Radiation Physics and Chemistry 79 (2010) 917–922

Contents lists available at ScienceDirect

Radiation Physics and Chemistry

journal homepage: www.elsevier.com/locate/radphyschem

A novel shielding material prepared from solid waste containing lead for gamma ray

Mehmet Erdem [a,*], Oktay Baykara [b], Mahmut Doğru [c,1], Fatih Kuluöztürk [c]

[a] Department of Environmental Engineering, Firat University, 23279 Elaziğ, Turkiye
[b] Division of Science Education, Firat University, 23119 Elaziğ, Turkiye
[c] Department of Physics, Faculty of Arts & Science, Bitlis Eren University, 13000 Bitlis, Turkiye.

ARTICLE INFO

Article history:
Received 23 February 2010
Accepted 14 April 2010

Keywords:
Shielding material
Gamma ray
Metallurgical waste
Lead

ABSTRACT

Human beings are continuously exposed to cosmogenic radiation and its products in the atmosphere from naturally occurring radioactive materials (NORM) within Earth, their bodies, houses and foods. Especially, for the radiation protection environments where high ionizing radiation levels appear should be shielded. Generally, different materials are used for the radiation shielding in different areas and for different situations. In this study, a novel shielding material produced by a metallurgical solid waste containing lead was analyzed as shielding material for gamma radiation. The photon total mass attenuation coefficients (μ/ρ) were measured and calculated using WinXCom computer code for the novel shielding material, concrete and lead. Theoretical and experimental values of total mass attenuation coefficient of the each studied sample were compared. Consequently, a new shielding material prepared from the solid waste containing lead could be preferred for buildings as shielding materials against gamma radiation.

1. Introduction

Radiation is a part of our daily life, so radiation and life are inseparable. Human beings are continuously exposed to background radiation from the sun and cosmic rays in the atmosphere, naturally occurring radioactive materials (NORM) within Earth, their bodies, houses and food. Exposure consists mainly of two distinct components, namely, internal and external exposure. Natural radiation significantly varies from place to place. It is not possible to be protected from sources of natural radiation which spreads throughout the environment. Nevertheless, when a source of radiation is concentrated and confined in a small area, the radiation dose people receive from that source can be limited by use of carefully planned structures and procedures.

There are some basic principles for radiation protection (e.g. shielding). Shielding is generally preferred due to its efficiency in intrinsically safe working conditions, whereas reliance on distance and time of exposure involves continuous administrative control over workers. The type and amount of shielding required depend on the type of radiation, the activity of radiation source and the dose rate that is acceptable for outside the shielding material. However, there are other factors for choice of shielding material such as their cost and weight. An effective shield will cause a large energy loss in a small penetration distance without emission of more hazardous radiation. Furthermore, the good shielding material should have high absorption cross-section for radiation and at the same time irradiation effects on its mechanical and optical properties should be small.

A number of experimental and theoretical works have been performed on radiation shielding, which has large different application areas with different materials (e.g. concrete, semiconductor, polymer, Lipowitz alloy, colemanite, etc.) (El-Sayed et al., 2003; Akkurt et al., 2007; Stewart et al., 2007; Osborn et al., 2006; Baltaş et al., 2005; Zeitlin, et al., 2006; Okuno, 2005; Tajiri et al., 2006).

Study of absorption of gamma and neutron radiations in shielding materials has been an important subject in the field of radiation physics. In order to design the protective shielding around the nuclear reactor, accelerators and high radiation region, the knowledge of the attenuation of high energy X-rays in shielding materials is essential. The methods for shielding in the applications of radiations are divided in two. First, shielding materials are furnished on the wall surface or directly in it (e.g. concrete); second, these materials are covered around the radiation source. In general, various materials have been used for the radiation shielding in different applications. For example, polyethylene, glasses, epoxy resin, colemanite, lead and concentrate have been used for neutron and gamma shield, and also,

* Corresponding author. Tel.: +90 424 2370000; fax: +90 424 2415526.
E-mail address: merdem@firat.edu.tr (M. Erdem).
[1] Permanent address: Department of Physics, Firat University, 23169 Elaziğ, Turkiye.

0969-806X/$ - see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.radphyschem.2010.04.009

lead, Lipowitz alloy and Cu–Ag polymer were used for electrons shield (Prasad et al., 1998). In order to win shielding properties to the concrete based composites, boron, lead or their compounds have been used (Demir and Keleş, 2006; El-Sayed et al., 2003). However, industrial wastes containing these elements or their compounds have not been evaluated for this purpose up to now.

Photon attenuation coefficient is an important parameter for characterizing the penetration and diffusion of X- and γ-rays in the multi-element materials. The scattering and absorption of gamma radiations are related to density and effective atomic numbers of material; knowledge of the mass attenuation coefficients is of prime importance. However, the linear (μ, cm^{-1}) or mass attenuation (μ/ρ ($cm^2\,g^{-1}$)) coefficient, which are defined as the probability of all possible interactions between γ-rays and atomic nuclei, has been described to investigate the radiation shielding properties of any shielding materials. These attenuation coefficients depend on the incident photon energy and the chemical composition of the absorbing materials' parameters such as their types, thickness and densities. The accurate values of mass attenuation coefficients (μ/ρ) of γ-rays in several materials are of great importance for industrial, biological, agricultural and medical studies. A number of related parameters can be derived from mass attenuation coefficient such as mass energy-absorption coefficient, the total interactions' cross-section, the molar extinction coefficient, the effective atomic number and the electron density (Singh et al., 2002).

An alternative or convenient method to experimental determination of mass attenuations coefficients is theoretical (Hubbell, 1982) or manual calculations using tabulated data that is generated using a computer program. For this purpose, a computer program was developed by Berger and Hubbell (Berger and Hubbell, 1999) (called XCOM) for calculating cross-sections and attenuation coefficients for any element, compound or mixture, at energies from 1 keV to 100 GeV. Afterwards, this program was updated and transformed to Windows platform and Windows version is being called WinXCom (Gerward et al., 2001, 2004).

The purpose of the present work is to investigate the γ-ray interactions with a novel shielding material prepared from a solid waste containing lead by measuring mass attenuations coefficient for different gamma energies (range from 88 to 1332.5 keV) by using different point radioactive sources (^{109}Cd, ^{60}Co, ^{57}Co, ^{133}Ba, ^{137}Cs and ^{54}Mn). Besides, for the comparison, mass attenuation coefficients of concrete and lead, which are commonly used for radiation shielding, have experimentally been obtained in the same experimental setup. We have obtained the mass attenuation coefficients of three distinct materials and compared these parameters to each other and to their theoretical calculations.

2. Theoretical calculations

In this section we summarize some theoretical relations that were used for determination of mass attenuation coefficient in the present work. The mono-energetic gamma rays or X-rays are attenuated in the target material according to Lamber–Beer law:

$$I = I_0 \exp(-\mu x) \tag{1}$$

where I and I_0 are the incident and transmitted intensities, respectively; x is the thickness of the absorbing material and μ is the linear attenuation coefficient. The basic data for mass attenuation coefficient (μ/ρ ($cm^2\,g^{-1}$)) were obtained by means of a proportional relationship with the total photon interaction cross-section per atom σ, the latter value being directly calculated for each element from theoretical models of interaction processes for photon with matter. The relationship between μ/ρ and σ is easily established. The macroscopic and microscopic cross-sections for γ- and X-rays are related by (El-Sayed et al., 2003; Wood,1982)

$$\mu = N_0 \sigma \tag{2}$$

and

$$N_0 = \rho N_A / A \tag{3}$$

Thus,

$$\frac{\mu}{\rho}(cm^2\,g^{-1}) = \frac{\sigma(b/atom)N_A}{A(atom/mol)(g/mol) \times 10^{-24}(cm^2/b)} \tag{4}$$

where N_0 is the number of atoms (or molecules) per unit volume of material, N_A is the Avagadro's constant, A is the atomic weight of the material and b shows the unit of barn (10^{-24} cm^2).

The mass attenuation coefficient, (μ/ρ), for a mixture or compound of known composition is given by

$$\frac{\mu}{\rho} = \sum_i w_i \left(\frac{\mu}{\rho}\right)_i \tag{5}$$

where ρ is the density of mass of the sample and w_i and $(\mu/\rho)_i$ are the weight fraction and the mass attenuation coefficient of the constituent element i, respectively. Total mass attenuation coefficients (μ/ρ) were calculated using the WinXCom computer program, which is the Windows version of XCOM (Gerward et al., 2001, 2004).

3. Materials and methods

3.1. Materials

In order to prepare shielding material, ordinary Portland cement (PC 42.5 type) and a solid waste containing lead were used in the study. Portland cement was obtained from Altinova Cement Factory in Elaziğ, Turkiye. Solid waste containing lead was supplied from Çinkur Zinc-Lead Metal Industry Inc. in Kayseri, Turkiye. The waste sample was dried at room temperature for 10 days prior to use, sieved by a 100 mesh standard sieve to obtain −100 mesh (150 μm) fractions and then stored in a tightly closed jar throughout the study. Its chemical and mineralogical compositions were determined by Philips PW-2404 electron X-ray fluorescence spectroscopy and Shimadzu XRD-6000 X-ray diffractometer.

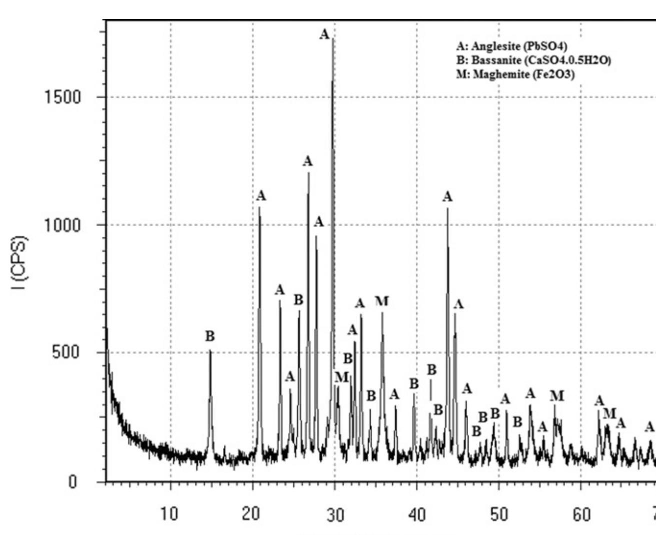

Fig. 1. XRD pattern of the solid waste.

Chemical and mineralogical compositions show that the waste sample contains 19.02% Pb, 12.25% S, 7.98% Zn, 6.19% Ca, 5.44% Fe and 6.74% Si as major elements. Varying amounts of other minor elements such as Al, Sr, Ti and Mn are also present in the material. X-ray diffraction pattern shows that the waste sample composed mainly of anglesite ($PbSO_4$), secondary of maghemite (Fe_2O_3) and bassanite ($CaSO_4 \cdot 0.5H_2O$) (Fig. 1).

3.2. Preparation of shielding material

Shielding material was prepared by mixing cement and the waste. For this, 630 g of cement and 1361 g of the waste sample were mixed homogenously. Mortar was prepared by addition of 378 ml of water (water/cement=0.60) into the mixture. The mortar was then poured into cylindrical polyethylene moulds (50 mm diameter and different heights in the range of 5–30 mm) and was compacted. The samples were cured for 28 days in a controlled moisture chamber (90% RH). The samples taken from moulds after 28 days were used to study their radiation permeability.

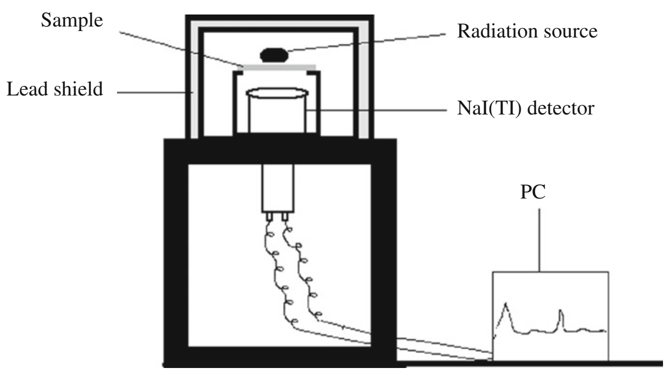

Fig. 2. Experimental setup.

3.3. Experimental procedures

Six different thicknesses (0.5, 1, 1.5, 2, 2.5 and 3 cm) of specimens of radiation shielding materials produced from the solid waste and ordinary Portland cement were prepared. The mass attenuation coefficients (μ/ρ) were determined by measuring the transmission of γ-rays through targets of those six different thicknesses individually. The experiments were performed using a low level gamma counting spectrometer including a 7.62 cm $\times$ 7.62 cm NaI(TI) detector by ORTEC Inc., connected to a multichannel pulse height analyzer. The necessary power for the detector as well as the acquisition of gamma spectra was achieved by an integrated spectroscopic system. This system is controlled by a personal computer. The control of acquisition parameters and analysis of the collected spectra are carried out using MAESTRO-32 (version 6.06) software package (ORTEC Inc.). The schematic view of experimental setup is displayed in Fig. 2. The detector is surrounded by a 5 cm thick lead shield to smooth the background γ-radiation. The detector has a resolution of about 7.6% at 662 keV of ^{137}Cs, which is capable of distinguishing the gamma ray energies for the experimental purpose. The experimental setup is shown in Fig. 2. The sample was placed between the standard gamma point source and the detector. The experiment was repeated with and without the sample for 5.000 s.

The samples were irradiated by photons emitted from ^{60}Co, ^{57}Co, ^{133}Ba, ^{109}Cd, ^{54}Mn and ^{137}Cs radioactive point sources in the range from 88 to 1332.5 keV. For each energy, the measurements were repeated three times for all the samples. This experimental

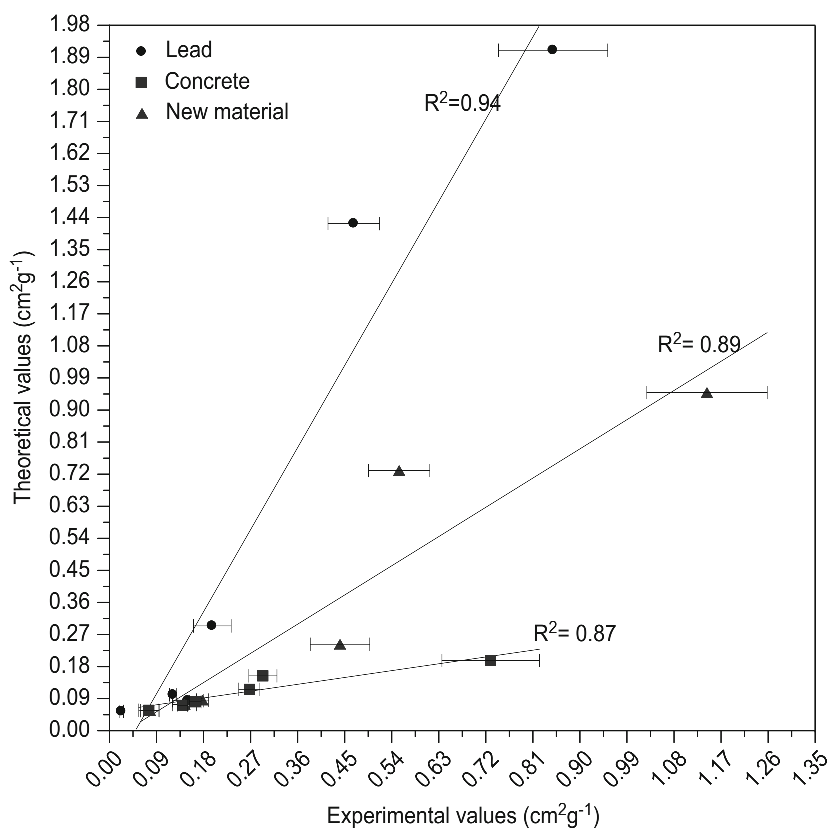

Fig. 3. The comparison of theoretical and experimental values of total mass attenuation coefficients.

procedure was checked out by measuring the mass attenuation coefficient of lead of thickness 0.5 cm (Fig. 3). It has been clearly seen that between theoretical and experimental values of the mass attenuation coefficients, values of lead have been observed in high correlation ($R^2=0.94$) in different gamma energies. The shape of this lead is similar to concrete and novel shielding materials. Nevertheless, theoretical calculation of mass attenuation coefficients of the lead, concrete and novel samples were carried out by using WinXCom software and then, the obtained values were compared.

Table 1
Chemical compositions of novel shielding material and concrete used as input parameters for WinXCOM.

Compound (%)	Novel shielding material	Element (%)	Concrete
Al_2O_3	3.502	Si	38.50
As_2O_3	0.010	Al	3.43
BaO	0.274	Fe	2.44
$CaSO_4$	21.060	Ca	9.72
CdO	0.027	Mg	0.09
Cr_2O_3	0.027	S	0.28
CuO	0.081		
Fe_2O_3	7.775		
K_2O	0.372		
MgO	0.669		
MnO	0.551		
Na_2O	0.812		
NiO	0.027		
P_2O_5	0.152		
$PbSO_4$	27.840		
SO_3	30.603		
Sb_2O_3	0.064		
SiO_2	14.427		
SrO	0.174		
TiO_2	0.185		
ZnO	9.930		

4. Results and discussion

The total mass attenuation coefficients (μ/ρ) for lead, concrete and the new material have been measured and calculated using WinXCom software at the energies from 10 keV to 10 MeV, which is in the border of the experimental studied energies (88 keV–1.3 MeV). Compound composition of novel shielding material and chemical analysis of concrete are shown in Table 1.

Oxygen and lead are higher constituent than others. At first, theoretical and experimental values of total mass attenuation coefficient of each studied sample were compared (Fig. 3), and the correlation theory is used to confirm the linearity of the theoretical and experimental values. The correlation coefficients for lead, concrete and the new material have been obtained to be 0.94, 0.87 and 0.89, respectively. It can be noted that the calculated and measurement values are in good agreement. The obtained and calculated values of the total mass attenuation coefficients are shown in Fig. 4 and 5, respectively. Both experimental and theoretical figures presented may be divided into two sections according to the applied photon energies. The first region (I) is from 0.03 MeV to about 0.6 MeV, and the second region (II) is from 0.63 MeV to about 1.15 MeV.

In region (I) (Fig. 4), the experimental total mass attenuation coefficients irregularly decrease with photon energy increase for new and concrete materials. Therefore, photoelectric effect and Compton scattering are dominant. Additionally, the discrepancy of the total mass attenuation coefficients of the three studied samples, especially new material and concrete, has been clearly seen in the low energy region and this may be attributed to the fact that the photoelectric effects are distinct due to different substance compositions of new material and concrete. But the total mass attenuation coefficients sharply decrease with the increase in photon energy for lead. This situation is the expected condition in the region (I).

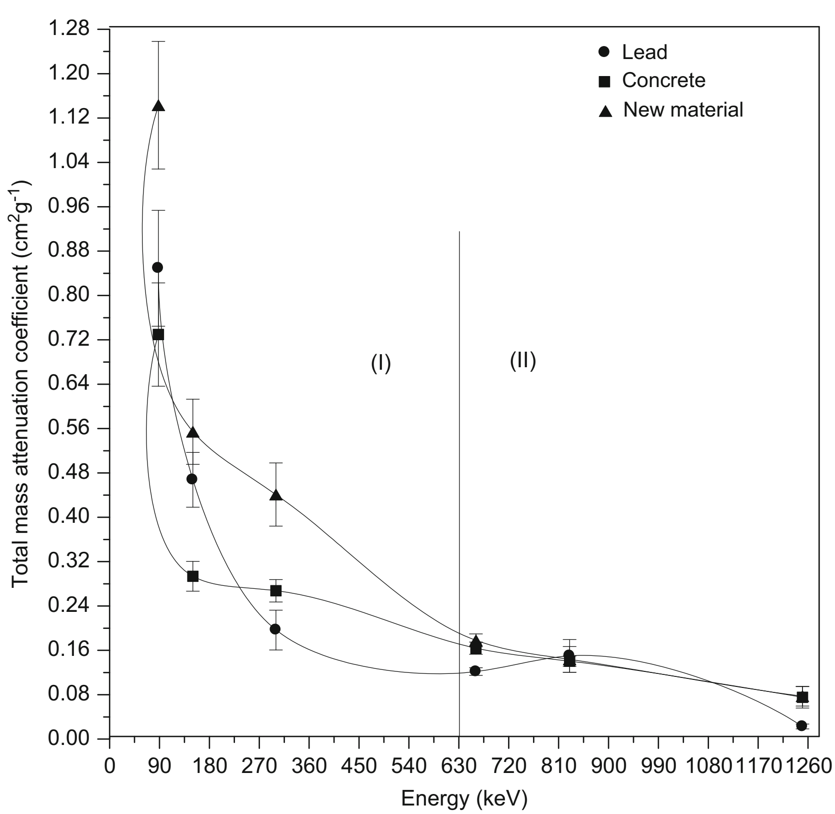

Fig. 4. The comparison of the experimental total mass attenuation coefficients for lead, concrete and the new material.

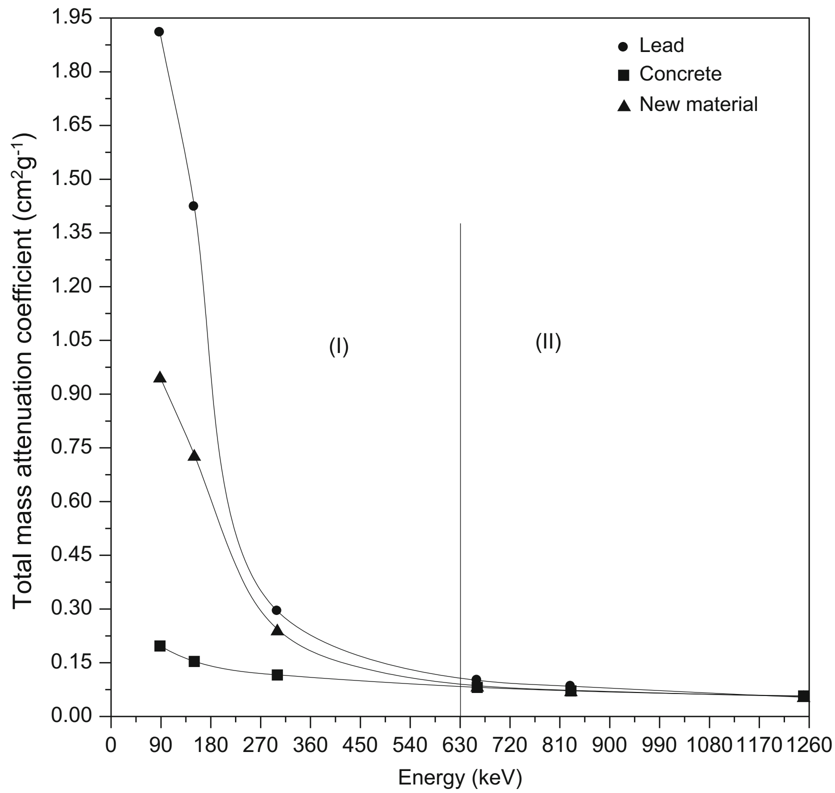

Fig. 5. The comparison of the theoretical total mass attenuation coefficients for lead, concrete and the new material.

In region (II), while the total mass attenuation coefficient sharply decrease in new and concrete materials, it reverts slightly to increase and then, slightly decreases again for lead with the increase in photon energy. The Compton scattering is a dominant reaction and its cross-section may be considered as the main interaction. Nevertheless, total mass attenuation coefficients are almost the same for the three samples in high energy. Generally, the obtained μ/ρ values of new material are higher than lead and concrete in spite of very low density.

In region (I) (Fig. 5), the calculated total mass attenuation coefficients sharply decrease for all the samples. These sharp decreases have continued through the region (I) and afterwards they slightly decrease. Therefore, photoelectric effect and Compton scattering are dominant in this region. Generally, total mass attenuation coefficients of lead are relatively higher in the low energy region. Therefore, it has been seen that there is a good consistency between the calculated values of total mass attenuation coefficients. As should be expected, density is the main parameter which affects the mass attenuation coefficient for the photon energy. However, in present work, it has been clearly seen that variations in the chemical composition of the materials are more significant. Despite the new material has low density ($2.19\,g\,cm^{-3}$), the total mass attenuation coefficient that is approximately equal to that of lead has been measured and calculated in high energy region in particular.

Total mass attenuation coefficients of the some shielding materials prepared from colemanite (Okuno, 2005), colemanite (15%) and clinker (85%), borogysum (10%) and clinker (90%) (Demir and Keleş, 2006), limestone (Awadallah and Imran., 2007) and the material called as FPPb, which contains 0.3237% C, 0.1288% O, 0.0435% H, 0.0008% Zn, 0.3537 Pb and 0.1491% B, have been reported to be 0.06551, 0.06093, 0.059 and 0.0546 $cm^2\,g^{-1}$, respectively (El-Sayed et al., 2003). When the total mass attenuation coefficient value of 0.89 $cm^2\,g^{-1}$ of the new shielding material prepared from the zinc extraction residue containing lead is taken into account, it can be stated that the new material possesses high shielding capacity.

5. Conclusion

A new shielding material was developed using a metallurgical solid waste containing lead in this study. The mass attenuation coefficients (μ/ρ) of the material in the high region of photon energy show that the novel shielding material prepared from a metallurgical solid waste containing lead would be preferred as shielding material for buildings against gamma radiation.

Acknowledgements

This work was supported by TUBITAK with project number of 107Y139. We would like to thank L. Gerward for providing the WinXCOM program.

References

Akkurt, I., Altindag, R., Onargan, T., Basyigit, C., Kılıncarslan, K., Kun, M., Mavi, B., Guney, A., 2007. The properties of various igneous rocks for γ-ray shielding. Constr. Build. Mater. 21, 2078–2082.

Awadallah, M.I., Imran, M.M.A., 2007. Experimental investigation of g-ray attenuation in Jordanian building materials using HPGe-spectrometer. J. Environ. Radioac. 94, 129–136.

Baltaş, H., Çevik, U., Tıraşoğlu, E., Ertuğral, B., Apaydın, G., Kobya, Aİ, 2005. Mass attenuation coefficients of YBaCuO and BiPbSrCaCuO superconductors at 511, 661 and 1274 keV energies. Radiat. Meas. 39, 33–37.

Berger, M.J., Hubbell, J.H., 1999. XCOM: Photon cross sections database, Web Version, 1.2. 〈http://physics.nist.gov./xcom〉. National Institute of Standards and Technology, Gaithersburg, MD 20899, USA. (Originally published as NBSIR 87-3597 "XCOM: Photon Cross Sections on a Personal Computer".)

Demir, D., Keleş, G., 2006. Radiation transmission of concrete including boron waste for 59.54 and 80.99 keV gamma rays. Nucl. Instrum. Meth. B 245, 501–504.

El-Sayed, A., Ali, M.A.M., Ismail, M.R., 2003. Natural fibre high-density polyethylene and lead oxide composites for radiation shielding. Radiat. Phys. Chem. 66, 185–195.
Gerward, L., Guilbert, N., Jensen, K.B., Levring, H., 2001. X-ray absorption in matter. Reengineering XCOM. Radiat. Phys. Chem. 60, 23–24.
Gerward, L., Guilbert, N., Jensen, K.B., Levring, H., 2004. WinXCom—a program for calculating X-ray attenuation coefficients. Radiat. Phys. Chem. 71, 653–654.
Hubbell, J.H., 1982. Photon mass attenuation and energy absorption coefficients from 1 keV to 20 MeV. Int. J. Appl. Radiat. Isot. 33, 1269–1290.
Okuno, K., 2005. Neutron shielding material based on colemanite and epoxy resin. Radiat. Prot. Dosim. 115, 259–261.
Osborn, J.C., Ersez, T., Braoudakis, G., 2006. Radiation shielding design for neutron diffractometers assisted by Monte Carlo methods. Physica B 385–386, 1321–1323.
Prasad, S.G., Parthasaradhi, K., Bloomer, W.D., Al-Najjar, W.H., 1998. Aluminum, copper, tin and lead as shielding materials in the treatment of cancer with high-energy electrons. Radiat. Phys. Chem. 53, 361–366.
Singh, K., Singh, H., Sharma, V., Nathuram, R., Khanna, A., Kumar, R., Bhatti, S.S., Sahota, H.S., 2002. Gamma-ray attenuation coefficients in bismuth borate glasses. Nucl. Instrum. Meth. B 194, 1–6.
Stewart, D.Y., Harrison, P.F., Morgan, B., Ramachers, Y., 2007. Radiation shielding for underground low-background experiments. Nucl. Instrum. Meth. A 571, 651–662.
Tajiri, M., Tokiya, Y., Uenishi, J., Sunoka, M., Watanabe, K., 2006. New shielding materials for clinical electron beams. Med. Phys. 80, 391–393.
Wood, J., 1982. Computational Methods in Reactor Shielding. Pergamon Press, New York, USA.
Zeitlin, C., Guetersloh, S.B., Heilbronn, L.H., Miller, J., 2006. Measurements of materials shielding properties with 1 GeV/nuc ^{56}Fe. Nucl. Instrum. Meth. B 252, 308–318.