

Epoksi Bazlı Metal ve Metal Oksit Katkılı Yeni Kompozit Nötron ve Gamma Radyasyon Moderatör Malzemesi

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Öz

Bu çalışmada beş farklı kompozit zırh malzemesi geliştirildi ve üretildi. Üretimlerde epoksi reçinesi taban malzeme olarak farklı özelliklere sahip [krom oksit (Cr_2O_3), lityum florür (LiF), nikel oksit (NiO), bizmut oksit (Bi_2O_3), manganez oksit (MnO), bakır oksit (CuO), titanyum oksit (TiO_2), kobalt oksit (CoO), gadolinyum oksit (Gd_2O_3), şelit (CaWO_4), demir oksit (Fe_2O_3), kurşun oksit (PbO)] gibi malzemeler kullanılmıştır.

Karışım oranları Monte Carlo Simülasyon programının GEANT4 kodu kullanılarak belirlenmiştir. Hızlı nötronlar için Toplam makroskopik tesir kesiti, ortalama serbest yol ve geçen nötron sayısı GEANT4 simülasyon kodu kullanılarak belirlendi. WinXCom yazılımı kullanılarak gamma radyasyonu için kütle zayıflama katsayısı ve yarı değer kalınlıkları (HVL) hesaplandı. Deneysel absorbtion doz ölçümleri yapıldı ve bu ölçümlerde ortalama nötron enerjisinin yaklaşık 4.5 MeV, 74 GBq aktiviteye sahip ^{241}Am -Be nötron kaynağı ve BF_3 gazlı dedektör kullanıldı. Hem simülasyon hem de deneysel ölçümler, geleneksel beton ve parafin ile karşılaştırıldı. Yeni kompozit zırh malzemelerinin bu referans malzemelerden çok daha iyi radyasyon emdiği tespit edildi. Bu yeni radyasyon zırh kompozit malzemelerin nükleer tıpta, radyoaktif atıkların taşınması ve depolanmasında ve nükleer santral gibi alanlarda, hem nötron hem de gama radyasyonu için kalkan malzemesi olarak kullanılabilirliği önerildi.

Anahtar Kelimeler: Nötron ve gama radyasyonu, epoksi, GEANT4, zırlama, metal oksit

Epoxy Based Metal and Metal Oxide Doped New Composite Neutron and Gamma Radiation Moderator Material

Abstract

In this study, five different composite shield materials were developed and produced. Epoxy resin based different proportions, materials such as [chromium oxide (Cr_2O_3), lithium fluorür (LiF), nickel oxide (NiO), bismuth oxide (Bi_2O_3), manganese oxide (MnO), copper oxide (CuO), titanium oxide (TiO_2), cobalt oxide (CoO), gadolinium oxide (Gd_2O_3), selit (CaWO_4), iron oxide (Fe_2O_3), lead oxide (PbO)] were used on production. The GEANT4 code of the Monte Carlo simulation program was used in determining mixing ratios. The total macroscopic cross section, mean free path and transmission neutron number were determined for fast neutron radiation by using GEANT4 simulation code. The mass attenuation coefficient and half-value layer (HVL) were calculated for gamma radiation by using WinXCom software. Experimental absorbed dose measurement was carried out and in these measurements ^{241}Am -Be neutron source with 74 GBq activity which average neutron energy is approximately 4.5 MeV and BF_3 gas detector were used. Both simulation and experimental measurements were compared with paraffin, conventional concrete. It was found that the new composite shielding material absorbed radiation much better than these reference materials. It has been suggested that this new radiation shielding composite material can be used in areas such as nuclear medicine, transport and storage of radioactive waste, nuclear power plants and as shield material for both neutron and gamma radiation.

Keywords: Neutron and gamma radiation, epoxy, GEANT4, shielding, metal oxide

2.1. Introduction

With the increasing usage of gamma and neutron radiation in industry, medicine, agriculture and military application, radiation safety studies have reached a significant level. There is always a need to develop new shielding material, which can be used both high resistant, chemical and environmental conditions and good radiation absorption power (Krocher et al., 1984). Polymer-based lightweight new composite materials such as polypropylene, polystyrene, polyethylene, polystyrene, and rubber are using the radiation safety application. The epoxy resin is occurring a chemical reaction between biphenol A and C_3H_5ClO as with more than one epoxy molecule which can be hardened into a usable plastic. The epoxy resin is used in many industrial applications because of excellent chemical corrosion resistance, high tensile, fatigue and compressive strength (Massingilljr et al., 2000). Epoxy resins that, combined with glass, carbon and aramid, metal and metal oxide new durable composite materials can produce. Epoxy-based composites are widely used in the aviation, marine, automotive and electronic industries (Tjong, 2010). Shielding materials such as heavy concretes, lead glass and stainless steel are using at the radiation therapy applications, however, they are neither aesthetic nor resistant to corrosion so epoxy can be used as alternative materials it has high chemical and mechanical properties. Epoxy resin based composites has become to be used due to its good chemical resistance and mechanical properties as shielding material in the nuclear applications. Nano carbon doped epoxy composites were produced as electromagnetic (EM) shielding

materials and on EM radiation shielding properties were determined (Vovchenko et al., 2008). Epoxy resin composites have both high mechanical and chemical properties so ultrahigh molecular weight polyethylene fiber Nano epoxy composites were produced for NASA spacecraft safety on cosmic radiation (Zhong et al., 2009). Boron compounds generally are used for neutron radiation shielding so to increase the shielding capacity of the epoxy resin, boron-containing ores three samples epoxy composites were development for slow neutron shielding (Li et al., 2011). Epoxy-Ferrochromium slag composites were designed and produced by using Monte Carlo Simulations for shielding X-ray, gamma and neutron radiation and the addition of FeCr slag in epoxy resin up to 50% percentages, improved shielding ability was reported for X-ray, gamma and neutron radiation (Korkut et al., 2013). Shielding capacity of the epoxy resin can improve adding heavy metal powders different weight percent such as tungsten. For example, tungsten/epoxy composites were prepared and shielding and mechanical characteristic of composites were investigated by using Co-60 gamma radiation source. Both shielding and mechanical property of composites increases were reported with the doped of tungsten (Chang et al., 2015). Since Epoxy resin flexible a material it is used, nuclear safety applications, but it must be powered. Epoxy resin strengthened composite neutron shield materials were produced by adding materials as Fe (Iron), Bi (Bismuth), Ta (Tantalum) and WC (Tungsten Carbide) and its shielding parameters were determined by GEANT4 and FLUKA simulation codes (Canel et al., 2019). Glass fibers/epoxy resin

composites are ideal materials for nuclear study. Because after the γ -ray irradiation still it have presented excellent fatigue strength, thermal conductivities, extraordinary, dimensional and thermal stabilities (Fang et al., 2018). In this study, to use epoxy resin as a shield material in nuclear applications, by adding based resin some materials such as (chromium oxide, lithium fluoride, nickel oxide, bismuth oxide, manganese oxide, copper oxide, titanium oxide, cobalt oxide, gadolinium oxide, selit, iron oxide, lead oxide) and five different types of new composite materials have been developed. Fast neutron and gamma shielding parameters were determined by using GEANT4 Monte Carlo calculations. Experimental absorbed dose measurements were carried out for fast neutrons.

2.2. Material and Method

2.1 Sample preparation

In this section, for epoxy resin doped different materials the neutron and gamma

radiation the shielding parameters were determined by using Geant4. In the enrichment of epoxy, chromium oxide, lithium fluoride, nickel oxide, bismuth oxide, manganese oxide, copper oxide, titanium oxide, cobalt oxide, gadolinium oxide, selit, iron oxide, lead oxide materials were used. Mass percentages of the materials to be incorporated into the epoxy were determined by the pre-simulations and the new composite materials were modelled. 3 scale epoxy ($C_{21}H_{25}ClO_5$) and 1 scale hardener ($C_9H_{10}O_3$) were used in the production of materials. The epoxy and added material the admixture were mixed until homogeneous. The obtained homogeneous mixture was poured into molds of 10 X10 X 4 cm. After casting, the molds were subjected to vibration in a vibration table and the air trapped in the mold was removed. The samples were then dried at $23^{\circ}C \pm 3^{\circ}C$ for 7 days. The chemical contents of the produced new composite samples are shown in Table 1.

Table 1 Chemical Compositions of New Composite Samples (% mass)

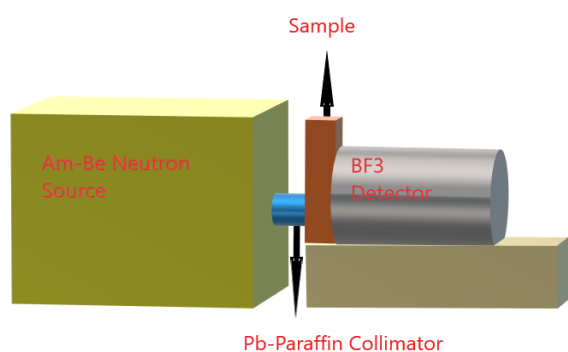
Compound	NCM 1 $\rho=3.25 \text{ g/cm}^3$	NCM 2 $\rho=5.57 \text{ g/cm}^3$	NCM 3 $\rho=6.15 \text{ g/cm}^3$	NCM 4 $\rho=3.044 \text{ g/cm}^3$	NCM 5 $\rho=3.32 \text{ g/cm}^3$
Epoxy+Hardened	65.0	64	51	65	55
Cr ₂ O ₃	15.0	10.0	15	13	10
NiO	10.0	-	-	7	-
TiO ₂	-	-	6	-	-
MnO ₂	5.0	-	-	-	-
CuO	-	-	3	-	-
LiF	-	-	13	-	12
Bi ₂ O ₃	5.0	12.0	-	-	-
CoO ₄	-	8.0	-	-	-
Gd ₂ O ₃	-	-	4	-	-
CdWO ₄	-	3	-	-	-
ZrO ₂	-	3	-	-	-
Fe ₂ O ₃	-	-	8	15	15
PbO	-	-	-	-	8

NCM: New composite material

$^{241}\text{Am}/\text{Be}$ neutron source (10 mCi) which emits 2–11 MeV neutron radiation and Canberra brand neutron detector were used for the neutron absorbed dose measurement. Experimental measurement design is shown in Fig. 2.



(a)



(b)

Figure 2: (a-b). Neutron experimental measurement system

2.2 Monte carlo simulation codes Geant4

The Geant4 simulation code is a software and it is used to simulate particle or radiation interactions with matter. Geant4 can be used nuclear application such as particle accelerators, high energy physics, medical physics, as well as detector system design,

computer science and space science (Agostinelli et al., 2003; Allison et al., 2006). This code can use to predict interaction with target material of neutrons such as high-Precision (HP) data-driven hadronic physics models (G4 Neutron HP Elastic, G4 Neutron HP Inelastic, G4 Neutron HP Capture, and G4 Neutron HP Fission). In addition to, this kit can used to simulate between many different energy range (from eV to TeV) radiation and target material which elements, compounds or mixtures. In this paper, this program was used to development new material. First, all components of the material were introduced into the program's library. The radiation absorption cross-sections of the components to be used in the material were determined. To find the best composition the base material of these metal oxide powder materials was by adding different percentages (from 3% to 15%) in the epoxy resin and the cross-sectional effect of radiation absorption was determined. Many simulations were performed by changing the components and their proportions in the base material. The these simulations were carried out both for the pure epoxy resin (100%) and for the resin+metal oxide which mix homogeneously.

Detailed information can be found at its www.Geant4.org.

2.3.Results and Discussion

In this study, five different types of epoxy resin based new composite samples are designed and produced; the chemical compositions are shown in Table 1. This new type epoxy based composite materials were content chromium 10-15 wt. %. It has been seen that by adding chromium oxide, lithium fluoride, nickel oxide, bismuth oxide, manganese oxide, copper oxide, titanium

oxide, cobalt oxide, gadolinium oxide, selit, iron oxide, lead oxide the composite materials structure has increased both heat resistance and radiation shielding capacity.

Neutron shielding parameters

The total macroscopic cross section, Mean free path and transmission number are basic parameters for neutron shielding design material. These parameters need to be determined in the radiation shielding material design study. In this study, these theoretically calculated by using Monte Carlo Simulation Geant4 code and obtained results are shown in Table 3.

3.1 Total macroscopic cross section

When interaction neutrons can perform with target materials either scattering (elastic or inelastic scattering) or absorption reactions. These interactions are expressed by microscopic section (σ) and macroscopic section (Σ). The microscopic cross-section represents the area of interaction of a particle with a target nucleus. It shows the possibility of a neutron interacting with the nuclei of light materials. And this is expressed by the barn or cm^2 . The possibility of interaction of neutrons with heavy materials such as concrete is indicated by a macroscopic section. The total macroscopic cross section (Σ) contains interaction, such as scattering, absorption, capture, fission, etc. The macroscopic cross-section is calculated as follows according to the microscopic cross-section and the atomic number.

$$\Sigma = N\sigma \quad (1)$$

$$N = \frac{\rho}{A} N_A \quad (2)$$

Here, N; atomic density in the interaction material, ρ ; density of the interaction material, N_A ; the number of Avogadro.

$$\Sigma_{\text{Total}} = \Sigma_{\text{scattering}} + \Sigma_{\text{absorption}} + \Sigma_{\text{capture}} + \Sigma_{\text{fission}} + \dots \quad (3)$$

If a material have high total macroscopic cross section values which good neutron shielding ability (Mclane et al., 1988).

3.2 Mean free path

The distance between the two collisions of a neutron particle with a material is expressed by the mean free path λ . When a neutron interacts with the target material, it has a dx travel distance in the material and this can be calculated by the following equation.

$$p(x)dx = \Sigma_t dx \cdot e^{-\Sigma_t x} = \Sigma_t \cdot e^{-\Sigma_t x} dx \quad (4)$$

The distance (x) that the neutrons can receive in the target material without any collision is expressed by the mean free (λ), which is calculated by the following formula.

$$\lambda = \int_0^{\infty} x \cdot p(x) dx = \Sigma_t \int_0^{\infty} e^{-\Sigma_t x} dx = \frac{1}{\Sigma_t} \quad (5)$$

Σ_t ; is specifies the possibility of a neutron collision with target material (Rinard. 1991). If the average free path for the material to which a neutron interacts is small, it indicates that the material has good shielding properties.

3.3 Neutron transmission number

The transmission is expressed of passing neutron numbers from the target material. If the number of neutrons passing through the target material is low, the neutron shielding, power of the material is so high. In this

study, to determine the number of neutrons passing through the samples, each sample was bombarded with 100000 neutron by using Monte Carlo simulation the Geant4 code. Passing neutron numbers were determined from to new composite and reference samples, obtained results discussed.

Shown in the Table 2 neutron Total Macroscopic Cross Section, Mean Free Path

and Transmission numbers were calculated for all studied materials and obtained results were compared with paraffin and conventional concrete which are used in the shielding radiation study generally. According to paraffin and conventional concrete, new composite samples have high Total macroscopic cross sections, low Mean Free Path value and Transmission number.

Table 2. Total macroscopic cross sections, Mean free path, Transportation values of the samples 4cm thick

Sample code	Total Macroscopic Cross Section (cm ⁻¹)	Mean Free Path (mm)	Transmission Number
Paraffin	0.8206	1.7223± 0.0135	44014
CC	0.6799	1.7783 ± 0.1429	50662
NCM1	1.339	1.5609 ± 0.0106	26199
NCM2	2.224	1.3075 ± 0.0296	10812
NCM3	2.459	1.2617 ± 0.0066	8501
NCM4	1.2625	1.5859 ± 0.1105	28292
NCM5	1.3526	1.5650 ± 0.1059	25856

CC: Conventional concrete, NCM: New composite material

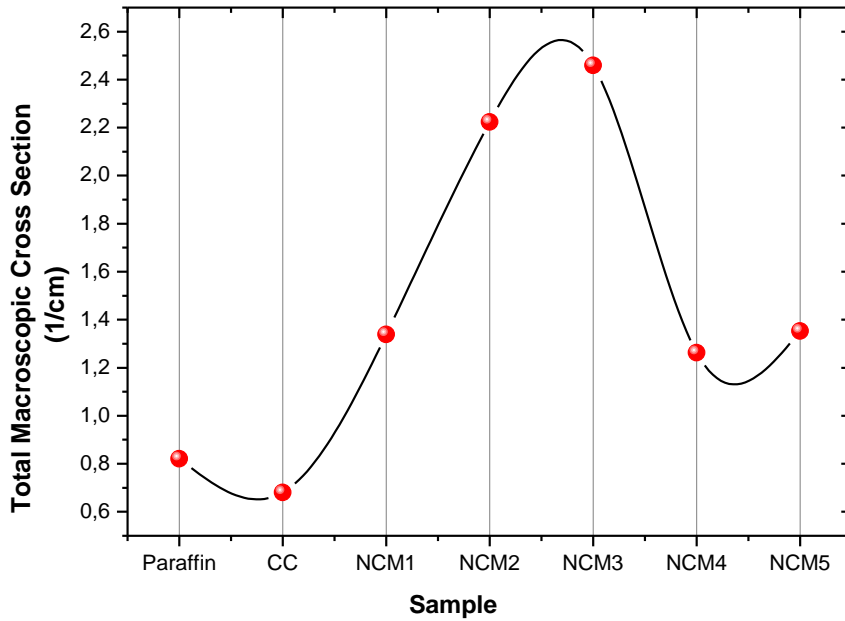


Figure 3. Total Macroscopic Cross Section (cm^{-1})

As shown in Fig. 3. it is determined that total Macroscopic Cross Sections values of new composite samples bigger than reference sample paraffin and conventional concrete. This result is an indication that the new composite materials are capacity of high shielding.

3.4 Experimental equivalent absorbed dose measurements

All samples were bombarded with a fast neutron source, equivalent dose rate measurements were carried out. Then absorbed dose amounts were determined of the samples. As shown in Table 2 and in Fig.3 obtained results were compared with reference sample paraffin and conventional concrete that commonly used for shielding neutron radiation and it was observed that all new composite samples were better than reference samples. NCM2-3 samples better than other samples for radiation-shielding capacity this is due to the fact that NCM2, Bi_2O_3 and NCM3, LiF concentration including.

Table 3. Experimental absorbed dose results

Sample	Equivalent Dose Rates Absorbed by Samples ($\mu\text{Sv/h}$)	Absorbed Dose Rate (%)
Background	1.3824	-
Paraffin	0.5215	37%
CC	0.4072	29%
NCM1	0.5801	41%
NCM2	0.7040	50%
NCM3	0.7109	55%
NCM4	0.6725	48%
NCM5	0.6801	49%

The measured absorbed dose rates of new composite and reference samples are compared in Fig.4.

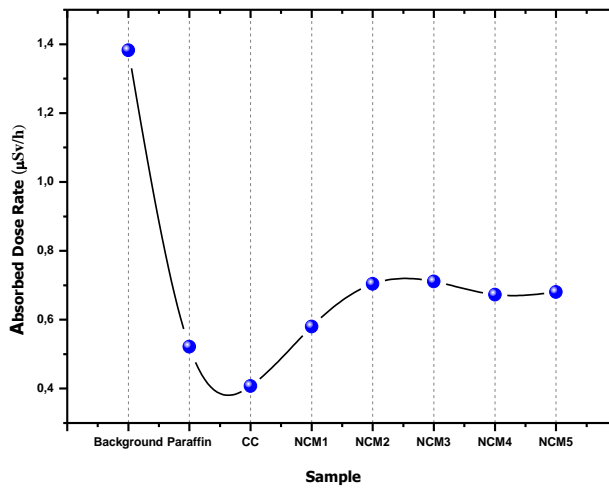


Fig 4: Experimental neutron equivalent dose rate measurement results

Looking at Fig 4, it is seen that all new composite samples show the highest absorption dose rate better than paraffin and conventional concrete. Which emphasize our produced that all new composite samples possesses highest shielding effective the paraffin and conventional concrete.

Gamma radiation absorption calculations

The gamma - ray interactions with of any material can be determined in terms of various parameters such as the mass attenuation coefficient, linear attenuation coefficient, mean free path, half value thickness, tenth value thickness, effective atomic number, effective electron number, etc. Determining all of these parameters is very useful, but the most important of these parameters mass attenuation coefficient and half value thickness so these must be calculated to determine the property of the interaction materials to be used for gamma-ray.

So, in this study, theoretical gamma-ray shielding parameters such as mass attenuation coefficient (MAC) and half value layer (HVL) of the samples were calculated by using the Beer-Lambert law and

WinXCom software (Berger et al., 1987; Singh et al., 2014). WinXCom software a computer program and it is used for the gamma ray shielding parameters calculation such as mass attenuation coefficients for any element, compound and mixture at energies ranging from 1 to 100 GeV.

When interaction with any shielding material mono energetic gamma ray, the energy decreases, depending on the thickness of the material and according to, the exponentially Lambert Beer law:

$$I = I_0 e^{-\left(\frac{\mu}{\rho}\right)t} \quad (6)$$

Where I_0 and I are the coming and transmitted gamma radiation energy of photons respectively. μ (cm^{-1}) and $\frac{\mu}{\rho}$ (cm^2g^{-1}) are the linear and mass attenuation coefficients (MACs), ρ ($\text{g}\cdot\text{cm}^{-3}$) is the density and t (cm) is the thickness of the target material.

The HVL is a thickness of shield material require to reduce the coming energy of the gamma radiation to its half and it can be defined as follow:

$$HVL = \frac{\ln(2)}{\mu} = \frac{0.639}{\mu} \quad (7)$$

At the in calculations the chemical compositions and densities of the samples were taken from Table 1. Half value level (HVL) is identified to be the thickness required to reduce the of gamma-ray intensity by a factor of 1/2 (Sayyed et al., 2019; Sayyed et al., 2017). MAC and HVL were calculated in the continuous energy range of 0.1 MeV to 100 GeV. The obtained results are shown in the Table 4 and Fig. 5.

Table 4. Mass attenuation coefficient (cm^2/g) values of samples

Energy (MeV)	Mass attenuation coefficient (cm^2/g)					
	Sample					
	Paraffin	NCM1	NCM2	NCM3	NCM4	NCM5
1.50E-02	55.478	13.018	23.326	15.746	15.998	18.971
2.00E-02	26.972	7.547	15.679	7.157	7.24	11.245
3.00E-02	8.6728	2.61	5.749	2.387	2.387	3.866
4.00E-02	3.865	1.27	2.75	1.141	1.131	1.853
5.00E-02	2.088	0.756	1.579	0.678	0.668	1.079
6.00E-02	1.284	0.515	1.025	0.79	0.457	0.716
8.00E-02	0.631	0.309	0.644	0.44	0.283	0.407
1.00E-01	0.39	0.419	0.878	0.302	0.216	0.594
1.50E-01	0.202	0.221	0.387	0.184	0.157	0.288
2.00E-01	0.148	0.159	0.242	0.145	0.134	0.195
3.00E-01	0.11	0.114	0.149	0.114	0.112	0.132
4.00E-01	0.094	0.096	0.116	0.099	0.099	0.108
5.00E-01	0.084	0.085	0.099	0.089	0.09	0.094
6.00E-01	0.077	0.077	0.089	0.082	0.083	0.085
8.00E-01	0.067	0.067	0.075	0.072	0.073	0.073
1.00E+00	0.06	0.06	0.066	0.064	0.065	0.065
1.50E+00	0.048	0.048	0.053	0.052	0.053	0.053
2.00E+00	0.042	0.042	0.046	0.045	0.046	0.045
3.00E+00	0.036	0.034	0.038	0.037	0.037	0.037
4.00E+00	0.033	0.03	0.034	0.032	0.032	0.033
5.00E+00	0.031	0.027	0.031	0.029	0.029	0.03
6.00E+00	0.03	0.025	0.029	0.027	0.027	0.028
8.00E+00	0.03	0.023	0.027	0.025	0.025	0.026
1.00E+01	0.03	0.021	0.026	0.023	0.023	0.024

According to Table 4 when MAC values are smaller than paraffin of new composite samples in the low energy region, it is bigger than, at the high energy region. It is clearly seen that, the theoretical results are highly dependent on the chemical compositions and mass percent ratio of the samples and incoming gamma ray energy. In the low energy region, it can be seen that the MAC values decreases with the increase in gamma ray energy all of the samples. This is because, as a result of the photoelectric absorption process. In this region the effective cross section of the samples is inversely proportional to the gamma-ray energy. Middle energy region (about 1 MeV) is the most common event Compton scattering. There is no sharp decrease in MAC values due to increasing gamma ray

energy in this region. When Table 4 is examined, it is seen that the NCM2 and NCM5 samples have the highest MAC values for all energies compared to other materials. This event may be caused of these samples contains different heavy metal. It is shown that, the samples of NCM2 and NCM5 has the both high effective atomic number and electron density. In addition to MAC values, the half value layer (HVL) of the samples was calculated in the energy range as shown in Fig. 5. The HVL indicates the thickness of the material required to halve the radiation intensity on a material. As it is seen from Fig. 5 the all samples has the HVL values smaller than in the paraffin. The small HVL value of a material means that higher gamma ray absorption ability (Ott et al., 1989).

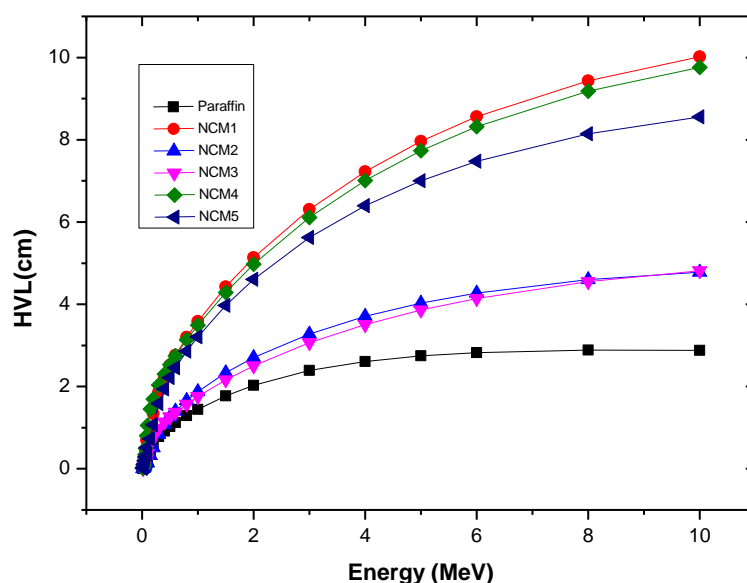


Figure 5: Variations of radiation shielding parameters in continuous energy range a; MACs b; HVLs

4. Conclusion

The new epoxy based composite samples were designed and produced at the five different content. Mass attenuation coefficients (cm^2/g) and half value layer were calculated all of the samples for the energy

range of 0.1 MeV to 100 GeV, gamma radiation. Obtained results compared with paraffin, it is seen that, all produced new composite samples have gamma-ray shielding capacity higher than paraffin. The equivalent absorbed dose was measured in all of the samples for fast neutron radiation. 4.5

MeV fast neutron particles shielding parameters were calculated such as total macroscopic cross section, mean free path and transmission number. It was founded both the total cross section, mean free path, transmission number values and absorption ability for fast neutron particles of the new composite samples better than the paraffin and conventional concrete. Obtained results compared with paraffin, it is seen that, all produced new composite samples have gamma-ray shielding capacity higher than paraffin. Metal oxide filled new epoxy composite samples have very high-quality shielding ability for both neutron and gamma radiation applications fields in measured energies. This materials can be used in nuclear applications for radiation safety.

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