Radiation and Environmental Biophysics (2020) 59:145–150
https://doi.org/10.1007/s00411-019-00824-y

ORIGINAL ARTICLE

Gamma-ray attenuation properties of some NLO materials: potential use in dosimetry

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Received: 23 August 2019 / Accepted: 10 November 2019 / Published online: 28 November 2019
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Abstract

Mass attenuation coefficients (μ_m) for some nonlinear optical materials such as potassium dihydrogen phosphate, ammonium dihydrogen phosphate, zinc tris-thiourea sulphate, and zinc thiourea chloride were measured using a 2×2 NaI(Tl) scintillation detector at gamma energies of 122 keV, 356 keV, 511 keV, 662 keV, 840 keV, 1170 keV, 1270 keV, and 1330 keV. In addition, GEANT4 simulations were carried out to mimic the experiment at these energies. As a result, good agreement between the experimental and GEANT4 results was observed. The measured μ_m values were used to compute effective atomic numbers (Z_{eff}) for the selected materials. It was found that the Z_{eff} values were in the range typical for dosimetric materials.

Keywords Attenuation coefficient · Z_{eff} · GEANT4 · NLO material

Introduction

Because ionsing radiation is widely used whether this is in a hospital, laboratory, nuclear power plant, or nuclear waste treatment location, it is very important to measure and quantify how much radiation is produced by any radioactive sources and how much radiation is absorbed by materials. This requires methods that allow determination of relevant radiation interaction parameters. For example, absorbed dose in matter can be obtained by solving radiation transmission equations. For gamma ray radiation, narrow beam geometry can be employed including the Beer–Lambert law (Swinehart 1962). In fact, narrow beam geometry is considered an essential setup to study the gamma ray interaction with matter by determining linear attenuation coefficients or mass attenuation coefficients. The latter coefficients are used to describe the probability of photon–matter interactions per unit mass and unit area (McNair 1981; Chantler 2000). The μ_m values are essential to derive atomic cross sections, electronic cross sections, and effective atomic numbers for a compound or mixture. The notion of effective atomic number was introduced first by Hine who noticed that the effective atomic number of a mixture or compound is not constant but variable with photon energy (Hine 1951). Nowadays, it is well known that μ_m and Z_{eff} are the most important parameters for selection of suitable materials for radiation dosimetry and radiation detection.

Many attempts have been made to identify materials to be used as radiation dosimeters. Reddy studied the radiation response of several liquid and solid materials (Kumar and Reddy 1997), using elemental data from Hubbell and Seltzer (1995), Chantler (1995). Shivaramu calculated the effective atomic numbers of some thermoluminescent dosimetric (TLD) materials such as LiF, $CaCO_3$, and $CaSO_4$ (Ramprasath et al. 2000). He concluded that effective atomic numbers vary linearly with photon energy. The mass attenuation coefficients of some TLD materials were also experimentally determined at different photon energies in a narrow beam geometry using a hyper pure germanium detector (Gowda et al. 2004). The radiological features of some gel materials were compared against water with respect to their Z_{eff} values (Taylor et al. 2008). In addition, gamma-ray buildup factors for TLD materials were reported and compared with ICRU standard dosimeters (Manohara et al. 2010; Kucuk et al. 2013; Singh and Badiger 2016). Recently, μ_m values for

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some polymers were determined using Monte Carlo simulation at different photon energies (Singh et al. 2015). In addition, there are numerous similar studies dealing with various materials using different methods including measurements, theoretical considerations, or simulations (Sidhu et al. 2012; Özdemir and Kurudirek 2009; Kurudirek 2014; Bootjomchai and Laopaiboon 2014; Singh et al. 2014).

Nonlinear optical NLO materials were found to be useful for many applications: for smart filtering (Dini et al. 2016), for photonics (Lin et al. 2014), and for biomedicine and radiation detection (Liu et al. 2017). NLO materials might possibly be used in biological dosimetry because of the following properties: their strength, lightness, low cost, and ease of fabrication. Very recently, Awasarmol studied some organic NLO materials in terms of μ_m and Z_{eff} (Awasarmol et al. 2017). He found that the NLO materials can support radiation measurement due to their low Z_{eff}. At present, fundamental gamma interaction parameters for selected NLO materials have not yet been published.

This prompted the present study in which dosimetric parameters of various NLO materials such as such as potassium dihydrogen phosphate, ammonium dihydrogen phosphate, zinc tris-thiourea sulphate, and zinc thiourea chloride were investigated. Both measurements and Monte Carlo simulations were performed in narrow beam geometry in an effort to determine mass attenuation coefficients. These mass attenuation coefficients were then utilized to compute the effective atomic numbers and effective electron densities of the selected materials. The present investigation is considered useful for selection of NLO materials to be used for biological dosimetry and other applications.

Experimental and computational methods

Experimental detials

Narrow beam transmission geometry was arranged, as shown in Fig. 1, to measure μ_m for the NLO materials shown in Table 1. Six radioactive sources were used, namely, ^{22}Na, ^{54}Mn, ^{57}Co, ^{60}Co, ^{133}Ba, and ^{137}Cs. Gamma ray spectrometry was performed by means of a scintillation

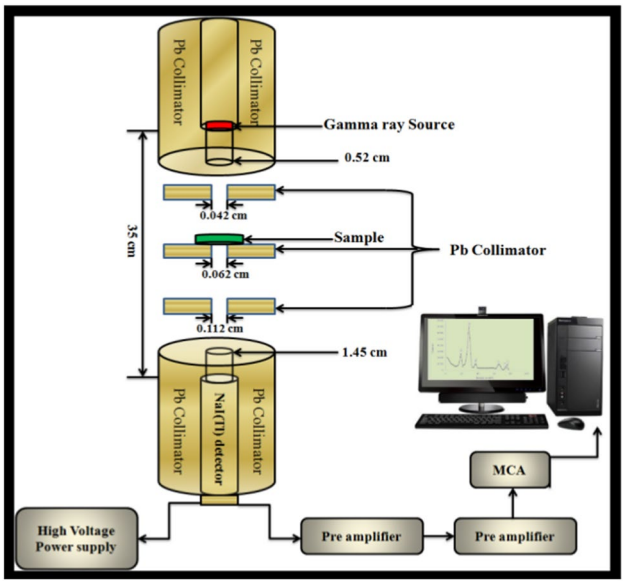

Fig. 1 Experimental setup for narrow beam geometry

detector (2 × 2 inch) to measure μ_m. The system showed an energy resolution of 8.2% at 662 keV and included a 16K multichannel analyzer (MCA). All samples were shaped as pellets with different thicknesses of 0.5–1.5 cm, and then irradiated by gamma ray energies of 122, 356, 511, 662, 840, 1170, 1275, and 1330 keV. The radioactive sources were within a lead cylinder with a 0.52 cm orifice to collimate and produce a pencil beam. The detector was also housed in a lead shield to minimize any background radiation. The spectrometer was calibrated using the 356.1 keV line emitted by ^{133}Ba, the 122 keV line emitted by ^{57}Co, the 511 and 1275 keV lines emitted by ^{22}Na, the 661.9 keV line emitted by ^{137}Cs, the 840 keV line emitted by ^{54}Mn, and the 1173.2 and 1332.5 keV lines emitted by ^{60}Co. The laboratory temperature was kept constant at 25°C (77°F). The distance between source and detector was kept 35 cm and the sample was placed in-between. Archimedes principle was utilized to obtain the density of the studied materials, where measurements were performed at room temperature with ethylene as the reference immersion liquid.

Table 1 Chemical composition of the investigated NLO materials

Sample	Sample code	Sample compositions (mol%)
Potassium dihydrogen phosphate	KDP	H (0.0148), O (0.4703), P (0.2276), K(0.2873)
Ammonium dihydrogen phosphate	ADP	H (0.0526), N (0.1218), O (0.5564), P (0.2693)
Zinc tris-thiourea sulphate	ZTS	H (0.0170), C (0.0505), N (0.1179), O(0.2694), S (0.2699), Zn (0.2752)
Zinc thiourea chloride	ZTC	H (0.0190), C (0.0565), N (0.1319), S (0.1510), Cl (0.3338), Zn (0.3078)

GEANT4 simulation

The GEANT4 code can be applied for simulation of a narrow beam geometry with various photon energies (Allison et al. 2016). Determination of mass attenuation coefficients for any compound or mixture requires knowledge on the chemical or elemental composition and density of the material. The GEANT4 simulation developed for the present study included a mono-energetic photon beam imposing on a sample, as shown in Fig. 2. The mass attenuation coefficients for the material under investigation were determined using the transmission method following Lambert's Beer law ($I = I_0 e^{-\mu_m x}$), where I_0 and I are the incident and attenuated photon intensity, respectively, μ_m (in $cm^2 g^{-1}$) is the mass attenuation coefficient and x is the thickness of the studied material in g/cm^2.

For the simulation of mass attenuation coefficients, an input file has been prepared including beam properties, irradiation parameters, sample compositions, and physical settings. A beam of 10^6 photons was simulated to attenuate in the NLO samples. The code was run to obtain the transmission values, I/I_0, for each of the investigated NLO samples.

Effective atomic number

Effective atomic numbers refer to the attenuation of photons, which occurs due to photon interactions with matter, is is given by the following ratio (Singh et al. 2014; Manohara et al. 2009):

$$Z_{eff} = \frac{\sigma_a}{\sigma_e}, \tag{1}$$

where σ_a is the total atomic cross section and σ_e is the total electronic cross section. The total atomic cross section can be obtained from the mass attenuation coefficient as follows:

$$\sigma_a = \frac{\mu_m}{N_A \sum_i w_i/A_i}, \tag{2}$$

where w_i is the weight fraction of the ith element in the sample. The total electronic cross section is given by the following equation:

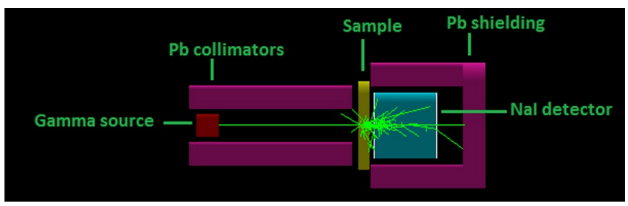

Fig. 2 GEANT4 setup for narrow beam geometry

$$\sigma_e = \frac{1}{N_A} \sum_i \frac{f_i A_i}{Z_i} (\mu_m)_i, \tag{3}$$

where N_A is Avogadro's number, f_i is the fractional abundance of the each constituent element given that $\sum_i f_i = 1$, Z_i is the atomic number, and A_i is the atomic weight.

Results and discussion

Mass attenuation coefficients for the selected NLO materials are shown in Fig. 3, while corresponding effective atomic numbers are shown in Fig. 4 . Effective atomic numbers as a function of effective electron density are shown in Fig. 5, and measured and simulated mass attenuation coefficients are compared in Table 2. In Table 3, effective atomic numbers of the investigated NLO materials, TLD materials, polymers, and various human tissues are compared.

Mass attenuation coefficients

Figure 3 shows the mass attenuation coefficients (μ_m) versus photon energy for the investigated NLO materials as obtained by measurement and simulation, with photon energies ranging from 1 to 1400 keV. It is to be noted that the μ_m values for NLO materials decrease with increasing photon energy down to about 0.06 cm^2/g at a photon energy of 1400 keV. It was found that the μ_m values are lowest for ADP and highest for ZTC, in the photon energy region of interest. The variation of the mass attenuation coefficients with photon energy can be explained using the photoelectric effect, Compton scattering and pair production (Chantler 2000; Singh et al. 2014; Tonguc et al. 2018; Özdemir and Kurudirek 2009).

The observed behaviour of the μ_m values with photon energy can be divided into three energy regions, i.e., low-energy ($E \leq 100$ keV), intermediate-energy ($0.1 \leq E < 1022$ keV), and high-energy ($E \geq 1022$ keV). It is observed that in the low-energy region, the μ_m values sharply decrease with increasing photon energy for all the selected NLO materials. In this low-energy region, the predominant interaction is described by the photoelectric effect, where photon interaction depends upon atomic number and photon energy. In the intermediate-energy region, the μ_m values only slightly decrease with increasing photon energy because of Compton scattering. In fact, Compton scattering is found to be roughly independent upon photon energy. In contrast, in the high-energy region, the μ_m values slightly increase and become constant due to pair-production being the dominant process. The highest μ_m values among the selected NLO materials were found for ZTC, which obviously provides superior shielding properties for the investigated photon energies.

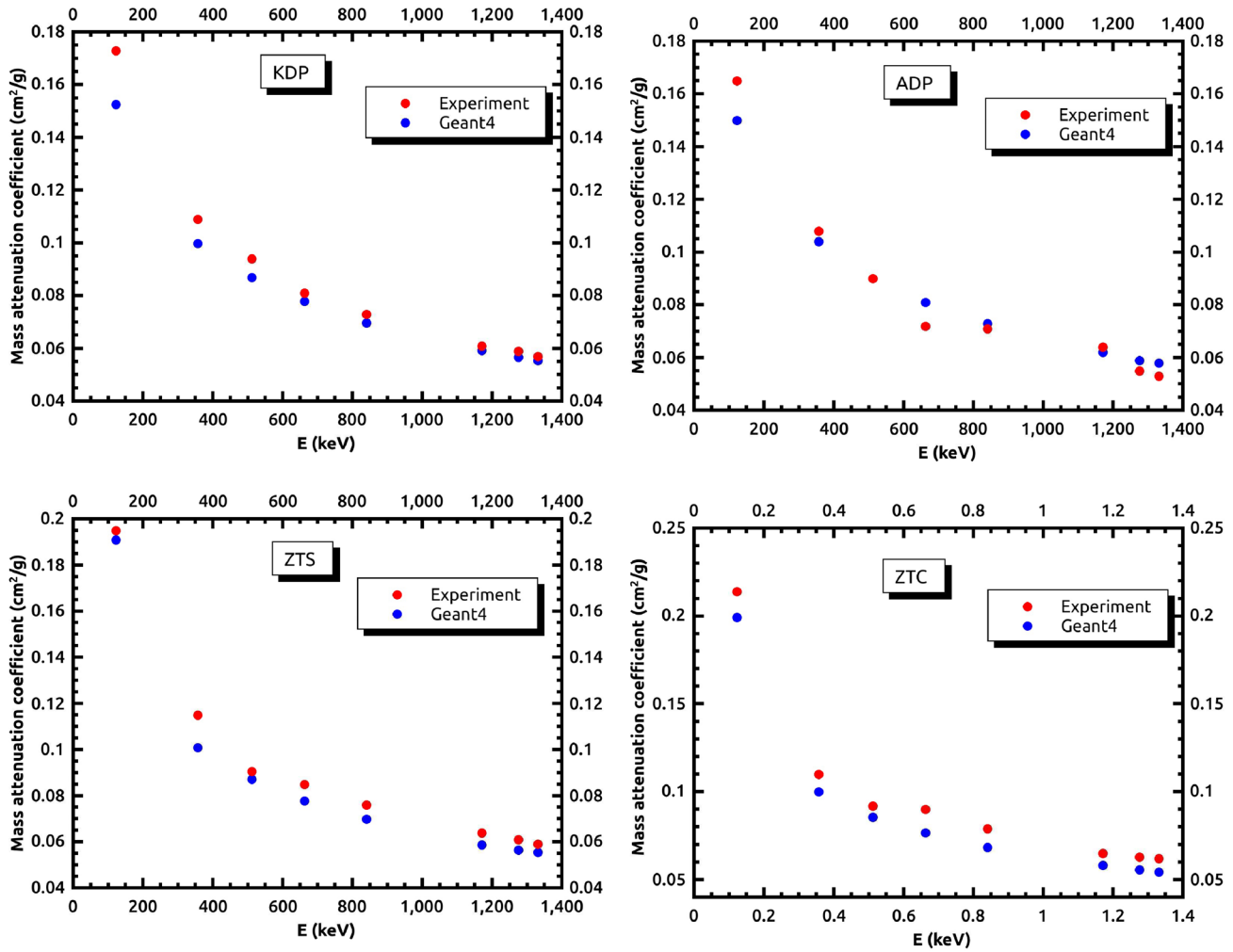

Fig. 3 Experimental and simulated values of mass attenuation coefficients as a function of photon enrgy for the investigated NOL materials

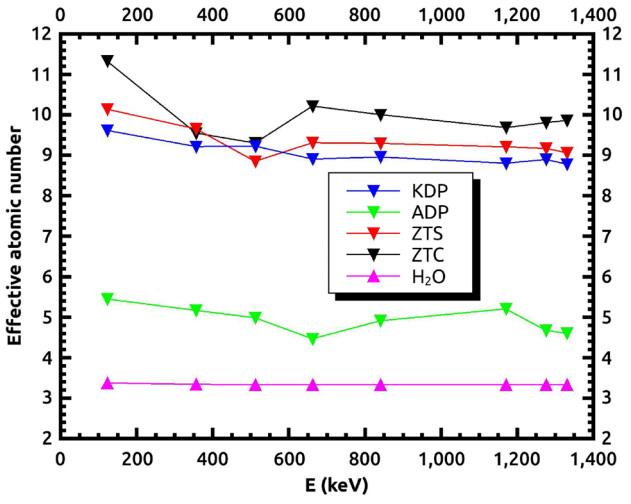

Fig. 4 Comparison of effective atomic number (Z_{eff}) values of NLO materials with those of water

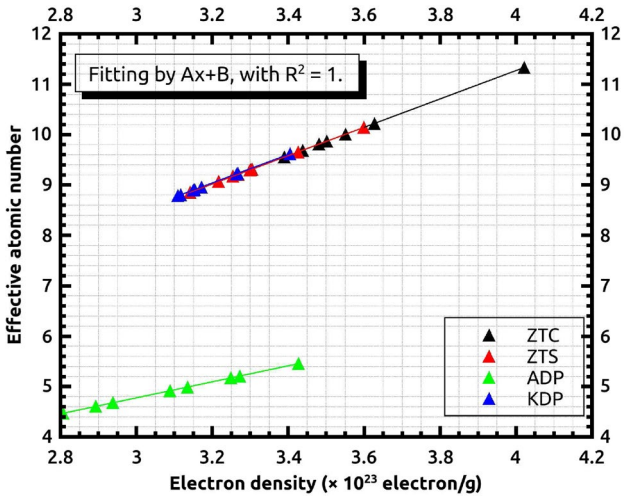

Fig. 5 Variation of effective atomic number (Z_{eff}) with effective electron density (N_{eff})

Table 2 Measured (Exp.) and simulated (GEANT4) mass attenuation coefficient (μ_m) values, for the investigated NLO materials

E (keV)	KDP			ADP			ZTS			ZTC		
	Exp.	GEANT4	%Dev.[a]	Exp.	GEANT4	%Dev.[a]	Exp.	GEANT4	%Dev.[a]	Exp.	GEANT4	%Dev.[a]
122	0.173	0.153	11.838	0.150	0.157	9.152	0.195	0.191	2.062	0.199	0.218	6.874
356	0.109	0.100	8.475	0.104	0.105	3.704	0.115	0.101	12.304	0.100	0.102	9.082
511	0.094	0.087	7.503	0.090	0.090	0.436	0.091	0.087	3.840	0.086	0.086	6.899
662	0.081	0.078	3.747	0.081	0.081	12.489	0.085	0.078	8.495	0.077	0.077	14.899
840	0.073	0.070	4.579	0.073	0.072	2.349	0.076	0.070	8.113	0.068	0.068	13.304
1170	0.061	0.059	2.977	0.062	0.061	3.533	0.064	0.059	8.114	0.058	0.058	10.558
1275	0.059	0.057	4.015	0.059	0.059	7.393	0.061	0.056	7.454	0.056	0.055	11.686
1330	0.057	0.055	2.796	0.058	0.058	9.021	0.059	0.056	5.912	0.054	0.054	12.185

[a]Deviation between experiment and simulation

Table 3 Effective atomic number (Z_{eff}) values of the investigated NLO materials as compared to those of TLD materials, polymers, and human tissues

E (keV)	NLO materials				TLD materials[a]			Polymers[b]		Human tissues[c]	
	KDP	ADP	ZTS	ZTC	LiF	$CaCO_3$	$CaSO_4$	ER	MC	Bone	Muscle
122	9.614	5.453	10.140	11.330	6.054	9.825	10.731	5.417	7.975	5.996	3.435
356	9.216	5.170	9.652	9.549	6.006	10.005	10.711	5.327	7.945	6.004	3.434
511	9.225	4.989	8.850	9.302	5.987	10.051	11.321	5.323	7.943	5.901	3.437
662	8.906	4.468	9.310	10.217	5.988	10.052	11.275	5.321	7.943	5.994	3.435
840	8.957	4.914	9.295	10.004	6.001	10.041	11.285	5.320	7.943	6.024	3.446
1170	8.802	5.208	9.207	9.684	5.924	10.025	11.344	5.320	7.942	6.104	3.478
1275	8.894	4.676	9.169	9.807	6.002	10.037	11.348	5.321	7.942	6.199	3.517
1330	8.781	4.605	9.063	9.863	5.966	10.042	11.351	5.319	7.942	6.297	3.559

[a] Z_{eff} of TLD matrials from Gowda et al. (2004)

[b] Z_{eff} of polymers from Singh et al. (2014)

[c] Z_{eff} of human tissues from Shivaramu (2002)

Table 2 compares measured and simulated mass attenuation coefficients. The table shows that both the measured and simulated coefficients are very close to each other within < 15% of standard deviation. Therefore, it is concluded that GEANT4 simulations allow for an investigation of photon interaction processes in compound or complex materials. It is also concluded that GEANT4 simulations can replace experimental investigations of photon interaction with materials.

Effective atomic number and electron density

Figures 4 and 5 show effective atomic numbers (Z_{eff}) as a function of photon energy and effective electron density (N_{eff}), respectively. It is observed that the Z_{eff} values for ZTC are the highest, while those for ADP are the lowest. The observed dependence of Z_{eff} on photon energy is due to the dependence of Z_{eff} on the atomic number of constituent elements and their compositions (see Eqs. 2 and 3). As compared to ADP, ZCT contains higher atomic number elements with higher chemical abundance, and therefore, Z_{eff} values are also found to be higher. It is to be noted that the relation between Z_{eff} and N_{eff} is linear and Z_{eff} values increase with increasing N_{eff}. Figure 4 shows a comparison of Z_{eff} values for the investigated NLO materials with those of water. It is found that the Z_{eff} values of ADP are comparable those of water. Therefore, in terms of dosimetry ADP can be considered as tissue equivalent.

In Table 3, Z_{eff} values of the invvestigated NLO materials are compared with those of TLD materials, polymers, and human tissues. It is observed that the Z_{eff} values of TLD materials, polymers, and human tissues are found comparable to those of the investiged NLO materials. Therefore, these NLO materials might be suitable for biological dosimetery and other radiation applications.

Conclusion

Mass attenuation coefficients for some NLO materials (potassium dihydrogen phosphate, ammonium dihydrogen phosphate, zinc tris-thiourea sulphate, and zinc thiourea chloride) were measured by using a NaI(Tl) detector for selected photon energies. GEANT 4 simulations were

carried out to investigate the same parameters. As a result, good agreement between the experimental and simulated results was observed. Effective atomic numbers of the NLO materials were compared with those of TLD materials, polymers and human tissues, and found to be comparable . This suggests that the investigated NLO materials could be used for dosimetric applications. In addition, it is concluded that GEANT4 simulations offer an alternative for experimental investigations of photon interaction with matter.

Acknowledgements The corresponding author would like to express his deep appreciation to Prof. Werner Ruhm, Institute of Radiation Protection, Helmholtz Zentrum München, Neuherberg, Germany for his an excellent corrections and suggestions to improve the present work.

Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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