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Abstract

X-ray production cross sections of Kα_1, Kα_2, K$\beta_1^{'}$ (= Kβß$_1$+ Kβ_3+ Kβ_5) and K$\beta_2^{'}$ (= Kβ_2 + Kβ_4 + transitions from higher levels) lines have been measured and theoretically calculated for six elements with $62 \leq Z \leq 74$ at excitation energy of 78.706 keV, the weighted avarage of K conversion x-rays emitted from Bi. The experimental results were compared with theoretically predicted values based on relativistic Hartree-Slater and Hartree-Fock theories, a comparison that was found to be in good agreement to within the experimental uncertainties.

Key Words: K shell, X-ray fluorescence, Cross section.

1. Introduction

The measurement of K x-ray production cross sections is important in the study of some basic phenomena in atomic, molecular and radiation physics and in the non-destructive elemental analysis of materials using energy-dispersive x-ray fluorescence (EDXRF). Also, comparison of measured cross sections with theoretical estimates provides a check on the validity of various physical parameters such as photoionization cross-section, fluorescence yield, K x-ray emission rates and jump ratios involved in the evaluation of theoretical estimates.

Photon-induced Kα and Kβ x-ray production cross sections have been measured by many researcher [1-14]. Whatson et al. [1] measured the K x-ray production cross sections for 14 elements relative to Cu, at 17.8 keV (Mo K x-rays). Chang et al. [2] and Naser et al. [3] measured the same cross sections using radioisotopes as excitation sources. Bhan et al. [4], Garg et al. [5] and Kumar et al. [6] have determined the K x-ray production cross sections for medium Z elements. Rani et al. [7] measured x-ray production cross sections for Kα x-ray lines for light elements relative to Ca at a photon excitation energy of 5.97 keV. Saleh et al. [8] measured Kα cross sections for 13 elements with medium atomic number relative to Ag at two excitation energy. Kα and Kβ x-ray production cross sections were measured for ten elements as dependent on excitation energy by Singh et al. [9] and Casnati et al. [10]. Rao et al. [11] also measured K x-ray production cross sections for medium Z elements at various excitation energy using an x-ray tube as excitation source and Durak et al. [12-14] experimentally determined K x-ray production cross sections for medium Z elements at induced photon energies of 59.5 and 122 keV.

On the other hand, Krause et al. [15] have theoretically calculated Kα, Kα_1, Kβ and Kβ_1 x-rays production cross sections. The production cross sections of Kα_2 and Kβ_2 x-rays were measured experimentally by Saleh et al. [16] for ten elements with $73 \leq Z \leq 82$ at 121.9 keV.

In this work, the measurements of Kα_1, Kα_2, Kβ_1' and Kβ_2' x-rays production cross sections were made for six elements with $62 \leq Z \leq 74$ at excitation energy of 78.706 keV, the weighted average of K conversion x-rays emitted from Bi. K x-rays from Bi excited by from a ^{57}Co radioisotope.

The results were then compared with the corresponding theoretically calculated values based on relativistic Hartree-Slater and Hartree-Fock theories. To the best of our knowledge, photon-induced K_i (i= α_1, α_2, β_1' and β_2') x-ray production cross sections for these elements at this excitation energy are being reported for the first time.

2. Experimental

The experimental arrangement is shown in Figure 1. In this arrangement, the samples were irradiated with Bismuth K x-rays, whose weighted average energy is 78.706 keV. To excite the K shell of Bi, an annular ^{57}Co radioisotope source of strength 10 mCi emitting gamma photons of 121.9 keV was used. In addition to the 121.9 keV,136 keV γ-rays are also emitted from the ^{57}Co. The 136 keV γ-rays can also excite the K shell of Bi, but there is a filter on the ^{57}Co annular source that almost completely absorbs all 136 keV emissions. It is worth mentioning that if we calculate the K XRF cross sections of a given element at the Kα and Kβ incident photon energies and then take the weighted average of cross sections according to the intensity of Kα and Kβ lines or calculate the K XRF cross sections directly at the weighted average energy, the difference between the two is not more than 1%. Our assumption of taking the weighted average energy of Kα and Kβ lines is therefore reasonable.

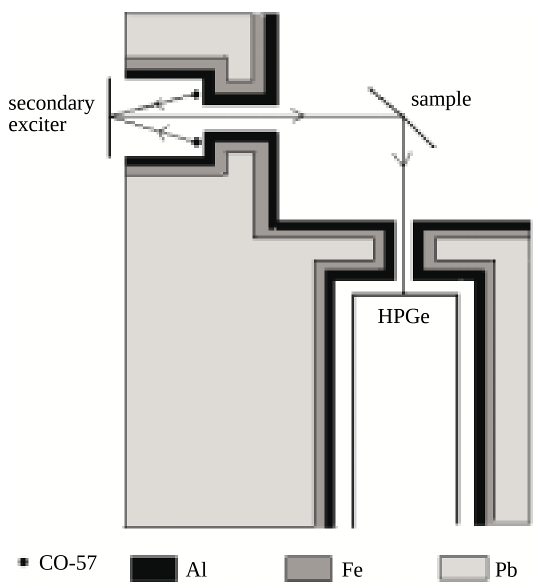

Figure 1. Schematic of the experimental set-up.

Spectroscopically pure (purity better than 99.99%) thin foils of Sm, Tb, Er, Yb, Ta and W of thickness ranging from 150 to 250 μg/cm^2 were used for the measurements. Each element was taken in its oxide form and the powdered specimen was uniformly pressed between two x-ray mylar films obtained from Spex Certi Prep, Inc. The thickness of the mylar films was 2.5 μm.

The samples were placed at a 45° angle with respect to the direct beam and the emitted fluorescent x-rays, which were recorded by a collimated HPGe detector (full width at half maximum = 170 eV at 5.96 keV, active area = 12.5 mm^2, sensitive depth = 3 mm) coupled to an ND 66B multichannel analyser.

The HPGe detector was setup with a 4 μs amplifier shaping time constant was selected to provide optimum resolution and the best signal-to-noise ratios at low count rates. This value of shaping time was used throughout the measurement.

The shielding in the arrangement was arranged so that the source can only see the secondary exciter. But the energy of gamma rays scattered coherently and incoherently from the secondary exciter is greater than the K threshold energy of all the samples and thus gamma rays scattered coherently and incoherently produce additional unwanted K x-rays. The correction for the unwanted contribution of K x-rays produced by scattered 121.9 keV gamma rays was applied to K x-rays counted in the detector by using an equivalent aluminium target in place of the sample under investigation as discussed by Man et al. [17]. The spectra of radiation emitted from the secondary exciter and equivalent Al target are matched to give almost the same scattering by adjusting the thickness of the equivalent Al target. A typical Er spectrum, with scattering effects subtracted, is shown in Figure 2. The spectra were analyzed using a quantitative x-ray analysis computer code Analysis of X-ray Spectra for Iterative Least-Squares Fitting (AXIL) [18-20].

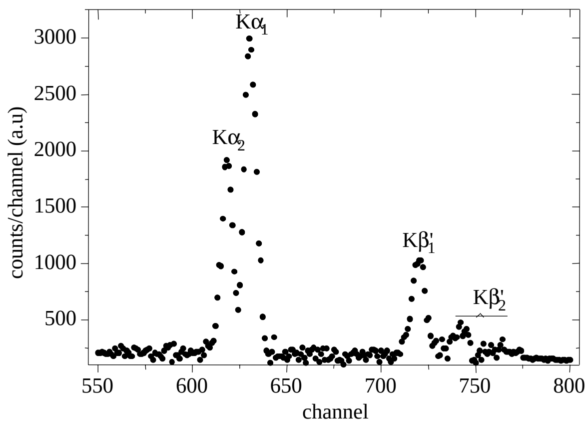

Figure 2. The spectrum of the K lines of Er excited by Bi K x-rays.

The experimental K x-ray production cross sections have been evaluated by using the relation

$$\sigma_{Ki} = \frac{N_{Ki}}{I_0 G \varepsilon_{Ki} \beta_{Ki} m}, \tag{1}$$

where N_{Ki} is the number of counts in unit time under the photopeak corresponding to the K_i x-rays; I_0G is the intensity of photons falling on the area of sample visible to the detector; ε_{Ki} is the detector efficiency at K_i x-ray energy; m is the mass thickness of the target in g/cm^2; and β_{Ki} is the absorption correction factor, which compensates absorption of the incident photons and the emitted characteristic K x-rays in the target material.

The values of the factor I_0G, which contains the terms relating to the flux of K x-rays emitted from the secondary exciter, were determined in a separate experiment. For this purpose, targets of Mo and Ag (in form thin foils) having the same diameter as in the main experiment were irradiated with K x-rays of the secondary exciter in the same experimental set-up and the intensities of the K x-rays emitted in each case were counted with the spectrometer used in the main experiment. I_0Gcan be expressed by the relation

$$I_0 G = \frac{N_K}{\sigma_K^x \varepsilon_K \beta_K t}, \tag{2}$$

where N_K, β_K and ε_K have the same meaning as in Equation (1). The theoretical values of K x-ray fluorescence cross sections (σ_K^x) were evaluated using the relationship

$$\sigma_K^x = \sigma_K(E)\omega_K, \tag{3}$$

where $\sigma_K$is the K shell photoionization cross section for given element at excitation energy (E), taken from the table of Scofield [21]; and ω_K is the K shell fluorescence yield taken from the recent standart fitted values of Hubbell et al. [22].

The relative efficiency curve for the HPGe detector in the energy range 15-75 keV was measured using Am-241, Ba-133, Cs-137 and Co-57 radioisotope testing sources according to an earlier method [23]. Although we used testing sources with low strength, peak/background ratios of all the determined spectra were found to be satisfactory and this can be attributed to having used an appropriate collimator in our setup to minimize the number of scattering photons. Therefore we could use the testing sources mentioned above to determine the efficiency of the HPGe detector. The self absorption correction factor β_{Ki} has been evaluated as described earlier [24].

3. Theoretical Evaluation

The theoretical values of K x-ray production cross sections σ_{Ki} were calculated using the equation

$$\sigma_{Ki} = \sigma_K(E)\omega_K F_{Ki}, \tag{4}$$

where $\sigma_K(E)$ and ω_K have the same meaning as in equation (3). F_{Ki} is the fractional x-ray emission rate for K_i x-ray and is defined as

$$F_{K\alpha_1} = \frac{I_{K\alpha_1}}{I_{K\alpha} + I_{K\beta}}$$

$$F_{K\alpha_2} = \frac{I_{K\alpha_2}}{I_{K\alpha_1}} F_{K\alpha_1}$$

$$F_{K\beta_1'} = \frac{I_{K\beta_1^1}}{I_{K\alpha_1}} F_{K\alpha_1}$$

and

$$F_{K\beta_2'} = \frac{I_{K\beta_2^1}}{I_{K\alpha_1}} F_{K\alpha_1}, \tag{5}$$

where I_{Ki} is the K_i x-ray intensity. In the present calculation, the value of σ_K(E) was taken from Scofield [21] and is based on a relativistic Hartree-Slater central potential; the value of ω_K was taken from the fitted values of Hubbell et al. [22]; and the values of intensity ratios were taken from Scofield based on relativistic Hartree-Slater theory [25] and the other on relativistic Hartree-Fock theory [26].

4. Results and Discussion

The present experimental results for K x-ray production cross sections are listed with theoretical values in Table 1. For comparison, the experimental results and those fitted alongside with the theoretical values of cross sections are plotted as a function of the atomic number, as shown in Figure 3. The overall error in the measured this cross sections is estimated to be less than 8%. The error associated in evaluating the photopeak area is 3%, the error in I_0G is 5%, the uncertainty in self-absorption correction factors is about 3%, the error in measurement of detector efficiency is 3% and the other systematic errors were about 3%. It is evident from Table 1 and Figure 3 that the experimental values of the K x-rays production cross sections for all elements are in general, agreement with theoretical values within experimental error. But the experimental values of $K\alpha_1$, $K\alpha_2$, and $K\beta_1^{'}$ x-rays production cross sections are higher than the theoretical estimates. In general the present experimental values are higher than theoretical values. This case can be attributed to a chemical effect since each element was taken in its oxide form. On the other hand, the experimental values of $K\alpha_1$ and $K\alpha_2$ x-rays production cross sections are more close to the theoretical values when the values of K x-ray intensity ratios based on relativistic Hartree-Slater theory and the experimental values of $K\beta_1^{'}$ x-ray production cross section are generally more close to the theoretical values when the values of K x-ray intensity ratios based on relativistic Hartree-Fock theory.

Table 1. Experimental and theoretical K x-rays production cross sections (b/atom).

Element	$\sigma_{K\alpha 1}$		$\sigma_{K\alpha 2}$		$\sigma_{K\beta'_1}$		$\sigma_{K\beta'_2}$	
	Present work	Theoretical	Present work	Theoretical	Present work	Theoretical	Present work	Theoretical
62 Sm	531.26 ± 42.50	515.17[a]	299.75 ± 23.48	284.89[a]	162.12 ± 12.16	100.81[a]	39.34 ± 3.15	24.88[a]
65 Tb	642.62 ± 51.41	607.50[a]	356.73 ± 28.03	339.55[a]	197.13 ± 15.37	184.68[a]	48.48 ± 3.39	45.82[a]
		601.26[b]		335.31[b]		191.51[b]		49.70[b]
68 Er	746.94 ± 59.45	708.63[a]	427.50 ± 34.20	393.68[a]	232.32 ± 17.42	219.62[a]	57.00 ± 4.46	54.78[a]
		701.44[b]		395.11[b]		227.22[b]		59.05[b]
70 Yb	765.10 ± 61.21	778.43[a]	468.30 ± 37.40	441.31[a]	257.67 ± 20.00	244.37[a]	64.02 ± 4.92	61.02[a]
		770.34[b]		437.04[b]		252.15[b]		65.75[b]
73 Ta	947.81 ± 76.00	884.83[a]	538.16 ± 43.05	507.89[a]	297.18 ± 22.99	282.18[a]	74.38 ± 4.01	73.08[a]
		876.09[b]		502.47[b]		291.10[b]		78.67[b]
74 W	971.88 ± 78.00	921.97[a]	564.06 ± 45.12	531.04[a]	308.43 ± 24.61	295.94[a]	79.51 ± 5.96	77.45[a]
		913.02[b]		525.56[b]		304.70[b]		83.30[b]

[a]The theoretical values based on relativistic Hartree-Slater theory
[b]The theoretical values based on relativistic Hartree-Fock theory

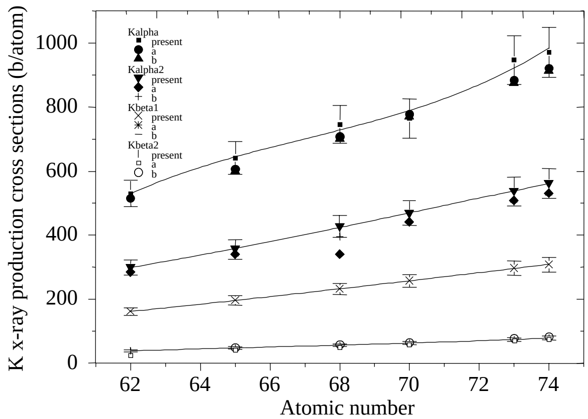

Figure 3. The K x-rays production cross sections versus atomic number: a) theoretical values based on relativistic HS, and b) theoretical values based on HF potential.

The present agreement between the theoretical and experimental values leads one to the conclusion that the data presented here could be of benefit to those using photon-induced x-ray fluorescence techniques for elemental analysis.

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