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Technical Note

A study of total mass attenuation coefficients, effective atomic numbers and electron densities for various organic and inorganic compounds at 59.54 keV

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ABSTRACT

The total mass attenuation coefficients μ/ρ, effective atomic numbers (Z_{eff}) and effective electron densities (N_e) were determined for 21 different compounds at 59.54 keV using a narrow beam good geometry set-up. The effective atomic numbers and electron densities have been determined on the basis of mixture rule using the values of total mass attenuation coefficients and compared with the calculated ones from theory. Also, the obtained values of effective atomic numbers have been compared with the ones calculated according to a different approach proposed by Hine.

1. Introduction

Since the gamma-active isotopes have been widely used in various fields, the photon mass attenuation coefficients, photon interaction cross sections, effective atomic numbers and effective electron densities have gained great importance with their use in medical diagnostic and therapy computations, as well as in diverse applications in other fields such as nuclear power plant shielding, health physics and industrial irradiation and monitoring, and in X-ray crystallography (Hubbell, 2006). Accurate values of those parameters are needed to establish the regions of validity of theory based parameterization, in addition to providing essential data in the above mentioned fields (Gowda et al., 2004). In order to determine the most suitable materials to be used in radiation shielding and dosimetry, the characteristics of the various materials based on radiation interaction with matter must be known (Shivaramu and Ramprasath, 2000).

Literature is rich in studies of photon interaction in the elements and also extensive data sets are available (Hubbell and Seltzer, 1995; Hubbell, 1982; Lide, 1996; Berger and Hubbell, 1987–1999; Gerward et al., 2001, 2004). However, limited attempts seem to have been made in compounds, mixtures or composite materials. The attenuation of X- and/or γ-photons is related to the density and atomic number of an element. In case of a multi-element material it is then related to the density and effective atomic number.

The effective atomic number and effective electron density are the basic and important quantities among the parameters determining the constitutive structure of an unknown object or material. Effective atomic number has a physical meaning and gives basic information about the characteristics of a multi-element material. Using the constants like effective atomic number (Z_{eff}) and effective electron density (N_e) of multi-element materials, one can calculate the energy absorption in a given medium via well established formulas. Effective atomic number (Z_{eff}) which is representing the radiation interaction with matter also takes place in some applications viz. designing radiation shielding, computing absorbed dose and build-up factor. In some cases, in order to have an initial information about the chemical composition of a material the Z_{eff} can be utilized. For example the materials having large Z_{eff} generally corresponds to the inorganic compounds and metals, while a small Z_{eff} ($\leqslant 10$) is an indicator of organic substances (Manohara et al., 2008). It was pointed out by Hine that the effective atomic number cannot be expressed by a single number and the various atomic numbers present in the compound have to be weighted differently (Hine, 1952).

Several attempts have been made to calculate Z_{eff} and N_e for different compounds of interest (Perumallu et al., 1984; Lingam et al., 1984; Singh et al., 1996; Kumar and Reddy, 1997; Gowda et al., 2004; Manjunathaguru and Umesh, 2006; Manjunathaguru and Umesh, 2007). However, the studies on effective atomic number and electron density for different compounds seem to be very scarce. Accurate values of the mass attenuation coefficients, photon interaction cross sections, effective atomic numbers and effective electron densities will throw light on the validity of the well-known mixture rule. Recently, de Jonge et al. have measured the mass attenuation coefficients and determined the imaginary component of the atomic form factor of molybdenum over the 13.5–41.5 keV energy range (de Jonge et al., 2005). Tran et al. have measured the X-ray mass attenuation coefficient of silver using the X-ray extended energy range with high accuracy (Tran et al., 2005). Keeping in mind the very limited number of effective atomic number and electron density study of different compounds, the present

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study aimed at the investigation of the photon interaction parameters of some compounds in terms of effective atomic numbers and electron densities as well as total mass attenuation coefficients. Therefore, we have found interesting to study those parameters at a specific energy for different compounds having constituent elements in the wide atomic range $1 \leqslant Z \leqslant 68$.

In the present study, twenty one substantially pure compounds have been chosen and an attempt has been made to determine total mass attenuation coefficients, effective atomic numbers and effective electron densities at 59.54 keV. Also, the effective atomic numbers have been calculated by using a different approach proposed by Hine and compared with the present results.

2. Theory

2.1. Total mass attenuation coefficient

According to the Lambert–Beer law, a parallel beam of monoenergetic X- and/or γ-photons is attenuated in matter by the following exponential attenuation equation

$$I = I_0 e^{-\left(\frac{\mu}{\rho}\right)t}, \tag{1}$$

where I_0 and I are the incident intensity of photons without attenuation and the attenuated intensity of photons in the sample, respectively, t is the mass thickness of the sample that corresponds to the mass per unit area in g/cm^2, μ/ρ (cm^2/g) is the mass attenuation coefficient which is a density independent quantity.

In case of a multi-element material (i.e. chemical compound or homogeneous mixture) constituting the sample, the mass attenuation coefficient can be obtained from the coefficients for the constituent elements which are assumed to be additive according to the weighted average

$$\mu/\rho = \sum\nolimits_i W_i(\mu/\rho)_i \tag{2}$$

where W_i is the proportion by weight of the ith constituent element. This well-known mixture rule is valid with the assumption that the effects of molecular binding and the chemical and crystalline environment are negligible.

2.2. Photon interaction cross sections, effective atomic number and effective electron density

The total atomic cross section can be obtained by dividing the mass attenuation coefficient μ/ρ (cm^2/g) of the compound by the total number of atoms present in one gram of that compound as follows:

$$\sigma_a = \frac{(\mu/\rho)_c}{N_A \sum_i \frac{w_i}{A_i}} \quad (barns/atom) \tag{3}$$

where $(\mu/\rho)_c$ is the mass attenuation coefficient of the compound, w_i is the fraction by weight of the element i, and A_i is the atomic weight of the ith element.

The attenuation cross section values of compounds were then interpolated in the attenuation cross section values of generated from WinXCom at the selected energy to calculate the effective atomic number (Z_{eff}) using the following logarithmic interpolation formula (Singh et al., 2007):

$$Z_{eff} = \frac{Z_1(\log\sigma_2 - \log\sigma) + Z_2(\log\sigma - \log\sigma_1)}{\log\sigma_2 - \log\sigma_1} \tag{4}$$

where σ_1 and σ_2 are the elemental cross section ($barns/atom$) in between which the atomic cross section σ of the compound lies and Z_1 and Z_2 are atomic numbers of the elements corresponding to the cross sections σ_1 and σ_2, respectively.

Also, the effective electron density is expressed by the following relation:

$$N_E = \left(\frac{Z_{eff}}{A_i}\right) N_A \sum\nolimits_i n_i \quad (electrons/g) \tag{5}$$

Thus, using the values of Z_{eff} obtained from logarithmic interpolation in the Eq. (5), one can calculate the values of N_E by using the Eq. (5).

Also, the effective atomic number can be obtained by the semi empirical relation suggested by Hine as follows:

$$Z_{eff} = \left(\sum\nolimits_i w_i Z_i^{3.1}\right)^{1/3.1} \tag{6}$$

3. Calculations

Mass attenuation coefficients of given materials have been calculated by the WinXCom program. This program which is based on the DOS-based compilation XCom (Berger and Hubbell, 1987–1999) provides total mass attenuation coefficient and total attenuation cross section data for about 100 elements as well as partial cross sections for incoherent and coherent scattering, photoelectric absorption and pair production at energies from 1 keV to 100 GeV (Gerward et al., 2004). The obtained values of mass attenuation coefficients were then used to calculate total interaction cross sections, effective atomic numbers and effective electron densities.

4. Experimental

4.1. Sample preparation

The compounds used in the present work are in the powder form. The samples were ground in a SPEX mill with 25 ml stainless-steel cup and balls to minimize the grain sizes and therefore to reduce the particle size effect on the X-ray intensity. Samples were sieved in <37 μm (–400 mesh) scaled sieves to ensure particle size homogeneity in the sample. Prior to analysis, samples were pressed into pellets having a diameter of 0.13 cm under the pressure of 10 ton per cm^2 in a SPEX Pmax hydraulic press. This progress provided a fairly smooth surface and so minimized the errors originate from improper surface of samples.

4.2. Measurements

In the present study, the total mass attenuation coefficients $[\mu/\rho(cm^2/g)]$ of the compounds were measured by performing transmission experiments in a narrow beam good geometry setup (Fig. 1). The beam path was aligned using a laser. The photons emitted from the radioactive point source ^{241}Am of 100 mCi (3700 MBq) strength were well collimated and suitably detected. The ^{241}Am gamma source was housed at the center of a cylindrical shield of 10 mm diameter and 36 mm depth. A high resolution liquid nitrogen cooled Si(Li) detector (FWHM = 160 eV for the Mn-Kα line at 5.9 keV, an active area of 12 mm^2, a sensitive crystal depth of 3 mm, a beryllium (Be) window of 0.025 mm thickness) coupled to 4 k multichannel analyzer was employed to detect 59.54 keV. The detector was shielded by a graded filter of Pb (–4.2 mm), Fe (–1.1 mm) and Al (–1 mm) to prevent radiation scattered by nearby objects and also radiation such as L X-rays from the Pb mask, environmental background and background arising from the scattered radiation due to sample holder. The spectra were recorded using a PC-based multi-channel analyzer, supplied by CANBERRA, USA. The measurements were performed three times for each sample. The each spectrum was recorded for suffi-

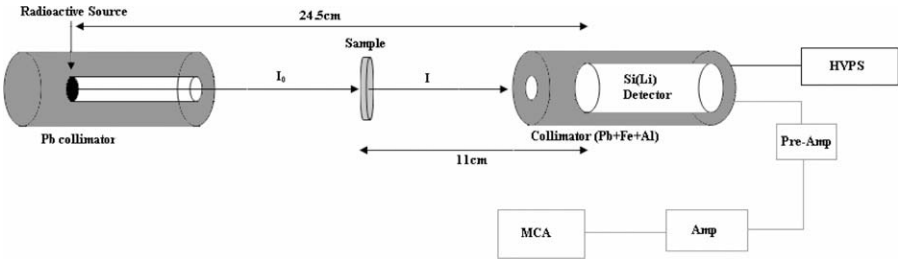

Fig. 1. The employed experimental set-up (dimensions are not to scale) used for transmission experiments.

cient time to accumulate an adequate number of counts under the photo peak to limit the uncertainty of <1%.

The compounds used as samples have purities of ⩾99.5%. The atomic weights of the elements constituting the compounds were taken from the values reported by IUPAC (IUPAC, 2007). The samples were placed one by one between the source and the detector. The sample diameter was measured by a micrometer which could measure down to 10^{-2} mm.

The maximum errors in total mass attenuation coefficients were calculated from errors in incident (I_0) and transmitted (I) intensities and areal density (t) using the propagation of error formula:

$$\Delta\left(\frac{\mu}{\rho}\right)=\frac{1}{t}\sqrt{\left(\frac{\Delta I_0}{I_0}\right)^2+\left(\frac{\Delta I}{I}\right)^2+\left(In\frac{I_0}{I}\right)^2\left(\frac{\Delta t}{t}\right)^2} \quad (7)$$

where t is the areal density (thickness that corresponds to the mass per unit area in g/cm^2), ΔI_0, ΔI and Δt are the errors in the intensities I_0, I and thickness t of the sample, respectively. Estimated error in the experimental measurement was less than 5%.

5. Results and discussion

The presented results have an uncertainty of less than 5%. This error arises mainly from counting statistics, peak area and thickness determination. The values of total mass attenuation coefficients of the compounds have been obtained using the Eq. (1). Table 1 lists the experimental and theoretical values of total mass attenuation coefficients of compounds considered at 59.54 keV. In general, the experimental values agree with the theoretical values which are calculated from the mixture rule (WinXCom program). It is evident from the table that the μ/ρ values tend to be higher in heavier compounds.

The experimental and theoretical values of effective atomic numbers and effective electron densities are tabulated in Table 2 and Table 3, respectively. It can be clearly seen that the variation in Z_{eff} depends on the range of atomic numbers of elements of which the compound is composed from the Table 2. The compounds used as samples in the present study have constituent elements in the atomic range $1 \leqslant Z \leqslant 68$. For this particular value of the incident photon energy, the Z_{eff} values were found to increase with the presence of high Z elements in the compound. However, with respect to the values of N_e, there was no similar trend with Z_{eff} that the variation of N_e values with regard to atomic number is not smooth (Table 3). Also, it was observed that the mean atomic numbers of the compounds are significantly different than the Z_{eff} values of the compounds except for the compounds consist of nearby Z elements (Table 2). In the present energy as mentioned above, the dominant photon interaction process is photoelectric effect. Accordingly, the Z_{eff} values are not close to the mean atomic number of the given compounds due to the Z-dependence of photoelectric process which is of the order of Z^{4-5}.

Table 1
Total mass attenuation coefficients of compounds along with the theoretical values at 59.54 keV.

Compound	Chemical formula	Total mass attenuation coefficient	
		μ/ρ(exp) (cm^2/g)	μ/ρ(theo) (cm^2/g)
Cobalt carbonate	$CoCO_3$	0.759	0.759
Cobalt thiocyanate	$Co(SCN)_2$	0.628	0.655
Molybdenum carbide	Mo_2C	4.29	4.12
Nickel (II) fluoride	NiF_2	1.04	1.01
Nickel (II) perchlorate hexahydrate	$Ni(ClO_4)_2{\cdot}6H_2O$	0.459	0.462
Nickel (II) phosphate heptahydrate	$Ni3(PO_4)_2{\cdot}7H_2O$	0.739	0.699
Nickel sulfide	Ni_3S_2	1.28	1.24
Ferrous sulfate heptahydrate	$FeSO_4{\cdot}7H_2O$	0.414	0.432
Zinc oxide	ZnO	1.47	1.48
Erbium chloride hexahydrate	$ErCl_3{\cdot}6H_2O$	6.21	6.27
Erbium oxalate hydrate	$Er_2(C_2O_4)_3{\cdot}10H_2O$	6.37	6.07
Erbium (III) sulfate octahydrate	$Er_2(SO_4)_3{\cdot}8H_2O$	6.51	6.19
Zirconium silicate	$ZrSiO_4$	2.10	2.02
Zirconium chloride octahydrate	$ZrOCl_2{\cdot}8H_2O$	1.23	1.28
Zirconium hydride	ZrH_2	3.82	3.75
Zirconium silicide	$ZrSi_2$	2.62	2.49
Copper (II) fluoride	CuF_2	1.10	1.09
Copper (II) selenate	CuO_4Se	1.42	1.47
Copper (II) sulfide	CuS	1.19	1.22
Copper (II) silicide	Cu_5Si	1.52	1.52
Manganese telluride	MnTe	5.15	5.35

It can be seen from the Table 2 that for compounds containing elements with the atomic numbers of which do not differ greatly, the present Z_{eff} values and the Z_{eff} values calculated according to the Hine's expression seem to have similar values. In contrast, for compounds containing very light elements such as hydrogen and for compounds containing elements of very wide spread atomic numbers, significance difference has been observed between Z_{eff} values. It should be noted that the Z_{eff} value of a compound lies between the lowest and highest atomic numbers of its constituent elements as limits. At a specific energy, the effective atomic number for total photon interaction can be specified uniquely but its magnitude depends on the numbers for partial processes and their relative contribution at that energy (Murty et al., 2000). In the present study, due to the availability of the present energy for which the dominant interaction process is photoelectric effect, we have found interesting to compare the present Z_{eff} values with those calculated by Hine's expression (Hine, 1952). It can be clearly seen from the Table 2 that for the compounds having low Z elements the agreement is quite satisfactory between the present Z_{eff} values and those evaluated using Hine's expression. However,

Table 2
Effective atomic numbers of the compounds along with the mean atomic numbers at 59.54 keV.

Compound	Chemical formula	$\langle Z \rangle$[a]	$(Z_{eff})_{exp}$	$(Z_{eff})_{theo}$	(Z_{eff})[b]
Cobalt carbonate	$CoCO_3$	11.4	17.74	17.74	16.46
Cobalt thiocyanate	$Co(SCN)_2$	12.14	16.96	17.19	16.23
Molybdenum carbide	Mo_2C	30	38.14	37.73	36.86
Nickel (II) fluoride	NiF_2	15.33	21.32	21.16	20.01
Nickel (II) perchlorate hexahydrate	$Ni(ClO_4)_2{\cdot}6H_2O$	7.75	11.52	11.56	11.24
Nickel (II) phosphate heptahydrate	$Ni3(PO_4)_2{\cdot}7H_2O$	7.29	14.87	14.57	13.57
Nickel sulfide	Ni_3S_2	23.2	25.24	25.02	24.61
Ferrous sulfate heptahydrate	$FeSO_4{\cdot}7H_2O$	5.33	9.84	10.07	10.25
Zinc oxide	ZnO	19	25.03	25.10	24.12
Erbium chloride hexahydrate	$ErCl_3{\cdot}6H_2O$	10.52	29.32	29.39	25.48
Erbium oxalate hydrate	$Er_2(C_2O_4)_3{\cdot}10H_2O$	7.36	33.12	32.70	24.2
Erbium (III) sulfate octahydrate	$Er_2(SO_4)_3{\cdot}8H_2O$	8.78	30.29	29.89	25.46
Zirconium silicate	$ZrSiO_4$	14.33	25.53	25.27	22.91
Zirconium chloride octahydrate	$ZrOCl_2{\cdot}8H_2O$	5.79	16.40	16.61	14.49
Zirconium hydride	ZrH_2	14	30.09	29.94	28.06
Zirconium silicide	$ZrSi_2$	22.67	30.73	30.33	28.75
Copper (II) fluoride	CuF_2	15.67	21.97	21.92	20.69
Copper (II) selenate	CuO_4Se	15.83	23.70	23.93	22.44
Copper (II) sulfide	CuS	22.5	24.72	24.88	24.31
Copper (II) silicide	Cu_5Si	26.5	27.77	27.78	27.53
Manganese telluride	MnTe	38.5	43.34	43.78	42.92

[a] Refers to the mean atomic number.
[b] Calculated by using Hine's expression.

Table 3
Effective electron densities of the compounds for total photon interaction at 59.54 keV.

Compound	Chemical formula	Effective electron density [(electrons/g) $\times 10^{23}$]	
		$(N_e)_{exp}$	$(N_e)_{theo}$
Cobalt carbonate	$CoCO_3$	4.49	4.49
Cobalt thiocyanate	$Co(SCN)_2$	4.08	4.14
Molybdenum carbide	Mo_2C	3.38	3.34
Nickel (II) fluoride	NiF_2	3.98	3.95
Nickel (II) perchlorate hexahydrate	$Ni(ClO_4)_2.6H_2O$	5.50	5.52
Nickel (II) phosphate heptahydrate	$Ni3(PO_4)_2{\cdot}7H_2O$	6.19	6.06
Nickel sulfide	Ni_3S_2	3.16	3.14
Ferrous sulfate heptahydrate	$FeSO_4{\cdot}7H_2O$	5.75	5.89
Zinc oxide	ZnO	3.70	3.71
Erbium chloride hexahydrate	$ErCl_3{\cdot}6H_2O$	10.17	10.20
Erbium oxalate hydrate	$Er_2(C_2O_4)_3{\cdot}10H_2O$	7.44	7.34
Erbium (III) sulfate octahydrate	$Er_2(SO_4)_3{\cdot}8H_2O$	9.75	9.62
Zirconium silicate	$ZrSiO_4$	5.03	4.98
Zirconium chloride octahydrate	$ZrOCl_2{\cdot}8H_2O$	8.58	8.69
Zirconium hydride	ZrH_2	5.83	5.80
Zirconium silicide	$ZrSi_2$	3.76	3.72
Copper (II) fluoride	CuF_2	3.91	3.90
Copper (II) selenate	CuO_4Se	4.15	4.19
Copper (II) sulfide	CuS	3.11	3.13
Copper (II) silicide	Cu_5Si	2.90	2.90
Manganese telluride	MnTe	2.86	2.89

with respect to the compounds having high Z elements which lead to a wide spread in the atomic range of elements constituting the compound, there is a disagreement between the the present Z_{eff} values and those evaluated using Hine's expression. Similar disagreement was observed by Prasad et al. (1998) and Murty et al. (2000) for alloys. It is worth mentioning that it should be reasonable to use Hine's expression to estimate Z_{eff} values in compounds which contain low and nearby Z elements whereas for compounds having elements of wide spread atomic numbers and having at least one high Z element it is not recommended to use Hine's expression due to the large variation in Z_{eff} values if compared to present Z_{eff} values.

6. Conclusions

(1) The photon interaction parameters have been investigated for 21 compounds in terms of effective atomic numbers and effective electron densities as well as total mass attenuation coefficient at 59.54 keV.
(2) The general agreement on the mass attenuation coefficients of compounds indicates that the mixture rule is applicable to these compounds at the incident photon energy of 59.54 keV.
(3) For this particular value of the incident photon energy of 59.54 keV, the effective atomic numbers of the given compounds are dependent on the range of atomic number of the constituent elements and are expected to be higher in heavy compounds whereas no well defined atomic number dependence occurs for the effective electron density in the compounds considered at 59.54 keV.
(4) The presented data on photon interaction parameters are expected to be helpful in dosimetry, radiation shielding and other radiation physics based applications.
(5) Further investigations on the photon interaction parameters in different compounds and/or composite materials are still needed to confirm the validity of the both interpolation method and mixture rule at different incident photon energies.

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