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Full Length Research Paper

Measurement of K_β/K_α x-ray intensity ratios and *K* to *L* shell total vacancy transfer probabilities for elements in the range $40 \leq Z \leq 50$

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The *K* shell x-ray intensity ratios K_β/K_α for 9 elements in the atomic range $40 \leq Z \leq 50$ have been determined for an excitation energy of 59.537 keV. The *K-L* total vacancy transfer probabilities (η_{KL}) for these elements have been determined, also. *K* x-rays emitted by samples were detected using a high resolution Si(Li) semiconductor detector. The measured values were compared with the theoretical values calculated using Scofield's tables based on the Hartree-Slater and Hartree-Fock theories, and available experimental values. The agreement of measured values of intensity ratios and vacancy transfer probabilities were quite satisfactory with theoretical calculations.

Key words: *K* shell intensity ratio, vacancy transfer probability, X-ray.

INTRODUCTION

Transition of an inner-shell electron from one shell to another will involve emission or absorption of high energy, short-wavelength radiation (x-rays). Usually, however, all of the energy levels of inner shells are occupied, so, in order to allow a transition to occur at all, a vacancy must be created in one of the inner shells. The energy required to do this can come from either the absorption of a sufficiently energetic photon, or from the impact with a high energy particle (Ewart, 2008).

The measurements of K_β/K_α x-ray intensity ratios are significant for comparison with theoretical predictions based on atomic models in order to test the validity of these models. The K_α x-rays arise from transitions from the *L* to the *K* shell. The K_β x-rays arise from transitions from the *M*-, *N*-, *O*-, etc. to the *K* shell. Atoms with vacancies in the inner shells are unstable and their stability can be regained by single or multiple electron transitions from the outer shells. When a single vacancy is created in an inner shell (for example, the *K* shell), the vacancy is filled up by an electron coming from some higher shell. As a result, the vacancy is shifted to the higher shell. The transfer coefficients, η_{KL} describes the mean number of vacancies produced in the *L* shell by one vacancy transfer probability and are important in many practical applications, such as nuclear electron capture, internal conversion of gamma rays, photoelectric effect, and atomic processes leading to the emission of x-rays, Auger electrons and computations for medical physics and irradiational process. X-ray fluorescence parameters such as intensity ratio and vacancy transfer probability are very important in understanding the ionization of atoms as well as for non-destructive elemental analysis in several fields such as material science, medical physics, industry and environmental science.

K to *L* shell vacancy transfer probabilities (η_{KL}) of molybdenum (Mo), Palladium (Pd), and Cadmium (Cd) have been measured by repeatedly exciting them with the *K* x-rays of Nickel (Ni) and Tin (Sn) induced by bremsstrahlung emanating from x-ray tube (Santra et al., 2005). Radiative vacancy transfer probabilities from the L_3 subshell to M-, N- and O- shells and subshells in the atomic range $72 \leq Z \leq 92$ have been obtained (Tuzluca et al., 2008). Vacancy transfer probabilities from *K* to *L* for high atomic number elements at 123.6 keV had been measured (Ertugral et al., 2005). *K* x-ray intensity ratios for Tantalum (Ta), Gold (Au), and Lead (Pb) targets by

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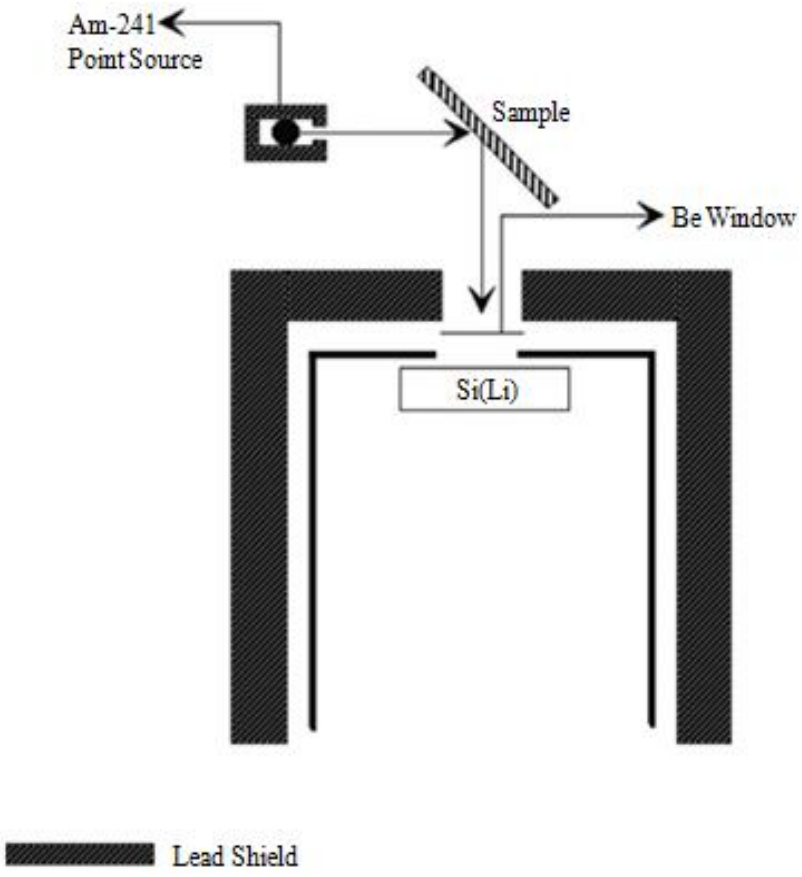

Figure 1. Experimental setup.

adopting a 2π geometrical configuration with weak radioactive ^{57}Co gamma source have been measured (Bennal and Badiger, 2006). Total vacancy transfer probabilities from *K* to *L* shell of selected elements in the atomic range 42 ≤ Z ≤ 82 have been determined using weak ^{57}Co gamma source (Bennal et al., 2010). The average vacancy distributions have been calculated by researchers (Rao et al., 1972). *K* to *L* shell and *L* to *M* shell vacancy transfer probabilities for elements in the atomic range 37 ≤ Z ≤ 42 and 18 ≤ Z ≤ 96 have been measured, respectively (Puri et al., 1993). *K* shell x-ray production cross-sections and fluorescence yields for Neodymium (Nd), Europium (Eu), Gadolinium (Gd), Dysprosium (Dy) and Holmium (Ho) have been measured using radioisotope x-ray fluorescence in the external magnetic field (Demir and Şahin, 2007).

In the present work, K_β/K_α x-ray intensity ratios, K shell fluorescence yields and radiative vacancy transfer probabilities from *K* to *L* shell for Zirconium (Zr), Niobium (Nb), Mo, Ruthenium (Ru), Rhodium (Rh), Pd, Silver (Ag), Cd, and Sn were investigated. The vacancies were produced by the 59.537 keV photons from an ^{241}Am point source radioisotope. *K* x-rays emitted by samples were detected using by a Si (Li) detector having a resolution of 180 eV full widths at half maximum at 5.9 keV.

Experimental setup and method of measurement

The experimental arrangement used for the present measurement shown in Figure 1. The parameters used for estimating $K_{\alpha,\beta}$ x-ray intensity ratios and total vacancy transfer probabilities are provided in Table 1. The samples were placed at a 45° angle with respect to the beam from the source and excited by 59.537 keV gamma rays from an ^{241}Am point source. The resultant fluorescent x-rays were detected by a Si(Li) detector (an active diameter = 6.2 mm, sensitive crystal depth = 5 mm, Be window thickness = 0.008 mm, FWHM = 180 eV at 5.9 keV) coupled to a analyzing system (Canberra-DSA1000). Spectroscopically high purity targets of Zirconium (Zr), Nb, Mo, Ru, Rh, Pd, Ag, Cd, and Sn foil samples of thickness ranging from 100 µm to 1 mm and Ru (powder sample) have been used for the measurement. A typical *K* x-ray spectrum of Zr is shown in Figure 2.

The K_α x-ray production cross-section at excitation energy are given by:

$$\sigma_{K_\alpha} = \sigma_K \omega_K f_{K_\alpha} \quad (1)$$

Where, σ_K is the total *K* shell ionization cross-section, w_K is the *K* shell fluorescence yield and $f_{K\alpha}$ is given by:

Table 1. Parameters used for estimating *K* x-ray intensity ratios and total vacancy transfer probabilities.

Sample	At. No	ρ_t (g/cm^2)	σ_K	$\beta_{K\alpha}$	$\beta_{K\beta}$
Zirconium (Zr)	40	0.163	463.77	0.9947	0.9946
Niobium (Nb)	41	0.214	528.29	0.9928	0.9927
Molybdenum (Mo)	42	0.334	557.76	0.9886	0.9885
Ruthenium (Ru)	44	0.126	663.73	0.9955	0.9954
Rhodium (Rh)	45	0.031	721.11	0.9988	0.9988
Palladium (Pd)	46	0.301	781.96	0.9889	0.9888
Silver (Ag)	47	0.105	845.96	0.9960	0.9959
Cadmium (Cd)	48	0.865	913.17	0.9677	0.9672
Tin (Sn)	50	0.552	1058.46	0.9786	0.9783

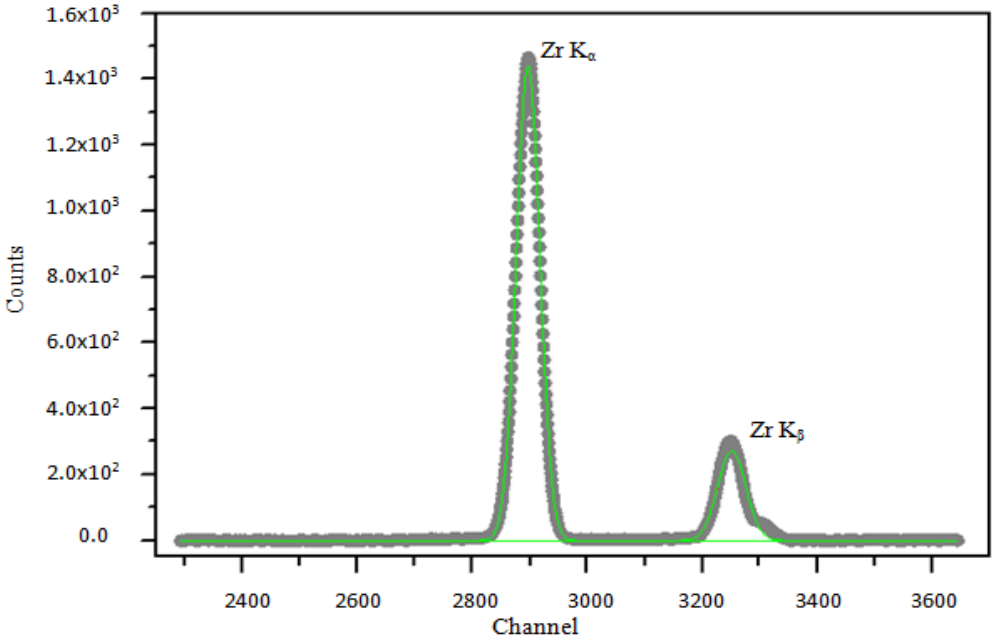

Figure 2. Typical *K* x-ray spectrum for Zr.

$$f_{Ka} = \left[1 + I_{K_\beta} / I_{K_\alpha}\right]^{-1} \quad (2)$$

The experimental K_β to K_α x-ray intensity ratios are evaluated using the following equation:

$$\frac{I_{K_\beta}}{I_{K_\alpha}} = \frac{N(K_\beta)}{N(K_\alpha)} \frac{\beta(K_\alpha)}{\beta(K_\beta)} \frac{\varepsilon(K_\alpha)}{\varepsilon(K_\beta)} \quad (3)$$

Where, $N(K_\alpha)$ and $N(K_\beta)$ are net counts observed under the peaks corresponding K_α and K_β x-rays, respectively, $\beta(K_\alpha)$ and $\beta(K_\beta)$ are the sample self-absorption corrections factors, and $\varepsilon(K_\alpha)$ and $\varepsilon(K_\beta)$ are photopeak efficiency values of the detector. The $\beta(K_\alpha)$ and $\beta(K_\beta)$ are the sample self-absorption corrections factors for the incident photon and emitted *K* x-rays photons. The values of β_K have been calculated by using the following expression obtained by assuming that the fluorescence x-rays are incident normally on the detector:

$$\beta_K = \frac{1 - \exp[-(\mu_1/\sin\theta + \mu_2/\sin\phi)t]}{(\mu_1/\sin\theta + \mu_2/\sin\phi)t} \quad (4)$$

Where, μ_1 and μ_2 are the absorption coefficients, obtained by means of a computer program named WINXCOM (Gerward et al., 2004), at the incident and emitted x-rays photon energy. The product $I_0 G \varepsilon$, containing the terms related to the incident photon flux, geometrical factor and the intrinsic absolute efficiency of the x-ray detector, was determined by collecting the K_α and K_β x-ray spectra of samples for ^{241}Am in the same geometry using equation:

Table 2. K_β/K_α x-ray intensity ratios.

Sample	Experiment		
	I_{K_β}/I_{K_α} Present	Theoretical	Other
Zr	0.195 ± 0.023	0.1913[a] 0.1737[b]	0.1898 ± 0.008[c] 0.191 ± 0.016[d]
Nb	0.197 ± 0.022	0.1772[b]	0.1993 ± 0.008[c]
Mo	0.201 ± 0.022	0.1981[a] 0.1809[b]	0.2016 ± 0.004[c] 0.193 ± 0.014[d]
Ru	0.204 ± 0.018	0.1875[b]	0.198 ± 0.016[d]
Rh	0.207 ± 0.018	0.1906[b]	0.212 ± 0.017[d]
Pd	0.211 ± 0.019	0.1933[b]	0.207 ± 0.014[d]
Ag	0.214 ± 0.022	0.1964[a] 0.1944[b]	0.2096 ± 0.004[c] 0.217 ± 0.015[d]
Cd	0.221 ± 0.024	0.1995[b]	0.2035 ± 0.006[c] 0.217 ± 0.015[d]
Sn	0.235 ± 0.033	0.2230[a] 0.2061[b]	0.2086 ± 0.011[c] 0.226 ± 0.020[d]

Sources: [a]Scofield (1974a), [b]Scofield (1974b), [c]Ertugral et al. (2007) and [d]Ertugral et al. (2001).

$$I_0 G \varepsilon_{K_i} = \frac{N_{K_i}}{\sigma_{K_i} \beta_{K_i} m_i} \quad (i = \alpha, \beta) \tag{5}$$

Where, N_{K_i} is the measured intensity, I_0 intensity of incident radiation, G a geometrical factor, ε_{K_i} detection efficiency, m_i and the thickness of target in g/cm^2 and β_{K_i} is the self- absorption correction for the target material.

The experimental *K* to *L* shell total vacancy transfer probabilities, η_{KL}, were calculated by using following equation:

$$\eta_{KL} = \frac{2 - \omega_K}{1 + (I_{K\beta}/I_{K\alpha})} \tag{6}$$

Where, ω_K is the fluorescence yield and $I_{K\beta}/I_{K\alpha}$ is the intensity ratio of the *K* x-rays. The quantities η_{KL_i} are defined as the averaged number of primary L_i subshell vacancies produced by one vacancy in the *K* shell through radiative $\eta_{KL_i}(\mathrm{R})$ and non-radiative $\eta_{KL_i}(\mathrm{A})$ transitions:

$$\eta_{KL_i} = \eta_{KL_i}(\mathrm{R}) + \eta_{KL_i}(\mathrm{A}) \tag{7}$$

The quantity $\eta_{KL_i}(\mathrm{R})$ is proportional to probability that a $K - L_i$ radiative transition takes place

$$\eta_{KL_i}(\mathrm{R}) = w_K \left[\frac{I(KL_i)}{I_K(R)} \right] \tag{8}$$

Where, $I(KL_i)$ is the $K - L_i$ x-ray intensity and $I_K(R)$ is the total intensity of *K* x-rays. The $I(KL_1)$ intensity is negligible because $K - L_1$ transitions are forbidden according to the selection rule $\Delta l = 0, \pm 1, l + l' \geq 1$ (l is the orbital angular momentum quantum number) for radiative transitions. Theoretical $\eta_{KL_i}(R)$ radiative vacancy transfer probabilities were calculated using the equation:

$$\eta_{KL_i}(R) = \frac{\Gamma_R(KL_i)}{\Gamma_K} \tag{9}$$

RESULTS AND DISCUSSION

The measured values of the K_β/K_α x-ray intensity ratios in the atomic range $40 \leq Z \leq 50$ are listed in Table 2 with the

Table 3. *K* x-ray fluorescence yields.

Sample	Experiment		
	w_K present	Theoretical	Other
Zr	0.729 ± 0.037	0.730[a] 0.734[b]	0.745 ± 0.060[c]
Nb	0.747 ± 0.038	0.747[a] 0.751[b]	0.747 ± 0.060[c]
Mo	0.765 ± 0.039	0.765[a] 0.767[b]	0.769 ± 0.011[d]
Ru	0.749 ± 0.038	0.749[a]	-
Rh	0.808 ± 0.042	0.808[a] 0.809[b]	-
Pd	0.820 ± 0.042	0.820[a] 0.820[b]	-
Ag	0.831 ± 0.043	0.831[a] 0.831[b]	0.834 ± 0.010[d]
Cd	0.843 ± 0.043	0.843[a] 0.842[b]	0.839 ± 0.067[c]
Sn	0.862 ± 0.044	0.862[a] 0.860[b]	0.858 ± 0.069[c]

Sources: [a]Krause (1979), [b]Hubbell (1989), [c]Han et al. (2007) and [d]Bennal et al. (2010).

theoretical and other experimental values. Theoretical values are based on relativistic Hartee-Fock and Hartree-Slater theories (Scofield, 1974a, b). As seen from Table 2, the agreement between the present results and theoretical prediction of Scofield is within the range 3%. From Table 2, our measured values closely agree with the values obtained from other researchers (Ertuğrul et al., 2001). The overall error in the experimental parameters is the sum of the uncertainties in different factors, namely, the evaluation of peak areas (2.25 to 6.20%), target mass thickness (1.65 to 4.60%) and statistical error (< 1.00%). Total errors affecting the experimental parameters are calculated between 2.96 to 7.78%. The errors in the evaluation of the areas under the X- and gamma-ray peaks have two main error sources, that is., the errors in the elimination of the background and in the peak fitting procedures.

The measured values of the *K* shell fluorescence yield w_K for elements in the atomic range $40 \leq Z \leq 50$ are compared with the theoretical values (Krause, 1979; Hubbell, 1989) and other experimental values (Han et al., 2007; Bennal et al., 2010) in Tables 3. As seen from Table 3, the measured w_K values increase with increasing atomic number. Also, our experimental values closely agree with the calculated values and other experimental results.

In Table 4, we present our values of *K* to *L* shell total vacancy transfer probabilities for the elements in the atomic range $40 \leq Z \leq 50$, together with theoretical and other experimental values. The error in η_{KL} values is less than 7.1% for all targets and it mainly comes from the uncertainty in w_K and K_β/K_α. As seen from Table 4, our measured values of η_{KL} agree with theoretical and other experimental values. We believe that the present experimental values may serve three purposes. Firstly, our values confirm the reliability of the existing theoretical values. Secondly, with the vacancy transfer probability values that we have obtained in this study, the elemental analysis of different samples can be calculated with confidence. Thirdly, the experimental values of these vacancy transfer probabilities can be used to calculate the experimental absorption jump factors and jump ratios of K shell, L shell-subshell. In addition, the agreement

Table 4. *K-L* total vacancy transfer probabilities.

Sample	Experiment		
	η_{KL} present	Theoretical	Other
Zr	1.063 ± 0.0011	1.064[d]	1.061 ± 0.07[c]
Nb	1.047 ± 0.0002	1.049[d]	1.044 ± 0.06[c]
Mo	1.028 ± 0.0001	1.030[d]	1.039 ± 0.005[a] 1.03 ± 0.08[b]
Ru	1.039 ± 0.0004	1.000[d]	-
Rh	0.988 ± 0.0001	0.984[d]	-
Pd	0.974 ± 0.0001	0.963[d]	0.99 ± 0.08[b]
Ag	0.962 ± 0.0002	0.958[d]	0.973 ± 0.006[a]
Cd	0.947 ± 0.0006	0.952[d]	0.964 ± 0.004[a] 0.99 ± 0.18[b]
Sn	0.925 ± 0.0008	0.933[d]	0.942 ± 0.005[a]

[a]Bennal et al. (2010), [b]Santra et al. (2005), [c]E.Öz (2006), [d]Rao et al. (1972).

between experimental and theoretical values indicates that the usage of XRF technique is beneficial for these studies.

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