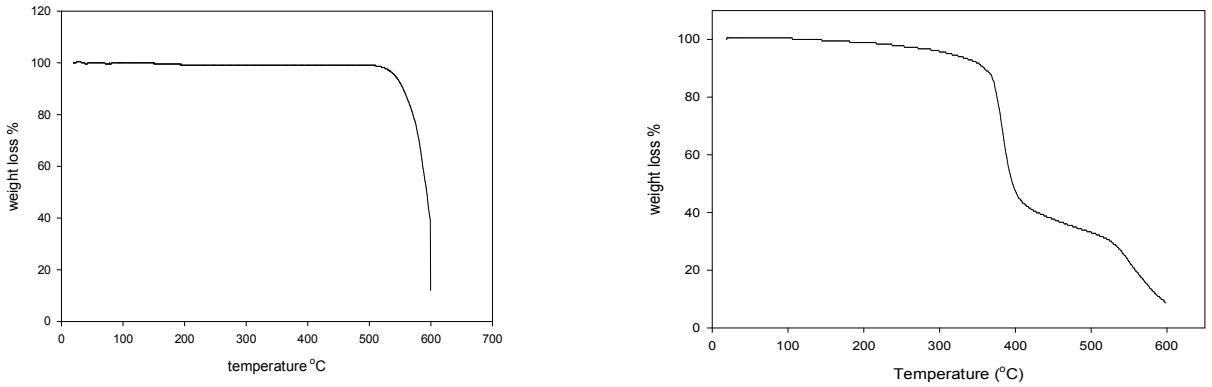
3.1.1. Morphological structure characterization



Scan electron microscope of lithium borate/ epoxy composite have been illustrated in figure 2. It showed that; scattering of white spots in regular through dark matrix [14,15]. It may due to homogenous distribution of lithium borate through the epoxy network. This observation conduct availability of using such composite into neutron shielding. There are some larger white spots distributed through the polymer matrix existed into some plates as seen. While in general there are regular distribution of lithium borate via the composite thickness which.

3.1.2.Thermal gravimetric analysis (TGA)

Temperature at which the loss in weight occurs is considered to be the thermal resistance of polymer [16,17] . Thermal characteristic was detected for both of the composite of Li_3BO_3 with epoxy and for the blank one. There are obvious difference in thermal stability between epoxy polymer and composite formation by incorporation lithium borate into them. Epoxy blank samples as seen in figure 2-a could be differentiated into three divisions. First division described the working temperature of epoxy showing high stability reached to 515 ^oC loss of weight through which did not exceed 2 % which may due to losing water content. Second division illustrated convex zone ended at 560 ^oc at which loss of weight was 13 %. The third division showed abrupt change in weight describing the isotherm's tail which ended at 13 % from the original weight at 600 ^oC.



a **b**

Figure 2:Thermogravimetric analysis of Epoxy/Li_3BO_3 composite.
a) Blank (no lithium borate), b) 20 % lithium borate

Figure 2-b represented thermogram of lithium borate with 20 % ratio to epoxy forming composites. The thermogram could be categorized into 5 divisions. First division described working temperature working temperature of the composites which reached to 280 ^oC at which weight loss did not exceed 2.1 %. This wide range of working temperature confirmed the composite usage into the desired applications. The second division was showing convex part ended at 376 ^oC while weight decreased to 17 %. Abrupt decrease of weight was seen in the third division reached to 46 % from the original weight by heating the composites to 405 ^oC. Forth zone illustrated gradual decrease of weight was 30 % from the original weight due to heating the composites to 530 ^oC. The tail of the thermogram have been observed at the fifth division through which loss of weight ended at 9 % of starting weight of the samples by heating the temperature to 600 ^oC. The step peak proved the reactivity of the epoxy towards lithium borate which reduced the thermal stability compared to the epoxy blank.

3.2. Electrical conductivity

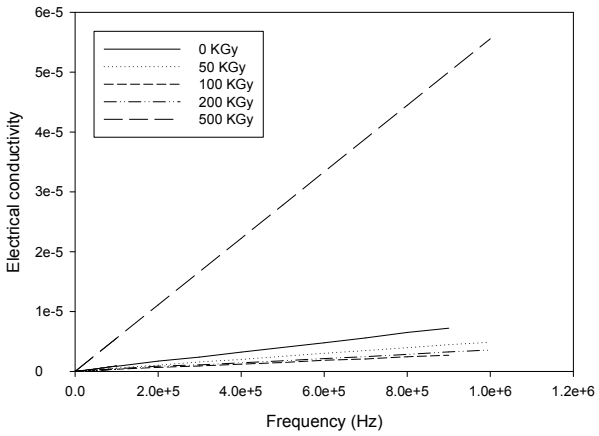


Figure 3: EC of Epoxy/Lithium borate at different Frequency

The conductivity in composite polymer electrolyte is not a linear function of the filler concentration [18, 19]. At low content level, the

dilution effect is efficiently contrasted by the specific interactions of the ceramic surface, which promote fast ion transport, and the overall effect is progressive enhancement of the conductivity. On the other hand, at high filler content, the dilution effect predominates and the conductivity decays. Similarly, concave effect on conductivity with inorganic/ceramic materials as fillers in solid polymer electrolyte [20–22], and ion pairing (insufficient salt dissociation) is identified as the origin of the later Deterioration

At very low frequencies the dipoles have sufficient time to align with the field before it changes direction and the dielectric constant is high. At very high frequencies the dipoles do not have time to align before the field changes direction and the dielectric constant is lower. At intermediate frequencies the dipoles move but have not completed their movement before the field changes direction and they must realign with the changed field. For non-polar plastics the dielectric constant is independent of the alternating current frequency because the electron polarization is effectively instantaneous.

The electrical conductivity of the gamma irradiated epoxy/ lithium borate composite at different frequencies have been observed at figure 3. The un-irradiated composites were semiconductor reached its value to 7.28196 x 10 -6 simon/ cm at highest value of frequency used 9 x 10^5 Hz. The electric conductivity at 50 KGy irradiation dose has been decreased by 38.8 % from its original value. The decrease of the electrical conductivity attempt the efficiency of the gamma irradiation for composites' cross-linking. The electrical conductivity's value was reduced to 63 % from the starting point upon rising the irradiation dose to 100 KGy. The electrical conductivity become constant while the composites was irradiated to 200 KGy. At 500 KGy irradiation dose the electrical conductivity increased dramatically to 540 % from the original value. It may be explained by maximum cross-linking of the composites have attained at 200 KGy irradiation dose. Over this value partial degradation of the composites has been performed resulted in loosing of some charged fragments and free radicals which has a very important role for rising the electrical conductivity value.

3.3. Neutron shielding

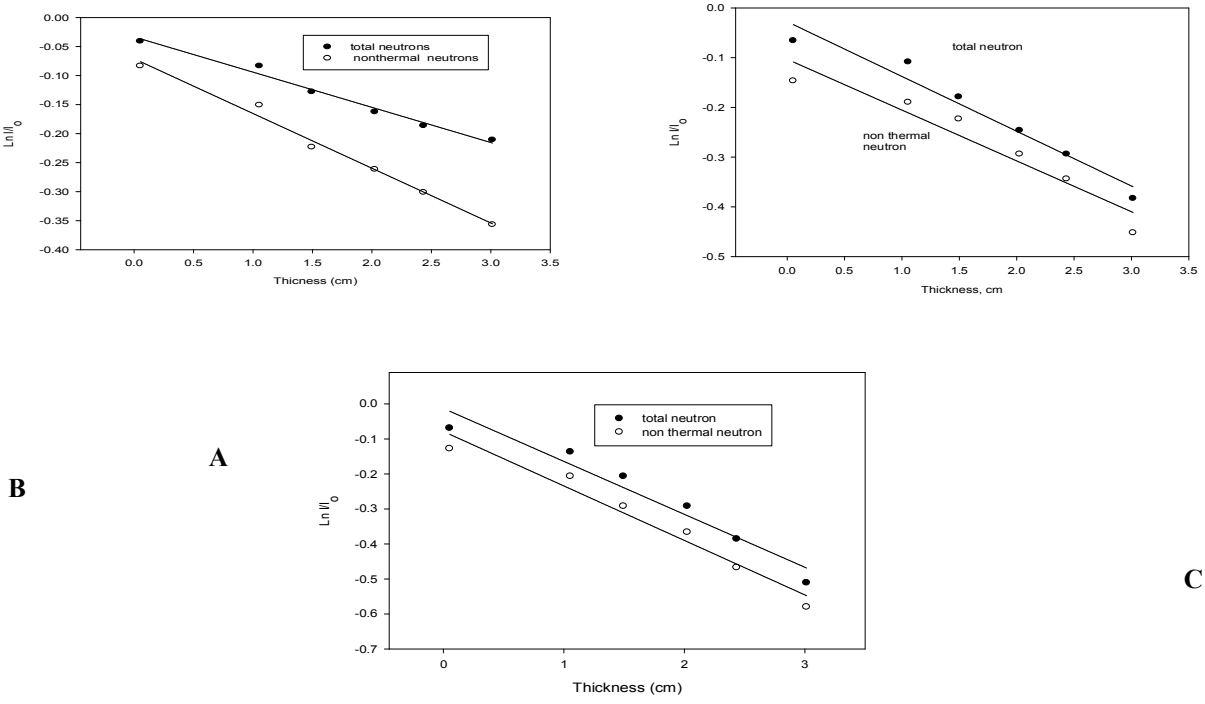


Figure 4: Effect of irradiation on neutron shielding of the epoxy/lithium borate composite. A) No irradiation, B) 50 KGy irradiation C) 50 KGy. irradiation

Table 1 :Effect of exposing gamma irradiation (KGy) on the $Li_3 BO_3$/ epoxy composite on the change of λ , Σ , HVL and TVL.

Symbol	0 KGy		50 KGy		100 KGy		500 KGy	
	Total neutrons	Non thermal neutrons	Total neutrons	Non thermal neutrc	Total neutrons	Non thermal neutrons	Total neutrons	Non thermal neutrons
Σ	0. 110	0.1022	0. 151	0.155	0.0956	0.0977	0.0606	0.0943
λ	9.09	9.78	6.62	6.45	10.46	10.25	16.5	10.6
HVL	6.3	6.78	4.58	4.47	7.248	7.09	11.43	7.3
TVL	20.9	22.5	15.23	14.38	24.058	23.54	37.95	24.39

The intensity of thermal neutron and gamma ray doses decrease almost exponentially with increasing the shield thickness. It is also noticed that, the rate of decrease for non thermal neutrons is more stable than that for total neutrons [23]. Neutron shielding using 20 % lithium borate within epoxy to synthesis composite was performed. Lithium and boron of borate have very important role for absorption (capture) of neutrons with different energies. Epoxy is rich with carbon and hydrogen which considered as efficient moderator applied for slowing down the neutrons. Attenuation efficiency increase with increasing the composites thickness as seen in figure 4. It was found that; the half value layer of the composites 6.3 cm for the total electron while that of the non-thermal was 6.78 cm. Different parameters were measured such as relaxation time (λ), tenth value layer (TVL) and macroscopic (Σ) as seen in table 1 which confirmed usage of the composites in the desired applications. As gamma irradiation doses exposed on the composites increased the neutrons attenuation decreased with slow rate. HVL decrease for the non thermal neutrons was 4.5 % while it reached to 6.7 % for total neutrons upon rising the exposing gamma irradiation to 200 KGy. These results strongly confirmed the composite application for wide range of irradiation dose exposing on the studied composite. Raising the irradiation dose to 500 KGy result in reduction of HVL of the total neutrons to 81 % from the original value while the change in HVL of the non-thermal neutrons was 7.7 % from the starting value. These results recommend the composites applications for shielding of non-thermal neutrons even with raising the irradiation dose to 500 KGy while it could be used for total neutrons till 200 KGy. Although the HVL of the non-thermal neutrons of un-irradiated composites was less than total one while its value decreased by gamma irradiation with slow manner and very small value. HVL of the total neutrons decreased dramatically especially after the composites were exposed to 200 KGy irradiation dose. The results attained refer to a reasonable agreement of the measured and calculated results. Also, the concerned composites have a good attenuation parameter comparing with the measured data for dolomite–sand [24] and Homogeneous and multilayered shields for neutrons [25]

3.4. Mechanical properties (hardness)

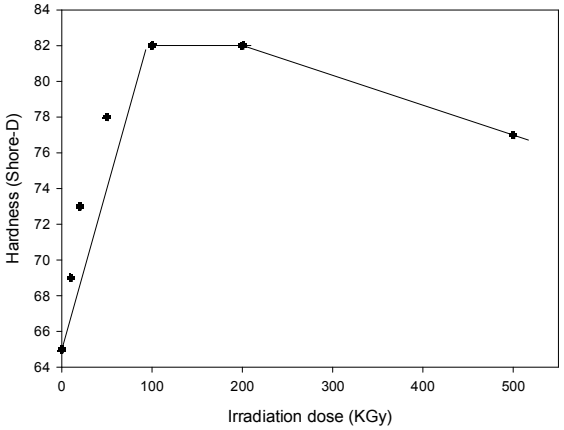


Figure 5: Effect of gamma irradiation dose (KGy) on hardness of the Li_3BO_4/epoxy composite.

Figure 5 illustrated the change in hardness of lithium borate / epoxy composite with 20 % ratio with different gamma irradiation doses. The hardness of the composite has accepted value used as mobile tiles for the purpose of neutron shielding (63 Shore-D). Increasing irradiation doses up to 100 KGy conducted increasing the hardness. The value of hardness may be increased due to increasing the composite cross-linking. the value of hardness kept constant while rising the irradiation doses to 200 KGy. The value of hardness decreased gradually with slow rate and very small extent (6 % of the highest value of hardness) upon increasing the irradiation dose till reached to 500 KGy. The results confirmed the application of such composite in neutron shielding for wide range of irradiation doses.

Conclusion

The goal of this work is synthesis of polymer composite used in medical application for neutrons' shielding. Mixing of lithium borate with 20 % ratio to epoxy was carried out prior adding the solidifying agent. Investigations of the composite were carried out morphologically, thermally and mechanically. Ageing of the composites was examined after gamma

irradiation with different doses up to 500 KGy. Electric conductivity of the irradiated samples showed increase their value with increase the irradiation doses reached to 90 % for 500 KGy dose while this value raised to 1310 % at 50 KGy. half value layer of the composites reached to 6.7 cm which recommend their application in the desired purpose. Increase of half value layer with irradiation dose did not exceed 7.7 % even with exposing irradiation to 500 KGy for non thermal neutron while this value increased to higher value reached to 81 % for thermal neutron.

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