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Review

# Determination of K shell X-ray intensity ratios for some heavy elements

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ABSTRACT

The K shell X-ray intensity ratios $K_{\alpha2}/K_{\alpha1}$, $K_{\beta1,3}/K_{\alpha1}$ and $K_{\beta2}/K_{\alpha1}$ for 21 elements with $65 \leqslant Z \leqslant 92$ have been measured using an incident photon energy of 123.6 keV. The X-rays have been measured with a Si(Li) semiconductor detector. $K_\beta$ and $K_\alpha$ X-rays have been analyzed into the components $K_{\beta1,3}$ and $K_{\beta2}$ and $K_{\alpha1}$ and $K_{\alpha2}$, respectively, using a computer program. The experimental results were compared with the theoretical values of Scofield and available experimental results. All X-ray intensity ratios values have been plotted versus atomic number.



**Contents**



## 1. Introduction

Information experimental result regarding the K X-ray intensity ratios for different elements is important because of its extensive use in atomic, molecular and nuclear physics, properties of superconductor, semiconductor thin films, etc., and nondestructive analysis of materials using energy dispersive X-ray fluorescence (ED-XRF) techniques. In earlier experimental investigations, Nelson and Saunders (1969) have measured $K_{\alpha2}/K_{\alpha1}$ X-ray intensity ratios for 36 elements from Sb to Am. Experiments have been performed using a Cauchois-type bent-crystal spectrometer and a Ge(Li) detector. Similarly, Salem and Wimmer (1970) have measured $K_{\alpha2}/K_{\alpha1}$ transition probabilities for 22 elements from Sc to Sn. K X-ray emission rates have been measured for some elements with $79 \leqslant Z \leqslant 92$ using a high-resolution Ge(Li) detector by de Pinho (1971). McCrary et al. (1971) have measured K X-ray relative intensities using Si(Li) and Ge(Li) detectors for elements ranging from Ca to Pu. Iwatsuki and Fukasawat (1987) have measured $K_{\beta2}/K_{\beta1,3}$ intensity ratios for As, Se and Br in various chemical states. The relative X-ray transition probabilities $K_{\beta1}/K_{\alpha1}$, $K_{\beta2}/K_{\beta1}$, $K_O/K_{\beta1}$, and $K_{\alpha2}/K_{\alpha1}$ have been measured for elements in the $47 \leqslant Z \leqslant 56$ region using HPGe detector by Martins et al. (1989). In the last few years some K X-ray relative intensity ratios the in atomic number region $40 \leqslant Z \leqslant 50$, $39 \leqslant Z \leqslant 68$ and $69 \leqslant Z \leqslant 92$, respectively, have been measured by some workers (Campbell, 2001; Ximeng et al., 2001; Ertuğrul and Şimşek, 2002). There have been various investigations on $K_\beta/K_\alpha$ X-ray intensity ratios for elements (Büyükkasap et al., 1994; Dhal and Padhi, 1994; Büyükkasap, 1997; Be et al., 1998; Ertuğrul et al., 2001 2007), compounds (Küçükönder et al., 1993, 2003; Raj et al., 1999a; Mukoyama et al., 2001), alloys (Bhuinya and Padhi, 1992; Söğüt et al., 1995; Raj et al., 1999b, 2001) and complex (Çevik et al., 2005).

In the present work, the $K_{\alpha2}/K_{\alpha1}$, $K_{\beta1,3}/K_{\alpha1}$ and $K_{\beta2}/K_{\alpha1}$ X-ray intensity ratios of 21 elements from terbium (Tb) to uranium (U) have been measured using a Si(Li) detector and $^{57}$Co radioisotope source. The experimental results are compared with theoretical and other experimental results.

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## 2. Experimental procedure and calculations

The geometry and the shielding arrangements of the experimental setup employed in the present work are as shown schematically in Fig. 1. The studied elements were given Table 1. The purity of commercially obtained materials was better than 99%. Powder samples were sieved to 400 mesh sizes and prepared by supporting the mylar film $\cong$20–40 mg/cm$^2$ mass thickness. All samples were irradiated by 123.6 keV photons emitted by an annular 925 MBq $^{57}$Co radioisotope source. (The radioactive source $^{57}$Co decays by electron conversion process into metastable states of $^{57}$Fe and in turn it gives $\gamma$ photons of energies 122 (85%), 136 (11%) and 14.4 (8.5%). Since the intensity of the 122 keV photons is predominant over that of the 136 keV photons and as they are close to each other, we can take the weighted average of 122 and 136 keV, i.e. 123.6 keV.) The samples were placed at 45° angle with respect to the direct beam and fluorescent X-rays emitted 90° to the detector. The incident beam and fluorescence X-rays emitted from the target were detected and analyzed with a Canberra Si(Li) detector (FWHM 160 eV at 5.9 keV, active area 13 mm$^2$, thickness 3 mm and Be window thickness 30 μm). The output from the preamplifier, with pulse pile-up rejection capability, was fed to a multi-channel analyzer interfaced with a personal computer provided with suitable software (Tennelec PCA II) for data acquisition and peak analysis program. The live time was selected to be 5000 s for all elements. Fig. 2 shows a typical K X-ray spectrum for thulium.

The $K_{\alpha2}/K_{\alpha1}$, $K_{\beta1,3}/K_{\alpha1}$ and $K_{\beta2}/K_{\alpha1}$ X-ray intensity ratios values have been calculated using the equation

$$\frac{I(K_i)}{I(K_j)}=\frac{N(K_i)}{N(K_j)}\times\frac{\varepsilon(K_j)}{\varepsilon(K_i)}\times\frac{\beta(K_j)}{\beta(K_i)}\quad (i=\alpha2,\ \beta1,3,\ \beta2 \text{ and } j=\alpha1), \qquad (1)$$

where $N(K_i)$ and $N(K_j)$ are the counts observed under peaks corresponding to K$i$ and K$j$ X-rays; $\varepsilon(K_i)$ and $\varepsilon(K_j)$ are the efficiencies of the detector for the K$i$ and K$j$ series of X-rays, respectively. $\beta(K_i)$ and $\beta(K_j)$ are the target self-absorption correction factors for both the incident and the emitted radiations. $I_0$ is the intensity of the incident radiation and $G$ is a geometrical factor. In the present study, the $I_0G\varepsilon(K_{i,j})$ values were determined as explained in our previous work (Ertuğral et al., 2005).

The self-absorption correction factor $\beta$ is calculated for both K$i$ and K$j$ separately by using the following expression:

$$\beta=\frac{1-\exp[-(\mu_{inc}/\sin\theta+\mu_{emt}/\sin\phi)t]}{(\mu_{inc}/\sin\theta+\mu_{emt}/\sin\phi)t} \qquad (2)$$

where $\mu_{inc}$ and $\mu_{emt}$ are the total mass absorption coefficients (from XCOM (Berger and Hubbell, 1999)) of target material at the incident photon energy and at the emitted average $K_i$ X-ray energy (Storm and Israel, 1970). $\theta$ and $\phi$ are the angles of incident photon and emitted X-rays respectively with respect to the normal at the surface of the sample. $t$ is the thickness of the target in g/cm$^2$.

## 3. Results and discussion

Experimental $K_{\alpha2}/K_{\alpha1}$, $K_{\beta1,3}/K_{\alpha1}$ and $K_{\beta2}/K_{\alpha1}$ X-ray intensity ratios values for $65\leqslant Z\leqslant 92$, measured for incident photon energies 123.6 keV, are presented in Table 1. These values have been plotted as a function of the atomic number as shown in Fig. 3(a–c). Experimental K X-ray intensity ratio values have been compared with theoretical estimates based on relativistic Hartree–Fock and Hartree–Slater theories calculated by Scofield (1974a, b) and other experimental values. The $K_{\alpha2}/K_{\alpha1}$ ratio experimental values are in general higher than the Scofield's values and the agreement between present results and theoretical predictions are within the range 1–8% and 0.5–9%. The $K_{\beta1,3}/K_{\alpha1}$, $K_{\beta2}/K_{\alpha1}$ ratio experimental values agree to within 0.6–11% and 0.6–8.3%, 1.2–25% and 4–19% with the theoretical values of Scofield (1974a, b), respectively.

The uncertainties in the K X-ray intensity ratios are estimated to be less than 6% and are found propagating the errors in various parameters used for the determination of the intensity ratios. The uncertainties in the parameters are listed in Table 2.

In this work, in order to reduce the absorption, thin samples were used as the target; furthermore, an absorption correction was also performed for each sample. In order to reduce the statistical error, the spectra were collected under the $K_{\alpha1,2}$ and $K_{\beta1,2,3}$ peaks. In the experimental determinations, spectral deconvolution is one of the main problems that arise when determining these parameters due to the strong peak overlapping in EDXRF system. Good statistics is not enough for this purpose

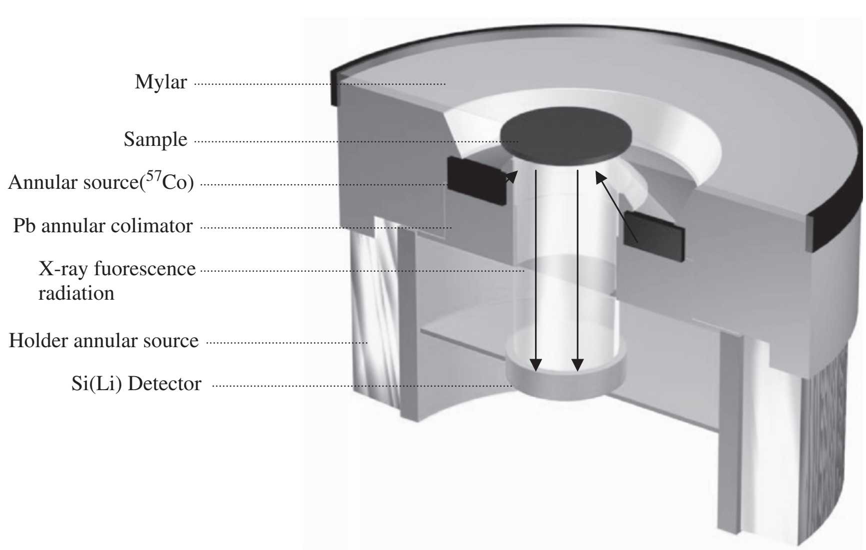


**Fig. 1.** Experimental geometry for Si(Li) detector.