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# Measurement of K X-ray fluorescence cross section of rare-earth element present in compounds

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**Abstract** : A new simple and direct method proposed by Jahagiradar *et al* to determine photoelectric cross section of elements at 123.6 keV by measuring K X-ray intensity has been used here to obtain the K X-ray fluorescence cross section. The K X-ray fluorescence cross section is determined by measuring the intensity of K X-ray radiations emitted by the rare-earth elements present in $La_2O_3$, $Nd_2O_3$, $Sm_2(CO_3)_3.8H_2O$, $Eu_2(CO_3)_3$, $Gd_2(CO_3)_3.8H_2O$, $Tb_2(CO_3)_3$ and $Tb_2O_3$ compounds at 123.6 keV by employing a NaI (Tl) spectrometer system in a $2\pi$ geometry set up. The measured K X-ray fluorescence cross sections are compared with their corresponding theoretical values and a good agreement between them is obtained

**Keywords** : X-ray fluorescence, rare-earth elements.

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## 1. Introduction

Measurement of X-ray fluorescence cross section is important in both basic and applied research. The accurate knowledge of fluorescence cross section data makes XRF suitable for quantitative elemental analysis.

Krause *et al* [1] have estimated K and L X-ray fluorescence cross section for a wide range of elements. Bhan *et al* [2] have measured the absolute values of K X-ray fluorescence cross section by using the theoretical values of photoelectric cross sections, fluorescence yield, and X-ray emission rate, employing strong source of 5–8 mci strength. Jahagirdar *et al* [3] have measured K X-ray fluorescence cross section from which the total photoelectric cross section of elements in the region $42 \leq Z \leq 82$ are evaluated.

In the present investigation, a simple method proposed by Jahagiradar *et al* [3] is used to obtain the K X-ray fluorescence cross section of rare-earth elements present in $La_2O_3$, $Nd_2O_3$, $Sm_2(CO_3)_3.8H_2O$, $Eu_2(CO_3)_3$, $Gd_2(CO_3.)_38H_2O$, $Tb_2(CO_3)_3$ and $Tb_2O_3$

compounds having atomic numbers in the range $57 \leq Z \leq 65$ by taking lead ($Z = 82$) as a standard element. The K X-ray fluorescence cross section can be obtained by the measurement of K X-ray intensity. The intensity of K X-ray photons produced by irradiating rare-earth compounds with 123.6 keV gamma-rays using 2" × 1" NaI(Tl) gamma-ray spectrometer system attached to 1K multichannel analyser (ORTEC) in a $2\pi$ geometrical configuration, is measured.

## 2. Basic relation

The K X-ray fluorescence cross section $\sigma_k(i)$ of an element $i$ at an excitation energy $\varepsilon_i$ is given by the equation [2–6]

$$\sigma_k(i) = I_k(i)/(I_0 G\varepsilon\beta t), \tag{1}$$

where $I_k(i)$–the observed intensity of the K X-ray line of the element,

$I_0$–the intensity of the exciting radiation,

$\varepsilon$–the efficiency of the detector at the K X-ray energy,

$G$-is the geometrical factor accounting for the geometrical arrangement of detector and sample set up [7],

$\beta$–the target attenuation correction factor for the incident as well as K X-ray radiations which is given by [8,9]

$$\beta = \left\{1 - \exp\left[-(\mu_i + \mu_e)t\right]\right\} \Big/ \left\{(\mu_i + \mu_e)t\right\}, \tag{2}$$

where $\mu_i$ and $\mu_e$ are the mass attenuation coefficients of incident and K X-ray radiations in the target, $t$ is the thickness of the target.

## 3. Experimental arrangement

In the present investigation, a Harshaw make 2" × 1" NaI(Tl) crystal attached to an RCA photomultiplier tube (6342 A) coupled to an 1K MCA (ORTEC) is used to measure K X-ray intensity $I_k$ of the target in a $2\pi$ geometrical configuration.

The experimental arrangement, involving the detector, source and the target system is shown in Figure 1 and the details of its arrangement is given by Jahagirdar *et al* [3]. Owing to the poor resolution of NaI(Tl) detector, the peaks of 122 keV and 136 keV photons from $^{57}$Co source are not separated and hence the weighted average of these two energies equal to 123.6 keV is considered. The $^{57}$Co radioactive isotope of about 10 μci strength standard gamma source, is used in this experiment.

The linearity and stability of the spectrometer were checked with a precision pulse generator and the room temperature was maintained at 25 ± 1°C. The gamma ray spectrometer was calibrated using $^{241}$Am (26, 59.6 keV), $^{141}$Ce (36.9, 145.4 keV) and $^{57}$Co (123.6 keV weighted average) sources.

The constant intensity is maintained throughout the experiment by fixing the source position and marking the orientation of the source. We have selected the live time mode because the strength of the source was so low (10 μci) and MCA showed no dead time losses even when it was placed on the face of the detector. In this method, we have used uniform circular pellets of 3 cm diameter of rare earth compounds prepared using a pellet making machine by applying a constant pressure of 400 kg/cm$^2$. The mass per unit area of each pellet was determined using an electrical balance and travelling microscope. The uniformity of each pellet was checked by measuring the transmitted intensity of the beam through different regions of the targets.

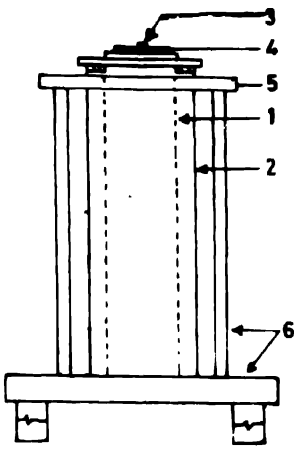


**Figure 1.** Experimental arrangement, involving the detector, source and the target system.
(1) (2" × 1") NaI(Tl) detector with pint, (2) Steel cylinder, (3) Position of source, (4) Target, (5) Aluminium plate and (6) Iron stand.

First, we scanned the background counts due to cosmic rays and other radioactive sources. Next, the source spectrum was scanned by placing the source directly on the head of the detector. The entire area under this spectrum is the sum of background intensity and the intensity of the source. This spectrum is stored in first half (say $H_1$) of the memory; then the pellet of known thickness was sandwitched between the source and the detector and the spectra is recorded. The total area under the entire spectrum consists of scattered radiation, secondary photons along with transmitted intensity of the primary radiation. This spectrum was stored in second half (say $H_2$) of the memory. These two spectra were recorded during suitable interval of times so as to reduce the statistical error below 1%. The subtraction of first spectrum from the second spectrum in the X-ray energy region gives the pure characteristic K X-ray fluorescence spectrum. The total area under this X-ray peak gives the K X-ray fluorescence intensity $I_k$. This has been followed for rest of the targets of different thickness. It is observed that the intensity of K X-ray fluorescence initially increases as a function of thickness $t$ and then decreases as the thickness of the target increases. We have taken the initial increasing portion, where the K X-ray production predominates over its attenuation for the evaluation of K X-ray fluorescence cross section $\sigma_k$.

The measured K X-ray intensity has been corrected using eq. (2) for self attenuation of fluorescent radiation in the target. The necessary theoretical mass attenuation coefficients are taken from Plechaty *et al* [10].

We have used eq. (1) to find the K X-ray fluorescence cross section $\sigma_k$ from the experimentally measured $I_k$. It is seen from eq. (1) that the quantity $(I_k/\beta)$ is proportional to $t$ and hence a plot of $I_k/\beta$ *vs* $t$ should be a straight line passing through origin and having slope equal to $\sigma_k I_0 G\varepsilon$. Using the least square fit value of the slope, we have determined the value of $\sigma_k$. We have plotted in Figure 2, the corrected intensity $(I_k/\beta)$ *vs* $t$ for all the seven rare-earth compounds including lead. The solid lines are least square fit straight lines, all are passing through origin, and experimental points lie very close to these lines.

**Table 1.** Values of the measured K X-ray fluorescence cross section $\sigma_k$ in $cm^2/gm$ for 123.6 keV gamma rays for rare-earth compounds

| Sl. No | Material used | Chemical formula | Element | Z | Measured value of $\sigma_k$ in $cm^2/gm$ | Theoretical value of $\sigma_k$ in $cm^2/gm$ [1] |
|---|---|---|---|---|---|---|
| 1 | Lanthanum oxide | $La_2O_3$ | La | 57 | 0.726 ± 0.001 | 0.714 |
| 2 | Neodymium oxide | $Nd_2O_3$ | Nd | 60 | 0.835 ± 0.002 | 0.844 |
| 3 | Samarium carbonate | $Sm_2(CO_3)_3$ $8H_2O$ | Sm | 62 | 0.937 ± 0.001 | 0.923 |
| 4 | Europium carbonate | $Eu_2(CO_3)_3$ | Eu | 63 | 0.956 ± 0.003 | 0.968 |
| 5 | Gadolinium carbonate | $Gd_2(CO_3)_3$. $8H_2O$ | Gd | 64 | 0.970 ± 0.003 | 0.994 |
| 6 | Terbium carbonate | $Tb_2(CO_3)_3$ | Tb | 65 | 1.085 ± 0.006 | 1.045 |
| 7 | Terbium oxide | $Tb_2O_3$ | Tb | 65 | 1.076 ± 0.008 | 1.045 |
| 8 | Lead | Pb | Pb | 82 | 1.832 ± 0.014 | 1.815 |

We have determined the $\sigma_k$ values of rare earth element present in compounds which are given in Table 1 along with the associated error. The theoretical cross sections which are calculated using log-log interpolation formula using theoretical cross sections of Krause *et al* [1] are also given for comparison. The two Tb compounds, namely $Tb_2O_3$ and $Tb_2(Co_3)_3$ with different chemical environment around the Tb atom in the molecule are used with the intention of knowing the effect of chemical environment in the K X-ray yield. The measured $\sigma_k$ values in both cases are little higher than the theoretical value which may be mainly due to the errors involved in the measurement. Although these higher values do not reveal any information about the chemical environment, this effect needs to be studied in detail.

## 4. Results and discussion

The measured values of $I_k/\beta$ are plotted against $t$ in Figure 2 for all the rare-earth elements present in compounds. The solid lines are the least square fit lines and experimental points

lie very close to the solid straight lines. It is also seen that all the straight lines are passing through origin, indicating that the $\beta$ correction factor applied for the self-attenuation of K X-ray in the increasing region of its production, is taking care of the necessary correction.

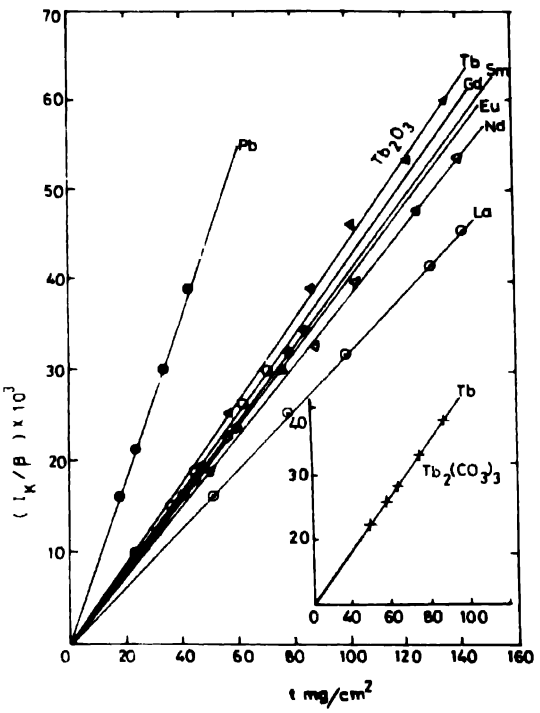


**Figure 2.** Variation of $(I_k/\beta)$ as a function of thickness of seven rare-earth compounds including lead. To avoid overlapping of lines due to $Tb_2(CO_3)_3$ and $Tb_2O_3$ compounds a separate line due to $Tb_2(CO_3)_3$ is shown in the

Also it is seen that the inclination of each line is different, indicating the strong dependence of production of K X-rays on $Z$-value of the element. From the slopes of these straight lines the value of $\sigma_k$ for all the elements have been obtained using eq. 1. The results of these $\sigma_k$ values are tabulated in Table 1. In Table 1, column 2 to 5 give the details of rare-earth compounds used with the $Z$-values of the rare-earth elements present in each compound. Column 6 gives our measured values of $\sigma_k$ along with their associated errors for elements in the range $57 \leq Z \leq 82$ including lead. These $\sigma_k$ values are the average values of the 6 independent measurements, performed on different days. Here, the $\sigma_k$ value of lead being an high-$Z$ element, is included to verify the reproducibility of the result and the accuracy of the method proposed by Jahagirdar *et al* [3]. The measured $\sigma_k$ values are given at the end of Table 1 for completeness. The errors in each case are due to statistical error in $I_k$ and $I_0$ measurement, measurement of $t$, determination of $\beta$ factor where theoretical values of $\mu_i$ and $\mu_e$ are used. The quoted error in $\mu_i$ and $\mu_e$ values [10] was found to be 2–3% in the energy range under consideration. The estimated error in $\beta$-factor was found to be less than 3% as $\mu_i$ and $\mu_e$ come under exponential term. The over all error in $\sigma_k$ values in all cases was found to be less than 5%. The measured values of $\sigma_k$ are compared with the corresponding theoretical values of Krause *et al* [1] given in column 7. It is seen that there

is a close agreement between the measured values of $\sigma_k$ with the corresponding theoretical values.

## 5. Conclusion

A simple, direct method proposed earlier for the determination of total photoelectric cross section by measuring K X-ray intensity has been successfully extended here to obtain the K X-ray fluorescence cross section of rare-earth elements present in their compounds having $Z$ in the range $57 \leq Z \leq 82$, including lead. The $\sigma_k$ value of each element is the average value of the six independent measurements made on different days. The error in each case is found to be less than 5% and a good agreement between the measured value with the theoretical value is seen, justifying the suitability of the method used. As far as our knowledge goes, the $\sigma_k$ values of the above seven rare earth elements measured here are the first values at 123.6 keV. This data is very much required as it has important application in wide veriety of fields.

**References**

[1] M O Krause, C W Jr Nestor, C J Jr Sparks and E Ricci *X-Ray Fluorescence Cross Section for K and L X-Rays of the Elements*. **ORNL-5399** (1978)

[2] C Bhan, S N Chaturvedi and N Nath *X-ray Spectrom*. **10** 128 (1981)

[3] H A Jahagirdar, B Hanumaiah and S R Thontadharya *Appl. Radiat. Isot*. **43** 399 (1992)

[4] R D Giauque and J M Jaklevic *Adv. X-ray Annal* **15** 164 (1972)

[5] Al-Saleh and N S Saleh *Appl. Radiat. Isot*. **38** 975 (1987)

[6] A Rani, R K Koshal, S N Chaturvedi and N Nath *X-ray Spectrom*. **17** 53 (1988)

[7] D K G De Boer *X-ray Spectrom*. **18** 119 (1989)

[8] H R Verma, D Pal, M L Garg and P N Trehan *J. Phys* **B18** 1133 (1985)

[9] M L Garg, R R Garg and K G Malmqvist *J. Phys* **B20** 3705 (1987)

[10] E F Plechaty, D E Cullen and R J Howerton *Tables and Graphs of Photon Interaction Cross Sections from 0.1 keV to 100 MeV dervied from LLL Evaluated Nuclear-Data Library*. **UCRL-50400** Vol 5 Rev 3 (1981)