

Determination of the effective atomic numbers of $\text{Fe}_x\text{Cu}_{1-x}$ binary ferroalloys using a nondestructive technique: Rayleigh-to-Compton scattering ratio

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Abstract: The aim of the present study is to determine effective atomic numbers (Z_{eff}) of $\text{Fe}_x\text{Cu}_{1-x}$ binary ferroalloys using scattering of 59.54 keV gamma photons at 130° ($x = 4.36\text{\AA}^{-1}$) scattering angle. For this purpose, the Rayleigh (R) and Compton (C) scattering intensities for the given alloys were measured using a monoenergetic beam of 59.54 keV gamma rays and the Rayleigh-to-Compton scattering ratio R/C was determined. Then the Z_{eff} values of these alloys were determined by interpolating the R/C of the material using the R/C data of adjacent elements in between the R/C of the alloys. For comparison, Z_{eff} s of alloys were also calculated using direct and interpolation methods in term of scattering R/C and attenuation of gamma photons. Good agreement in relative differences between Z_{eff} s (experimental and theoretical) was found (maximum of $\leq 3.3\%$ for total att.-direct method) for the chosen alloys and the Rayleigh-to-Compton scattering ratio was shown to be a complementary approach to determine the effective atomic number of binary ferroalloys. In addition, a polynomial equation between the experimental R/C values and the mass attenuation coefficients of alloys was developed to estimate the mass attenuation coefficient of different materials.

Key words: Effective atomic number, Rayleigh-to-Compton scattering ratio, alloy

1. Introduction

X and/or gamma rays are widely used in different fields such as industry, nuclear engineering, atomic and molecular physics, medicine, biology, and materials science because it is a nondestructive technique. Thus, when multielement materials are analyzed under X and/or gamma ray experiments, total photon attenuation and scattering cross-sections provide reliable knowledge to determine some properties of materials like effective atomic number (Z_{eff}), which represents the interaction of gamma photons with targets [1]. Determining the effective atomic number of a composite material can also help to calculate the energy absorption, absorbed dose, and build-up factor in a given medium when the gamma radiation beam is incident on it via well-established formulas.

Alloys, and particularly ferroalloys, are of extensive use in various applications of modern science such as electronic technology and industrial applications in heavy industry and they have been investigated in terms of radiation application to date. It is quite reasonable to define a Z_{eff} to describe the properties of an alloy in terms of an equivalent element since the Z_{eff} of an alloy could be a very useful parameter for many fields of radiation-related applications. The literature contains many studies regarding the calculation of Z_{eff} of alloys using attenuation or scattering methods. Transmission condition or geometry, based on attenuation of

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photons, has been widely used to determine Z_{eff} of alloys [2–13]. On the other hand, the Rayleigh-to-Compton scattering ratio is a nondestructive technique that provides useful data about the crystal structure and physical properties of materials and is based on the measurement of Rayleigh and Compton scattered photons emerging from the sample of composite material in a suitable source–sample–detector arrangement. It has also been used to obtain the Z_{eff} of alloys in different energies and scattering angles, although not as extensively as for transmission [14–17].

In the present study, Fe_xCu_{1-x} ferroalloys have been investigated with respect to the Z_{eff} for scattering of 59.54 keV gamma rays at a scattering angle of 130° . An Am-241 annular monoenergetic radioactive source emitting 59.54 keV photons has been used in this work at 130° scattering angle, i.e. $x = 4.36\text{\AA}^{-1}$ momentum transfer. Therefore, the scattering angle has been kept constant but the concentrations of elements constituting the alloy have been varied. In addition, an alternative method, namely a direct method, has been used to obtain Z_{eff} for scattering and attenuation of gamma rays to compare with the experimental results determined by the interpolation procedure. The Z_{eff} s of alloys have also been obtained by using the interpolation method for photon attenuation to compare with the experimental results. The measured values of the effective atomic numbers have been then compared with the theoretical values calculated using heterogeneous radiation sources via the Auto- Z_{eff} program. On the other hand, the experimental R/C ratios of alloys were plotted as a function of theoretical mass attenuation coefficients obtained from WinXCom [18,19] and fitted to a polynomial equation. The experimental R/C values of alloys were then used to calculate the mass attenuation coefficient of a different alloy, $Fe_{0.5}Ni_{0.5}$, using this fit equation and agreement with WinXCom and this work was found to be quite satisfactory.

2. Theoretical procedure

The Rayleigh-to-Compton scattered photon intensity ratios can be used to determine Z_{eff} of the chosen alloys. At a scattering angle of θ , the numbers of Rayleigh (N_R) and Compton (N_C) photons are directly proportional to the areas of their respective peaks in the measured spectrum. The scattered intensity ratios can be theoretically calculated using the following equation:

$$R/C(xZ) = \frac{N_R}{N_C} = \frac{N_0\eta_{at} [d\sigma/d\Omega]_R \Delta\Omega V \epsilon A_R}{N_0\eta_{at} [d\sigma/d\Omega]_C \Delta\Omega V \epsilon A_C}, \quad (1)$$

where N_0 is the initial fluence, η_{at} is the number of atoms per volume of sample, $\Delta\Omega$ is the solid angle subtended by the detector, ϵ is the detector efficiency, A_R and A_C are the self-attenuation correction factors for each scattering process, and $[d\sigma/d\Omega]_R$ and $[d\sigma/d\Omega]_C$ are differential cross-sections [20]. If N_R and N_C are measured in the same geometric conditions of irradiation and detection, then N_0 , η_{at} , $\Delta\Omega$, V , and ϵ become constant values. Eq. (1) can be rewritten considering the Thomson, $[d\sigma/d\Omega]_{Th}$, and the Klein–Nishina, $[d\sigma/d\Omega]_{KN}$, differential cross-sections, the atomic form factor F , and the incoherent scattering function S , [21], which are dependent on the momentum transfer ($x = \sin(\frac{\theta}{2})/\lambda$), as follows:

$$R/C(xZ) = \frac{[d\sigma/d\Omega]_R A_R}{[d\sigma/d\Omega]_C A_C} = \frac{[(\frac{d\sigma}{d\Omega})_{Th} F^2(x, Z)] A_R}{[(\frac{d\sigma}{d\Omega})_{KN} S(x, Z)] A_C}. \quad (2)$$

The self-attenuation factors for the Rayleigh and Compton intensities can be calculated as

$$A_R = 1/V \int v e^{-[\mu(E_0)L_i + \mu(E_0)L_s]} dV \quad \text{and} \quad A_C = 1/V \int v e^{-[\mu(E_0)L_i + \mu(E_C)L_s]} dV, \quad \text{respectively.} \quad \mu(E_0) \quad \text{and} \quad \mu(E_C) \quad \text{are}$$

the linear attenuation coefficients for the incident (E_0) and Compton (E_C) scattered energies, L_i is the distance from the surface of the sample to the elemental scattering volume (dV) and L_S from this element to the surface of the sample, in the direction of the detector [20,22]. For a fixed experimental condition, when a small energy shift occurs between Rayleigh and Compton scattering the ratio of A_R and A_C becomes 1 ($A_R/A_C \approx 1$) [20,22,23]. In this case, Eq. (2) can be reduced. However, in the present work there is an energy shift between Rayleigh and Compton scattering, and thus the condition $A_R/A_C \approx 1$ can not be satisfied. Therefore, self-attenuation correction factors were calculated and properly used in Eqs. (1) and (2). If molecular weight and elemental composition fractions of the given compounds or composite materials are known, R/C is calculated by weighting the atomic percentages α_j^{at} of elements as in the following equation:

$$R/C = \left[\frac{(d\sigma/d\Omega)_{Th}}{(d\sigma/d\Omega)_{KN}} \right] x \left[\frac{\sum_{j=1}^n \alpha_j^{at} [F(q, Z_j)]^2}{\sum_{j=1}^n \alpha_j^{at} S(q, Z_j)} \right] x \frac{A_R}{A_C}, \quad (3)$$

where α_j^{at} is defined by weight percentage w_j and atomic mass A_j of the j th element as [23]:

$$\alpha_j^{at} = \frac{(w_j/A_j)}{\sum_j (w_j/A_j)}. \quad (4)$$

After determination of R , Z_{eff} can be calculated using a well-known interpolation procedure shown below, at the same as scattering angle and energy [14,24,25]:

$$Z_{eff} = \frac{Z_1(\log R_2 - \log R) + Z_2(\log R - \log R_1)}{\log R_2 - \log R_1}, \quad (5)$$

where R_1 and R_2 are the R/C ratios in between the R of the material and Z_1 and Z_2 are atomic numbers of the elements corresponding to the ratios $(R/C)R_1$ and R_2 , respectively. This procedure was also used for total photon attenuation based on the total atomic cross-sections ($\sigma_a = \frac{(\mu/\rho)_{comp}}{N_A \sum_i w_i/A_i} (b/atom)$) obtained by dividing the mass attenuation coefficients of the alloys. Here, $(\mu/\rho)_{comp}$ is the mass attenuation coefficient of the alloy, N_A is the Avogadro constant, w_i is the fraction by weight of element i , and A_i is the atomic weight of the i th element.

The Z_{eff} s of alloys were calculated by using a direct method (for total photon attenuation) utilizing the following formula [26]:

$$Z_{eff} = \frac{\sum_i f_i A_i \left(\frac{\mu}{\rho}\right)_i}{\sum_j f_j \frac{A_j}{Z_j} \left(\frac{\mu}{\rho}\right)_j} \quad (6)$$

where f_i is the fraction by mole of each constituent element provided $\sum_i f_i = 1$, Z_j is the atomic number, and $(\mu/\rho)_i$ is the mass attenuation coefficient from WinXCom [18,19]. This method has been also used for determining Z_{eff} s of alloys using the Rayleigh-to-Compton scattering ratio (R) of each constituent element instead of the mass attenuation coefficient (μ/ρ) of element. On the other hand, in order to determine Z_{eff} s of alloys the Auto- Z_{eff} program (based on photon attenuation) was used for multienergetic photons emitted through heterogeneous radiation sources such as Pd-103, Tc-99, Ra-226, I-131, Ir-192, Co-60, 30 kVp, 40 kVp, 50 kVp (Intrabeam, Carl Zeiss Meditec), and 6 MV (Mohan-6 MV) sources as well [27].

3. Experimental procedure

The alloys used in this study were in fine powder form. The samples were pelleted using a pressing machine to form tablet samples prior to measurements. The pellets were pressed at 8 t/cm^2 in a Spex hydraulic press. Table 1 lists the weight fractions, thicknesses, and densities of the chosen alloys. The experimental setup is shown in Figure 1. In order to obtain the Rayleigh and Compton scattering intensities, all alloys were excited using 59.54 keV gamma rays emitted from an Am-241 annular radioactive source (100 mCi). The scattering peaks (Rayleigh and Compton peaks) emitted from the targets were detected by a Si(Li) detector (effective area 12 mm^2 , thickness 3 mm, Be window thickness 0.025 mm, Canberra SL30165, with energy resolution of 165 eV at 5.9 keV) and analyzed using Genie-2000 software. The data were collected into 4096 channels of the MCA and further analyzed by the demo version of Origin 8 software. A peak fitting procedure was applied in Origin software based on fitting the data to the appropriate Gaussian function. The counting time for each measurement was kept at 10,800 s in order to reduce the statistical uncertainties arising from counts, both for the Rayleigh and Compton peaks. The scattering angle is $\theta = 130^\circ$ from sample to excitation source and excitation source to detector distances are 10 cm.

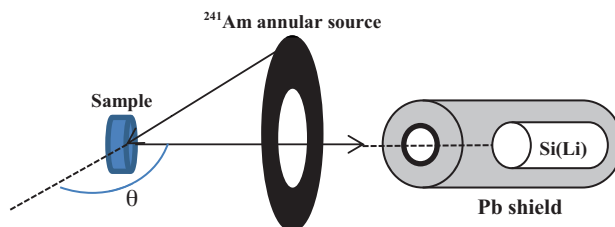


Figure 1. The experimental setup (including scattering angle, incident, and scattered beam).

Table 1. The weight fractions, thicknesses and densities of the given alloys.

	Alloy	Density (g/cm^3)	Thickness (cm)
A 1	$\text{Fe}_{0.1}\text{Cu}_{0.9}$	8.84	0.22
A 2	$\text{Fe}_{0.2}\text{Cu}_{0.8}$	8.72	0.24
A 3	$\text{Fe}_{0.3}\text{Cu}_{0.7}$	8.60	0.26
A 4	$\text{Fe}_{0.4}\text{Cu}_{0.6}$	8.49	0.29
A 5	$\text{Fe}_{0.5}\text{Cu}_{0.5}$	8.37	0.30
A 6	$\text{Fe}_{0.6}\text{Cu}_{0.4}$	8.27	0.30
A 7	$\text{Fe}_{0.7}\text{Cu}_{0.3}$	8.16	0.32
A 8	$\text{Fe}_{0.8}\text{Cu}_{0.2}$	8.06	0.33
A 9	$\text{Fe}_{0.9}\text{Cu}_{0.1}$	7.96	0.35

4. Results and discussion

The various sources of error in the measurements are due to counting statistics, mass thickness determination, evaluation of photopeak areas by peak fitting, etc. The error in the counting statistics was reduced to $<1\%$ by collecting at least 10^4 counts under the Compton and Rayleigh peaks. The error associated in evaluating the area of the scattered peak by the peak fitting routine was less than 3%. The uncertainty in estimating mass thickness of the targets was about 1%. The uncertainty in the scattering angle was approximately 1%. By using the R/C ratio, the sensitivity to sample thickness variation and positioning can be reduced, thus leading to lower experimental uncertainties [23]. In addition, the uncertainties in $F(xZ)$ and $S(xZ)$ were found to be less than 1% [21].

In this work, the Rayleigh (R) and Compton (C) scattering intensities for $\text{Fe}_x\text{Cu}_{1-x}$ binary ferroalloys have been measured using a monoenergetic beam of 59.54 keV gamma rays and a scattering angle of 130° (momentum transfer $x = 4.36\text{\AA}^{-1}$), and good agreement has been obtained between experimental and theoretical R/C values (via Eqs. (1)–(3)) of alloys ($<4\%$). The scattered spectra of some alloys are given in Figure 2 as an example and the Compton scattering intensity increases as the concentration of the lowest Z element in the alloy, i.e. Fe, increases, as seen in the figure. In contrast, it decreases when the highest Z element is more abundant in the alloy. This is because the Compton scattering is predominant for materials of low and medium Z elements and the Rayleigh scattering has a Z^2 dependence, which makes it predominant for high Z elements.

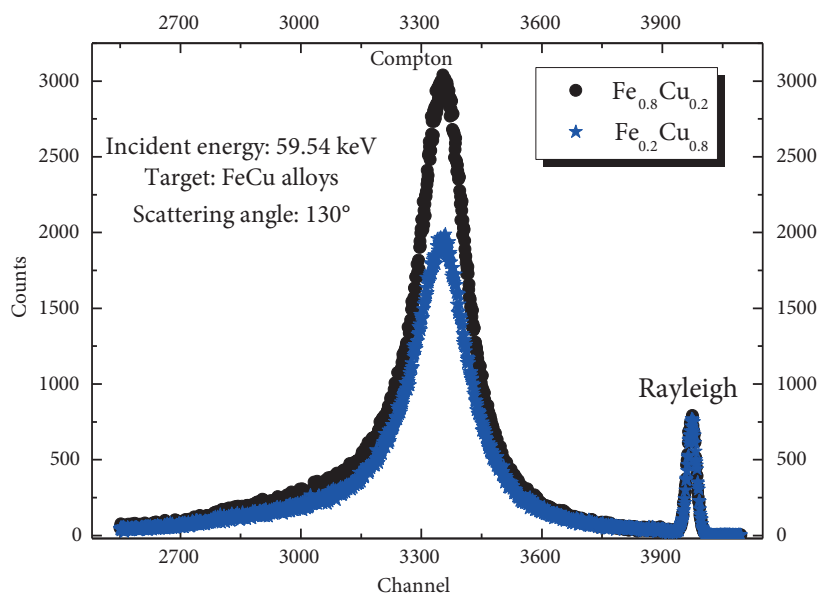


Figure 2. A typical observed spectrum at 130° from two different targets when irradiated by 59.54 keV incident photon energy.

In the present study, the experimental R/C of the material and the data of adjacent elements in between the R/C of the material were used to interpolate the Z_{eff} of the chosen alloys. On the other hand, alternative approaches have been employed to show its availability for estimating Z_{eff} for scattering and attenuating of gamma rays. For this purpose the Z_{eff} s of the chosen alloys were calculated for total photon attenuation and scattering of gamma rays using the direct and interpolation methods as given in Table 2. The highest values of Z_{eff} were obtained for attenuation of gamma rays for the direct method and it was observed that Z_{eff} increases as the weight fraction of high Z elements increases in the alloy in general for all results, as shown in Table 2. Figure 3 shows the relative difference (%) between the experimental and theoretical values of Z_{eff} . The maximum relative differences in Z_{eff} were observed (for $\text{Fe}_{0.6}\text{Cu}_{0.4}$ alloy) as 2.89% for scattering (direct), 3.02% for total attenuation (interpolation), and 3.24% for total attenuation (direct). Since excellent agreement with highest relative difference of 2.89% has been observed between the experimental and direct methods, the direct method was found to be an alternative and practical method for calculation of Z_{eff} for scattering of gamma rays. The highest relative difference in Z_{eff} between direct and interpolation methods was also observed (for $\text{Fe}_{0.2}\text{Cu}_{0.8}$) as 1.28% with respect to total photon attenuation, and thus the Z_{eff} for the methods showed a very good agreement. On the other hand, it has to be noted that the total attenuation has higher relative differences

of Z_{eff} according to experimental results than the scattering ratio (R/C) method. The total attenuation refers to the sum of attenuations due to each partial interaction process, namely the photoelectric effect, Compton scattering, and pair production, and it has been somewhat problematic to arrange the optimum distance between source and sample and sample and detector to avoid the divergence of the incident beam, which may cause buildup of photons. Meanwhile, in the R/C method based on Rayleigh and Compton scattering intensities, the sensitivity to sample thickness variation and positioning can be reduced, thus leading to lower experimental uncertainties. This could be the reason for observing differences in Z_{eff} between scattering and attenuation of photons. Besides, for scattering of gamma rays, Z_{eff} values of the chosen alloys have been calculated for photon radiation sources of heterogeneous in energy using Auto- Z_{eff} . Results are given in Table 3 for heterogeneous radiation sources. It can be clearly seen that the alloys with higher Cu content possess the higher values of Z_{eff} for all heterogeneous radiation sources. Figure 4 shows the relative difference (%) between the experimental and theoretical values of Z_{eff} for photon radiation sources and the relative differences were found to be $\leq 7.09\%$ between the experimental and theoretical values of Z_{eff} .

Table 2. Z_{eff} of the chosen alloys for different methods for photon scattering and attenuation.

Alloy	Interpolation Scattering (Exp)	Total Att. (Th)	Direct Scattering (Th)	Total att. (Th)
Fe _{0.1} Cu _{0.9}	28.67	28.56	28.71	28.74
Fe _{0.2} Cu _{0.8}	28.35	28.11	28.42	28.48
Fe _{0.3} Cu _{0.7}	28.01	27.86	28.12	28.20
Fe _{0.4} Cu _{0.6}	27.62	27.67	27.83	27.92
Fe _{0.5} Cu _{0.5}	27.03	27.47	27.53	27.63
Fe _{0.6} Cu _{0.4}	26.44	27.26	27.23	27.33
Fe _{0.7} Cu _{0.3}	26.35	27.05	26.92	27.01
Fe _{0.8} Cu _{0.2}	26.21	26.74	26.62	26.69
Fe _{0.9} Cu _{0.1}	26.07	26.41	26.31	26.35

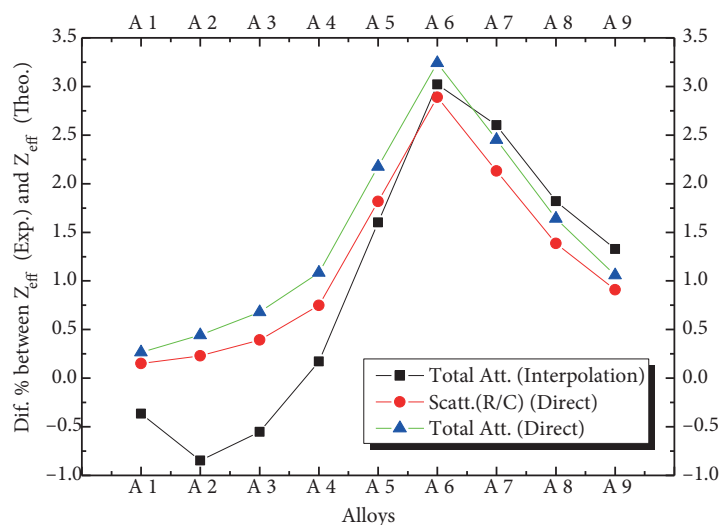


Figure 3. Differences (%) between experimental and theoretical values obtained using different interactions (scattering-attenuation) and methods.

Table 3. Z_{eff} of the chosen alloys for various photon sources heterogeneous in energy (multienergetic).

	^{192}Ir	^{60}Co	^{131}I	^{103}Pd	^{226}Ra	^{99}Tc	30 kVp	40 kVp	50 kVp	6 MV
A 1	28.16	28.62	28.50	26.77	28.69	27.80	27.01	27.22	27.49	28.66
A 2	27.85	28.33	28.19	26.50	28.38	27.51	26.73	26.94	27.20	28.34
A 3	27.55	28.02	27.88	26.23	28.08	27.23	26.46	26.66	26.91	28.02
A 4	27.25	27.71	27.58	25.96	27.78	26.94	26.21	26.37	26.62	27.71
A 5	26.95	27.40	27.28	25.69	27.48	26.64	25.80	26.08	26.33	27.41
A 6	26.66	27.11	26.98	25.43	27.18	26.36	25.43	25.80	26.05	27.12
A 7	26.37	26.82	26.69	25.16	26.89	26.07	25.16	25.52	25.77	26.83
A 8	26.09	26.54	26.40	24.88	26.58	25.77	24.88	25.23	25.47	26.54
A 9	25.81	26.27	26.12	24.62	26.29	25.48	24.51	24.95	25.19	26.27

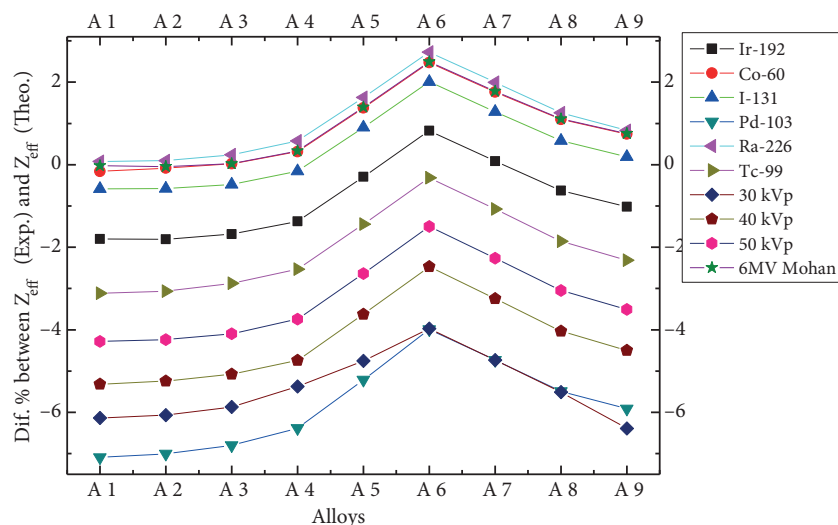


Figure 4. Differences (%) between experimental and theoretical values obtained from Auto- Z_{eff} for various photon sources heterogeneous in energy (multienergetic).

Generally transmission measurement is used to determine the mass attenuation coefficients of materials. However, a different method is to use the Rayleigh-to-Compton scattering ratio versus the mass attenuation coefficient curve obtained with a fitting program to determine the mass attenuation coefficients of materials [28]. For this purpose, the experimental R/C values of alloys obtained by 59.54 keV gamma ray incident energy were adjusted to a polynomial function of the 5th order by the Origin 8 curve fitting software against the theoretical mass attenuation coefficients of alloys as shown Figure 5. This fit equation was then used to calculate the mass attenuation coefficient of a different alloy, $\text{Fe}_{0.5}\text{Ni}_{0.5}$, which has 0.0545 Rayleigh-to-Compton scattering ratio (R/C) as determined in the same experimental conditions, i.e. 59.54 keV gamma photons and scattering angle of 130° . The mass attenuation coefficient of the alloy was then calculated using this fit equation as $1.285 \text{ cm}^2 \text{ g}^{-1}$ and the relative difference between WinXCom and this work was found to be 7.29%. Therefore, it is very important to note here that this fit equation shows quite agreement between the experimental and theoretical values of the mass attenuation coefficient under the relative difference of 7.29%.

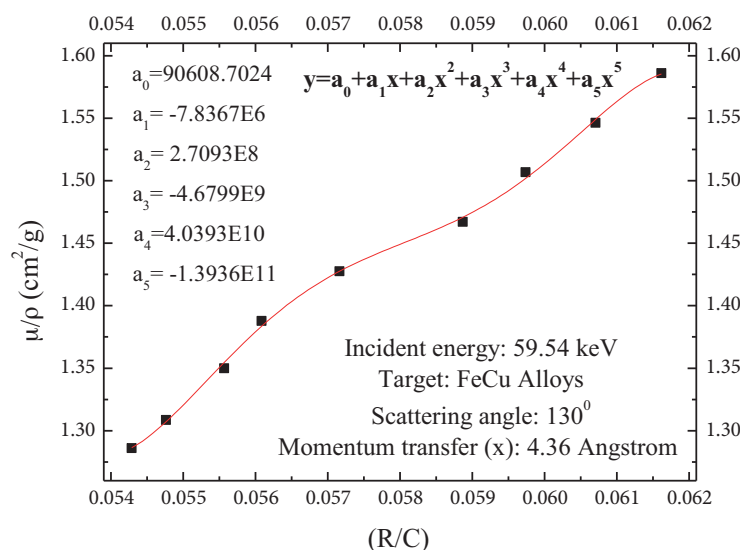


Figure 5. Rayleigh-to-Compton scattering ratio (R/C) versus mass attenuation coefficient (μ/ρ) obtained by WinX-Com for the 0.054–0.062 R/C range for 59.54 keV.

5. Conclusion

In the present work, the effective atomic numbers (Z_{eff}) of Fe_xCu_{1-x} ferroalloys were experimentally obtained for scattering of 59.54 keV γ -rays at an angle of 130° and 4.36\AA^{-1} momentum transfer. Z_{eff} s of alloys were also calculated using direct and interpolation methods in term of scattering (R/C) and attenuation of gamma photons. The agreement was found to be quite satisfactory between the experimental and theoretical values of Z_{eff} . The obtained Z_{eff} s for photon scattering were then compared to the Z_{eff} s for total photon attenuation obtained using the Auto- Z_{eff} program for various photon sources and the results showed good agreement (relative differences of $\leq 7.09\%$). The results led to the conclusion that for determining the Z_{eff} of materials, both the used method and the type of radiation interaction process, i.e. scattering or attenuation, even for the same type of radiation, should be taken into account and thus cannot be considered as a true constant. On the other hand, the Rayleigh-to-Compton scattering ratio was shown to be a complementary approach to obtain the mass attenuation coefficient and effective atomic number of ferroalloys. In addition, the fit equation developed in this study can be used to calculate the mass attenuation coefficient of known and unknown materials in this R/C region, especially for alloys between Fe and Cu such as FeCo, FeNi, CoNi, or FeCoNi via the relative difference of $\leq 7.29\%$.

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