

Mass Attenuation Studies of Rare Earth Elements at K-Absorption Edge by Attenuation of Compton Scattered Gamma Rays.

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Abstract

The mass attenuation coefficient of rare earth elements Eu, Gd, Tb and Er were determined at the K-shell absorption edge by the attenuation of the Compton scattering peak. The gamma rays of energy 59.54 keV from Am²⁴¹ source were scattered by aluminum target to get a broad span of the Compton scattered peak. After attenuation of Compton peak by absorber was detected using an high resolution intrinsic HPGe detector. A complex histogram obtained was unfolded to best iterative fit by using a Monte Carlo simulation (MCPNX) and Maximum Likelihood Extreme Maximum (ML-EM) method.

The measured experimental values of the mass attenuation coefficient $(\mu/p)_k$ at lower and higher K-edge energy were compared with theoretical values shows good agreement reported by Hubble (1995) NISTIR 5632, XCOM Table.

Keywords: mass attenuation coefficient, k-edge absorption, jump ratio.

Introduction

The accurate measurement of the mass attenuation coefficient near the k-absorption edge is important in numbers of fields of **medical dosimetry, radiation Physics, material sciences, and the nuclear industry.**

Generally energy dispersive X-ray fluorescence (EDXRF) and Compton scattered photons and photons/X-rays sources are used for attenuation purposes: However, the Compton peak attenuation method is preferred for studying k-shell jumps. In this study, Compton-scattered photons from the Am²⁴¹ radionuclide are used.

The X-ray jump by attenuation of Er was studied by Alejandro P. Ayla (1996) using a parameterized attenuation method with a least squares approach, achieving an error of due to absorber thickness.

Masaya Tamura (2002) used parametric X-rays (PXR) to estimate the mass attenuation coefficient near the absorption edge for rare earth elements. However, the spectrum available in LINAC to generate PXR degraded the counting statistics and resolution.

Recap Polat (2004) measured K-shell absorption jump ratios in rare earth elements based on simultaneous measurements of fluorescence radiation and scattered radiation. Though the method is rapid, it has large variation with theoretical values.

Budak (2004) measures x-ray jump factors for rare earth elements by attenuating the Compton peak using a transmission geometry with an annular source.

Nacati Kaya et al. (2007) derived K-shell absorption jump ratios by conducting attenuation measurements using an energy dispersive X-ray fluorescence spectrometer.

K.K. Abdulla (2008) studied the attenuation coefficient by attenuating the Compton peak near k-absorption edges using pellets of rare earth oxide targets.

A.S. Bennal (2007) proposed an alternative method to determine K-shell absorption jump factors by adopting 2π geometrical configuration, considering the self-attenuation factor and closer distances between the detector, target, and gamma source.

Raul T. Mainardi (2010) comments on methodologies and geometric setups used in previous works on the measurement of K-absorption jump factors and ratios.

Recap Polat (2013) and Baltej singh (2011) used the EDXRF technique to estimate k-shell absorption jump factors for lanthanide elements without thin foils or point sources.

K.M. Niranjana (2013) determined k-shell photoelectric parameters around k-shell binding energies ranging from 8 - 52 keV by Compton scattering by silver foil.

M.R. Kacal (2014) determined K-shell absorption jump factors and jump ratios for 3d transition metals using EDXRF techniques.

Gupta (2014) studied the effect of the chemical environment on k-shell absorption jump ratios and edge parameters.

Ferdi Akman (2015) determined K-shell absorption jump ratios using two different methods: EDXRF and Compton-scattered photon attenuation.

Sangam et al. (2019) discussed the Maximum Likelihood Expectation Maximization (MLEM) deconvolution method to unfold doublet spectra, gives accurate net counts by considering the response function of a NaI(Tl) detector.

In the present study, the k-absorption jump factor and jump ratio for rare earth elements europium (Eu), Gadolinium (Gd), Terbium (Tb) and Erbium (Er) were measured by attenuating Compton-scattered photons from the gamma source (Am^{241}) of energy 59.54 keV near the absorption edge. The absorption at good geometry setup, the unfold spectra with best fit in the region of interest (Sangam, 2019) improved the measurement accuracy at both sides of k-shell.

Experimental Setup

The schematic experimental setup adopted in the present work is shown in Figure 1. When a gamma rays of energy E_i by a Al target, produces inelastic scattered photons a high-purity aluminum target to produce scattering of photons of energy E_e , then the energy of the scattered photon E_e is related to the scattering angle θ between the incident and scattered photons as given by:

$$E_e = \frac{E_i}{1 + \left(\frac{E_i}{m_0 c^2}\right)(1 - \cos\theta)} \quad (1)$$

A collimators of having inner diameter of 4 mm is used to focus the beam and to minimize the internal scattering of photons. A perfect fabricated sheet stand is prepared to hold the source, target, and detector in a vertical goniometer geometry, ensuring that the gamma rays are perfectly aligned to the 25.2 mm beryllium window of the HPGe detector head. The entire assembly is shielded in lead castle to minimize background counts.

The scattered photons are detected using a Canberra made HPGe detector, which is coupled to a 16k multi-channel analyzer (MCA). The detector head is vertically positioned in a liquid nitrogen (LN₂) dewar.

In the present study, the K-absorption jump factor and jump ratio for rare earth elements Eu, Gd, Tb and Er were measured by attenuating the Compton scattered photons from the 59.5 keV energy near the absorption edge (Table-1). The absorption of a good geometry setup and the unfolding of spectra in the k-edge region of interest (Sangam, 2019).

For oxide based rare earth elements in the form of pellet absorber, the mass attenuation coefficient is calculated according by Bragg's mixture rule given by

$$\frac{\mu}{\rho} = \sum w_i \left(\frac{\mu}{\rho} \right)_i \quad (2)$$

where w_i and $(\mu/\rho)_i$ are the weight fraction and mass attenuation coefficient of the i_{th} constituent element respectively.

Table 1. Scattering data of absorber elements used in experiment.

Atomic No.	Name of absorber element	Scattering angle (°)	k-edge Energy	Compton Photon Energy (keV)
63	Eu	162°	48.510	57.287
64	Gd	126°	50.239	57.140
65	Tb	103°	51.995	56.988
68	Er	48°	57.486	56.496

Method of Computation

When a beam of photon energy of E is scattered by a aluminum target and when attenuated by a absorber having mass thickness ($x=\rho t$) gives scattered spectra $S(E)$ detected by a multichannel analyzer can be written in integral form as:

$$M(n) = \int_{E_0}^E S(E) \cdot (n, E) dE \quad (3)$$

where $R(n, E)$ is the resolution of detector in lower (E_0) and upper (E_1) energy of Compton scattered peak. The actual values uses in analysis are estimated for individual absorbers.

When the observer foil at mass thickness x is placed in collimated Compton spectra and detector, then the attenuation coefficient for each channel is given by,

$$A(E, x) = \exp[-\mu(E) \cdot x] \quad (4)$$

Where (E) is mass attenuation coefficient of observer of mass thickness (x) and $A(E, x)$ is the intensity of photons at respective energy channels.

The attenuated histogram is therefore expressed in terms of integration as,

$$(n, x) = \int_{E_0}^{E_1} (E) \cdot R(n, E) \cdot A(E, x) \cdot dE \quad (5)$$

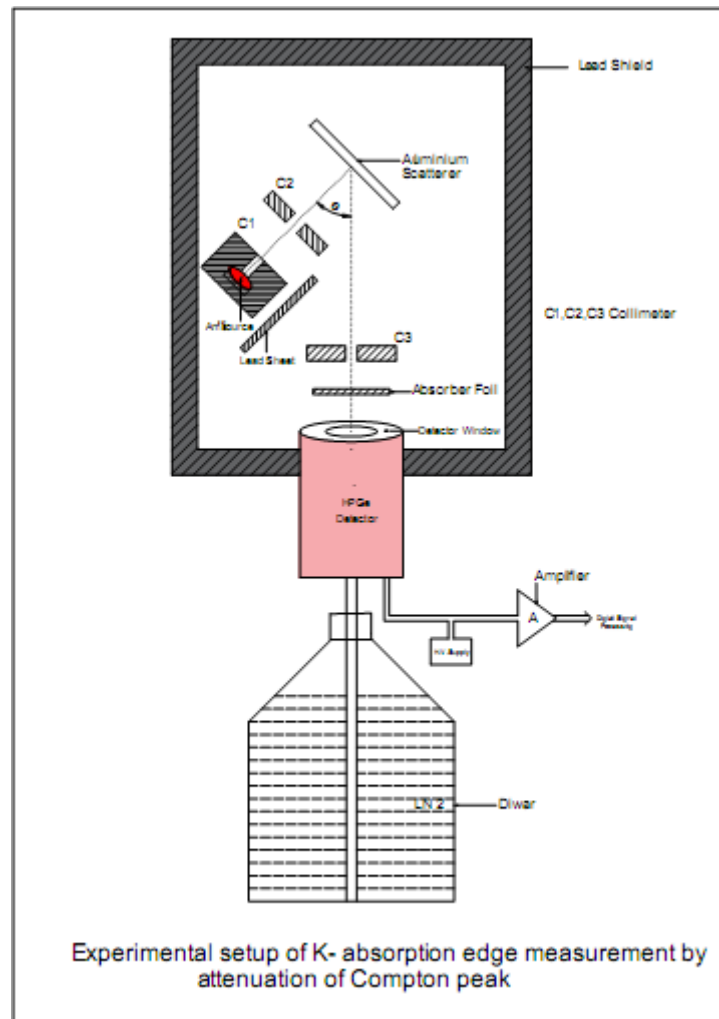


Fig. 1. : Experimental geometry setup of k-absorption edge measurement.

The (n, x) can be expressed as

$$\begin{bmatrix} M_1 \\ \vdots \\ M_2 \end{bmatrix} = \begin{bmatrix} R_{11} & \cdot & \cdot & R_{1j} \\ \cdot & \cdot & \cdot & \cdot \\ \cdot & \cdot & \cdot & \cdot \\ R_{i1} & \cdot & \cdot & R_{ij} \end{bmatrix} \begin{bmatrix} S_1 \\ \vdots \\ S_2 \end{bmatrix}$$

$$M_i(E) = \sum_{i,j=1}^n R_{ij} S_j$$

Where M_i is the true detector counts, R_{ij} is the response function of detector and S_j is discretized incident function.

Multiplying eqⁿ (5) by inverse of matrix R_{ij} gives

$$S = R^{-1}M \quad (7)$$

As a resolution (response) function matrix has large span of energy channels arrives number of problem like geometrical factor, negative results and quantization error in signal conditioner etc., contributing to the photo-peak width. The spectra $M_i(E)$ attenuated (Graph -1) above bellow and above k-edge the channels are folded with

counts caused by wider full width half maximum (FWHM) peak obtained and counting statistics.

$$A(E, x) = \exp[-(\mu_1(E_k) \cdot x) \times \left[1 - x \left(\frac{d\mu_1}{dE} \right)_{E_k} \cdot (E - E_k) - \frac{x^2}{2} \left(\frac{d^2\mu_1}{dE^2} \right)_{E_k} \cdot (E - E_k)^2 \right]] \quad (8)$$

Thus for $E < E_k$

$$A(E, x) = a + b \cdot (E_k - E) + f \cdot (E_k - E)^2$$

For $E > E_k$

$$A(E, x) = c + d \cdot (E_k - E) + g \cdot (E - E_k)^2$$

By putting the value of known energy E & E_k , the equation (4) it becomes,

$$M(n, x) = \int_{E_0}^{E_k} [a + b(E_k - E) + f(E_k - E)^2] \cdot S(E) \cdot R(n, E) \cdot dE + \int_{E_0}^{E^1} [c + d(E - E_k) + g(E - E_k)^2] S(E) \cdot R(E) \cdot dE$$

$$M(n, x) = aM_1 + bM_2 + cM_3 + dM_4 + eM_5 + fM_6 \quad (9)$$

The integral are evaluated using the MCNPX software and ML-EM deconvolution procedure L.B. Lucy (1974) and Sangam(2019) absorption jump ratio and jump factors are the measures at amount of photons absorbed by the atomic level. The measured mass attenuation coefficient obtained graphically (Graph-2) is used to calculate the absorption jump ratio and jump factors of elements.

$$\text{Jump ratio } (J_k) = \frac{\ln(c)}{\ln(a)} \quad \text{or} \quad = \frac{[(\mu/\rho)_h]}{[(\mu/\rho)_l]} \quad (10)$$

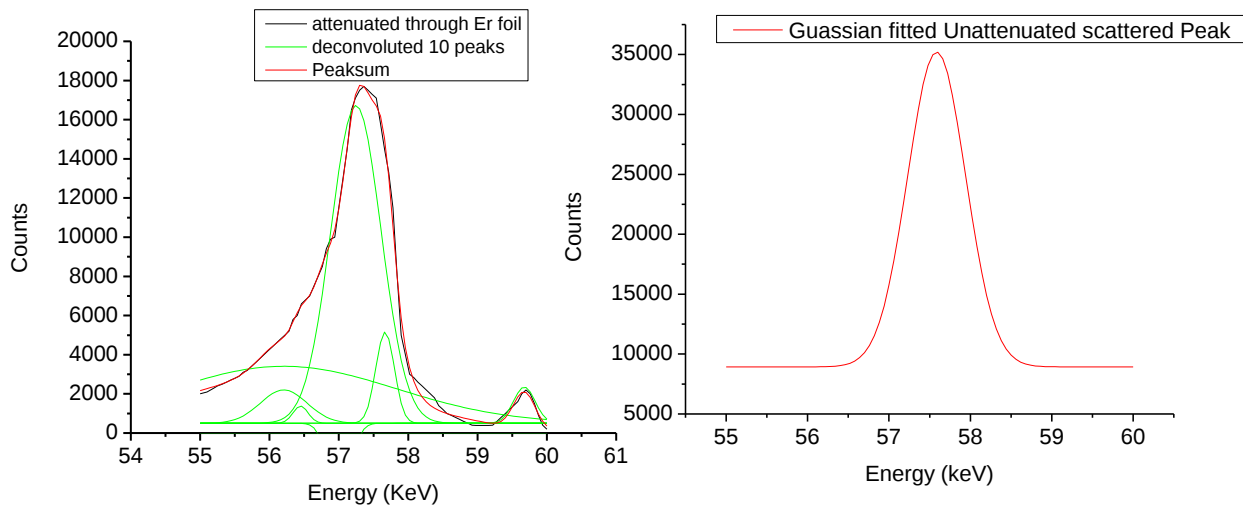
$$\text{and jump factor } (J_f) = \frac{(\mu/\rho)_h - (\mu/\rho)_l}{(\mu/\rho)_h} \quad (11)$$

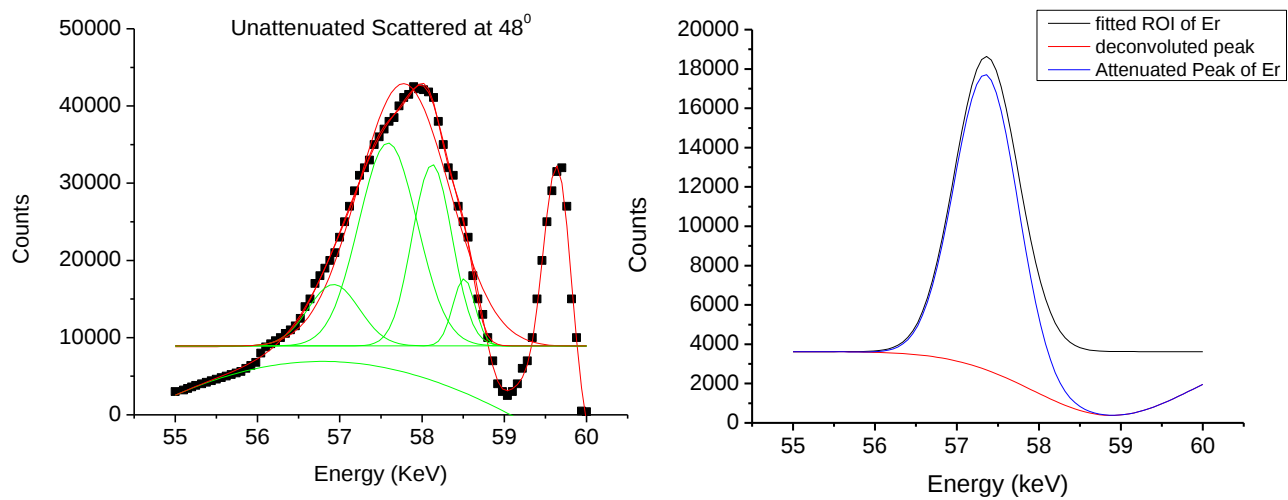
The calculated values of jump ratio (J_k) and jump factor (J_f) for absorber elements for Gb, Tb, Eu, and Er are tabulated in Table:2

Table 2: Experimental measured mass attenuation coefficient, jump ratio and jump factor for absorber used in experiment.

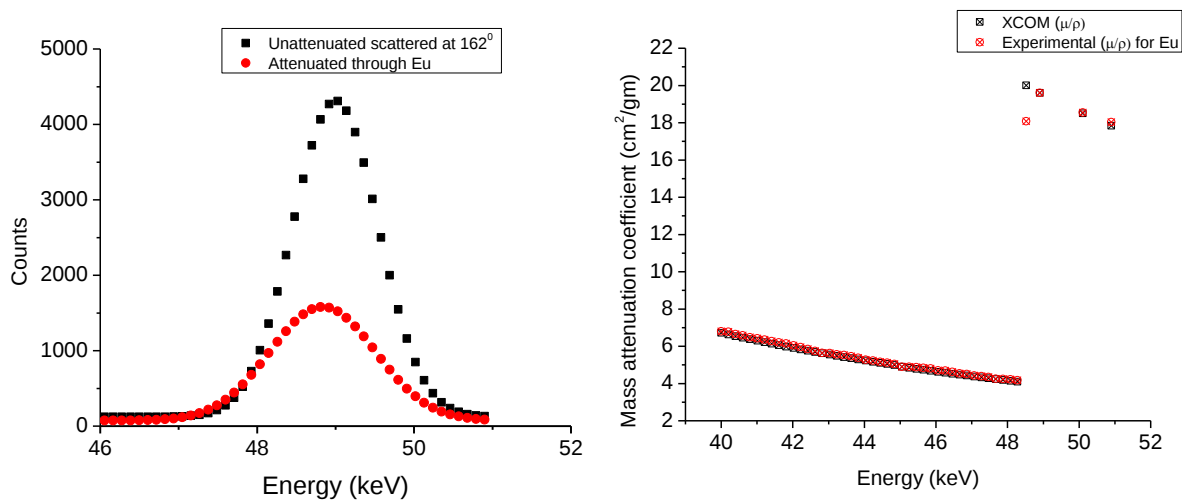
Name of absorber element	Iterative Parameter						Mass Attenuation coefficient		Jump Ratio		% error Jump parameter
	a	b	c	d	e	f	At lower edge $\left(\frac{\mu}{\rho}\right)_l$ $\left(\frac{\text{cm}^2}{\text{gm}}\right)$	At higher edge $\left(\frac{\mu}{\rho}\right)_h$ $\left(\frac{\text{cm}^2}{\text{gm}}\right)$	Jump Factor	Jump Ratio	
Eu	1.2200	0.7950	2.3710	0.0190	0.9490	0.0020	4.1632 4.0600 ^a	18.0800 20.0100 ^a	4.3428 4.9286 ^a 4.85 ^e	0.7697 0.7971 ^a 0.820 ^{c,d} 0.794±0.018 ^e	3.5553
Gd	1.2250	0.7960	2.6580	0.0180	0.9480	0.0020	3.8202 3.8200 ^a	17.5400 18.5000 ^a	4.8377 4.8429 ^a 5.305±0.4 56 ^c 4.846 ^d	0.7822 0.7935 ^a 0.824 ^{c,d}	1.4242
Tb	1.2300	0.7970	2.3900	0.0170	0.9470	0.0030	3.728 3.7080 ^a	15.7685 17.7700 ^a	4.2297 4.7923 ^a 5.46 ^e	0.7636 0.7913 ^a 0.814 ^{c,d} 0.817±0.028 ^e	3.6347
Er	1.1218	0.7928	2.3610	0.3590	0.9460	0.0020	3.2400 3.2330 ^a	13.2430 15.1300 ^a	4.0873 4.6799 ^a 5.4710±0.077 ^b 4.032 ^d		3.9395

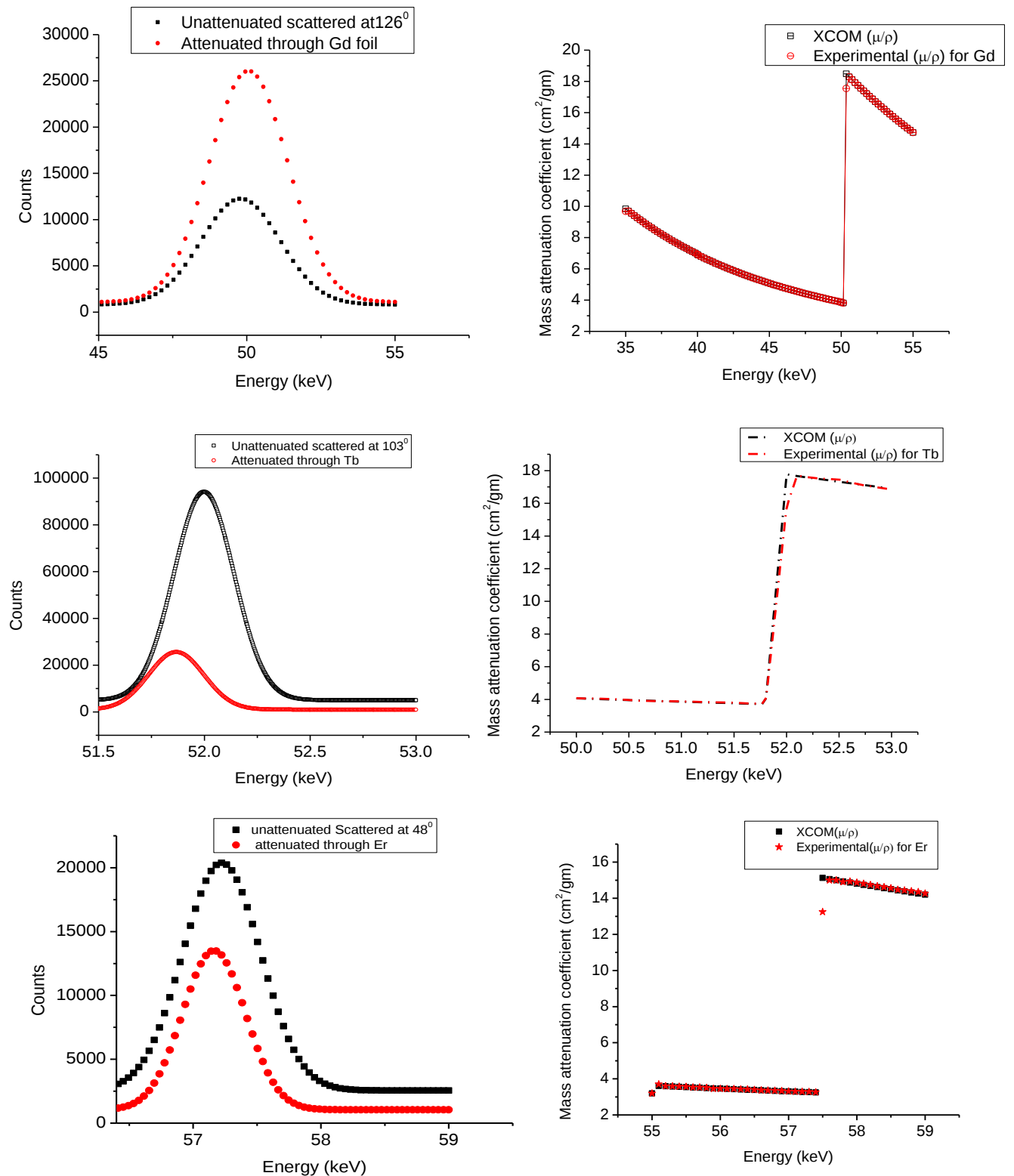
a) XCOM b) A. P. Ayala et. al 1996 c) F. Akman et. al. 2015 d) G. Budak et al. 2003 e) R. Polat et. al. 2013





Graph-1: Unattenuated Scattered, attenuated spectra before and after deconvolution for erbium absorber.





Graph -2 Experimental Gauss fitted unattenuated and attenuated HPGe spectra of mass attenuation coefficients curve nature for Eu,Gd,Tb and Er absorbers at the K-edge energy.

Result and discussion

In this present work, the total mass attenuation coefficient of the rare earth elements Eu, Tb, Gd, and Er near the K-absorption region is studied. The scattering geometry in Figure 1 has a simple yet sophisticated design to adjust the required scattering angle.

The histogram of unattenuated and attenuated channels $S(E)$ observed by the detector is shown in the span of the lower and higher sides of the K-absorption edge energies, governed by the scattering angle (Table 1).

The attenuated spectra, along with the best-fit results obtained using Monte Carlo simulation software, are further refined using the Maximum Likelihood Expectation-Maximization (MLEM) deconvolution method. This approach provides the best-fit interactive parameters a , b , c , d , e , and f , and the total mass attenuation coefficient values estimated from these computational methods are listed in Table 2.

A key parameter at the K-absorption edge is the jump ratio and jump factor, which are compared with theoretical values from Hubbell's (1995) NISTIR 5632 XCOM database and values reported by other researchers. The extracted values of the best-fit total attenuation coefficients are shown in Graphs 1 and 2, where the dotted line represents experimental data, while the solid line corresponds to Hubbell (1995) NISTIR 5632 XCOM theoretical values.

The measured K-shell absorption jump ratio and jump factors are within a 3% to 4% agreement with theoretical values. The observed deviations may be attributed to counting statistics, geometrical errors, and other experimental uncertainties.

The agreement between the experimental and theoretical values near the K-absorption edge leads to the conclusion that the present methodology is useful for fluorescence studies and for estimating the concentration of high-Z element impurities in materials.

References :

1. Hubble & Seltzar (1995) , Tables of Xray mass attenuation coefficients and mass absorption coefficient 1keV to 20 MeV for elements Z1 to 92 and 48 additional substances for dosimetric interest. NISTIR 5632.
2. Sandstorm, Handbuck. Physik. ,1957,Volume.30 P.207.
3. S. Manninen , T. Paakkari & K. Kajantie, "Gamma ray compton profile of aluminium",Philosophical Magazine,1974,29:1, 167-178.
4. P. Paatero, S. Manninen & T. Paakkari, "Deconvolution in Compton profile measurements", Philosophical Magazine,1974,30-6,Page no1281-1294.
5. Malcolm Cooper , Philip Pattison & Jochen R. Schneider,"Compton profile measurements with 412 keV γ -radiation",Philosophical Magazine,1976,34-2,243-257.
6. T.R. Canada, R.C.Bearse and J.W. Tape, "An Accurate Determination Of The Plutonium K-Absorption Edge Energy Using Gamma-Ray Attenuation", Nuclear Instruments and Methods,1977,142,609-611.
7. Robert G. Ouellet and L. John Schreiner, "A parameterization of the mass attenuation coefficients for elements with $Z=1$ to $Z=92$ in the photon energy range from approximately 1 to 150 keV",Physics in Medicine & Biology,1991,36-7,987-999.
8. Alejandro P. Ayala and Raul T. Mainardi, "Measurement of the K X-Ray absorption jump ratio of erbium by attenuation of a Compton peak", Radiation Physics & Chemistry, 1996, 47-2, 177-181.
9. H. A. Jahagirdar, B. Hanumaiah and S. R. Thontadarya , "Measurement of total And Photoelectric Cross Sections Of Rare Earth Elements Present In Compounds For 123.6 Kev And 145.4 Kev Photons", Radiation Physics & Chemistry,1996,47-6,801-805.
10. G. Budak , A. Karabulut, O. Simsek , M. Ertugrul,"Measurement of total atomic attenuation, total atomic photoelectric and total atomic scattering cross sections in the range $40 < Z < 52$ ",Nuclear Instruments and Methods in Physics Research ,1999,149,379-382.
11. M. Ertugrul, A. Karabulut, G. Budak,"Measurement of the K shell absorption jump factor of some

- elements", Radiation Physics and Chemistry, 2002, 64,1-3.
12. M.L. Mallikarjuna, S.B. Appaji Gowda, R. Gowda, T.K. Umesh, "Studies on photon interaction around the K-edge of some rare-earth elements", Radiation Physics and Chemistry,2002,65,217-223
13. Masaya Tamuraa, Tadashi Akimotoa, Yohei Aoki, Jiro Ikeda, Koichi Sato, Fumiyuki Fujita, Akira Homma, Teruko Sawamura, Masakuni Narita, "Measurement of mass attenuation coefficients around the K absorption edge by parametric X-rays", Nuclear Instruments and Methods in Physics Research A, 2002,484,642-649.
14. G. Budak, A. Karabulut, M. Ertugrul, "Determination of K-shell absorption jump factor for some elements using EDXRF Technique", Radiation Measurements, 2003, 37-2,103-107.
15. G. Budak, R. Polat, "Measurement of the K X-ray absorption jump factors and jump ratios of Gd, Dy, Ho and Er by attenuation of a Compton peak", Journal of Quantitative Spectroscopy & Radiative Transfer, 2004, 88, 525-532.
16. R. Polat, G. Budak ,A. Gürol ,A. Karabulut ,M. Ertugrul, "K-shell absorption jump factors for the elements Ag,Cs,Ba and La derived from new mass attenuation coefficient measurements using EDXRF technique", Radiation Measurements, 2005,39,409-415.
17. Necati Kaya, Engin Tirasoglu, Gokhan Apaydin, Volkan Aylıkçı, Erhan Cengiz, "K-shell absorption jump factors and jump ratios in elements between Tm (Z = 69) and Os (Z = 76) derived from new mass attenuation coefficient measurements", Nuclear Instruments and Methods in Physics Research B ,2007,262,16-23.
18. N.Kaya, E.Tirasoglu, G. Apaydin And A.I. Kobya, "Measurements of total atomic attenuation cross sections of Tm, Yb, Lu, Hf, Ta, W, Re and Os Elements at 122keV and 136keV",Sixth International Conference of the Balkan Physical U,2007,173-174.
19. S. Bennal and N. M. Badiger, "Measurement of K shell absorption and fluorescence parameters for the elements Mo, Ag, Cd, In and Sn using a weak gamma source",Journal Of Physics B: Atomic, Molecular And Optical Physics, 2007, 40, 2189-2199.
20. K.K. Abdullah, N. Ramachandran, K. Karunakaran Nair, B.R.S. Babu, Antony Joseph, Rajive Thomas and K.M. Varier, "Attenuation studies near K-absorption edges using Compton scattered 241Am gamma rays", Pramana Journal of Physics, 2008, 70-4,633-641.
21. N. Kaya, E. Tirasoglu, G. Apaydin, "Determination of K shell absorption jump factors and jump ratios in the elements between Tm(Z = 69) and Os(Z = 76) by measuring K shell fluorescence parameters", Nuclear Instruments and Methods in Physics Research B,2008,266,1043-1048.
22. K.M. Niranjana, N.M. Badiger, Daisy Joseph and S. Kailas, "Measurement of K absorption edge energies of silver and tin targets using a weak beta source", X-Ray Spectrom,2010,39,359-363.
23. Raul T. Mainardi, "On measurement of the K X-ray absorption jump factors and jump ratios of Gd, Dy, Ho and Er by attenuation of a Compton peak", Journal of Quantitative Spectroscopy & Radiative Transfer, 2010, 111, 243-244.
24. Raul T. Mainardi, "Comments on Polat, R., Budak, G., Gurol, A., Karabulut, A., Ertugrul, M.,K-shell absorption jump factors for the elements Ag, Cs, Ba and La derived from new mass attenuation coefficient measurements using EDXRF technique",Radiation Measurements, 2010, 45,880-881.
25. R. Polat, "Reply to comments on Polat, R., Budak, G., Gürol, A., Karabulut, A., Ertugrul, M., 2005, K-shell absorption jump factors for the elements Ag, Cs, Ba and La derived from new mass attenuation coefficient measurements using EDXRF technique",Radiation Measurements, 2010, 45, 882-883.
26. Sunil Kumar, Muhanad Alrakabi, Sanjeev Kumar, D. Mehta, S.C. Bedi, Nirmal Singh,"Contribution of near-edge processes to attenuation of the characteristic X-rays in elements with $48 < Z < 83$ ",Nuclear Instruments and Methods in Physics Research B, 2010, 268,431-439.
27. Baltej Singh Sidhu, A.S. Dhaliwal, K.S. Mann, K.S. Kahlon,"Measurement of K-shell absorption edge jump factors and jump ratios of some medium Z elements using EDXRF technique", Radiation Physics and Chemistry, 2011, 80,28-32.
28. P. Latha, A.M. Vinodkumar, K.M. Varier , B.R.S. Babu , Antony Joseph, K.K. Abdullah, M.P. Unnikrishnan, "Effective atomic numbers for gamma ray interaction at 59.54 keV in heterogeneous layers of materials using 241 Am gamma rays", Radiation Physics and Chemistry,2012,81,1817-1822
29. K.M. Niranjana, Krishnananda and N.M. Badiger, "Determination of K shell parameters of silver using high resolution HPGe detector spectrometer" Int. J. Nuclear Energy Science and Technology, 2013, 7-3, 179-190.

30. R. Polat O. Iceli, Z. Yalcin, E. Pasen, S. Orak, "Measurement of K-shell absorption edge jump factors and jump ratios of some medium Z elements using EDXRF technique", *Annals of Nuclear Energy*, 2013, 54, 267-273.
31. F. Akman,, R. Durak, M. R. Kaçal,"Determination of K shell absorption parameters for some lanthanides using the X-ray attenuation method" *Canadian Journal of Physics*, 2015, 93-12,1532-1540
32. Ferdi Akman, ,Rıdvan Durak , Mustafa Recep Kaçal, Mehmet Fatih Turhan,Fatma Akdemir, "Determination of K-shell absorption jump factors and jump ratios for La₂O₃, Ce and Gd using two different methods," *Canadian Journal of Physics*, 2015, 97-7, 786-790.
33. Manoj Kumar Gupta, Gurinderjit Singh, A.S. Dhaliwal, K.S. Kahlon, "Measurement of absorption edge parameters near K edge for the compounds of Zn, Zr, Cd, Ba and Pb", *Journal of Alloys and Compounds*,2015, 623-25,407-412
34. K.M. Niranjana, N.M. Badiger, "K shell parameters of some lanthanide elements using bremsstrahlung", *Radiation Physics and Chemistry*, 2015, 107,59-64.
35. Mustafa Recep Kacal, Ibrahim Hanand Ferdi Akman, "Measurements of K shell absorption jump factors and jump ratios using EDXRF technique", *European Physics Journal*, 2015, 69-103,1-7.
36. F. Akman, M.R. Kaçal and R. Durak,"Chemical effect on the K shell absorption parameters of some selected cerium compounds", *JINST*,2016,111-14(P08006).
37. Adnan Küçükönder and Hatice Topuz, "Determination of K shell absorption jump factors for As, Se, Sr, Ag, Ba, La and Ce compounds", 9th International Physics Conference of the Balkan Physical Union.
38. K shell absorption jump ratios and jump factor measurements for some elements in the range of $40 \leq Z \leq 50$ *Canadian Journal of Physics*, 2018,97, 0481.
39. **Sangam N. N., A. H. Deshpande,** C.S. Mahajan, Pravina P. Pawar, "A deconvolution of Gamma-Ray spectra for accurate attenuation studies" *Letters in High Energy Physics*, 2019, 2(4),14-20.
40. K.S. Kahlon, A.S. Dhaliwal, Baltej Singh Sidhu and Manoj Kumar Gupta,, "Calculated values of jump factor and jump ratios of Lanthanum compounds in K shell and L1, L2 and L3 subshells", *Advanced Materials and Radiation Physics*,2020,50029,1-3