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A Study on the Gamma-Ray Attenuation Coefficients of Al_2O_3 and $Al_2O_3.TiO_2$ Compounds

Nimet ZAIM*[1], Ozkan BAYHATUN[1]

[1]Trakya Universitesi, Faculty of Science, Department of Physics, 22030, Edirne

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$Al_2O_3.TiO_2$,
NaI (Tl) detector

Abstract: In this study, micro and nanometers-sized powders of Al_2O_3 and $Al_2O_3.TiO_2$ were prepared using a hydraulic cold press and their gamma-radiation shielding properties were assessed at photon energies of 661.7, 1173.2 and 1332.5 keV. A sample counting process was implemented using a gamma-ray spectrometer with a 3×3 NaI (Tl) detector and 13384-channel multichannel analyser. The results we obtained indicated that the gamma-shielding properties of the materials increased with TiO_2 addition and also, the particle size found to be an important effect on the properties. The results were compared with each other and also compared with the results of similar studies reported in literature.

Al_2O_3 ve $Al_2O_3.TiO_2$ Bileşiklerinin Gama Zayıflama Katsayıları Üzerine Bir Çalışma

Anahtar kelimeler
Gama ışını zayıflaması katsayıları,
Al_2O_3,
$Al_2O_3.TiO_2$,
NaI (Tl) detektor

Özet: Bu çalışmada parçacık boyutları mikrometre ve nanometre düzeyinde olan toz halindeki Al_2O_3 ve $Al_2O_3.TiO_2$ bileşikleri, soğuk hidrolik pres metodu kullanılarak deneye hazırlandı ve 661.7, 1173.2 ve 1332.5 keV foton enerjileri dikkate alınarak gama-radyasyonuna karşı zırhlama özellikleri incelendi. Örnekler için sayım işlemi 3×3 NaI (Tl) dedektörü ve 13384-kanalı olan çok kanallı gama-radyasyonu spektrometresi kullanılarak gerçekleştirildi. Elde edilen sonuçlardan, malzemelere TiO_2 katkısının, malzemelerin gama zırhlama özelliklerini arttırmakta olduğu ve bu bulguya ilaveten malzemelerin parçacık boyutlarının da zırhlama özelliklerini etkilemekte olduğu anlaşıldı. Elde edilen sonuçlar kendi aralarında ve de literatürdeki mevcut benzer çalışmaların sonuçları ile kıyaslandı.

1. Introduction

The increasingly widespread usage of radioisotopes in recent years has created a radiation-protection problem, owing to which shielding solutions had to be developed. We conducted a literature search to obtain data on the radiation-attenuation properties of of some materials [1-10]. Bagheri et al. examined the mass attenuation coefficients of silicate glasses containing different concentration of Bi_2O_3, PbO, and BaO using XCOM and XMuDat programs, in the energy range of 10 keV-10 MeV [1]. The mass absorption coefficients of alloys such as brass, bronze, steel, aluminium-silicon and lead-antimony have been measured by El-Kateb et al. [2]. Bulk of aluminium close-cell composite metal foams and

open-cell aluminium foam infiltrated with variety of second phase materials were investigated by Chen et al. [3]. And Buyuk et al. studied the composite materials titanium diboride (TiB_2) reinforced boron carbide-silicon carbide, with particle size 3.851 µm and 170 nm for titanium diboride [5].

From this survey, it could be concluded that the development of novel materials to protect against the destructive effects of radioactivity is important. In order to achieve this objective, in this study, aluminium oxide and aluminium titanate were investigated to determine their suitability to be used as shielding materials.

Although high-Z materials and composite materials show high resistance to radiation, because of the difficulties in their use and high price, we focused on aluminium compounds in this investigation. Aluminium alloys have several advantageous properties, such as low density and cost. Apart from these well-known properties of Al_2O_3, the following features can be listed in its benefits column – good electrical insulation (1×10^{14} to 1×10^{15} Ω cm), moderate to extremely high mechanical strength (300 to 630 MPa), high compressive strength (2,000 to 4,000 MPa), high hardness (15 to 19 GPa), moderate thermal conductivity (20 to 30 W/mK), high corrosion and wear resistance, good gliding properties, operating temperatures of 1,000 to

* Corresponding author: nnimetzaim@yahoo.com.tr

1,500°C without mechanical loading, bioinert nature, and food compatibility. [11]. $Al_2O_3.TiO_2$ also has several advantageous features, such as an excellent thermal shock resistance (0–1,000 °C), very low thermal expansion (<1×10^{-6} K^{-1} between 20 and 600 °C), high thermal insulation (1.5 W/mK), low Young's modulus (17 to 20 GPa), good chemical resistance, and poor wettability [12].

Different parameters related to the gamma radiation-attenuation properties of the materials were studied. The linear attenuation coefficient (μ, cm^{-1}) is an important property in terms of radiation protection. The mass attenuation coefficient (μ_m, cm^2 g^{-1}), on the other hand, is more often used, as it is independent of the density of the material. The mass attenuation coefficient of a material is determined as μ/ρ, where ρ is the density of the absorbing sample. This attenuation parameter is not dependent on the density of sample and hence it can be more conveniently measured in different phases of the same medium, which naturally have different densities. In this study, the linear and mass attenuations coefficients of the samples were measured. Theoretical mass attenuation coefficients were obtained using the 'Photon Cross Section Database' (XCOM) computer code [13]. The experimental and theoretical results were compared; further, the obtained results were evaluated against those reported in literature. Furthermore two parameters that describe the gamma-ray shielding effectiveness of a material are its half-value layer (HVL) and tenth-value layer (TVL); these were calculated from the linear attenuation coefficients of the studied samples.

We investigated the total linear and mass attenuation coefficients and the HVL and TVL of aluminium oxide and aluminium titanate samples. Results were compared with those reported in the literature.

2. Material and Method

The gamma-transmission technique is a non-destructive method to measure the gamma-shielding properties of a material. The calculations are based on the data obtained by comparing the intensity of the radiation incident on the material and the intensity of the radiation passing through the material. We designed an experimental setup for conducting these measurements [14]. The gamma source, detector, and the sample were placed on the same vertical axis. The positions of the source and sample containers (lead) were adjustable. Two collimators were placed on both sides of the sample; collimator 1 was placed above it and collimator 2 below it. The diameter of the collimators was 12 mm. Thus, the photon beam could be obtained with a narrow beam geometry. The entire setup was surrounded by a 3 mm-thick lead cylinder. In all the measurements, the distances between detector-sample was 4 cm and sample-source was 12 cm exactly.

The linear attenuation coefficient of a material with respect to gamma rays is calculated from the exponential attenuation law, $I = I_0 e^{-\mu x}$, where I and I_0 are the transmitted and incident (incoming) intensities, respectively, and x is the thickness of the sample. The linear attenuation coefficients (μ), (R^2) and standard error for each linear coefficients were obtained from the graphs plotted using the Origin 9 computer program.

The gamma-ray photon relative transmission rates through the samples were measured using a gamma-ray spectrometer with a 3×3 NaI (Tl) detector and 13384-channel multichannel analyser. The energy resolution of the spectrometer was 2.1% with respect to the 1332.5 keV gamma-ray line of Co-60 (full-width at half maximum (FWHM) of 70.44%). Analysis of the spectrum was carried out using a spectrum-receiving and analysing software called ORTEC. For energy calibration of the system, three gamma lines were used – a Co-60 point source (1173.2 and 1332.5keV) and a Cs-137 point sources (661.7 keV). The overall uncertainty in the counted values was calculated by the software and was found to be in the range of 2%–4%.

The prepared samples were investigated against Co-60 and Cs-137 radioisotope sources. Co-60 has two gamma peaks at 1173.2 keV and 1332.5 keV, while Cs-137 has a single gamma peak at 661.7 keV. The gamma radioisotope sources exhibit an activity of 1 μCi. The measurement time was 1000 s for both Co-60 and Cs-137 gamma sources. All the measurements were carried out thrice for each sample.

The samples used in this study, aluminium oxide and aluminium titanate, were in the powder form and their dimensions were in the micrometers and nanometers range. These samples were examined as disc-shaped compressed tablets, which is very practical for measurement studies. Disc-shaped compressed tablets were prepared from powders using stainless steel evacuable pellet dies and a hydraulic cold press at a pressure of 40 MPa (fusion manual pressing method) in average 5 minutes. To our knowledge, this is the first time that aluminium oxide and aluminium titanate samples in the tablet form have been used to analyse gamma-ray attenuation properties.

The samples were denoted as $Al_2O_{3;}$mm, $Al_2O_{3;}$nm, $Al_2O_3.TiO_{2;}$mm, and $Al_2O_3.TiO_{2;}$nm. The radius of each sample was 1.5 cm and their thickness ranged from 0.492 cm to 4.046 cm. While $Al_2O_{3;}$mm and $Al_2O_3.TiO_{2;}$mm were prepared using a cold press, $Al_2O_{3;}$nm and $Al_2O_3.TiO_{2;}$nm could not be obtained without binding materials (linkers). This may be due to the increased surface tension in these samples;

when the particle size decreases, the average particle surface area increases. Ethyl alcohol with a natural resin solution, which was used for binding, was prepared by dissolving 3 g of a natural resin in 50 mL of ethyl alcohol (95%). New $Al_2O_{3;}$nm and $Al_2O_3.TiO_{2;}$nm samples were prepared by adding 40 mL of this solution to the specimen powder (nanometers size, 40 g) and allowed to dry for 2 days, at room temperature. The dried mixtures were then pressed at 40 MPa pressure, similar to other samples. These samples were then used for further analysis.

The scanning electron microscope (SEM) images of the samples are shown in Figure 2.a, b, c, d. These images show that nanometres size particles get closer to each other and build a more compact material with fewer and smaller gaps than micrometres size particles do. This may explain the differences observed in the measured parameters of the samples.

The HVL and TVL of a sample are attenuation parameters that describe its gamma-ray shielding strength. The HVL and TVL are defined as the thicknesses of a sample at which the intensity of the primary photon beam is reduced to half and one-tenth of its original value, respectively. They can be calculated as follows [15].

$$\text{HVL} = \frac{ln2}{\mu_t}, \text{TVL} = \frac{ln10}{\mu_t} \quad (1)$$

3. Results

The linear attenuation coefficients, mass attenuation coefficients, HVL values, and TVL values of $Al_2O_{3;}$mm, $Al_2O_{3;}$nm, $Al_2O_3.TiO_{2;}$mm, and $Al_2O_3.TiO_{2;}$nm were calculated at three different photon energies and are listed in Table 1, Table 2 and Table 3.

As expected, the linear attenuation coefficients of all the studied samples decreased with an increase in the photon energy, as shown in Figure 3. It can be seen that the linear attenuation coefficients of $Al_2O_3.TiO_{2;}$mm and $Al_2O_3.TiO_{2;}$nm are higher than those of $Al_2O_{3;}$mm and $Al_2O_{3;}$nm in the energy range of 662–1332 keV (Figure 3). The dependence of the linear attenuation coefficient on Z can be observed in the results; an increased attenuation can be obtained at a high Z value. [2, 15, 16]. It is noted that linear coefficients (μ) of $Al_2O_3.TiO_{2;}$mm and $Al_2O_3.TiO_{2;}$nm are high at 661.7 keV (in a region with Compton Effect dominance).

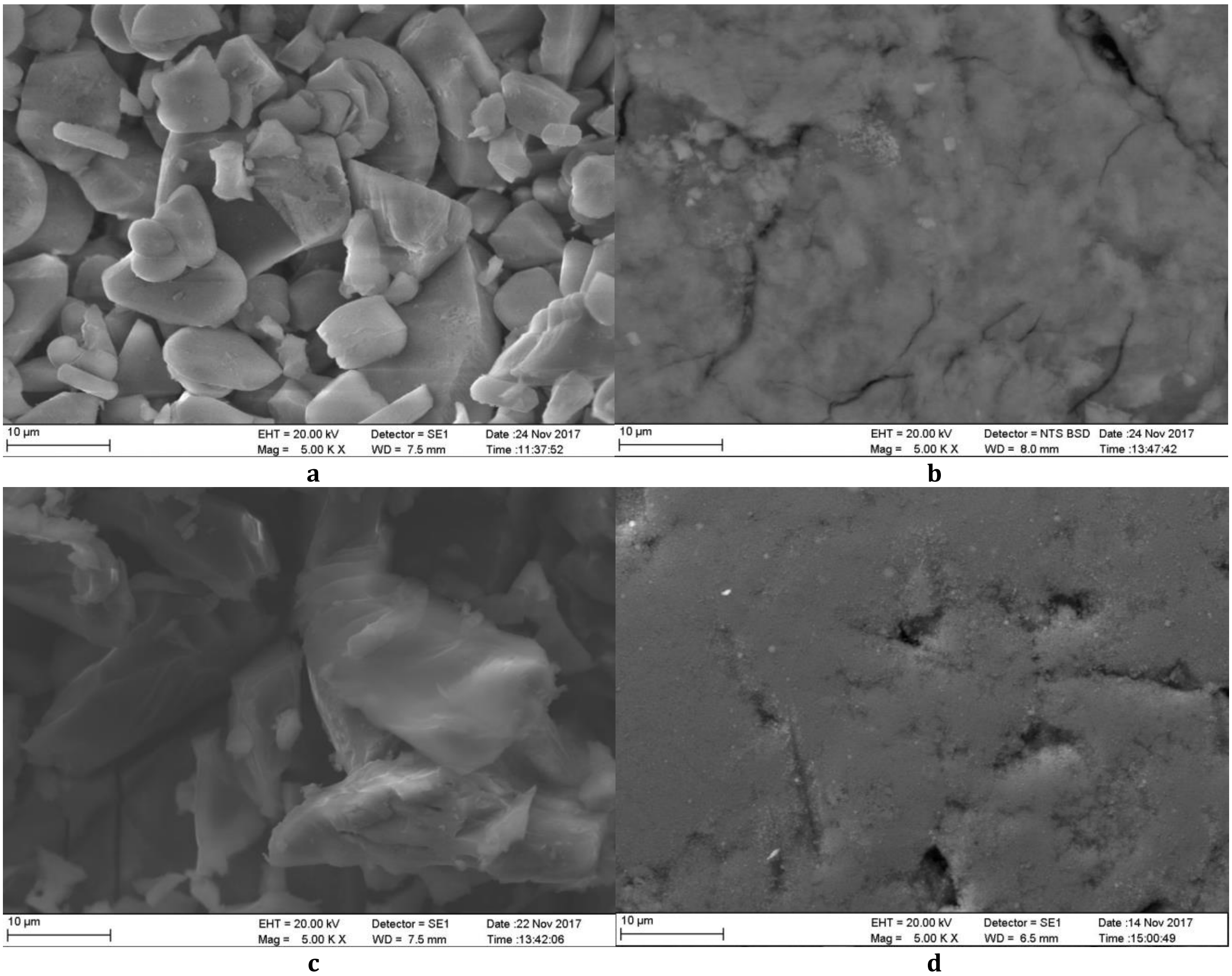

Figure 2. a. The SEM views of $Al_2O_{3;}$mm sample, **b.** The SEM views of $Al_2O_{3;}$nm sample, **c.** The SEM views of $Al_2O_3.TiO_{2;}$mm sample, **d.** The SEM views of $Al_2O_3.TiO_{2;}$nm sample

We should draw attention to the fact that in the case of Al_2O_3;mm, at 1173.2 keV, an unexpected result is obtained that the attenuation is higher than others. This can be explained by the cold fusion method, which produces samples with highly heterogenous properties.

The relative transmission rates (I/I_0) of the samples were determined at different thicknesses and the results are illustrated in Figure 4.a, b, c, d. The transmission rates were found to decrease as the sample thickness increased. While the R^2 of plot lines for three measurements has values of 0.85, 0.88 and 0.89, the R^2 values of nine other measurements varied between 0.91–0.99.

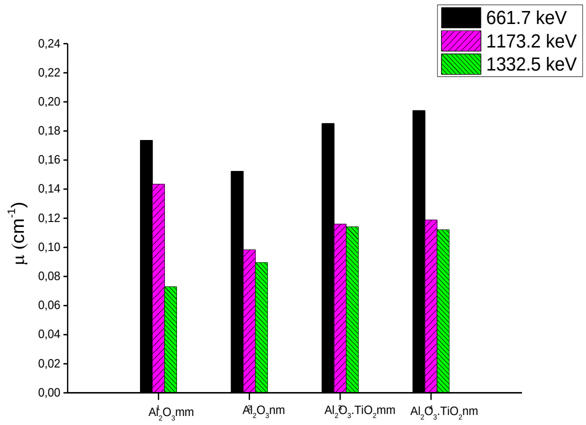

Figure 3. The variation of linear attenuation coefficients versus gamma ray energies

The mass attenuation coefficient is an important parameter in determining the gamma-ray attenuation properties of a material. The web version of it, the computer program, XCOM, developed by Berger and Hubbel [17] was used to calculate the theoretical mass attenuation coefficients **[13]**. These were then compared with the experimental values. It is determined that photon energy increases when the mass attenuation coefficient decreases.

The compared experimental and theoretical values, summarized in Table 1 and Table 2, shows more than 10% difference for four results out of twenty. Once again, we should note that the discrepancy between experimental and theoretical values may be attributed to the structural impurities in the samples, which might be generated during sample preparation or due to other conditions, such as the physical conditions of the material and the environment (pressure, humidity, temperature, and intensity of source) [1]. However, we can determine that all the parameters exhibit similar tendencies; as shown in Table 1 and Table 2, the attenuation values decrease with increasing energy.

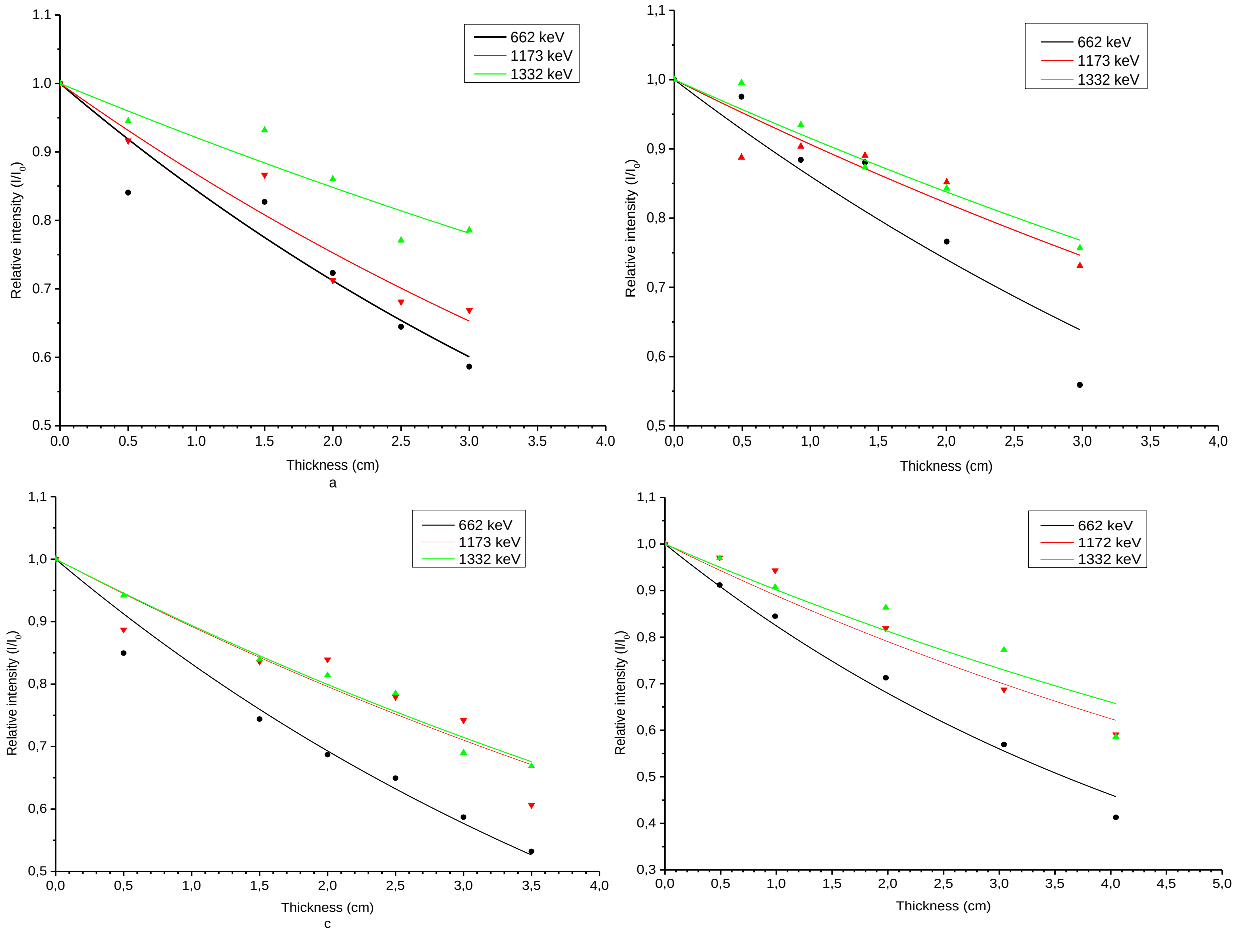

Figure 4. a, b, c, d. Transmission rate as a function of thickness for four samples (a, for Al_2O_3;mm; b, for Al_2O_3;nm; c, for $Al_2O_3.TiO_2$;mm; d, for $Al_2O_3.TiO_2$;nm) at different photon energies

The HVL and TVL values of the samples tested with 661.7, 1173.2, and 1332.5 keV gamma lines are listed in Table 2. The attenuation parameters corresponding to $Al_2O_3.TiO_2$;mm and $Al_2O_3.TiO_2$;nm are lower than those of Al_2O_3;mm and Al_2O_3;nm.

The results we obtained were further compared to the values reported in literature (Table 3) and the two sets of values were found to be compatible with each other.

Table 1. The measured linear and mass attenuation coefficients of Al_2O_3;mm, Al_2O_3;nm and comparison with theoretical values of mass attenuation coefficients for 661.7, 1773.2 and 1332.5 keV

Al_2O_3;mm (ρ=2.2276 g cm^{-3})			
Energy (keV)	μ (cm^{-1})	μ_m (cm^2 g^{-1})	μ_m (XCOM)
661.7	0.17355±0.00946	0.07791	0.07593
1173.2	0.14341±0.01053	0.06438	0.05774
1332.5	0.07292±0.00587	0.03274	0.05412
Al_2O_3;nm (ρ=1.6692 g cm^{-3})			
Energy (keV)	μ (cm^{-1})	μ_m (cm^2 g^{-1})	μ_m (XCOM)
661.7	0.15234±0.02392	0.09123	0.07593
1173.2	0.09835±0.00616	0.05892	0.05774
1332.5	0.08923±0.00587	0.05364	0.05412

Table 2. The measured linear and mass attenuation coefficients of $Al_2O_3.TiO_2$;mm, $Al_2O_3.TiO_2$;nm and comparison with theoretical values of mass attenuation coefficients for 661.7, 1773.2 and 1332.5 keV

$Al_2O_3.TiO_2$;mm (ρ=2.2481 g cm^{-3})			
Energy (keV)	μ (cm^{-1})	μ_m (cm^2 g^{-1})	μ_m (XCOM)
661.7	0.18513±0.00862	0.08235	0.07513
1173.2	0.11596±0.00891	0.05158	0.05704
1332.5	0.11411±0.00369	0.050759	0.05347
$Al_2O_3.TiO_2$;nm (ρ=2.7272 g cm^{-3})			
Energy (keV)	μ (cm^{-1})	μ_m (cm^2 g^{-1})	μ_m (XCOM)
661.7	0.19398±0.00762	0.071127	0.07513
1173.2	0.11881±0.00727	0.04356	0.05704
1332.5	0.1121±0.0115	0.0411	0.05347

The linear attenuation coefficients of Al_2O_3;mm and Al_2O_3;nm are similar to those of TiB_2-reinforced B_4C-silicon carbide composites [5] and spark plasma-sintered B_4C-Al [9]; however, they are higher than the values reported for h-BN and h-BN-$TİB_2$ composites [4] and lower than those of Al-4% Cu/B_4C metal matrix composites [6], B_4C Al metal matrix composites [7], and some stainless and BS_s steels [8] at 661.7 keV. At gamma energies of 1173.2 and 1332.5 keV, Al_2O_3;mm and Al_2O_3;nm samples have lower linear attenuation coefficients than Al-4% Cu/B_4C metal matrix composites [6] and TiB_2-reinforced B_4C-SiC [10].

When we compare the mass attenuation coefficients of Al_2O_3;mm and Al_2O_3;nm with the values reported in literature, we can determine that the results of the samples tested in this study are close to or better than the results of Reza Bagheri et al. (2018) [1], A.H. El-Kateb et al. (2000) [2], Shuo Chen et al. (2014) [3], B. Buyuk, A. B. Tugrul, A. Okan, Addemir et al. (2014) [4], B. Buyuk and A. B. Tugrul (2014) [5], A. Akkaş et al. (2015) [7], B. Buyuk (2015) [8], and B. Buyuk, A.B. Tugrul, M. Cengiz, et al. (2015) [9] at 661.7 keV. The mass attenuation coefficients of the same samples are almost similar to those reported by Reza Bagheri et al. (2018) [1], Shuo Chen et al. (2014) [3], and Büyük and A.B. Tugrul (2015) [10] at 1173.2 keV; further, they are similar to or slightly lower than the results obtained by Reza Bagheri et al. (2018) [1], A.H. El-Kateb et al. (2000) [2], Shuo Chen et al. (2014) [3], and Büyük and A.B. Tugrul (2015) [10] at 1332.5 keV.

While the linear attenuation coefficient values of $Al_2O_3.TiO_2$;mm and $Al_2O_3.TiO_2$;nm are higher than those of h-BN and h-BN-$TİB_2$ composites [4], TiB_2-reinforced B_4C-silicon carbide composites [5], and spark-plasma sintered B_4C-Al [9], they are lower than the values reported for Al-4% Cu/B_4C metal matrix composites [6], and some stainless and BS_s steels [8]. Furthermore, they are similar to the values reported for B_4C Al metal matrix composites [7] at a gamma energy level of 661.7 keV. However, the linear attenuation coefficients of $Al_2O_3.TiO_2$;mm and $Al_2O_3.TiO_2$;nm are lower than those of Al-4% Cu/B_4C metal matrix composites [6] and TiB_2-reinforced B_4C-SiC [10] at 1173.2 and 1332.5 keV.

The mass attenuation coefficients of $Al_2O_3.TiO_2$;mm and $Al_2O_3.TiO_2$;nm are slightly higher than the values reported by Büyük and A.B. Tugrul (2015) [2], Shuo Chen et al. (2014) [3], B. Buyuk, A. B. Tugrul, A. Okan, Addemir et al. (2014) [4], B. Buyuk and A. B. Tugrul, (2014) [5], A. Akkaş et al. (2015) [7], B. Buyuk (2015) [8] and B. Buyuk, A.B. Tugrul, M. Cengiz, et al. (2015) [9] and Reza Bagheri et al. (2018) (except lead oxide silicate glass sample) [1] at 661.7 keV gamma energy. However, at gamma energies of 1173.2 and 1332.5 keV, the obtained values are similar to the results of A.H. El-Kateb et al. (2000) [2], Shuo Chen et al. (2014) [3], and Büyük and A.B. Tugrul (2015) [10], but lower than those reported by Reza Bagheri et al. (2018) [1] for lead oxide silicate glass samples.

The best HVL value of 3.57 cm obtained in this study is better than the results of [9], but significantly less than the values reported for some stainless and boron steels [8].

Conclusion

- The gamma-radiation attenuation ability of four different samples, Al_2O_3;mm, Al_2O_3;nm, $Al_2O_3.TiO_2$;mm, and $Al_2O_3.TiO_2$;nm, have been investigated at gamma energies of 661.7, 1173.2, and 1332.5 keV.
- The titanium-containing compounds, $Al_2O_3.TiO_2$;mm and $Al_2O_3.TiO_2$;nm exhibited better shielding effectiveness and radiation-attenuation properties than aluminium oxide-containing compounds, which is a result of the higher Z number of Ti.
- This result may be attributable to Compton scattering, which is a dominant interaction

Table 3. Linear and mass attenuation coefficients, HVL, TVL values of the studied materials, comparison with the literature values

Samples	661.7 keV				1173.2 keV				1332.5 keV			
	μ (cm^{-1})	μ_m ($cm^2 g^{-1}$)	HVL (cm)	TVL (cm)	μ (cm^{-1})	μ_m ($cm^2 g^{-1}$)	HVL (cm)	TVL (cm)	μ (cm^{-1})	μ_m ($cm^2 g^{-1}$)	HVL (cm)	TVL (cm)
Al_2O_3;mm ([present work)	0.17355	0.07791	3.99	13.27	0.14341	0.06438	4.83	16.06	0.07292	0.03274	9.51	31.58
Al_2O_3;nm (present work)	0.15234	0.09123	4.55	15.11	0.09835	0.05892	7.05	23.412	0.08953	0.05364	7.74	25.72
$Al_2O_3.TiO_2$;mm (present work)	0.18513	0.08235	3.74	12.44	0.11596	0.05158	5.98	19.86	0.11411	0.05076	6.07	20.18
$Al_2O_3.TiO_2$;nm (present work)	0.19398	0.07113	3.57	11.87	0.11881	0.04356	5.83	19.38	0.1121	0.0411	6.18	20.54
[1]		0.074-0.0776 (BaO glass) 0.0863-0.0986 (Bi_2O_3 glass) 0.0861-0.0975 (PbO glass) 0.0975 (lead oxide silicate glass)				0.0604 (lead oxide silicate glass)				0.052 (lead oxide silicate glass)		
[2]		0.0724 (Brass) 0.076 (Bronz) 0.0736 (Steel) 0.0749 (Alıminium-silicon) 0.1091 (Lead-antimony)								0.0509 (Brass) 0.0512 (Bronz) 0.0524 (Steel) 0.0529 (Alıminium-silicon) 0.0555 (Lead-antimony)		
[3]		0.0806 (steel-steel composit metal foam (CMF)) 0.0779 (Al-steel CMF) 0.0733 (Al A356) 0.0707-0.0869 (Open-cell Al foam)				0.0525 (steel-steel CMF) 0.0561 (Al-steel CMF) 0.0549 (Al A356) 0.0547-0.0771 (Open-cell Al foam)				0.0524 (steel-steel CMF) 0.0481 (Al-steel CMF) 0.0489 (Al A356) 0.0514-0.0727 (Open-cell Al foam)		
[4]	0.1073 (h-BN) 0.1565 (h-BN-TiB_2)	0.07153 (h-BN) 0.07069 (h-BN-TiB_2)	6.460 (h-BN) 4.429 (h-BN-TiB_2)									
[5]	0.1618 0.1662 0.1700 0.1669 0.1747	0.06760 0.06880 0.07022 0.06711 0.07000										
[6]	0.233 0.244 0.240 0.230				0.189 0.185 0.181 0.173				0.158 0.157 0.140 0.159			
[7]	0.1919 0.1874 0.1839 0.1805	0.07274 0.07191 0.07175 0.07146										
[8]	0.539-0.609	0.06917-0.07584	1.138-1.286									
[9]	0.157 0.168 0.174 0.180	0.06359 0.06763 0.06977 0.07163	4.415 4.126 3.984 3.851									
[10]					0.1351 0.1371 0.1411	0.05604 0.05622 0.05613			0.1223 0.1232 0.1280	0.05064 0.05047 0.05095		

echanism in the medium energy range of 500–1000 keV.

- Decreasing the average particle size of $Al_2O_3.TiO_2$ from micrometre scale to nanometre scale led to better attenuation properties.
- These results shed light on the properties of nanoparticles, one of the foci of the current research scenario.
- Improvements in the sample preparation method can probably increase a sample's shielding ability.
- The good shielding properties of aluminium compounds with respect to gamma radiation, along with their other advantageous physical, chemical, and economical properties can be successfully exploited in several industries.
- On the other hand, $Al_2O_3.TiO_2$ compounds are economically more viable and can be used in nuclear applications.

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