NEUTRON AND PHOTON SHIELDING PARAMETERS OF SOME HYDRIDES AND BOROHYDRIDES

[a]*Olarinoye, I.O., [b]Suraju, K.O.

[a]*Department of Physics, Federal University of Technology, Minna. Nigeria.

[b]Department of Physics/Geology/Geophysics, Federal University, Ndufu Alike Ikwo, Ebonyin State.

leke.olarinoye@futminna.edu.ng; lekeola2005@yahoo.com

Abstract

The mass attenuation coefficients and effective atomic numbers of three hydrides (MgH_2, TiH_2, and VH_2), seven borohydrides ($NaBH_4$, $Mg(BH_4)_2$, $Al(BH_4)_3$, KBH_4, $Ca(BH_4)_2$, $Mn(BH_4)_2$, and $RbBH_4$) and water were calculated and compared for photon energies in the range 0.015-15 MeV. The fast neutron removal cross-section of the 11 substances were also estimated. The mass attenuation coefficients and effective atomic numbers of the materials were observed to vary with photon energy and chemical composition. The effective atomic number and photon shielding capacity was found to be highest for $RbBH_4$ and least for water throughout the photon energy spectrum. The total fast neutron removal cross section for MgH_2, TiH_2, VH_2, $NaBH_4$, $Mg(BH_4)_2$, $Al(BH_4)_3$, KBH_4, $Ca(BH_4)_2$, $Mn(BH_4)_2$, $RbBH_4$ and water were 0.111, 0.1652, 0.1998, 0.109, 0.1322, 0.0725, 0.0867, 0.0781, 0.0737, 0.0847 and 0.1029, respectively. Neutron shielding capacity of the materials depended on their hydrogen partial fraction. The hydrides are better neutron shields compared to the borohydrides and water.

Keywords: Photons; Neutrons, Interaction cross-section; Hydrides; Borohydrides

1. Introduction

Over the years, research in nuclear science and technology has been very active. This is due to the wide applications of nuclear energy in diverse fields such as agriculture, medicine, industries and for research. Nuclear radiations are often released in many nuclear processes involved in these applications. Although, nuclear energy presents immense benefits for mankind, however, uncontrolled exposure of man and the biota to components of nuclear radiation has dangerous health and environmental implications. The continuous and effective use of nuclear energy and technology consequently depends to a large extent on the ability to confine the radiations in such a way that it does not cause harm to man and his environment.

One of the methods of radiation protection is through the use of radiation shield. Radiation shielding involves confining the nuclear radiation to a volume of space. Shielding requires no administrative control unlike other forms of radiation protection. An ideal shield would not only attenuate considerably the primary radiation, but should also not be a source of secondary radiation. The effectiveness of any shield depends on its chemical composition and radiation parameters such as type (particulate or electromagnetic) and energy. In a nuclear reactor facility, neutrons and photons (gamma rays are of major concern to nuclear engineers when designing structures for the purpose of radiation shielding. This is due to their massless (photons only) and uncharged nature which enables them to penetrate deeper into any given medium. Materials for gamma rays attenuation are required to be of high density, on the other hand, fast neutron shields require low density hydrogenous materials as moderators and materials rich in elements (B, Eu, Pu, Cd) that have high neutron absorption cross-section for the absorption of the moderated neutrons. However, low density materials emit gamma rays whose energy is in the range 0.10- 10 MeV [1] when used for neutron shielding. Obviously, a combination of low density hydrogen rich materials and high atomic number materials are required for the construction of an effective absorber for neutrons [2-4]. This would not only provide adequate absorption for neutrons as well as for the secondary radiation (captured gamma rays).

Traditionally, radiation shield materials include lead, water, depleted uranium, polythene, light and heavy concretes [5-10]. Nevertheless, some of these materials have major drawbacks- the toxicity of lead has limited its use; depleted uranium is relatively less abundant, and also has radiation issues; concrete has durability and space issues as a result of its bulkiness [11, 12]. In addition to these, concrete loose its water and hence its hydrogen content as temperature increases [12]. Water on the other hand, is a liquid and thus require a container. These and many more problems have made research into alternative shielding materials very active and necessary.

Hydrides and borohydrides have been considered as good hydrogen storage materials due to their high hydrogen densities. Consequently, they have attracted much research interest especially in the area of energy storage [13-15]. Their high hydrogen content has also made them potentially effective moderators and absorbers of fast neutrons but they may not be effective in shielding the secondary photons that accompanies such interactions due to their low atomic masses. This report thus aims at evaluating the shielding effectiveness of three heavy hydrides (VH_2, MgH_2 and TiH_2), seven borohydride ($NaBH_4$, $Mg(BH_4)_2$, $Al(BH_4)_3$, KBH_4, $Ca(BH_4)_2$, $Mn(BH_4)_2$, and $Rb(BH_4)_2$) materials and water with respect to gamma rays and fast neutrons. The relationship between their shielding capacity and hydrogen content is explained. Shielding parameters such as; fast neutron removal cross section, mass attenuation coefficient, effective atomic numbers of the 11 hydrogen rich materials were also estimated and compared.

2. Theory and Calculations

2.1. Fast neutron removal cross section: The macroscopic effective removal cross section is a parameter that can be used to characterise neutron penetration in a material. It represents the probability that a fast neutron is removed from incident neutron beam [16, 17]. For fast neutron energy in the range 2-12 MeV, the effective removal cross section is a constant [18]. A lot of formulae has been suggested for the evaluation of the macroscopic effective removal cross-section (Σ_R) of fast neutrons (19, 18-22). These expressions have also been used for the evaluation of Σ_R for many elements [18-20]. For homogenous materials, Σ_R can be evaluated using the mixture rule [16]:

$$\Sigma_R = \sum_i w_i \left({}^{\Sigma_R}/_{\rho} \right)_i \qquad (1)$$

Where, w_i and ${}^{\Sigma_R}/_{\rho}$ represent the partial density and mass removal cross section of the i^{th} element in the compound respectively.

2.2. Mass Attenuation Coefficients

The mass attenuation coefficient (μ_m) of a medium is a quantity that characterises its interaction with photons. According to the Beer- Lambert's law the transmission photon flux (I) through a medium of thickness t, when photon flux (I_o) is incident on it is predicted by the equation:

$$I = I_o e^{-\mu_m t} \qquad (2)$$

The mass attenuation coefficient depends on atomic number of the medium and the photon energy. For composite material (chemical compound or homogeneous mixture), μ_m is expressed as:

$$\mu_m = \sum_i w_i (\mu_m)_i \qquad (3)$$

Where w_i and $(\mu_m)_i$ are the weight fraction and mass attenuation coefficient of the *i*th elemental constituents in the compound. The μ_m of any medium at a specified energy also depends majorly on the photoelectric, Compton scattering and the pair production interaction coefficients [23]. The dominance of any of these three interaction modes dictates the magnitude of μ_m. Generally, μ_m measures the degree to which a material can attenuate photons.

2.3. Effective atomic number (Z_{eff})

The interaction cross section of photon with any medium is a function of its chemical constituents. For pure elements, their chemical characteristics may be described by their atomic number. However, for a composite material (mixture or compound), the effective atomic number (Z_{eff}) is used to define their response to electromagnetic radiation. The Z_{eff} is a parameter that conveniently represents the interaction of a medium with photons for the purpose of radiation absorbed dose measurement, shielding construction and identification of tissue equivalent material. Unlike the atomic number of pure elements, Z_{eff} is not a constant but rather depends on photon energy.

3. Methodology

Three hydrides (VH_2, TiH_2, and MgH_2), 7 borohydride ($NaBH_4$, $Mg(BH_4)_2$, $Al(BH_4)_3$, KBH_4, $Ca(BH_4)_2$, $Mn(BH_4)_2$, and $Rb(BH_4)_2$) and water were analysed for their neutron and photon shielding capacity. The chemical formulae, physical density and hydrogen density in these materials were obtained from literature [13, 18, 24] and presented in Table 1.

3.1 **neutron removal cross section:** The macroscopic effective removal cross section (Σ_R) for the hydrides and borohydrides were evaluated through the use of a computer program- WinNC-toolkit [22]. The program was designed for theoretical determination of attenuation coefficients of fast neutrons in composite materials. The results from the toolkit has been verified to agree with good accuracy to experimental data [22].

3.2 **Mass attenuation coefficient:** The mass attenuation coefficients of the materials were estimated theoretically via the use of the WinXCom computer code [25] for photon energies from 0.01-15 MeV. This code is the window version of an earlier program (Xcom) developed by Berger and Hubbel [26]. The program can be used to calculate photon interaction cross section for element, compound and mixture in the energy spectrum of 1 keV-1 GeV.

3.3. **Estimation of the Effective atomic number (Z_{eff}):** the values of $(\mu_m)_i$ for each compound obtained from WinXCom was used to calculate the total molecular cross section according to the equation:

$$\sigma = \mu_m \frac{M}{N_A} \qquad (4)$$

.Where, M and N_A is the molecular weight of each compound and the Avogadro's number respectively. The total atomic cross section (σ_t) was then calculated using the equation:

$$\sigma_t = \sigma / \sum_i n_i \qquad (5)$$

Here**,** n_i is the number of formula unit of the i^{th} element. The effective atomic number of the water, halides and boro-hydrides was then calculated using the equation [27]:

$$Z_{eff} = {}^{\sigma_t}/_{\sigma_e} \qquad (6)$$

Where, σ_e is the total electronic cross section of the material and it is given as [23]:

$$\sigma_e = \frac{1}{N_A}\sum_i \frac{(fA)_i}{Z_i}\mu_i \quad (7)$$

The weight proportion of the *i*th element with atomic mass number in a compound is represented by *f* and *A* respectively in equation x.

4. Results and Discussions

The fast neutron removal cross-section for the hydrides is presented in Figure 1. The cross section was 0.111, 0.1652 and 0.1998 for MgH_2, TiH_2 and VH_2 respectively. The increase in neutron removal ability was consistent with their hydrogen (content neutron removal cross section) partial density as depicted in the same figure (Figure 1). This is due to the fact that hydrogen has higher neutron cross section compare to any of Mg, Ti or V. Consequently higher hydrogen partial density implies higher affinity for neutron removal. The removal cross section for the considered borohydrides, partial removal cross section of hydrogen and boron in the compounds are shown in Figure 2. The removal cross section was higher for borohydride with higher combination of boron and hydrogen removal cross sections irrespective of the physical densities and atomic molar masses of the borohydrides. The total fast neutron removal cross section for $NaBH_4$, $Mg(BH_4)_2$, $Al(BH_4)_3$, KBH_4, $Ca(BH_4)_2$, $Mn(BH_4)_2$, and $RbBH_4$ are: 0.109, 0.1322, 0.0725, 0.0867, 0.0781, 0.0737, and 0.0847 respectively, while that of water was found to be 0.1029. From the results, MgH_2, VH_2, $NaBH_4$, and $Mg(BH_4)_2$ are better fast neutron moderators than water and thus could be used for the construction of neutron shield in a nuclear facility. The partial neutron removal cross section for hydrogen which is directly linked with the hydrogen partial density is a vital factor for the relative shielding efficacy of neutron of all the considered materials.

The mass attenuation coefficients (μ_m) for the eleven substances in the low energy (0.015-0.1 MeV) and high energy (0.2-15 MeV) ranges are presented in Figures 3a and 3b respectively. The changes in the mass attenuation coefficients of the hydrides, borohydrides and water behave similarly within the presented photon energy spectrum. The μ_m were higher at lower energy region and vice versa for all the materials. Between 0.015 and 0.04 MeV, mass attenuation was highest. Beyond this energy, the μ_m decreased rapidly with energy up to 0.1 MeV. From 0.1 MeV to 4 MeV the mass attenuation coefficients remain relatively stable for all considered substances after which a rise in value begins (Figure 3b). This trend is consistent with theoretical argument which predicted high attenuation coefficients for low energy photons in any material due to the removal of low energy photon from photon beam after interaction with any medium [23]. This removal can be attributed to two main reasons; firstly, low energy photons have high interaction coefficient thus possess more tendency to loss a good part of their energy during collision with the atoms of the interacting medium. This leads to further loss of photon energy and ultimately their absorption. Secondly, photoelectric effect and Compton scattering are two major interaction modes that takes place within the region below 2 MeV. These two modes of interaction depletes the population of photons in a beam incident on a material medium. While the photoelectric effect removes completely photons from a beam, incoherent scattering reduce considerably the energy of the interacting photons. These two events account for the loss of photons at the lower end of the spectrum and thus accounts for the relative high attenuation coefficient for a material when compared to the high energy region. At the higher energy region, the photons are very energetic and penetrate deeper into the material without interacting. This explains why the attenuation is low at low energy. although pair production takes place at energies greater than 1.02 MeV, and also annihilate photons, this is not enough to cause a rapid increase in the attenuation but rather gradual as shown in the figure beyond 4 MeV. Comparatively, from the figures, and Table 1, it appears that the physical density of the substances play no major role in their photon shielding efficacies as may be expected. This is probably due to the fact that the use of physical density for selecting photon shielding materials is only applicable to mixtures of similarly compound such as concretes with different mixture composition. For pure compounds, the chemical constituents and their concentrations play a more vital role. In the low energy region, the mass attenuation coefficients for the substances in the following order: $RbBH_4$ > VH_2 > TiH_2 > $Mn(BH_4)_2$ > KBH_4 > $Ca(BH_4)_2$ MgH_2 > $NaBH_4$ > $Al(BH_4)_3$ >H_2O> $Mg(BH_4)_2$. However, in the high energy region, there seems to be no noticeable difference between the mass attenuation coefficients of all the substances considered.

The effective atomic numbers and their variations with photon energy within the range 0.015-15 MeV is shown in Figure 4. Obviously, the changes in Z_{eff} with respect to photon energy is almost similar for all the eleven substances considered. From the figure, the Z_{eff} decreased steadily from their maximum value in the low energy region (0.015-0.1 MeV) of the spectrum. Thereafter, the values become relatively stable before increasing slightly towards the end of the energy spectrum. The peak value of Z_{eff} for all the materials was obtained at 15 KeV while the minimum values were observed at different energies for different material between photon energy range of 0.5 MeV and 1 MeV. The observed changes in Z_{eff} with energy of photons can be attributed to photon interaction modes with each substances. Within the energy region of interest in this study, the photoelectric effect, Compton (incoherent) scattering and pair production are the three dominant interaction modes for photons. In the low energy region (0.015-0.04 MeV), photoelectric effect dominates, while Compton scattering and pair production dominated at intermediate (0.05-1 MeV) and high energy (above 1.5 MeV) regions respectively. The photoelectric effect (τ), Compton scattering (σ) and pair production (κ) interaction coefficients depends on energy and atomic number according to the following equations [27]:

$$\tau = a\frac{Z^5}{E^3} \quad (8)$$

$$\sigma = b\frac{Z}{E} \quad (9)$$

$$\kappa = cZ^2(E - 1.02) \quad (10)$$

Where *a*, *b*, *c* are constants. Consequently, the effective atomic numbers of the substances are higher in the lower energy region where photoelectric effect is dominant and least in the region of Compton scattering as predicted by the equation 1-3. The final rise in the Zeff at the later energy region is predicted nby equation 2 due to the predominance of pair production.

From the results, for each of the eleven materials, the upper and the lower limits of the Zeff depend on the atomic composition of the compounds. The upper and lower limits did not exceed the atomic number of the heaviest and lightest atoms respectively in the composite material. This explains why compound with high atomic number atoms have higher effective atomic number at all energies and vice versa. $RbBH_4$ had the highest Zeff throughout the considered energy spectrum while water and Al(BH4)3 had the least in the low energy region and high energy region respectively. The Zeff of the materials varied from 4.66; 8.01; 8.34; 3.33; 2.73; 2.50; 18.26; 3.46; 3.92; 7.76; and 3.33 to 11.38; 21.80; 22.82; 9.28; 9.37; 9.76; 4.67; 18.70; 24.07; 35.14 and 6.74 respectively for: MgH_2; $TiH_{2;}$ $VH_{2;}$ $NaBH_4$; $Mg(BH_4)_2$; $Al(BH_4)_3$; KBH_4; $Ca(BH_4)_2$; $Mn(BH_4)_2$; $RbBH_4$;and water.

Table 1. Mass number, physical density and hydrogen density of the hydride, borohydride and water.

Material	Mass Number	Density (gcm^{-3})	Hydrogen Density (10^{22} cm^{-3})
MgH_2	26.32	1.45	6.6
TiH_2	49.88	3.77	9.1
VH_2	52.96	4.62	10.5
$NaBH_4$	37.83	1.08	6.9
$Mg(BH_4)_2$	53.99	1.48	13.2
$Al(BH_4)_3$	71.51	0.79	8
KBH_4	53.94	1.17	5.3
$Ca(BH_4)_2$	69.76	1.07	7.5
$Mn(BH_4)_2$	84.62	1.24	7.1
$RbBH_4$	100.31	1.92	4.6
Water	18.02	1	6.7

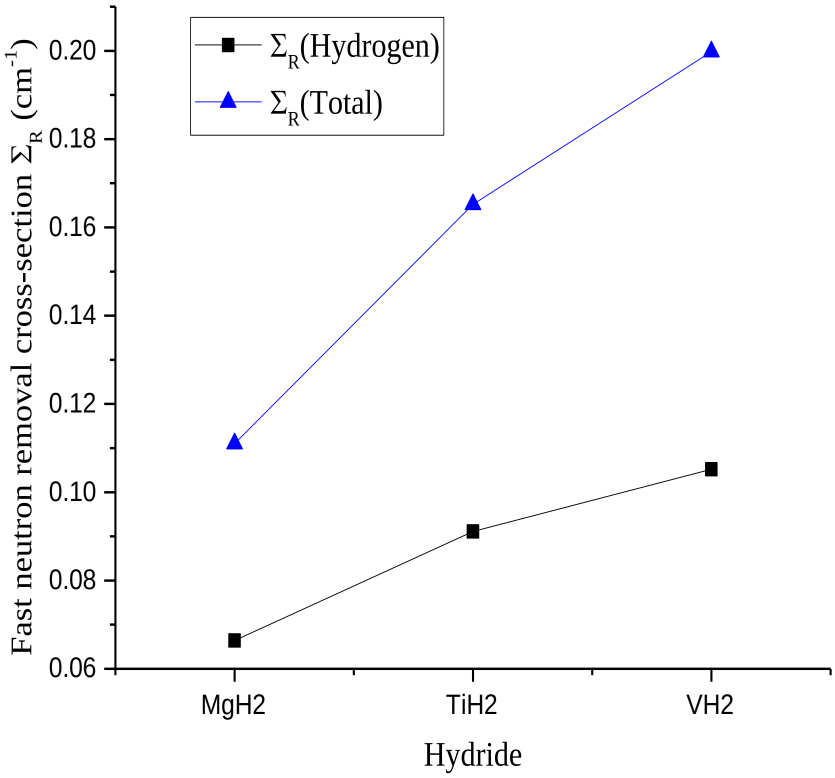

Figure 1. Fast neutron removal cross-section for the hydrides and hydrogen content

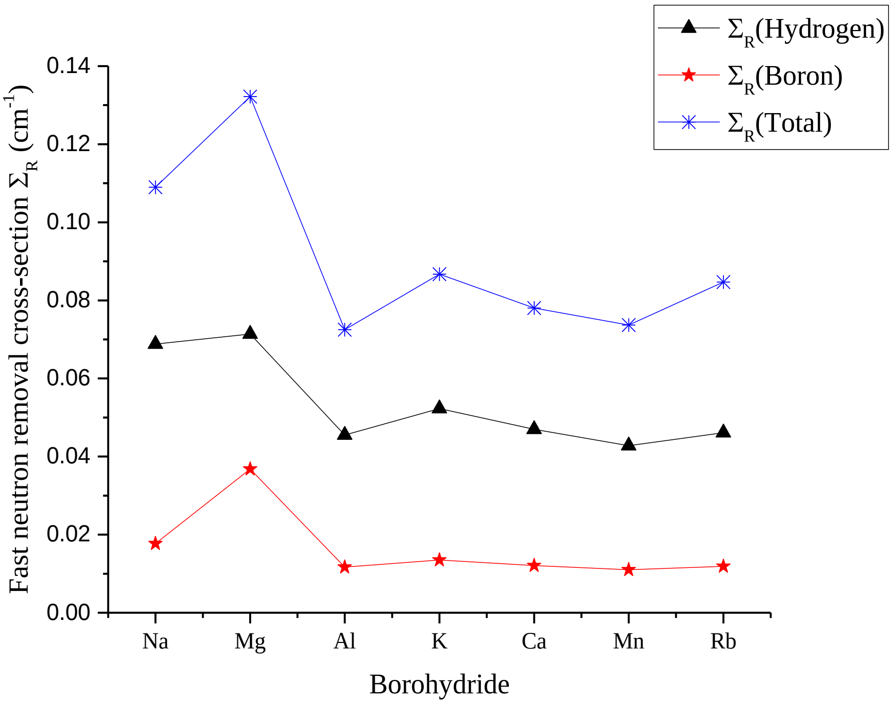

Figure 2. Fast neutron removal cross section for the borohydrides and boron and hydrogen content.

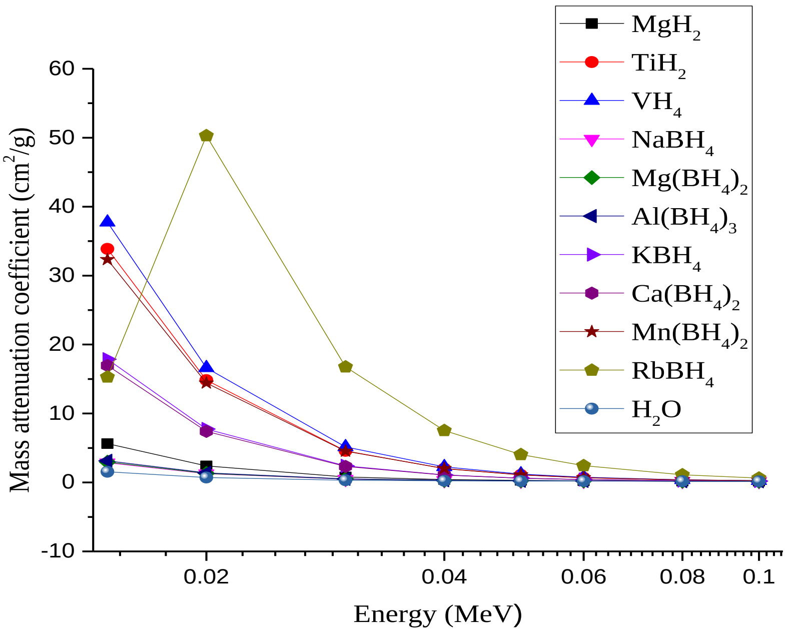

Figure 3a. Mass attenuation Coefficieents of the substances in the low energy region.

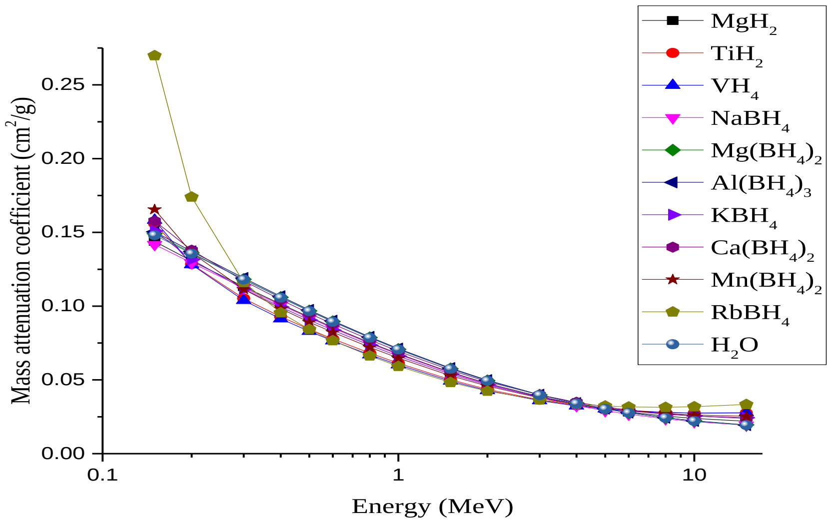

Figure 3b. Mass attenuation coefficients of the substances in the high energy region.

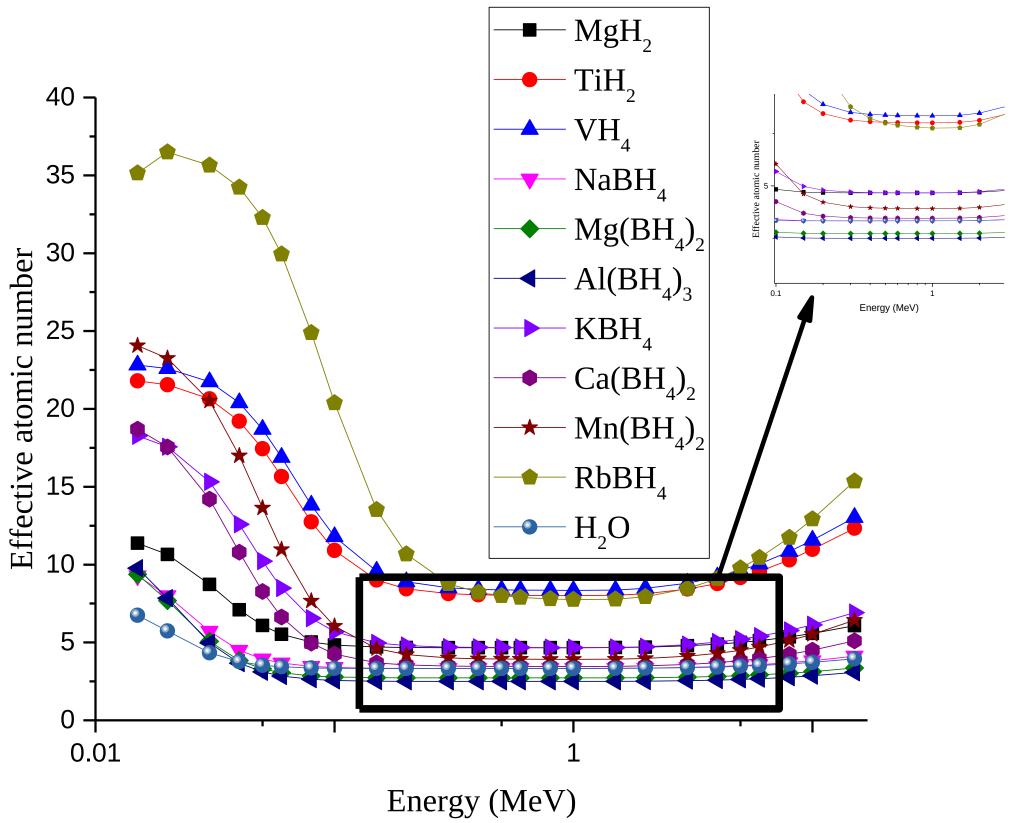

Figure 4. Effective atomic number of the substances.

5. Conclusion

The neutron and photon shielding capacities of three hydrides (MgH_2, TiH_2, and VH_2), seven borohydrides ($NaBH_4$, $Mg(BH_4)_2$, $Al(BH_4)_3$, KBH_4, $Ca(BH_4)_2$, $Mn(BH_4)_2$, and $RbBH_4$) and water were calculated and compared. For the purpose of fast neutron moderation or shielding, the partial hydrogen density and neutron removal cross section plays a vital role irrespective of physical densities of the materials. The calculated mass attenuation coefficients of the materials vary with photon energies in similar version but differ in magnitude from one material to the other. The mass attenuation coefficient was higher for compound containing denser atoms and smaller for compound with less heavy atoms. From the results, $RbBH_4$ showed better photon shielding capacity compared to other compounds while water was the least effective for photon shielding. The effective atomic number varied with photon energy as predicted by the partial photon interaction modes (photoelectric effect, Compton scattering and pair production) dominant at a particular energy. Although the effective atomic number is an approximation and similar in function to the atomic number of elements for the purpose of radiation control and measurements, the observed variation with photon energy thus suggest that a single digit cannot be used to represent it for compounds and mixtures. Rather, the value to be used for this purpose will depend greatly on the spectrum of photon energy of interest.

Generally, the requirement for good neutron shield are not the same for photon shield, consequently, the same material cannot be used for the same function with equal efficiencies. From the results of this research, $RbBH_4$ shield photons better while VH_2 presents a better neutron shield out of all the materials considered. However in a mixed radiation environment comprising fast neutron and photons, a combination of shielding materials to serve for both photons and neutrons are recommended. A combination of VH_2 and $RbBH_4$ or a material containing both compounds could be a perfect shield for radiation protection of man and the biota against the harmful effects of the radiations.

References

[1]. V.P. Singh, M.E. Medhat, N.M. Badiger, Nuclear Engineering Design, 270 (2014) 90-100.

[2]. S.W. Gary, Fundamental of Radiation Material Science, Springer-Verlag, Berlin, 2007.

[3]. T. Hayashi, K. Tobita, Y. Nakamori, S. Oriomo, Journal of Nuclear Materials 386-388 (2009) 119-121.

[4]. T. Hayashi, K. Tobita, S. Nishino, et al., Fus. Eng. Des. 81 (2006) 1285.

[5]. A.S. Makarious, et al., Ann. Nucl Eng. 23 (1996) 3, 195-206.

[6]. S. M. J. Mortazavi, Radiat. Prot. Dosimetry, 142 (2010), 2-4, 120-124.

[7]. I.I. Bashter, Ann. Nucl. Energy, 24 (1997), 17, 1389-1401.

[8]. Oyeleke O.I. (2009). *Journal of science, education and technology, 2(1). 197-202.*

[9]. M.R. Shavers, N. Zapp, R.E. Barber, J.W. Wilson, G. Qualls, L. Toupes, S. Ramsey, V. Vinci, G. Smith, F.A. Cucinnotta. Adv. Space Res. (2004) 1333-1337.

[10]. J. Miller, C. Zethlm, F.A. Cicinotta, L. Heilbronn, D. Stephens, J.W. Wilson. Radiat. Res. 159 (2003) 381-390.

[11]. Elbio Calzada et al., Nucl. Instruments and Methods in Physics Research, A 651 (2011), 77-80.

[12]. A. Akkurt et al., Radiat. Meas. 45 (2010) 827-830.

[13]. S. Oriomo, Y. Nakamori, J. Eliseo, A. Zuttel, C.M. Jensen.Chem. Rev. 107 (2007) 4111-4132.

[14]. Y. Nakamori, K. Miwa, A. Ninomya, H. W. Li, N. Ohba, S. Towata, Z. Zuttel, S. Orimo, Phys. Rev. B. 74 (2006) 045126 (1).

[15]. S. Oriomo, Y. Nakamori, A. Zuttel, Mater. Sci. Eng. B 108 (2004) 51.

[16]. E.P. Blizard, L.S. Abbott, Reactor Handbook, Vol. III, part B, Shielding Wiley, New York, 1962.

[17]. J.J. Duderstadt, L.J. Hamilton, Nuclear Reactor Analysis (Wiley, New York) 1976.

[18]. M. F. Kaplan, concrete Radiation Shielding, Wiley, New York, 1982.

[19]. V.P. Singh, N. M. Badiger, J. Fusion Energ. 2014 DOI10.1007/S10894-014-9679.4

[20]. A.E. Profio, Radiation Shielding and Dosimetry, Wiley, New York, 1979.

[21]. A.B. Chitton, J.K. Shultis, R.E. Faw, Principles of Radiation Shielding, Prentice-Hall, Eaglewood Cliffs, N.J. 1984.

[22]. K.S. Mann., Toolkit for fast neutron removal cross-section, 3rd international Conference Advancements in Engineering and Technology, Bhai Gurdas Institute of Engineering and Technology, Sangrur (Puanjab), India.

[23]. Olarinoye, I.O. Res. J. Chem Sci. 1(2) (2011) 64-69.

[24]. K. Tobita, S. Konishi, S. Nishio, K. Kosako, T. Tabaro, Letters 10, (2001) 1035-1039.

[25]. L. Gerward, N. Guibert, K.B. Jensen, H. Levring. 2004. WinXCom: a program for calculating X-ray attenuation Coefficients. Radiat. Phys. Chem. 71, 653-654.

[26]. M.J. Berger, J.H. Hubbel, 1987 NBSIR 87-3597, XCOM: photon cross section on personal computer. National institute of standards. Gaithersberg, M.D. 20899, U.S.A.

[27]. E.M. James (2006)Physics for radiation protection: a handbook. Wiley-VCH, Verlag GmbH and Co. Weinheim, 822.