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Measurement of mass attenuation coefficient, effective atomic number and electron density of thermoluminescent dosimetric compounds

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Abstract

Photon mass attenuation coefficients of some thermo luminescent (TLD) compounds, such as $CaCO_3, BaCO_3 CaSO_4, ZnSO_4, and MnSO_4$ were determined at 59.5-1332kev in a Gamma ray transmission set up using a NaI (TI) detector biased to a high voltage. The attenuation coefficient data were then used to compute the effective atomic number and the electron density of TLD compounds in photon energy range 51.5-1332.5 keV emitted by radioactive sources ^{241}Am, ^{133}Ba, ^{57}Co ^{137}Cs, ^{54}Mn, and ^{60}Co. The values of effective atomic numbers and electron densities of TLD compounds were in agreement with the other available data at energies of interest.

Keywords: Gamma-ray transmission, Single channel analyzer, attenuation coefficient, TLD compounds, photoelectric interaction, Compton scattering and Pair production

1. Introduction

Thermo luminescent Dosimeter (TLD) is a type of radiation dosimeter that is used for both environmental and personnel monitoring in facilities involving radiation exposure.

A TLD measures exposure to ionizing radiation by measuring the amount of visible light emitted from a crystal in the detector when the crystal is heated.

One property of thermo-luminescent dosimeter that determines its interaction with photons is the atomic number Z provided the thermo-luminescent material inside the dosimeter is a pure element.

When such a dosimeter is made up of compounds or mixtures, then the dependence of photon interaction gives rise to the notion of effective atomic number Z_{eff} because Z_{eff} is considered as being representative of the attenuating properties of the heterogeneous medium.

Effective atomic number and electron density also provide a convenient means of interpreting X-ray attenuation data generated by a complex medium like biological tissue, which provides input data for the calculation of dose in radiography and radiation dosimetry etc., (Shivaramu*et al.*, 2001) [7].

In several applications relating to determination of Z_{eff} and δe in composite materials, measurements are not directly carried out, but are normally determined from the value of photon attenuation coefficients of such materials.

Thus accurate experimental measurement of photon attenuation coefficient is central to determination of Z_{eff} and δe of composite substances used in thermo luminescent dosimeters.

Such experimental measurement should take cognizance of the fact that photoelectric effect, Compton scattering and pair production processes are the dominant interaction processes between the photons and atoms over a wide range of energies.

Gamma transmission measurements has been used for studying penetration of gamma rays in soil in order to evaluate different properties of soil and soil-water diffusion processes (Baytas and Akbal, 2002; Vaz*et al.*, 1999; Oliveira *et al.*, 1997) [1].

Gamma ray transmission measurements has been used in solid phantoms to investigate the radiological equivalency of these phantoms and their water using transmission curves and compared with Monte Carlo calculations and standard published data (Hill *et al.*; 2008)[5].Apart from its application in determination of Z_{eff}, data on attenuation coefficient of different multi-element materials are also applied in development of shield against harmful radiations and in various fields of nuclear science, technology and medicine.

In this work, accurate measured values of total attenuation coefficient, effective atomic numbers and electron densities of TLD compounds at energies: 51.5-1332.5keV was obtained by experiment.

These attenuation coefficient values were used to determine the effective atomic number and electron density of TLD compounds.

Owing to the growing use of TLD for personnel and environmental monitoring, there is need for local production of TLD badges. This work is aimed at providing appropriate experimental data for the choice and specification of TLD compound for use in subsequent design and production of TLD badges for personnel radiation monitoring.

The purpose of this research work is to set up γ-transmission arrangement with the view:

To carry out accurate measurement of mass attenuation coefficient of selected thermo-luminescent dosimetric. Compounds, to determine the effective atomic number and electron density of these compounds and to evaluate the efficiency these TLD compounds for radiation exposure monitoring in terms of the photon energies.

The present study covers experimental determination of mass attenuation coefficient of TLD compounds including $CaCO_3, BaCO_3, ZnSO_4.7H_2O$ and $MnSO_4$ at energies 51.5, 136.5, 661.6, 854, 1173.2 and 1332.5keV, belonging to various radioisotope sources LISTED IN Table 1.1. Z_{eff} and electron density were determined from the measured mass attenuation coefficients of these compounds using theoretical formula relating these two quantities to mass attenuation coefficient.

Table 1.1: Standard Gamma Sources Used For Gamma Transmission Measurement

Parent radio-nuclide	Dateof Production	Activity (kBq)	Half-Life $T_{1/2}$(Years)	Gamma ray Energy (KeV)
^{241}Am	1st July 2004	36.50	432.2	59.5
^{57} Co	August 2008	36.50	0.745	122.1
^{137} Cs	July 2008	36.50	30.00	661.6
^{54}Mn	Sept. 2008	36.50	312.5	834.8
^{60} Co	August 2008	36.50	5.263	1173.6
^{60} Co	August 2008	36.50	5.263	1332.5

Theoretical Framework

Thermo-luminescent dosimetric materials are usually composed of various elements. It is therefore assumed that the contribution of each element to the total photon interaction is additive, yielding the well-known "mixture rule" (Umesh*et al.*, 1992) [8] that presents the total mass attenuation coefficient (μ/ρ) of any compound as the sum of the appropriately weighted proportions of the individual atoms. Thus,

$$\left(\frac{\mu}{\rho}\right)_c = \sum_i W_i \left(\frac{\mu}{\rho}\right)_i \qquad (1)$$

Where $(\mu/\rho)_c$ is photon mass attenuation coefficient for the compound, $(\mu/\rho)_i$ is the photon mass attenuation coefficient for the individual elements of the compound. For any compound, a quantity called the effective atomic cross-section σ_a can be calculated by averaging over atoms of all the elements in the compound. Thus, we have,

$$\sigma_a = \frac{\left(\mu/\rho\right)_C}{N_A \sum_i \frac{W_i}{A_i}} \qquad (2)$$

Where; N_A is the Avogadro's number, A_i is the atomic weight of the constituent element and W_i is the fractional weights of the elements in the compound. Similarly, the average electronic cross-section, σ_{el}, is given by;

$$\sigma_{el} = \frac{1}{N_A} \sum_i f_i \frac{A_i}{Z_i} \left(\frac{\mu}{\rho}\right)_i \qquad (3)$$

Where:

$$f_i = \left(n_i / \sum_j n_j\right) \qquad (4)$$

And f_i is the fractional abundance such that n_i is total number of atoms of the constituent element and $\sum_j n_j$ are the total number of all types present in the compound as per its chemical formula. The effective atomic number, Z_{eff}, can now be written as:

$$Z_{eff} = \frac{\sigma_a}{\sigma_{el}} \qquad (5)$$

Other expressions for the effective atomic numbers are found in [El-katel*et al.*, 1991 and Jackson *et al.*, 1981] [3]. The effective electron number or density, N_{el} (number of electrons per unit mass) can be found from;

$$N_{el} = \left(\mu/\rho\right)_c \sigma_{el} \qquad (6)$$

2. Materials and Method

The thermo-luminescent compounds that was used in this workinclude $MnSO_4, ZnSO_4.7H_2O, BaCO_3, CaCO_3, BaSO_4$ and $CaSO_4$. These compounds were converted to 2.5 cm discs by adding few drops of locally made binder to a weighed quantity of finely ground powder and pressing in a hand operated hydraulic press. Each sample thus prepared was weighed in a digital balance capable of weighing up to a fraction of a milligram. The weighing was repeated a number of times to obtain consistent values of the mass. The mean of this set of values was taken to be the mass of the sample. The density thickness corresponding to mass of loose powder from each compound per unit area was also determined

The radioactive sources ^{241}Am, ^{133}Ba, ^{57}Co ^{137}Cs, ^{54}Mn, and ^{60}Co were used in the present investigation. Each γ -ray of energy 51.5, 132.07, 661.6, 834.8, 1173.2 and 1332.5 keV emitted by the above radioactive isotopes were used for the measurement. Each of these sources are standard (low activity) γ -ray sources that emit reasonably monochromatic γ -rays of specific energy. The half-lives, abundance and date of production of these sources were earlier listed in Table 1.1.

A schematic diagram of experimental arrangement used for γ -ray attenuation measurement is shown in Fig 1.1.It consists of 7.6 × -7.6cm NaI(TI) scintillation detector, which has a resolution of 7% at662keV line of ^{137}Cs (Jonah *et al.*, 2001) mounted on a photomultiplier tube. The output of the detector was connected directly to the input of the amplifier coupled to a single channel analyzer (Model TC 246, Oxford instruments Inc, 601 Oak Ridge USA). The choice of single channel analyzer (instead of multi-channel) analyzer was predicated on its high sensitivity for discrete photon energy with additional compensation for inadequacy in the design of gamma ray shielding for the NaI (TI) detector.

The intensities of the incident and transmitted photons when the detector was biased to 950V were determined by choosing the counting time such that, at least over a three hundred counts were recorded for each gamma ray energy. This ensures that the statistical uncertainty was kept below 0.2% in most cases. However, these requires the positioning of the sources closer to the detector.

The γ-rays emitted by these sources were detected by the NaI (TI) detector when biased to a high voltage of 950V.

For each γ-ray source, thirty (30) replicate counting of γ-ray transmitted through 2.5 cm diameter discs made from the various thermo-luminescent compounds of mass thickness of about 3.8 g/cm^{2}was carried out for a preset time of 20 seconds. Three sets of measurements were taken in order to determine the mass attenuation coefficient of the samples. These are: the incident beam intensity, the attenuated beam intensity and the background. The sample/source holder was marked to indicate the position where the sample and source was placed in order to obtain a reproducible geometry for measurements involving only the gamma ray source and gamma ray/sample arrangement.

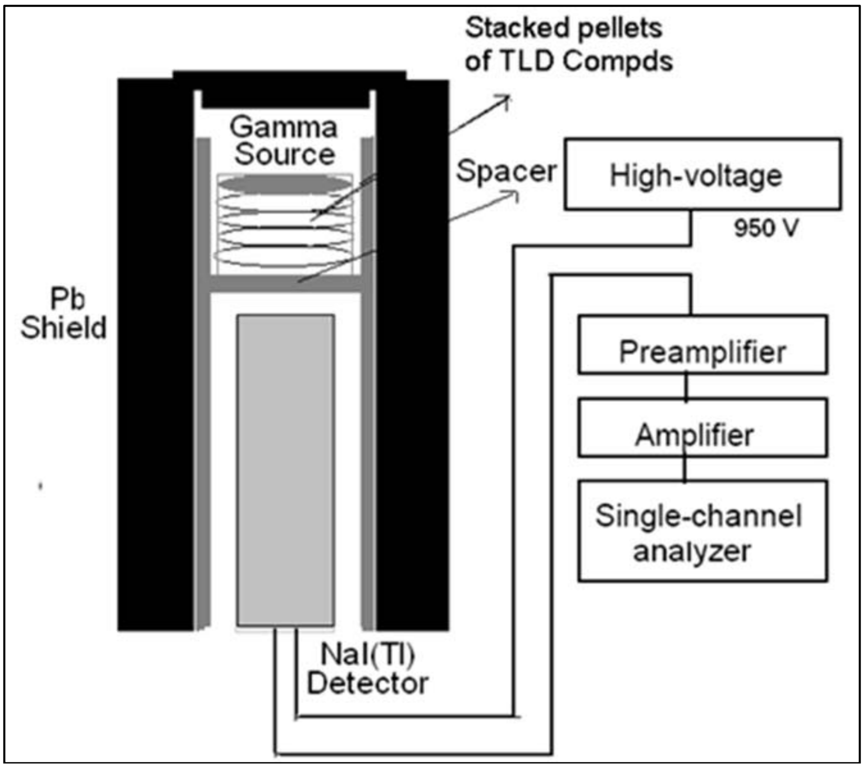

Fig 1.1: Experimental Arrangement for Gamma Transmission Measurement.

Table 2.1: Gamma Ray Intensity Data for $CaCO_3$Themo-luminescent Compound

Gamma ray source	Gamma ray Energy (keV)	Sample thickness (g/cm^2)	Mean Background Count (I_{BG})	Mean Source Count(I_o)	Mean Sample Count (I_s)
^{241}Am	51.5	4.83	6.40±2.39	957.5±30.37	310.33±23.10
^{57}Co	136.5	4.83	35.60±4.98	1649.33±41.97	977.30±42.37
^{137}Cs	661.6	3.24	24.95±5.91	5904.20±73.25	4637.00±40.64
^{54}Mn	834.5	3.24	19.25±4.55	529.40±28.02	436.75±19.54
^{60}Co	1173.2	3.24	7.45±3.75	2145.40±62.45	1821.40±54.58
^{60}Co	1332.5	3.24	16.65±4.15	2226.20±46.36	1931.65±53.19

The source was positioned normal to the NaI (TI) detector and the distance between the source/sample maintained at 5cm. Appropriate number of pellets were stacked together to obtain overall density thickness of about 3.8 g/cm^2 which represents the optimum thickness required to obtain accurate values of mass attenuation coefficient during the preliminary experiments. The choice of an optimum density thickness has the effect of reducing errors due to small angle scattering.

Table 2.2: Gamma Ray Intensity Data for $BaCO_3$Themo-luminescent Compound

Gamma ray source	Gamma ray Energy (keV)	Sample thickness (g/cm2)	Mean Background Count (I_{BG})	Mean Source Count(I_o)	Mean Sample Count (Is)
^{241}Am	51.5	0.81	6.67±2.30	976.76±35.20	456.67±23.04
^{57}Co	136.5	1.69	36.13±4.96	1687.30±34.99	912.87±28.75
^{137}Cs	661.6	3.24	24.89±5.91	5688.15±73.25	4648.00±54.23
^{54}Mn	834.5	3.24	19.25±4.55	529.40±28.02	436.75±19.54
^{60}Co	1173.2	3.24	7.45±3.75	2226.20±46.36	1903.10±42.14
^{60}Co	1332.5	3.34	16.65±4.15	2145.40±62.45	1836.80±54.58

Table 2.3: Gamma Ray Intensity Data for $MnSO_4.4H_2O$ Themo-luminescent Compound

Gamma ray source	Gamma ray Energy (keV)	Sample thickness (g/cm2)	Mean Background Count (I_{BG})	Mean Source Count(I_o)	Mean Sample Count (Is)
^{241}Am	51.5	2.43	7.53±2.45	927.07±31.19	491.27±24.04
^{57}Co	136.5	3.25	35.60±4.99	1651.73±46.58	1126.83±32.60
^{137}Cs	661.6	3.24	24.95±5.91	5769.45±460.40	4626.60±244.45
^{54}Mn	834.5	3.25	19.25±4.55	529.40±28.02	422.50±18.36
^{60}Co	1173.2	3.25	7.45±3.75	2145.40±62.45	1789.00±44.54
^{60}Co	1332.5	3.24	16.65±4.15	2226.20±46.36	1836.80±54.57

Table 2.4: Gamma Ray Intensity Data for $ZnSO_4$Themo-luminescent Compound

Gamma ray source	Gamma ray Energy (keV)	Sample thickness (g/cm2)	Mean Background Count (I_{BG})	Mean Source Count(I_o)	Mean Sample Count (Is)
^{241}Am	51.5	2.43	6.43±2.43	944.30±36.78	398.40±16.79
^{57}Co	136.5	3.25	36.13±4.96	1672.63±46.65	1186.93±42.19
^{137}Cs	661.6	3.24	24.95±5.91	5769.45±460.35	4600.45±244.25
^{54}Mn	834.5	3.24	19.25±4.55	529.40±28.20	441.10±20.01
^{60}Co	1173.2	3.24	16.65±4.14	2226.20±46.36	1903.40±51.57
^{60}Co	1332.5	3.24	16.65±4.14	2226.20±46.95	1866.40±59.27

Table 2.5: Gamma Ray Intensity Data for $CaSO_4$Themo-luminescent Compound

Gamma ray source	Gamma ray Energy (keV)	Sample thickness (g/cm2)	Mean Background Count (I_{BG})	Mean Source Count(I_o)	Mean Sample Count (Is)
^{241}Am	51.5	2.43	6.13±2.20	928.00±31.05	501.00±25.05
^{57}Co	136.5	3.25	36.13±4.96	1674.50±46.56	1142.10±49.89
^{137}Cs	661.6	3.24	24.95±5.91	5887.20±73.25	4485.20±54.22
^{54}Mn	834.5	3.24	19.25±4.55	529.40±28.12	436.74±19.54
^{60}Co	1173.2	3.24	16.65±4.15	2226.20±46.36	1931.65±53.19
^{60}Co	1332.2	3.24	7.45±3.65	2145.40±60.87	1825.35±53.19

3. Results and Discussion

Using the values of attenuated intensity I and the un-attenuated intensity Io displayed in Table 2.1-2.5 the value of the mass attenuation coefficient in the different TLD compounds were determined using equation(7) These are displayed in Table 3.1 for the different gamma energies.

$$\left(\frac{\mu}{\rho}\right)_c = \left(\frac{\ln(I_o / I}{\rho t}\right)_c \qquad (7)$$

Before the actual measurement, a comparison was made for the different absorbers with the different thickness. It was found that the number of count for the photon detected by the detector decreases as the thickness of the absorbers increases. Several centimeters of the pellet thickness made from the thermo-luminescent compounds results in a huge reduction in the peak intensity of gamma rays because it has higher density compare to others. At a certain critical thickness each thermo-luminescent compound produce a value of the mass attenuation coefficient that is closest to the corresponding XCOM values calculated using the XCOM/WINCOM software package. The XCOM/WINCOM package was developed by Berger and Hubbell (1987/99) for calculation of mass attenuation coefficients or photon interaction cross-sections for any element, compound or mixture at energies from 1 keV to 100 GeV. Recently, this well-known and much used program was transformed to the Windows platform by Gerward *et al.* (2001).

The Windows version, called WinXCom, runs under the Windows operating system and provides an interface that facilitates defining, re-defining and saving substances in a substance definition list which comes with a predefined list of the first hundred elements in the periodic table.

The optimum value of density thickness serves to reduce effects due to small angle scattering. Thus Table 3.1 also contains the calculated value of mass attenuation coefficient and mass attenuation of some compounds using the XCOM software at the energies being considered.

Apart from errors due to small angle scattering, other factors that affect the accuracy of measurements include (i) sample impurity, (ii) non-uniformity of the sample, (iii) photon build-up effects, (iv) dead time of the counting instrument and (v) pulse pile-up effects. The error due to sample impurity can be high, but only when a large percentage of high-Z impurities is present in the sample. In all the compounds used in the present study, the content of high-Z impurities was low as evidenced in the low value of density thickness for each compound. Hence, sample impurity corrections were not applied to the measured data. In the high purity samples and in other cases, non-uniformity of the sample was avoided by pulverizing the samples into very fine powder using agate mortar and pestle. This ensures homogenous pellets whose constituent portions are representative of the entire pellet. Photon build up effects, dead time and pulse pile up effects are naturally eliminated by using low activity sources in these measurements. The disagreement between experimental and theoretical values is in the region of 2-3% for the dosimetry samples, the percent deviation being the difference between the experimental and theoretical μ/ρ values divided by theoretical value.

The effective atomic number for each sample was determined from the ratio of the effective atomic cross-section represented by equation (2) and the electronic cross-section represented by equation (3).

Application of equation (2) involves averaging over atoms of all the elements in the compound. This requires the values of mass attenuation coefficient of individual compounds, weight fraction of individual elements, atomic number of individual elements and Avogadro's number as input data. The values of atomic cross section as a function of gamma ray energy determined for the individual dosimetry compounds used in this work are displayed in Table 3.1. The electronic cross-section, σ_{el} , also requires the above input data in addition to mass attenuation coefficients of the constituent elements of each dosimeter compound and the fractional abundance that requires total number of atoms of the same type for each constituent element and the total number of all types present in the compound indicated by the chemical formula of the thermo luminescent compound of interest. The values of the electronic cross section determined in this work are displayed in Table 3.2. Finally, the effective atomic number and the effective electron density were determined from the ratio of atomic and electronic cross sections and from the product of the effective electron number and mass attenuation coefficient of each thermoluminescent compound respectively. The effective atomic number and the effective electron density for the thermoluminescent dosimetry compounds determined in this work are displayed in Table 3.4 and 3.5 respectively

Table 3.1: Experimentally measured photon mass attenuation coefficient of some TLD compounds in cm^2/g (with some of the XCOM values in parenthesis)

Themo-luminescent Compound	**Measured Mass attenuation /XCOM values at Gamma Ray Energy (keV)**					
	51.5	**136.5**	**661.6**	**834.5**	**1173.2**	**1332.5**
$CaCO_3$	0.2362 ±0.0144	0.1115 ± 0.0190	0.0749 ±0.0028 (0.0775)	0.0619 ± 0.0074	0.0507 ± 0.0017 (0.0589)	0.0441 ±0.0022 (0.0552)
$CaSO_4$	0.2562 ±0.0133	0.1211 ±0.0111	0.0843 ±0.0031 (0.0792)	0.0627 ±0.0075 (0.0775)	0.0543 ±0.0018 (0.602)	0.0442 ±0.0022 (0.0564)
$BaCO_3$	0.9472 ±0.0525	0.3735 ±0.0375	0.0626 ±0.0023	0.0618 ±0.0074	0.0485 ±0.0015	0.0481 ± (0.0016)
$MnSO_4$	0.2648 ± 0.0167	0.1201 ± 0.0114	0.0684 ± 0.0025	0.0725 ±0.0088	0.0563 ± 0.0018	0.0601 ±0.0030
$ZnSO_4.7H_2O$	0.3597 ± 0.0205	0.1085 ± 0.0100	0.0702 ± 0.0026	0.0586 ± 0.0070	0.0548 ± 0.0027	0.0601 ± 0.0030

Table 3.2: Effective atomic cross section σ_a (b/atom) of TLD compounds

Themo-luminescent Compound	**Gamma ray energy (keV)**					
	51.5	**136.5**	**661.6**	**834.5**	**1173.2**	**1332.5**
$CaCO_3$	7.845 ±0.479	3.723 ±0.632	2.493 ±0.093	2.061 ±0.246	1.695 ±0.0565	1.463 ±0.073
$BaCO_3$	15.465 ±3.440	7.339 ±2.460	4.849 ±0.151	4.063 ±0.485	3.342 ±0.098	2.883 ±0.011
$CaSO_4$	8.893 ±0.501	4.220 ±0.418	2.826 ±0.117	2.336 ±0.128	1.923 ±0.067	1.658 ±0.083
$MnSO_4$	9.863 ±0.698	4.681 ±0.476	3.135 ±0.104	2.591 ±0.368	2.131 ±0.075	1.839 ±0.125
$ZnSO_4.7H_2O$	10.546 ±0.916	5.005 ±0.447	3.352 ±0.116	2.771 ±0.313	2.279 ±0.121	1.966 ±0.134

Table 3.3: Effective Electronic cross section σ_{el} (b/atom) of TLD compounds

Themo-luminescent Compound	**Gamma ray energy (keV)**					
	51.5	**136.5**	**661.6**	**834.5**	**1173.2**	**1332.5**
$CaCO_3$	0.259	0.233	0.198			0.184
$BaCO_3$	0.642	0.453	0.259	0.233	0.198	0.184
$CaSO_4$	0.642	0.453	0.259	0.233	0.198	0.184
$MnSO_4$	0.642	0.453	0.259	0.233	0.198	0.184
$ZnSO_4.7H_2O$	0.642	0.453	0.259	0.233	0.198	0.184

Table 3.4: Effective Atomic Number Z_{eff} of TLD compounds

Themo-luminescent Compound	Gamma ray energy (keV)					
	51.5	**136.5**	**661.6**	**834.5**	**1173.2**	**1332.5**
$CaCO_3$	12.229 ±0.746	8.213 ±1.393	9.624 ±0.359	8.865 ±1.060	8.579 ±0.286	7.954 ±0.398
$BaCO_3$	96.76 ±5.363	53.98 ±5.421	15.841 ±0.582	17.476 ±2.900	16.129 ±0.497	17.179 ±0.570
$CaSO_4$	15.052 ±0.781	9.989 ±0.916	12.260 ±0.451	10.109 ±1.210	10.425 ±0.343	9.041 ±0.451
$MnSO_4$	17.251 ±1.088	11.072 ±1.043	11.041 ±0.403	13.083 ±1.570	11.916 ±0.381	13.605 ±0.681
$ZnSO_4.7H_2O$	25.061 ±1.428	10.062 ±0.978	12.113 ±0.449	11.249 ±1.340	12.433 ±0.611	14.606 ±0.729

Table 3.5: Electron density N_{el} of TLD compounds

Themo-luminescent Compound	Gamma Ray Energy (keV)					
	51.5	**136.5**	**661.6**	**834.5**	**1173.2**	**1332.5**
$CaCO_3$	3.679 ±0.225	2.471 ±0.419	2.895 ±0.108	2.667 ±0.318	2.581 ±0.086	2.393 ±0.119
$BaCO_3$	3.679 ±0.818	2.471 ±0.827	2.857 ±0.088	2.667 ±0.318	2.581 ±0.076	2.393 ±0.087
$CaSO_4$	3.679 ±0.207	2.453 ±0.243	2.895 ±0.119	2.652 ±0.321	2.581 ±0.091	2.392 ±0.091
$MnSO_4$	3.679 ±0.260	2.453 ±0.249	2.895 ±0.097	2.652 ±0.376	2.581 ±0.091	2.392 ±0.163
$ZnSO_4.7H_2O$	3.679 ±0.319	2.453 ±0.219	2.895 ±0.100	2.652 ±0.299	2.581 ±0.136	2.392 ±0.163

Generally, Z_{eff} for each of the substances considered is not a constant but varies with photon energy. The same applies to electron density of TLD compounds. Generally, the behavior of Z_{eff} with energy for all the substances considered in this work is similar. It decreases steadily as energy increases then becomes almost constant and later decreases again. These variations can be attributed to the photon interaction dominating at the energies considered.

For all the TLD substances, Z_{eff} was highest at the lower end of the energy spectrum considered and lowest at intermediate energies. This behavior is attributed to the photoelectric effect and Compton scattering dominating at the low energy and intermediate energies respectively.

4. Conclusion

In the present work, the Z_{eff} values and the electron densities of the TLD compounds were obtained using the measured values of their mass attenuation coefficients in the photon energy range 51.5 to 1332.5 keV. The values of effective atomic numbers and the electron densities of TLD compounds are in agreement with the other available data at energies of interest.

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