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# Determination of average number of *M* shell vacancies produced on decay of an $L_3$ subshell vacancy in Th and U

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**Abstract** : The average number of *M* shell vacancies $\bar{n}_{L3M}$ produced on the decay of an $L_3$ subshell vacancy in Th and U has been determined from the measurement of *M* X-ray intensity in photon induced fluorescent X-ray emission spectra of Th and U respectively. $L_3$, *M* and higher subshell electrons of Th and U are selectively photoionized, in turn and the number of the resultant *M* shell vacancies is determined from the measured *M* X-ray intensity corrected for *M* shell fluorescence yield. To the best of our knowledge the experimental determination of the quantity has been made for the first time.

**Keywords** : Fluorescent X-rays, *M* shell vacancies, photoionization

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## 1. Introduction

The intensity of *M* X-rays in photon induced fluorescent X-ray emission spectrum of an element depends upon the total number of *M* shell vacancies resulting from the interaction of the photons with the atoms of the elements and the *M* shell fluorescence yield. When $L_3$, *M* and higher subshell electrons of the element are selectively photoionized by proper choice of photon energy, the total number of resultant vacancies produced in *M* shell is the sum of the vacancies produced by direct photoionization of *M* shell electrons and decay of $L_3$ subshell vacancies to *M* shell through radiative and non-radiative transitions. Information about the production of vacancies in *M* shell from the decay of vacancies in $L_3$ subshell, can thus be obtained by subtracting the calculated contribution of *M* shell vacancies produced by direct photoionization of *M* shell electrons from the measured total number of *M* shell vacancies produced by selective photoionization of $L_3$, *M* and higher shell electrons. The method and the results of the measurement of the average number of *M* shell vacancies produced on decay of an $L_3$ subshell vacancy in Th and U are presented in this paper.

## 2. Method of measurement

The $K$ conversion X-rays of weighted mean energies [1] 16.896 and 17.781 keV are produced by irradiating, in turn, primary targets ($P$) of Nb and Mo with a collimated beam of 59.57 keV gamma rays obtained from $^{241}$Am radioactive source ($R$) and used to irradiate further secondary targets ($S$) of Th and U, respectively. All the components $K_{\alpha1}$, $K_{\alpha2}$, $K_{\beta1}$, $K_{\beta2}$ *etc* of $K$ conversion X-rays of Nb and Mo have energies between the $L_3$ and $L_2$ edge energies of Th and U, respectively. Therefore, $K$, $L_1$ and $L_2$ subshell electrons are not ionized but $L_3$, $M$ and higher shell electrons in each of the Th and U targets are ionized. The photoionization of $L_3$ and $M$ subshell electrons give rise to vacancies in $L_3$ and $M$ subshells. Most of the $L_3$ subshell vacancies decay to $M$ shell and add to $M$ shell vacancies produced by direct photoionization of $M$ shell electrons. The total number of the resultant $M$ shell vacancies produced both by direct photoionization of $M$ shell electrons and decay of $L_3$ subshell vacancies to $M$ shell in Th and U is determined by measuring the intensity of $M$ X-rays emitted from the Th and U targets, respectively corrected for the respective $M$ shell fluorescence yields.

The experimental arrangement of $^{241}$Am source ($R$), primary target ($P$), secondary target ($S$) and detector ($D$) is shown in Figure 1. Graded shielding of Pb, Fe and Al was so arranged that the source ($R$) and detector ($D$) can only see the primary target ($P$) and secondary target ($S$), respectively. The source ($R$) cannot see the secondary target ($S$) and detector ($D$) directly. The primary target ($P$) and secondary target ($S$) can see each other. 59.57 keV gamma rays from ≈ 1 Curie $^{241}$Am source purchased from Radio Chemical Centre, England were collimated on self-supporting primary targets of Nb and Mo and radiations emitted from them were collimated on the secondary target of Th and U. All the targets were in the form of circular discs of 4 cm diameter. The targets of Mo, Th and U were metallic foils purchased from Reactor Experiments Inc USA. However, self-supporting target of Nb was prepared using Nb metal powder with a technique described in an earlier paper [2]. The $M$ shell fluorescent X-rays emitted from Th and U secondary targets were counted by a Si(Li) detector with active diameter 10 mm and sensitive depth 4.66 mm of crystal and Be window of thickness 0.0254 mm coupled to an ND 600 multichannel analyzer. The resolution of the Si(Li) X-ray spectrometer was 170 eV at 5.9 keV.

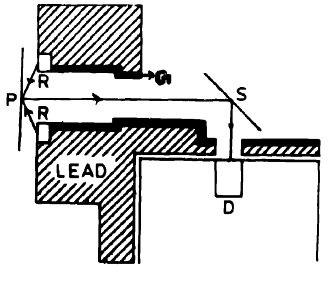


**Figure 1.** Schematic diagram of the experimental arrangement used in present measurements. R–Radioactive source $^{241}$Am, P–Primary target, S–Secondary target, D–Si(Li) X-ray detector, G–Graded shielding of Al and Fe.

Typical spectra of radiations emitted from secondary target U with Mo and equivalent Al primary targets are shown by curves A and B, respectively in Figure 2. The various peaks shown in spectra are labelled. The individual lines of the *M* and *L* shell X-rays of U are not resolved due to the limited resolution of the spectrometer but well known groups of *M* and *L* X-ray lines appear under familiar peaks. Peaks due to scattering of Mo *K* X-rays from U target are also seen. 59.57 keV gamma rays which are scattered from primary targets of Mo and equivalent Al are again scattered from secondary target U and appear under the last two peaks in the spectra A and B. The first peak corresponds to incoherent scattering and the second to coherent scattering. The matching of the scattering peaks in the spectra A and B shows that the gamma ray scattering from Mo and equivalent Al primary targets is almost same. *M* X-ray peaks are well resolved from all other peaks. The difference spectrum A – B = C is also shown in the Figure 3. It consists of peaks corresponding to *M*, $L_l$, $L_\alpha$ and $L_\beta$ groups of X-rays emitted from the U target when it is irradiated with 17.781 keV external conversion X-rays of Mo only. The $L_\gamma$ X-ray peak is missing showing that $L_1$ and $L_2$ subshell electrons are not ionized because firstly the $L_1$ and $L_2$ edge energies of U are higher than 17.781 keV and secondary the contribution due to gamma ray scattering from Mo

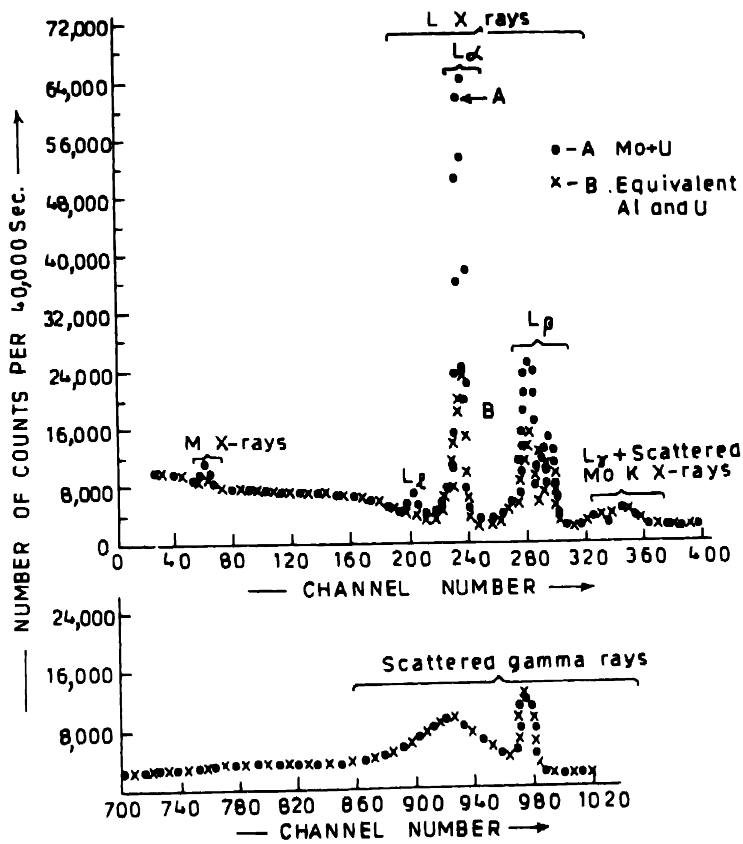


**Figure 2.** Spectra recorded with Si(Li) detector : A-Mo primary and U secondary target; B-equivalent Al primary and U secondary target.

which has energy higher than the $L_1$ and $L_2$ edge energies of U is adequately compensated by the use of equivalent Al scatterer. The counts under the $M$ X-ray peaks seen in the difference spectrum A – B correspond to the intensity of $M$ X-rays emitted from the U target when it is irradiated with $K$ conversion X-rays of Mo of weighted mean energy 17.781 keV only.

The experimental cross sections for the emission of $M$ X-rays produced by the photoionization of $M$ and $L_3$ subshell electrons was determined from the relation :

$$\sigma_M^{*} = \frac{N_M M_M}{t_M \beta_M \left[N_K(P) w \varepsilon_M N/4\pi\right]}, \tag{1}$$

where $N_M$ is the number of counts per unit time under the peaks due to $M$ X-rays, $M_M$ is the atomic weight of the secondary target element, $t_M$ is the thickness of the secondary target, $N_K(P)$ is the number of $K$ X-rays of primary target incident on the secondary target in unit time, $w$ is the secondary target-detector solid angle, $\varepsilon_M$ is the photopeak detection efficiency of the detector at the energy of $M$ X-rays, $N$ is the Avogadro's number and $\beta_M$ is the self-absorption correction factor which accounts for the absorption of the incident primary $K$ X-rays and emitted secondary $M$ X-rays in the secondary target. The target self-absorption correction factor $\beta_M$ [3] was calculated from the relation :

$$\beta_M = \sum_{i=1}^{n} P_i \left[\frac{1 - \exp\left[-\left(\mu_K + \mu_M(x_i)\right)\right] t_M/\cos\theta}{\left[\mu_K + \mu_M(x_i)\right] t_M/\cos\theta}\right], \tag{2}$$

where $\mu_K$ is the absorption coefficient of the secondary target element at the weighted mean energy of incident $K$ conversion X-rays of primary target element, $\mu_M(x_i)$ is the absorption coefficient of the secondary target element at the energy of the $i$-th component of fractional

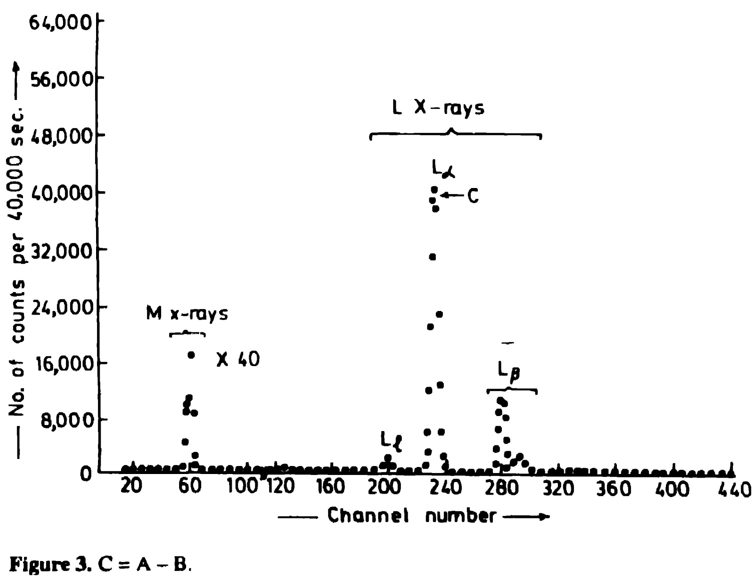


**Figure 3.** C = A – B.

intensity $P_i$ in a mixture of $n$ components of emitted $M$ X-rays and $\theta$ is the angle of incidence of $K$ X-rays as well as angle of emergence of $M$ X-rays which is equal to 45° in the present experiment. For this purpose the values of absorption coefficients generated from XCOM computer program of Berger and Hubbell [4] and fractional intensities of Jenkins [5] were used.

The value of $N_M$ were determined from the area under the peaks corresponding to $M$ X-rays of secondary target in the difference spectrum in Figure 3. Sufficient number of runs for 40,000 sec were made for each combination of primary and secondary targets (Mo-U, Eq.Al-U, Nb-Th, Eq.Al-Th) so as to achieve statistical accuracy of $\approx 2\%$ in counting rates of $M$ X-rays. The value of the factor $[N_K(P)\, w\, \varepsilon_M\, N/4\pi]$ which contains terms relating to the flux of primary target $K$ conversion X-rays incident on the secondary target, geometrical factor and absolute efficiency of X-ray detector was determined by irradiating targets of S, Cl, K, Ca, Ti and V with $K$ conversion X-rays of Nb and Mo in the same experimental set-up and counting fluorescent $K$ X-rays emitted in each case with the same spectrometer. The number of $K$ X-rays $N_K(S)$ emitted from the targets as counted under the photopeak per unit time is given by a relation similar to (1) above which can be rewritten as

$$\frac{N_K(P)\, w\, \varepsilon_K\, N}{4\pi} = \frac{N_K(S)\, M_K}{t_K\, B_K\, \sigma_K^*}. \tag{3}$$

All the terms in eq. (3) have the same meaning as in eq. (1) except that they correspond to $K$ X-rays instead of $M$ X-rays. The $K$ X-rays production cross sections $\sigma_K^*$ were calculated from theoretical values of $K$ shell photoionization cross sections [6] and $K$ shell fluorescent yields [7]. Using measured values of $N_K(S)$ and calculated values of $\sigma_K^*$ and $\beta_K$, the values of factor $[N_K(P)\, w\, \varepsilon_K\, N]/4\pi$ were determined at weighted mean energies [1] of $K$ X-rays of elements $16 \le Z \le 23$. The values of this term were then interpolated at the weighted average energies [5] of $M$ X-rays of U and Th to calculate the values of the average cross sections $\sigma_M^*$ for the production of $M$ X-rays due to direct photoionization of $M$ shell electrons and decay of $L_3$ subshell vacancies to $M$ shell.

Evidently, we have

$$\sigma_M^* = \sigma_M^P\, \bar{w}_M + \sigma_{L3}^P\, \bar{n}_{L3M}\, \bar{w}_M, \tag{4}$$

where $\sigma_M^P$ and $\sigma_{L3}^P$ are the total $M$ shell and $L_3$ subshell photoionization cross sections respectively, $\bar{n}_{L3M}$ is the average number of total $M$ shell vacancies produced on decay of $L_3$ subshell vacancy and $\bar{w}_M$ is the average $M$ shell fluorescence yield. The first term in eq. (4) corresponds to $M$ X-ray emission by direct photoionization of $M$ shell electrons while the second term gives $M$ X-ray emission due to decay of $L_3$ subshell vacancy to $M$ shell. The measurement of $\sigma_M^*$ and knowledge of $\sigma_M^P$, $\sigma_{L3}^P$ and $\bar{w}_M$ allows the determination of $\bar{n}_{L3M}$ from eq. (4).

## 3. Results and discussion

The measured values $\sigma_M^*$ of the total $M$ X-ray production cross sections following photoionization of $M$ and $L_3$ subshell electrons in Th and U by photon of weighted mean energies 16.896 and 17.781 keV respectively are given in column 3 of Table 1. Theoretical

**Table 1.** Measured values of average number of $M$ shell vacancies on decay of an $L_3$ subshell vacancy, $\bar{n}_{L3M}$, compared with available data.

| Target elements | Energy (keV) | $\sigma_M^*$ ($b$ / atom) | $\sigma_M^P$ ($b$ / atom) | $\sigma_{L3}^P$ ($b$ / atom) | $w_M$ | $n_{L3M}$ |
|---|---|---|---|---|---|---|
| Th | 16.896 | 2090 ± 160 | 1.23E + 04 | 2.29E + 04 | 0.044 ± .004[a] | 1.53 ± 0.21[d] |
| | | | | | 0.045[b] | 1.23[c] |
| U | 17 781 | 2210 ± 110 | 1.17E + 04 | 2.14E + 04 | 0.051 ± .005[a] | 1 48 ± 0.21[d] |
| | | | | | 0 050[b] | |

(a) Experimental values of $\bar{w}_M$ of Shatendra *et al* [8], (b) Semi-empirical values of Hubbell [7], (c) Theoretical values of $\bar{n}_{L3M}$ of McGuire [9], (d) Present experimental values of $\bar{n}_{L3M}$

values of $\sigma_M^P$ and $\sigma_{L3}^P$, the photoionization cross sections of $M$ shell and $L_3$ subshell, respectively, are given in columns 4 and 5. In column 6 semi-empirical [7] as well as experimental values [8] of average $M$ shell fluorescence yields $\bar{w}_M$ are listed. The values of $\bar{n}_{L3M}$ as determined from eq. (4) are compared with the available theoretical values [9] in last column of Table 1.

The overall error in the measured values of $\sigma_M^*$ cross sections is estimated to be less than 8% which arises due to uncertainties in the various physical parameters required to evaluate the experimental results using eq. (1). The uncertainty in all the parameters are listed in Table 2. Theoretical values of $M$ and $L_3$ subshell photoionization cross sections

**Table 2.** Details of uncertainties involved in the various quantities used for the evaluation of $M$ X-ray emission cross sections.

| Sl. No. | Quantity | Nature of uncertainty | uncertainty |
|---|---|---|---|
| 1 | $N_M$ | Statistical and other possible errors in area evaluation | ≈ 2% |
| 2. | $\beta_M$ | Due to errors in the absorption coeff. at incident and emitted photon energies and in the measurement of target thicknesses | ≈ 5% |
| 3. | $\frac{N_K(P)\, w N \varepsilon_K}{4\pi}$ | Statistical, error in area evaluation, errors in absorption coeff. at incident and emitted photon energies and in the values of $\sigma_K$ and $w_K$ and target thickness measurements | ≈ 5% |

**Table 2.** *(Cont'd.)*

| Sl. No. | Quantity | Nature of uncertainty | uncertainty |
|---|---|---|---|
| 4. | $\sigma^P_{L3/M}$ | Errors in the calculated values of $L_3$ and total $M$ shell photo-ionization cross section | ≈ 0.1% |
| 5. | $\bar{w}_M$ | Errors as quoted in the measured values of average $M$ shell fluorescent yield | ≈ 9% |

have been interpolated from latest available tabulations [6] which have a calculational error of less than 0.1%. Experimental values of $M$ shell and $L_3$ subshell photoionization cross sections for some elements and photon energies have also been shown [10–12] to agree with theoretical calculations within experimental errors of 5–10%. The overall error in the measured values of $\bar{n}_{L3M}$ is estimated to be ≈ 14%. No other experimental data on measurement of $\bar{n}_{L3M}$ is available in literature. However, theoretically calculated value of the parameter, which is available in case of Th only, has been found to be 20% lower than that of the presently determined values. Keeping in view the scaricity of the experimental and theoretical data for these parameters and their need for analysis of $M$ X-ray spectra when $L$ shell is also ionized along with $M$ shell, more determinations of these have to be made. Measurements of these parameters can be fruitfully made over a wider 'Z' range using Synchrotron radiations.

## References

[1] E Strom and I Israel *Nucl. Data Tables* **A7** 565 (1970)

[2] K L Allawadhi, S K Arora and B S Sood *Pramana* **10** 511 (1978)

[3] S K Arora, K L Allawadhi and B S Sood *Pramana* **16** 467 (1981)

[4] M J Berger and J H Hubbell *NBSIR Report No. 87-3597* (Department of Commerce, Gaithersburg MD-20899, USA) (1987)

[5] R Jenkins *An Introduction to X-ray Spectroscopy* (London : Heyden) (1976)

[6] J H Scofield *Lawerence Livermore Laboratory Report* **No. 51326** (1973)

[7] J H Hubbel *NISTIR Report No. 89-4144* (US Department of Commerce, Gaithersburg MD-20899, USA) (1989)

[8] K Shatendra, K L Allawadhi and B S Sood *Physica* **124C** 279 (1984)

[9] E J McGuire *Phys. Rev.* **A3** 587 (1971)

[10] S K Arora, K L Allawadhi and B S Sood *J Phys. Soc. Jpn.* **50** 251 (1981)

[11] K Shatendra, N Singh, R Mittal, K L Allawadhi and B S Sood *X-ray Spectrometry* **14** 195 (1985)

[12] K S Mann, N Singh, R Mittal, K L Allawadhi and B S Sood *J. Phys.* **B23** 2497 (1990)