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Measurements of radiative vacancy transfer probabilities from L_3 subshells to M, N and O shells and subshells in the atomic range $72 \leqslant Z \leqslant 92$

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ABSTRACT

The probabilities of vacancy transfer, η_{L3M}, ηL_3M_1, ηL_3M_4, ηL_3M_5, η_{L3N}, ηL_3N_1, ηL_3N_4, ηL_3N_5, ηL_3O_1 and $\eta L_3O_{4,5}$ from L_3 subshell to M, N and O shells and subshells for the elements Hf, Ta, W, Re, Pt, Au, HgO, Tl, Pb, Bi, Th, U have been measured using L shell fluorescence yields and X-ray intensity ratios. These experimental values were obtained from samples excited by 59.5-keV γ-rays, which were emitted from an ^{241}Am radioisotope source. Also K and L X-rays emitted from samples were measured by means of Si(Li) detector with a resolution of 155 at 5.9 keV. The results obtained from this study are compared with the results of other studies.

1. Introduction

When a vacancy is created in the L shell by either the primary excitation X-ray or by the previous event, an electron from the M or N shell "jumps in" to occupy this vacancy. In this process, atom emits a characteristic X-ray unique to this element and, in turn, produces a vacancy in the M, N and O shells (McGuire, 1971). When an atom has been ionized, an inner-shell radiative or Auger transitions occurs. Knowledge of vacancy transfer is important for the study of such processes as nuclear electron capture, internal conversion of γ-rays, photoelectric effect, characteristic X-ray production, radiative and radiationless transition probabilities (McGuire, 1971).

Söğüt (2006) has obtained the experimental values of the vacancies transfer probabilities from K to L and M shell in the Cr_xNi_{1-x} and Cr_xAl_{1-x} alloys using ^{241}Am a radioactive source. Rao et al. (1972) have acquired the theoretical values of the vacancy transfer probabilities from L_i to M, η_{LiM} ($i = 1, 2, 3$) for the elements in the atomic region $26 \leqslant Z \leqslant 92$. The values of L_i-to-M-shell vacancy transfer probabilities, η_{LiM}, for eight elements in the atomic number region $50 \leqslant Z \leqslant 90$ were reported by (McGuire, 1971). Puri et al. (1993a, b) have evaluated K- to L-shell and L- to M-shell vacancy transfer probabilities for elements in the atomic number range $37 \leqslant Z \leqslant 42$ and $18 \leqslant Z \leqslant 96$, respectively. In addition, Puri et al. (1993b) have computed values and fitted them versus atomic number Z values and fitted coefficients for vacancy transfer probabilities of K to L_1, K to L_2, K to L_3, K to L (average), L_1 to M, L_2 to M and L_3 to M shells. Ertugrul et al. (1997a) have measured the radiative vacancy transfer probabilities from the K to L_2, L_3 subshell and to M shell in the atomic regions $69 \leqslant Z \leqslant 92$. Scofield (1974a) has calculated radiative decay rates of L-shell vacancy states using relativistic Hartree–Fock solutions for a set of elements $Z = 18$–94 in their initial and final states. Besides, Scofield (1974b) has made the same calculations for K X-ray emission rates. The K and L X-ray emission rates in the atomic number range $5 \leqslant Z \leqslant 104$ were calculated using relativistic Hartree–Slater theory by Scofield (1974c). Bonzi (2006) has determined the radiative vacancy transfer probabilities from L_3 to M shell, η_{L3M} (R) and L_3 to N shell, η_{L3N} (R) for W, Re and Pb. Şimsek (2002) has reported the probabilities, η_{L3J} (R) (J = M, N and higher shells), for Pb, Th and U, deduced using the measured L_3 subshell X-ray intensities from the targets excited by K X-ray of Rb, Nb and Mo secondary exciters used in combination with ^{241}Am source. The method used for the estimation of additional contribution to the L_3 subshell X-rays due to excitation by the scattered 59.5-keV photons is not trustworthy (Kumar et al., 2005). Kucukonder et al. (2004) have measured L X-ray fluorescence cross-sections, and intensity ratios for elements in the atomic range $70 \leqslant Z \leqslant 92$ at the excitation energy 59. 5 keV using a Si (Li) detector. Therefore, there are a lot of works concerning with this area (Sharma et al., 2005; Ertugrul, 2001, Ertugrul, 2003; Dogan and Ertugrul, 2004 and Ozdemir et al., 2005). K shell X-ray production cross-section and K to L-and M-shell radiative vacancy transfer probabilities for Nd, Eu, Gd, Dy and Ho at excitation with 59.5-keV photons in an external magnetic field have been measured by Demir and Şahin (2007).

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In this study, the vacancy transfer probabilities from L_3 subshell to M, N and O shells and subshells for the elements Hf, Ta, W, Re, Pt, Au, HgO, Tl, Pb, Bi, Th, U have been evaluated using L shell fluorescence yields and X-ray intensity ratios.

2. Experimental procedure

A typical experimental setup are shown in Fig. 1. The samples were excited with 59.5-keV γ-photons emitted from a 75 mCi ^{241}Am source. Spectroscopically pure samples of Hf, Ta, W, Re, Pt, Au, HgO, Tl, Pb, Bi, Th, U of thickness ranging 37×10^{-3}–$553\times10^{-3}\,\mathrm{g\,cm^{-2}}$ have been used. The purity of commercially obtained specimens was better than 99.9%. In order to minimize the matrix effects, all powder samples were sieved to 400 mesh and supported on Mylar film. The L X-rays emitted from the samples were detected with a Si(Li) detector (FWHM = 155 eV at 5.96 keV, active area = 12.5 mm^2, sensitivity depth = 3.5 cm, Be window thickness = 12.5 μm) and coupled to S 100 card with ADC and spectroscopy amplifier. The L X-rays were counted at the emission angle of 90° because the anisotropic emissions of L_3 X-rays (L_l, L_α and $L_{3\beta}$) at angles smaller and greater than 90° are shown to compensate one another (Mann et al., 1994; Ertugrul et al., 1997b). The experimental intensity ratios have been obtained by using the following equations (Baydaş et al., 2001).

$$\frac{IL\beta}{IL\alpha}=\frac{N(L\beta)\varepsilon(L\alpha)\beta(L\alpha)}{N(L\alpha)\varepsilon(L\beta)\beta(L\beta)} \tag{1}$$

$$\frac{IL\beta}{IL\iota}=\frac{N(L\beta)\varepsilon(L\iota)\beta(L\iota)}{N(L\iota)\varepsilon(L\beta)\beta(L\beta)} \tag{2}$$

$$\frac{IL\iota}{IL\alpha}=\frac{N(L\iota)\varepsilon(L\alpha)\beta(L\alpha)}{N(L\alpha)\varepsilon(L\iota)\beta(L\iota)} \tag{3}$$

$$\frac{IL\iota}{IL\beta}=\frac{N(L\iota)\varepsilon(L\beta)\beta(L\beta)}{N(L\beta)\varepsilon(L\iota)\beta(L\iota)} \tag{4}$$

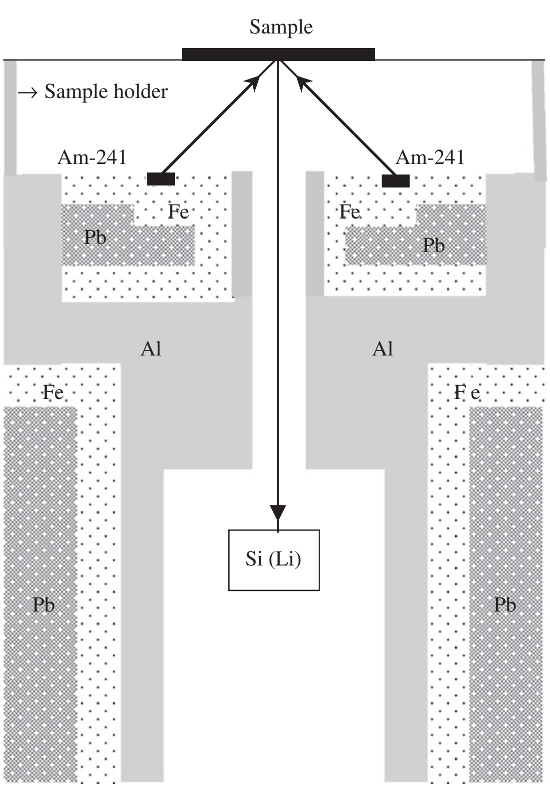

Fig. 1. A typical experimental setup.

$NL\beta/NLi$ ($i=\alpha,\iota$) represents the ratio of the counting rates under the $L\alpha$ and Li peaks. $\beta L\beta/\beta Li$ ($i=\alpha,\iota$) is the ratio of the self-absorption correction factor of the target for $L\beta$ and Li and $\varepsilon L\beta/\varepsilon Li$ ($i=\alpha,\iota$) is the ratio of the detector efficiency values of $L\beta$ and Li X-rays, respectively. The other ratios have the same meaning as mentioned above. In addition, the L_3 subshell cross-section and fluorescence yield have been calculated using the following equations (Ertugrul et al., 1997a):

$$\sigma_{L_i}=\frac{I_{L_i}}{I_0G\varepsilon_{L_i}\beta_{L_i}t}\quad(i=\iota,\alpha,\beta) \tag{5}$$

$$\sigma^x_{L_3}=\frac{\sigma_{L\alpha}}{F_{3\alpha}} \tag{6}$$

To calculate the L_3 subshell fluorescence yield (Ertugrul et al., 1997a) the following equation was used:

$$\omega_3=\frac{\sigma^x_{L_3}}{\sigma_3+\eta_{KL_3}\sigma_K+(\sigma_2+\eta_{KL_2}\sigma_K)f_{23}+(\sigma_1+\eta_{KL_1}\sigma_K)(f_{13}+f_{12}f_{23})} \tag{7}$$

But the stimulating source cannot excite K shell of sample. Therefore, at this state the following equation has been used (Dogan and Ertugrul, 2004):

$$\omega_3=\frac{\sigma^x_{L_3}}{\sigma_3+\sigma_2f_{23}+\sigma_1(f_{13}+f_{12}f_{23})} \tag{8}$$

The experimental radiative vacancy transfer probabilities from L_3 subshell to M, N and O subshells have been obtained by using the following equations (Dogan and Ertugrul, 2004):

$$\eta_{L_3M}=\omega_3\left[\left(1+\frac{I_{L\alpha}}{I_{L\iota}}+\frac{I_{L\beta}}{I_{L\iota}}\right)^{-1}+\left(1+\frac{I_{L\iota}}{I_{L\alpha}}+\frac{I_{L\beta}}{I_{L\alpha}}\right)^{-1}\right] \tag{9}$$

$$\eta_{L_3N}=\omega_3\left(1+\frac{I_{L\iota}}{I_{L\beta}}+\frac{I_{L\alpha}}{I_{L\beta}}\right)^{-1}\left[\left(1+\frac{I_{L\beta_{2,15}}}{I_{L\beta_6}}+\frac{I_{L\beta_{5,7}}}{I_{L\beta_6}}\right)^{-1}+\left(1+\frac{I_{L\beta_6}}{I_{L\beta_{2,15}}}+\frac{I_{L\beta_{5,7}}}{I_{L\beta_{2,15}}}\right)^{-1}\right] \tag{10}$$

$$\eta L_3M=\omega_3\left[1+\frac{IL_\iota}{IL_\beta}+\frac{IL_{3\alpha}}{IL_1}\right]^{-1} \tag{11}$$

$$\eta L_3M_4=\omega_3\frac{\Gamma L_{\alpha2}}{\Gamma L_\alpha}\left[1+\frac{IL_l}{IL_\alpha}+\frac{IL_{3\beta}}{IL_\alpha}\right]^{-1} \tag{12}$$

$$\eta L_3M_5=\omega_3\frac{\Gamma L_{\alpha1}}{\Gamma L_\alpha}\left[1+\frac{IL_l}{IL_\alpha}+\frac{IL_{3\beta}}{IL_\alpha}\right]^{-1} \tag{13}$$

$$\eta L_3N_1=\omega_3\frac{\Gamma L_{\beta6}}{\Gamma L_{3\beta}}\left[1+\frac{IL_\alpha}{IL_{3\beta}}+\frac{IL_l}{IL_{3\beta}}\right]^{-1} \tag{14}$$

$$\eta L_3N_4=\omega_3\frac{\Gamma L_{\beta15}}{\Gamma L_{3\beta}}\left[1+\frac{IL_\alpha}{IL_{3\beta}}+\frac{IL_l}{IL_{3\beta}}\right]^{-1} \tag{15}$$

$$\eta L_3N_5=\omega_3\frac{\Gamma L_{\beta2}}{\Gamma L_{3\beta}}\left[1+\frac{IL_\alpha}{IL_{3\beta}}+\frac{IL_l}{IL_{3\beta}}\right]^{-1} \tag{16}$$

$$\eta L_3O_1=\omega_3\frac{\Gamma L_{\beta7}}{\Gamma L_{3\beta}}\left[1+\frac{IL_\alpha}{IL_{3\beta}}+\frac{IL_l}{IL_{3\beta}}\right]^{-1} \tag{17}$$

$$\eta L_3O_{4,5}=\omega_3\frac{\Gamma L_{\beta5}}{\Gamma L_{3\beta}}\left[1+\frac{IL_\alpha}{IL_{3\beta}}+\frac{IL_l}{IL_{3\beta}}\right]^{-1} \tag{18}$$

where ω_3 is L_3 subshell fluorescence yield and IL_i/IL_j is the ratio of intensities of L_i and L_j X-rays (Dogan and Ertugrul, 2004). In addition, $IL_{3\beta}$, $IL_{\beta2}$, $IL_{\beta5}$, $IL_{\beta7}$, $IL_{\beta7'}$, $IL_{\beta15}$ and $IL_{\beta6}$ have been semi-empirically computed by the following equations (Dogan and