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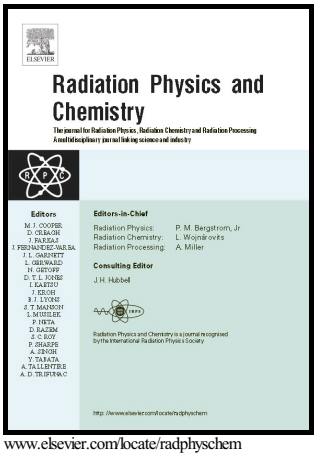


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# Determination of effective atomic numbers, effective electrons numbers, total atomic cross-sections and buildup factor of some compounds for different radiation sources

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Abstract

The photon interaction parameters such as mass attenuation coefficient, effective atomic number, effective electron density, buildup factor have been measured for $Fe(NO_3)_3$, $V_4O_2$, $NaCO_3.H_2O$, $C_6H_5FeO_7.H_2O$ and CuCI compounds using $^{137}Ba$, $^{157}Gd$ and $^{241}Am$ γ-rays sources in stable geometry. The mass attenuation coefficients have been determined experimentally via Energy Dispersive X-ray Fluorescence Spectroscopy (EDXRF) system and theoretically by using WinXCom computer program. Then, effective atomic numbers, $Z_{eff}$, and electron densities, $N_{eff}$, have been calculated by using the mass attenuation coefficients. The obtained values of effective atomic numbers have been compared with the ones calculated according to a different approach proposed by Hine and the calculated ones from theory. Also, photon buildup factors were obtained by changing collimator diameters in the different photon energies. We observed that the buildup factor increased as the collimator diameter increased for all sources used.

Keywords:

Buildup factor, EDXRF, effective atomic number, effective electron density.

## 1. Introduction

In recent years, the study of photon matter interaction parameters such as mass attenuation coefficient, effective atomic number, effective electron density, buildup factor and photon interaction cross-section in compounds (or materials) have attained significant increasing use of radioactive sources in gamma-ray fluorescence studies, medicine, radiation physics and chemistry, industrial applications and agriculture etc. Also, the dependence of photon interaction upon atomic number Z has many applications in radiation studies and other fields involving radiation matter interaction. Thus, the knowledge of these parameters is absolutely

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necessary. These parameters have determined for Fe $(NO_3)_3$, $V_4O_2$, $NaCO_3.H_2O$, $C_6H_5FeO_7.H_2O$ and CuCI compounds in this study.

Because radiation used in the many areas is dangerous for living, it is important to eliminate the effects of primary and secondary radiation in shielding technology. The photon forwarding in the material exposes many scattering (Compton scatter and Photoelectric effect, e.g.) and the secondary radiations occur as a result of the scattering. The precise determination of the radiation rate depends on the correct measurement of total absorbed photon for studies involving radiation in nuclear areas and dosimeter. Calculations of the energy absorbed in a medium include not only the additive of the uncollided photons from the source, but also include the additive from collided and secondary photons. The energy absorption buildup factor is the product of the Compton Scattering known as the ratio of the total energy absorbed due to uncollided, collided, and secondary photons to the energy absorbed due to only uncollided photons (Overcamp, 2009). There are many studies on the effective atomic number and the buildup factors in the literature (Brar and Mudahar, 1995, Sharma et al., 2012, Singh et al., 2003, Manohara et al., 2010, İçelli et al., 2013). Shimizu et al. (2004) calculated the ratio of buildup factor depending different thickness and energy for some elements. Variations of buildup factor with absorber thickness were reviewed for different values of scatter acceptance angle (Changing the diameter of collimator) in high volume flyash concrete and water by Singh et al. (2008).

The mass attenuation coefficient $(\mu/\rho)$ is the parameter which evaluates probability of interaction of photon with the matter and is measured in $cm^2/g$. It is the essential parameter to provide many other photon interaction parameters such as atomic cross-section, electronic cross-section, equivalent and buildup factor, electron density, effective atomic numbers etc. In this direction, one of the first studies on the mass absorption coefficient was made for 32 elements $Z = 1$ to 90 and the photon energy range between 30 eV to 2.5 MeV by Allen (1935). In later years, measurements of $(\mu/\rho)$ values are made for different energy ranges and Z atomic numbers (Brown, 1966, Biggs and Lighthill, 1971, Veigele, 1973). Mass attenuation coefficients of some compounds and mixtures of dosimetric in the energy range 1 keV – 20 MeV were determined by Hubbell (1982). Berger and Hubbell (1987) developed a computer program that calculates theoretically attenuation coefficients for any element, compound or mixture. Then Gerward et al. (2001, 2004) improved the program and its name as WinXCom.

It can calculate attenuation coefficients of elements, compounds and mixtures atomic numbers from 1 to 100 and energy of up to 100 GeV.

The atomic numbers of compounds, alloys and composite materials, as in the pure elements cannot represented by a single number in the different energy regions. This number for compounds, alloys or mixtures is named as the ‘‘effective atomic number”, $Z_{eff}$, and it varies with the photon energy. This concept has been introduced for the very first time by Hine (1952). Effective electron density, $N_{eff}$, is a measurement the number of electrons per unit mass of the interacting substance. The increase in the electron density increases the chances of photon interactions. Literature is rich of experimental as well as theoretical studies regarding effective atomic number and electron density in which radiation sources of X- and/or gamma-rays are used (El-Kateb et al., 2000; Gowda et al., 2004; Manjunathaguru and Umesh, 2006; Kaewkhao et al., 2008; Cevik et al., 2008; Manohara et al., 2008, 2009; Han and Demir, 2009; Sidhu et al., 2012; Elmahroug et al., 2015).

In present study, mass attenuation coefficient, effective atomic number, effective electron density, buildup factor of $Fe(NO_3)_3$, $V_4O_2$, $NaCO_3.H_2O$, $C_6H_5FeO_7.H_2O$ and CuCI compounds have been determined experimentally for different radiation sources emitting photons of different energy. These parameters (except for buildup factor) of the compounds have been calculated for the same energies. Also we have investigated change of buildup factor with increasing photon interaction in material for different sources. Chemical compounds used in the study are especially useful in the manufacture of some special steels, analytical chemistry, glass production, as a wetting elements in brick-making and applications related of humidity indicator.



## 2. Theoretical formulation

### *2.1. Mass attenuation coefficient*

The mass attenuation coefficient is a measurement of scattering or absorption of incident light depending on the chemical properties of the material, per unit mass. If the intensity of the incoming radiation on materials is known, the mass attenuation coefficient is a useful parameter for finding the radiation leaving from the materials. This process is described by the following equation:

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

where $I_0$ is the original intensity of the beam ; $I$ is the intensity of the beam after attenuation into the substance; $\mu/\rho$ is the mass attenuation coefficient *(cm²/g)* and $t$ is sample mass thickness *(g/cm²)* (the mass per unit area). If $I$ and $I_0$ are the measured count rates in detector respectively with and without the absorber of thickness *x (cm)* of the absorber, the linear attenuation coefficients ($\mu$) (and consequently the mass attenuation coefficients) can be extracted by Lambert-Beer Law:

$$\mu = \frac{1}{x}\ln\left(\frac{I_0}{I}\right) \quad (2)$$

The same experimental procedure (and condition) was repeated for different radiation sources to determine the mass attenuation coefficients. After determining the mass attenuation coefficients of compounds experimentally, it has been obtained theoretically by using WinXCom computer program. The experimental and theoretical values of the mass attenuation coefficients of compounds have been used to determine the total atomic cross-section, $\sigma_a$.

### *2.2. Effective atomic number ($Z_{eff}$)*

The atomic numbers of chemical compounds or mixtures cannot be represented by a single number for all energies as in the elements. So the effective atomic number, $Z_{eff}$, is the term used for composite samples. It can be obtained by using of the atomic number of each elements and the total atomic cross-section. The total atomic cross-section ($\sigma_a$) is given by the following formula;

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

where $(\mu/\rho)_{comp}$ is the mass attenuation coefficient of the compound, $N_A$ is the Avogadro constant, $w_i$ is the fraction by weight of the element *i*, and $A_i$ is the atomic weight of the *i*th element. After determining the total atomic cross-sections of compound experimentally, it has

been obtained theoretically by using WinXCom computer program. The experimental and theoretical values of the total atomic cross-section, $\sigma_a$, of compounds have been used to determine the effective atomic number, $Z_{eff}$. The total atomic cross-section values of samples were interpolated in the total atomic cross-section values of elements generated from WinXCom (Gerward et al., 2004) at the selected energy to calculate the effective atomic number ($Z_{eff}$) using the following logarithmic interpolation formula (Singh et al., 2007):

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

where $\sigma_1$ and $\sigma_2$ are the atomic cross-sections (barns/atom) in between which the atomic cross-section $\sigma$ of the material lies and $Z_1$ and $Z_2$ are atomic numbers of the elements corresponding to the cross-sections $\sigma_1$ and $\sigma_2$, respectively.

The mean atomic number ($\bar{Z}$) has been suggested by Müller (1954) and Lloyd (1987).

$$\bar{Z} = \sum_{i=1}^{n} w_i Z_i \quad (5)$$

Another approach of effective atomic number ($Z_{eff}$) put forward by Hine is as follows.

$$Z_{eff} = (\sum_i w_i (Z_i)^{3.1})^{1/3.1} \quad (6)$$

where $w_i$ is the fraction by weight of the element *i*, and Z is atomic numbers of the element.

### *2.3. Effective electron density ($N_{eff}$)*

The electron density ($n_e$) is expressed as number of electrons per unit mass for the elements.

$$n_e = \frac{N_A Z}{A} \quad (7)$$

This expression can be generalized to a compound, and one has

$$N_{eff} = N_A \frac{nZ_{eff}}{\sum_i n_i A_i} = N_A \frac{Z_{eff}}{<A>} \left(\frac{electrons}{g}\right), \quad (8)$$

where $< A >$ is the average atomic mass of materials (Manohara et al., 2008). The values of $N_{eff}$ (Eq.8) can be calculated using $Z_{eff}$ obtained in Eq. (4).

$$< Z >= \frac{n_i Z_i}{n} \tag{9}$$

where $< Z >$ is the mean atomic numbers.

*2.4. Buildup factor*

The Lambert-Beer Law is applicable only if the following three conditions are realized: *i)* Radioactive source must be mono-energetic, *ii)* Target/interacting material must be thin (in order to have a single interaction), *iii)* Narrow beam geometry must be used. In case of being violated of these conditions, then the modified Lambert-Beer Law ($I = BI_0e^{-\mu x}$) is used for the intensity relation, where B represents the Buildup factor. The incident ($I_o$) and transmitted ($I_{measured}$) intensities of photons were recorded for various compounds. The value of transmitted intensity ($I_{calculated}$) for incident intensity ($I_o$) was obtained using Lambert-Beer Law $I = I_0e^{-\mu x}$. It was observed that the value of $I_{measured}$ is always greater than $I_{calculated}$ and the buildup factor:

$$B = \frac{I_{measured}}{I_{calculated}} \tag{10}$$

as mentioned before (Singh et al., 2008).

3. Experimental details

The transmission experiments were carried out for compounds. A schematic diagram of geometry used in present investigation is shown in Fig. 1. The samples were irradiated by 32.2, 36.4, 42.9, 48.7 and 59.5 keV photons emitted by $^{137}$Ba, $^{157}$Gd and $^{241}$Am radioactive point source ($^{137}$Ba and $^{157}$Gd were used as secondary sources and excited by $^{241}$Am), respectively. Incident ($I_o$) and transmitted beam intensity ($I$) for each sample were measured by a Si(Li) detector (FWHM = 160 eV at 5.9 keV, active area 12 mm$^2$, thickness 3 mm, Be window thickness 0.025 mm) coupled with a multi-channel analyzer system and spectroscopy amplifier. Two collimators were used for the formation of a narrow beam of photons from the source. Also the surroundings of experimental geometry were covered with lead shield to

prevent other directly, scattering or background radiation from the environment and source. The compounds used in the experiment were pressed in the pressure of 5 tons/cm$^2$ by a hydraulic press (Spex, Cat. B25). This process provides the formation of rather a flat surface and may be minimized errors resulting from uneven surfaces. Masses of the samples made into tablet were weighed by a scale with precision $10^{-5}$g. The experimental geometry and environmental conditions were kept as constant as possible for all measurements. Firstly, both collimators were kept constant for measurements ($I_0$, $I$) to calculate the effective atomic number and the effective electron density. Secondly, the diameter of the collimator2 in the experimental geometry was changed by using collimators which have 4 different diameters in the range of 2.0–8.9 mm for observe the changes depending on the collimator diameter of buildup factor. The measurements ($I_{measured}$, $I_{calculated}$) were taken for each collimator. The peak areas were calculated from the spectrum obtained for each measurement. Measurement was repeated three times for all samples. The spectrums were analyzed by using Microcal Origin 7.5 Demo Version software program with least-squares fit method.

### *3.1.Errors*

The error margin for experimental values obtained in this study is due to from the mass attenuation coefficient. The maximum errors in total mass attenuation coefficients were calculated from errors in incident ($I_o$) and transmitted ($I$) intensities and areal density ($t$) using the propagation of error formula (Özdemir and Kurudirek, 2009):

$$\Delta\left(\frac{\mu}{\rho}\right) = \frac{1}{t}\sqrt{\left(\frac{\Delta I_0}{I_0}\right)^2 + \left(\frac{\Delta I}{I}\right)^2 + \left(ln\frac{I_0}{I}\right)^2\left(\frac{\Delta t}{t}\right)^2} \tag{11}$$

where $t$ is the areal density (thickness that corresponds to the mass per unit area in g/cm$^2$), $\Delta I_0$, $\Delta I$ and $\Delta t$ are the errors in the intensities $I_o$, $I$ and areal density $t$ of the sample, respectively. Estimated error in experimental measurement is determined of approximately 7%. The Vernier calipers were used to determine the diameter of the collimator. Each measurement for the diameter of the collimator was repeated three times to minimize errors. The uncertainty of measurement was less than 1%.

## 4. Results and Discussion

The experimental and theoretical values of total atomic cross-sections of chemical compounds at different energies are given in Table 1. The experimental values were obtained by using the measurement data and the theoretical values were obtained by administering the mixture ratio from WinXCom program. These results of fairly close together, the experimental values were smaller than the theoretical value. This may be attributed to the higher $I_{measured}$ due to the buildup effect. It is seen that the primary photons leads to secondary scattering in the samples and increase of the total number of photons. Thus, the $I_{measured}$ (experimental) was found higher than the $I_{calculated}$ (theoretical) value calculated from Eq. (1). As a result of this evaluation, small differences between the theoretical and experimental values are formed. Moreover, this difference may have been due to experimental conditions and uncertainty such as oxidation of the chemical compounds, counting statistics and evaluation of the peak area. It is clearly seen that the values of all $\mu/\rho$ depends on the photon energy and decrease with the increasing photon energies.

The experimental and theoretical values of effective atomic numbers ($Z_{eff}$) and effective electron densities ($N_{eff}$) of given compounds are given in the Table 2 and Table 3 for different energies. It can be clearly seen that the variation in $Z_{eff}$ depends on the range of atomic numbers of elements from the Table 2. The $Z_{eff}$ values increased with the growth of the atomic number of the Z elements in the compound. The experimental and theoretical $Z_{eff}$ values for all compounds were found close to each other and the theoretical values were higher than the experimental values narrowly. In addition, it was observed that the values are consistent with the calculation used by Hine's expression but the mean atomic numbers are quite different than the $Z_{eff}$ values of the compounds in the Table 2 because of the fact that Hine's expression and mean atomic number equation do not depend on the growth of the atomic number of the Z elements of compounds or incident energy, while effective atomic number depends on both the two parameters. Unfortunately, $N_{eff}$ values in the Table 3 didn't show a significant change with the growth of the atomic number of the Z elements. Generally, it can be seen that the $Z_{eff}$ and $N_{eff}$ values of the compounds decreased with increasing incident photon energies from the Table 2. The photoelectric effect is dominant for energy range used in this study and photoelectric absorption cross section is proportional to $Z^{4–5}$.

It is well known that the $Z_{eff}$ value of a compound lies between lower and upper limit atomic numbers of its constituent elements. Also it was observed that the $Z_{eff}$ value depends on the

photon energy and buildup factor in previous studies (Mann et al., 2015; Kaewjang et al., 2014). In the present work, due to the use of low-energy photons region, the photoelectric absorption is the dominant process. The reason of the deviation for some values in this study is considered to be related to the absorption K-edge of the high Z-element (Elmahroug et al., 2015).

The photon forwarding in the material exposes many scattering, and the secondary radiations occur as a result of the scattering. After interacting with the substance, the total number of photons will cause increase. The amounts of the total photons include both uncollided photons from the source and additive from collided and secondary photons. The $I_{calculated}$ that is calculated using theoretical mass attenuation coefficient ($\mu/\rho$) indicates less number of photons reaching the detector. Therefore, buildup factor, the photon correction factor is needed to adapt the theoretical measurements with experimental measurements. The buildup, *B*, varies depending on the absorbing medium, photon energy and experimental design and it is always $B \geq 1$ (Babapour et al., 2008). The buildup factor in our study was found to be dependent on the photon energy and the collimator diameter. The buildup factor decreased with increasing energy, and in particular has taken fairly large value at low energy. Figs. 2-6 shows the values of buildup factors of compounds with collimator diameter for different energies, respectively. It can be seen that the value of buildup factor increases with the increase in the collimator diameter. This becomes clear as more numbers of multiple scattered photons are produced with the increase in collimator diameter, and simultaneously the exposure of the detector to the transmitted radiation increases, the probability of multiply scattered photons reaching the detector also increases. These results are in line with the previous findings of Singh et al. (2004, 2008).



## 5. Conclusions

In this work, the values of the total atomic cross-sections (barns/atom), the effective atomic number $Z_{eff}$ and the effective electron density $N_{eff}$ for five compounds were determined experimentally and theoretically in the different energies. The results obtained from this study indicate that these parameters depend on the incident photon energy as mentioned by previous authors and experimental and theoretical values of these parameters have good agreement in the energies; differences between experimental and theoretical values of effective atomic

numbers is under 4.87%. The values of buildup factors of compounds were determined in the same energies and the results show that the buildup factors depend on collimator diameter. Also, we showed the effect of collimator diameter on the buildup factors at different photon energies. The buildup factors obtained for all chemicals used in this study were found greater than 1 and maximum value in the low energies. It is clear that significant errors may be made if the buildup factor does not included to calculation radiation and shielding technology. The results of this work maybe useful to determine correctly the dose of radiation and shielding technology.

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Fig. 1.

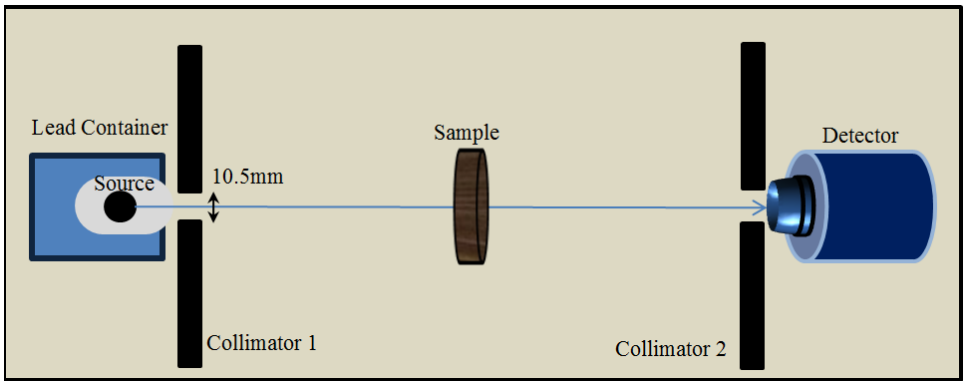


Fig. 2.

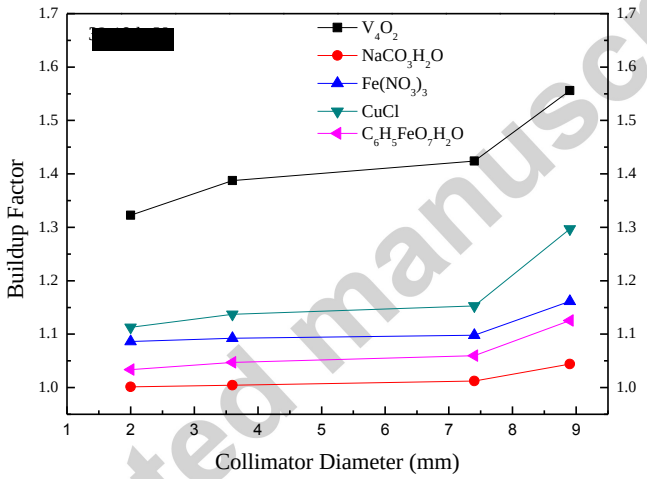


Fig. 3.

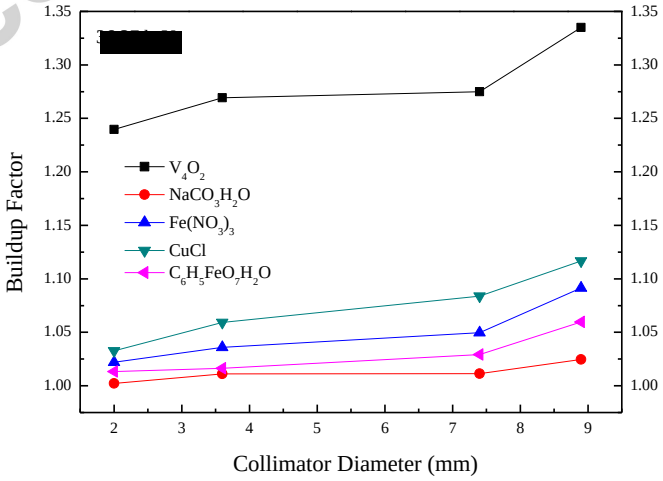

Fig. 4.

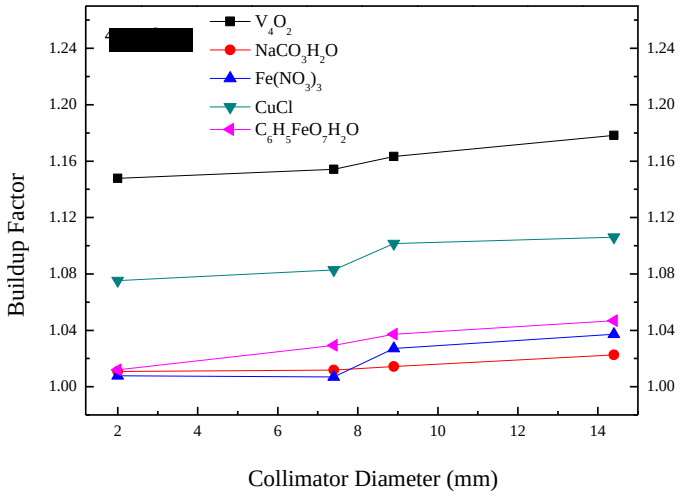


Fig. 5.

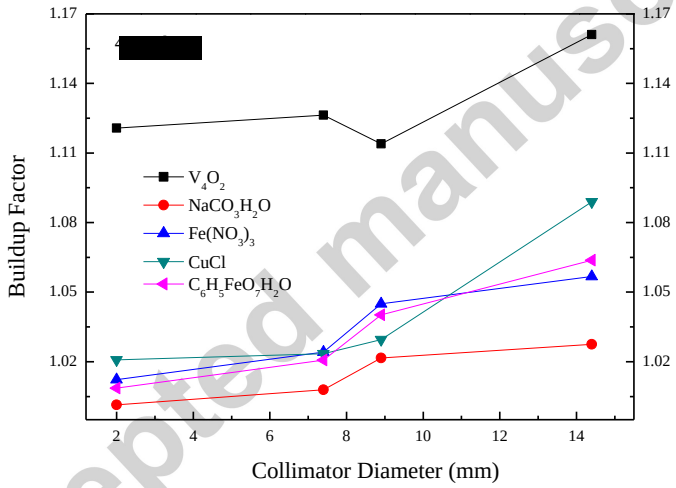


Fig. 6.

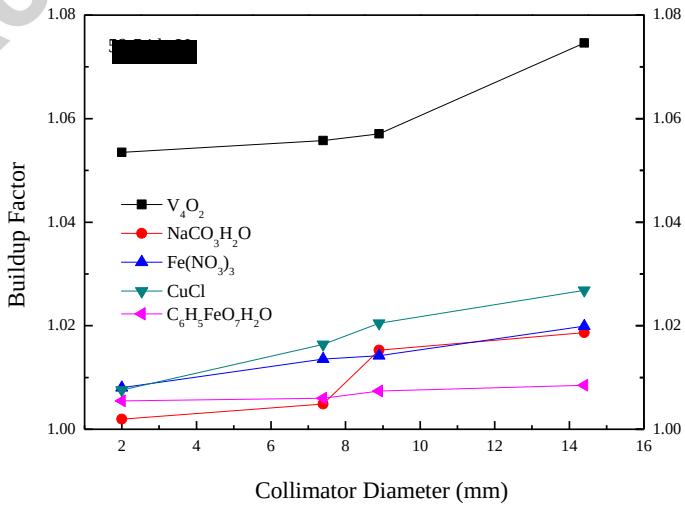

Table 1. Total atomic cross-sections of given compound at different energies (barns/atom).

| Compounds | 32.2 (keV) | | 36.4 (keV) | | 42.9 (keV) | | 48.8 (keV) | | 59.5 (keV) | |
|---|---|---|---|---|---|---|---|---|---|---|
| | Exp. | Theo. | Exp. | Theo. | Exp. | Theo. | Exp. | Theo. | Exp. | Theo. |
| $Fe(NO_3)_3$ | 47.50 ± 0.56 | 55.54 | 39.84 ± 2.64 | 40.45 | 25.77 ± 1.40 | 26.79 | 18.26 ± 0,84 | 20.06 | 12.21 ± 0,57 | 13.29 |
| $V_4O_2$ | 234.72 ± 11.85 | 259.98 | 164.10 ± 7.68 | 184.39 | 98.04 ± 2.63 | 116.51 | 71.34 ± 2.01 | 83.35 | 42.30 ± 0.55 | 50.20 |
| $NaCO_3.H_2O$ | 8.12 ±0.18 | 8.18 | 6.65 ± 0.57 | 6.77 | 5.35 ± 0.40 | 5.52 | 4.80 ± 0.27 | 4.89 | 4.11 ± 0.10 | 4.22 |
| CuCI | 510.52 ± 19.84 | 531.15 | 370.74 ± 4.70 | 376.99 | 223.23 ± 5.54 | 237.29 | 164.44 ± 5.98 | 168.39 | 97.43 ± 1.34 | 98.91 |
| $C_6H_5FeO_7.H_2O$ | 30.18 ± 2.05 | 33.02 | 22.99 ± 0.72 | 24.14 | 15.09 ± 0.92 | 16.12 | 11.42 ± 0.45 | 12.17 | 7.68 ± 0.62 | 8.16 |

Table 2. Experimental and theoretical values of effective atomic numbers ($Z_{eff}$) of given compounds at different energies.

| Compounds | 32.2 (keV) | | 36.4 (keV) | | 42.9 (keV) | | 48.8 (keV) | | 59.5 (keV) | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Exp. | Theo. | Exp. | Theo. | Exp. | Theo. | Exp. | Theo. | Exp. | Theo. | $Z_{eff}{}^{a}$ | $Z_{eff}{}_{b}$ |
| $Fe(NO_3)_3$ | 13.42 | 13.99 | 13.90 | 13.96 | 13.68 | 13.84 | 13.26 | 13.68 | 12.84 | 13.30 | 16.61 | 9.15 |
| $V_4O_2$ | 20.28 | 20.81 | 20.20 | 20.81 | 19.92 | 20.80 | 19.82 | 20.78 | 19.70 | 20.71 | 21.94 | 18 |
| $NaCO_3.H_2O$ | 7.67 | 7.70 | 7.50 | 7.57 | 7.24 | 7.37 | 7.13 | 7.21 | 6.84 | 6.97 | 8.69 | 6.38 |
| CuCI | 24.73 | 24.97 | 24.89 | 24.99 | 24.63 | 25.02 | 24.87 | 25.03 | 24.92 | 25.02 | 25.97 | 23 |
| $C_6H_5FeO_7.H_2O$ | 11.84 | 12.15 | 11.85 | 12.02 | 11.48 | 11.75 | 10.13 | 10.42 | 10.29 | 10.63 | 16.14 | 6.05 |

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

Table 3. Experimental and theoretical values of effective electron densities ($N_{eff}$) of given compounds at different energies (electrons/g*$10^{23}$).

| Compounds | 32.2 (keV) | | 36.4 (keV) | | 42.9 (keV) | | 48.8 (keV) | | 59.5 (keV) | |
|---|---|---|---|---|---|---|---|---|---|---|
| | Exp. | Theo. | Exp. | Theo. | Exp. | Theo. | Exp. | Theo. | Exp. | Theo. |
| $Fe(NO_3)_3$ | 4.34 | 4.53 | 4.50 | 4.52 | 4.43 | 4.48 | 4.29 | 4.43 | 4.15 | 4.30 |
| $V_4O_2$ | 3.11 | 3.19 | 3.09 | 3.19 | 3.05 | 3.19 | 3.04 | 3.18 | 3.02 | 3.17 |
| $NaCO_3.H_2O$ | 3.66 | 3.67 | 3.58 | 3.61 | 3.45 | 3.52 | 3.40 | 3.44 | 3.26 | 3.32 |
| CuCI | 3.01 | 3.04 | 3.03 | 3.04 | 3.00 | 3.04 | 3.03 | 3.04 | 3.03 | 3.04 |
| $C_6H_5FeO_7.H_2O$ | 5.96 | 6.12 | 5.97 | 6.06 | 5.78 | 5.92 | 5.10 | 5.25 | 5.18 | 5.35 |

## Highlights

- The photon interaction parameters have been measured for some chemical compounds.
- The effective atomic numbers ($Z_{eff}$) depends on the range of atomic numbers of elements in the compound.
- The buildup factor increased with the increasing collimator diameter.
- The effective atomic numbers ($Z_{eff}$) value depends on the photon energy and buildup factor.