X-ray fluorescence in some rare earth and high Z elements excited by 661.6 keV γ-rays

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Abstract. The K-shell X-ray fluorescence cross sections are determined experimentally for 10 elements such as Pb, Hg, Ir, W, Lu, Tm, Dy, Tb, Gd and Nd at excitation energy of 661.6 keV associated with γ-rays of ^{137}Cs radioisotope. The technique employed involves the measurement of total intensity of fluorescent K X-rays that follow the photoeffect absorption of a known flux of γ-rays using a well type NaI(Tl) detector. The obtained results are compared with the available theoretical values and other measured values.

Keywords. Photoeffect; X-ray fluorescence cross section; K-shell fluorescence yield.

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1. Introduction

Studies on the interaction of radiation with matter provide a wealth of valuable information on radiation shielding and biological applications. The X-ray fluorescence (XRF) cross section and X-ray fluorescence yield values for different elements at various photoionization energies are required in a variety of applications including atomic, molecular and radiation physics studies, X-ray fluorescence surface chemical analysis, dosimetric computations for health physics, cancer therapy and industrial irradiation processing (Hubbell *et al* [1]). Quantitative knowledge of the emission of characteristic K-radiation is still of great interest for both fundamental and applied physics.

Vacancies in the shells can be produced by charged particle impact, photoionization, internal conversion, orbital electron capture or by higher order effects in nuclear decay. In the present method, the K-shell vacancies are created by photons and filled by outer electrons leading to the emission of K X-rays. Estimation of these fluorescent X-rays that follow the photoeffect interaction provides a measure of the probability of the interaction.

Some theoretical studies on K-XRF cross sections for several elements at low excitation energies have been reported. Krause *et al* [2] have calculated theoretically K-XRF cross sections for elements with $5 \leq Z \leq 101$ at photon energies ranging from 10 to 60 keV. Puri *et al* [3] have calculated K and L shell XRF cross sections for elements with $13 \leq Z \leq 92$

and $35 \leq Z \leq 92$ respectively at 1 to 200 keV incident photon energy range. McGuire [4], Walters and Bhalla [5] and Chen *et al* [6] obtained theoretical values of K-shell fluorescence yield (ω_K) for various elements by using different approaches. An annotated bibliography of measurements, analysis, fits and tables of K, L and higher atomic shell X-ray fluorescence yield, from 1978 to 1993 were presented by Hubbell *et al* [1]. They obtained values of ω_K for the elements $11 \leq Z \leq 99$ by fitting the available experimental data and compared these values with the data based on theoretical models presented by Bambynek *et al* [7] and Krause *et al* [8]. Later, in 1984, Bambynek *et al* [9] in their review article presented a further re-evaluation of ω_K values incorporating new measurements. Some measurements of K-XRF cross sections for low and medium Z elements at low excitation energies have been reported (see [10–16]). Several measurements of K as well as higher shell fluorescence yields have been reported [17,18]. The data cover several elements at incident photon energies below 300 keV. In spite of these, data are very scarce in the rare earth as well as high Z element region at higher excitation energies. Therefore, we felt it worthwhile to measure the intensity of K-shell fluorescent X-rays emitted as a result of photoeffect interaction of 661.6 keV γ-rays with the K-shell electrons of experimental target elements Pb, Hg, Ir, W, Lu, Tm, Dy, Tb, Gd and Nd using a well type NaI(Tl) scintillation detector.

2. Experimental details

The experimental set up for the measurement of spectral intensity of X-rays is shown in figure 1. A 10 mCi ^{137}Cs source in the form of radiographic capsule emitting 661.6 keV γ-rays was obtained from Bhabha Atomic Research Centre, Mumbai, India. A well-collimated beam of 661.6 keV photons was incident on the target placed inside the well of the detector. The apertures of the collimating system, source and the detector are all adjusted to be in the same coaxial line for obtaining good geometry condition thereby avoiding the external secondary radiation from nearby objects and the interfering effects.
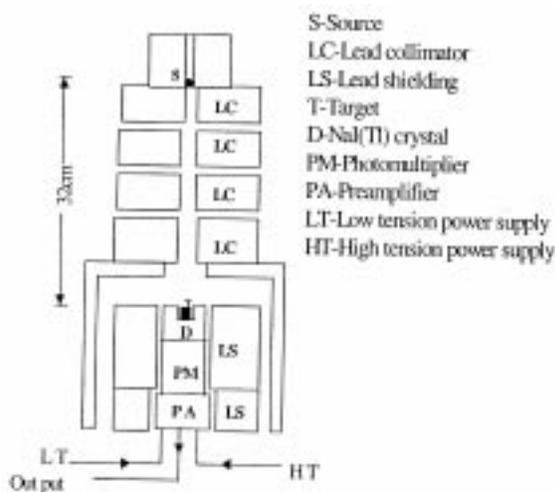

Figure 1. Experimental setup (not to scale).

Targets were prepared by using spectroscopically pure compounds $Pb(HCOO)_2$, $HgCl_2$, $IrCl_3$, $NaWO_4$, Lu_2O_3, Tm_2O_3, DY_2O_3, Tb_4O_7, Gd_2O_3 and Nd_2O_3 of thickness ranging from 124.4 to 295.2 mg/cm^2. Each compound in fine powder form was filled in cylindrical plastic containers. The K X-ray spectra from various targets for different masses of the compounds were recorded with a scintillation spectrometer consisting of a $1\frac{3}{4}''$ diameter $\times$ 2'' thick well type NaI(Tl) crystal. The well had a diameter of 16.6 mm and a depth of 39.2 mm. The detector was optically coupled to a suitable photomultiplier tube. The detector resolution was 70 keV at 661.6 keV. The detector signal was amplified by a suitable linear amplifier and the spectrum was analysed in a 1024 channel EG&G ORTEC 7150 model multichannel analyser. The detector was sufficiently shielded to reduce the radiation from the background source reaching the detector. The well type detector provides nearly 4π geometry and in this geometry, the detector measures the radiation from all sides of the target, which has the effect of increasing the detection efficiency of low energy photons emitted from the target. Also the NaI(Tl) crystal provides relatively high Z and high density with highest currently available luminescent efficiency. Hence it reduces the scattering of the incoming radiation which favours the complete absorption of γ-rays. The spectrum recorded was corrected for the contribution from the target container and surrounding air by recording another spectrum without target in position and subtracting it from the former. A typical resolved K X-ray spectrum of Hg from the $HgCl_2$ sample is shown in figure 2. The area under the peak, obtained by fitting the Gaussian, gives the total number of K X-rays. Since the sample thicknesses are considerably large, the X-rays emitted may be absorbed by the sample itself. Therefore, in order to correct for the absorption in the sample, an extrapolation technique has been employed using five different masses of the same sample. Then the intensity of X-rays per unit mass was plotted against the corresponding masses of the sample. This was found to be best represented by a second order polynomial. Using this best fit, this was extrapolated to zero mass. Thus, from the extrapolated value, the number of K X-rays per atom was obtained. The number of atoms of the element of interest was obtained by considering the weight fraction of that element in the compound.

To determine the number of photons that are incident on the sample a separate experiment was conducted using scintillation spectrometer consisting of a $1\frac{3}{4}''$ diameter $\times 2''$ thick well type NaI(Tl) crystal. The spectrum of the source ^{137}Cs used in the present work was recorded in a good geometry setup. The photoelectric events due to 661.6 keV γ-rays were recorded and this number was corrected for photopeak efficiency and geometric efficiency by determining the peak to total ratio experimentally. The intrinsic efficiency for the crystal was obtained using a ^{137}Cs standard source of known strength. This was further corrected for the geometry of the set up.

3. Results and discussion

The experimental K X-ray fluorescence cross sections σ_K^x for a given element at an energy E were calculated using the relation

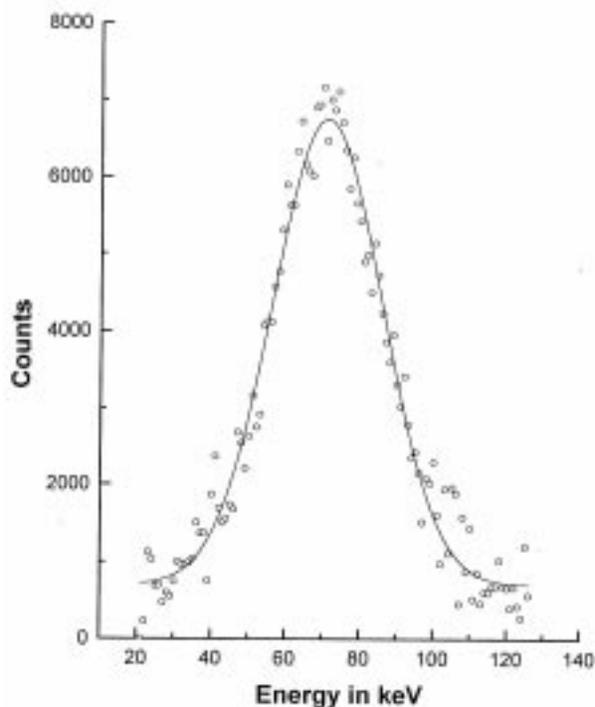$$\sigma_K^x(E) = \frac{N_K}{NSG\varepsilon_K}, \tag{1}$$

Figure 2. Typical resolved K-XRF spectrum of Hg.

where N_K is the count rate per unit area under K X-ray peak of the given element, S is the source strength, G is the geometry factor, ε_K is the efficiency of the detector for K X-rays and N is the number of atoms per unit area of the target, calculated using the relation

$$N = \frac{mA}{M}. \tag{2}$$

Here m and M are mass per unit area and atomic weight of the target respectively and A is the Avogadro number. The mass of the target was determined by using an analytical balance correct to 1 mg. The area of each sample was determined by measuring the inner diameter of the corresponding containers using a travelling microscope. The mass and area so determined were then used to calculate the mass per unit area m of each sample.

The K-XRF cross sections determined by the present method for different elements at excitation energy of 661.6 keV are listed in table 1. A plot of the present values of cross section versus the atomic number of the elements is shown in figure 3. It was found by the polynomial regression that the trend of variation correspond to a second order polynomial fit in Z. The experimental errors are to the extent of 3% to 4%. The error due to counting statistics was kept below 0.3% by accumulating 10^5–10^6 counts within the photopeak. Apart from the counting statistics, the other possible sources of error in the present method are: (a) multiple scattering; (b) small angle scattering; (c) non-uniformity of the sample; (d) sample impurity; (e) dead time of the counting instrument and (f) pulse pile up effects.

Table 1. Experimental K-XRF cross sections (barns per atom) and K-fluorescence yield values (experimental errors are to the extent of 3% to 4%).

		Present cross sections	ω_K	
Z	Element	(b/atom)	Present	Others
60	Nd	2.96	0.932	0.941[a],0.920[c],0.918[d] 0.921[e], 0.947[f]
64	Gd	3.98	0.946	0.946[a],0.922[b],0.934[c] 0.932[d], 0.935[e],0.966[f]
65	Tb	4.27	0.949	0.938[e],0.969[f]
66	Dy	4.58	0.952	0.956[a],0.954[b],0.940[c] 0.938[d], 0.941[e],0.972[f]
69	Tm	5.59	0.959	0.947[c],0.945[d],0.949[e] 0.979[f]
71	Lu	6.35	0.963	0.952[c],0.949[d],0.953[e] 0.981[f]
74	W	7.61	0.966	0.968[a],0.956[b],0.957[c] 0.954[d], 0.958[e],0.983[f]
77	Ir	9.08	0.973	0.962[c],0.958[d],0.962[e] 0.982[f]
80	Hg	10.71	0.975	0.971[a],0.966[c],0.962[d] 0.965[e], 0.980[f]
82	Pb	11.88	0.974	0.973[a],0.961[b],0.968[c] 0.963[d], 0.967[e],0.978[f]

[a]Durak [15], [b]Balakrishna [18], [c]Bambynek [7], [d]Bambynek [9], [e]Krause [8], [f]Hubbell [1].

3.1 *Multiple scattering*

The effects of multiple scattering have been corrected for in the present study by extrapolation technique described earlier.

3.2 *Small angle scattering*

In the present experimental set up, a distance of 32 cm was maintained between the detector and the source. Maximum angle of scattering from sample to detector is about 40 min. According to theoretical estimates the contribution of coherent as well as incoherent scattering at such small angles to the measured cross section at the energy of interest is negligible. Hence, small angle corrections were not applied to the measured data.
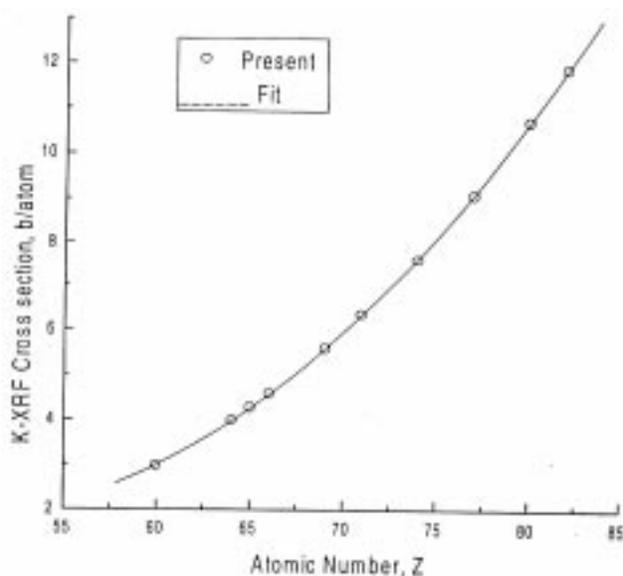
Figure 3. K-XRF cross sections.

3.3 *Non-uniformity of the sample*

The error due to non-uniformity of the sample material has been corrected for in the present method by extrapolation technique described earlier.

3.4 *Sample impurity*

The error due to sample impurity could be a significant factor only when large percentages of high-Z impurities or very close Z elements are present in the sample. As per the manufacturers specification the content of high Z impurity in rare earth samples used in the present study is less than 0.005%. The contribution of these impurities to the X-ray peaks of interest was insignificant. Hence, impurity corrections were not applied to the present data.

3.5 *Dead time of the counting instrument*

The dead time of the multichannel analyser was a combination of the rise time of the pulse, the conversion time in the analog-to-digital converter and the data processing time. In the multichannel analyser used in the present study, there was a built-in provision for dead time correction. The percentage dead time correction was always less than 2% in the present study.

3.6 *Pulse pile up effects*

The pulse pile up effects were kept to a minimum by selecting an optimal count rate and counting time.

Thus, the overall error in the present measurements was about 3% to 4%.

It may be argued that in addition to the photoeffect, Compton scattering from bound electrons of the target as well as the non-radiative transitions may contribute to the measured intensity of fluorescent X-rays. However, in the energy region of interest, the photoeffect for high Z elements is more predominant than the Compton scattering process and also because the elements used for the measurements have large K-shell fluorescence yield, the contribution of Compton scattering and radiative transitions is considered negligible in the present study.

Since theoretical and measured K-XRF cross sections were not available for all elements at excitation energy used in the present work, we have evaluated experimental ω_K values using the relation

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

where $\sigma_K^x(E)$ is the present measured K-XRF cross section and $\sigma_K^P(E)$ is the K-shell photoeffect cross section for the given element at excitation energy E. $\sigma_K^x(P)$ values were taken from the table presented by Scofield [20]. Scofield treated the electrons relativistically and electrons were assumed to be moving in the same Hartree–Slater-central potential, both before and after the absorption of photons. In this model, all the multipole contribution and retardation effects are included in the treatment of the radiation field. Of all the currently available K-shell photoeffect cross sections, Scofield values are considered to be most complete and reliable and are available in tabular form for all elements of the periodic table for photon energies up to 1500 keV. The ω_K values calculated using eq. (3) are listed in table 1 along with the other available experimental and theoretical values. It is expected that the experimental errors are carried over to the calculated values. It can be noted that there is a reasonably good agreement among the present values and the other available theoretical and experimental values within the range of experimental errors.

4. Conclusions

This study has enabled a systematic determination of K-XRF cross sections and fluorescence yields at a high excitation energy for elements in the medium and high Z range by making use of targets containing their respective simple chemical compounds. The results are in good agreement with the other available data.

Acknowledgements

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