Chemical effect on the *K* shell x-ray fluorescence parameters of some Ce compounds

A. Turşucu*, D. Demir

Department of Physics, Faculty of Sciences, Ataturk University, 25240 Erzurum, Turkey

HIGHLIGHTS

► The samples were excited by 59.54 keV γ-rays from a 5Ci [241]Am annular radioactive source.
► The chemical environment produces mainly energy shifts and relative intensity variations of emission lines.
► Different bond distances and energies cause different interaction between ligand and central atoms.
► More experimental data are clearly needed, particularly for different symmetries and for chemical compounds.

ARTICLE INFO

ABSTRACT

Chemical effects on the K_β/K_α x-ray intensity ratios, fluorescence yields w_K and vacancy transfer probabilities η_{KL} for some Ce compounds were investigated. In this study, the samples were excited by 59.54 keV γ-rays from a 5Ci [241]Am annular radioactive source. *K* x-rays emitted by samples were counted by an HPGe detector with a resolution of 182 eV at 5.9 keV. The experimental values were compared with the theoretical and other experimental values. The aim of these measurements was to characterize the dependence of the line position and line width with the chemical environment changes.

1. Introduction

X-ray photons are a kind of electromagnetic radiation produced by irradiation of an inner shell electron and following transition of atomic orbital electrons from high states to low energy states. The photoelectric effect, the Compton scattering and the pair production process are the predominant interactions between the photons and atoms over a wide range of energies. Below 100 keV photon energies, the photoelectric effect is the predominant process especially with high *Z* elements.

The K_α x-rays arise from transitions of the *L* to the *K* shell. The K_β x-rays arise from transitions of the *M*-, *N*-, *O*-, etc. to the *K* shell. Measurement of the K_β/K_α intensity ratio is of prime importance in x-ray emission techniques for elemental analysis in which electron, proton, and photon beams are widely employed. The de-excitation of an atomic shell is characterized by the fluorescence yield and it is defined as "the probability that a vacancy in the shell or sub-shell is filled through a radiative transition". Atoms with vacancies

in the inner shells are unstable and their stability can be regained by single or multiple electron transitions from outer shells. As a result, the vacancy is shifted to the higher shell. The transfer coefficient η_{KL} describes the mean number of vacancies produced in the *L* shell by one vacancy transfer probability are important in many practical applications, such as nuclear electron capture, internal conversion of γ-rays, photoelectric effect, atomic processes leading to the emission of x-rays, Auger electrons and computations for medical physics and irradiational process.

A ligand that is connected to the central atom with a chemical bond may influence the valance electron configuration and oxidation state. Different compounds have different bond energies and these energies give us information of bond distance which is between ligand and central atoms. Different bond distances and energies cause different interactions between ligand and central atoms. Therefore, *K* x-ray spectra of elements and their compounds are significantly influenced by the chemical environment. The chemical environment produces mainly energy shifts and relative intensity variations of emission lines (Büyükkasap, 1997).

There are many studies that have been devoted to a better understanding of the chemical effect on the *K* and *L* x-ray structure. Söğüt et al. (2002) have determined chemical effect

---

* Corresponding author. Tel.:+90 442 231 41 40.
E-mail addresses: ahmettursucu@atauni.edu.tr,
ahmetursucu@hotmail.com (A. Turşucu).

variation on K_β/K_α x-ray intensity ratios in 3d elements. Baydaş et al. (1999) have investigated chemical effects on L x-ray fluorescence cross-section of Ba, La and Ce compounds. Aylikci et al. (2007) have investigated chemical effects on L x-ray production cross-sections and the average fluorescence yield for Hf compounds. Büyükkasap (1997) have investigated chemical effect on L x-ray fluorescence cross-section of Hg, Pb and Bi compounds. Porikli et al. (2008) investigated K_β/K_α x-ray intensity ratio and line shape with the effects of external magnetic field and chemical combination. Baydaş et al. (1998) have measured chemical effects on the L_α/L_β x-ray intensity ratios of Ba, La and Ce compounds. Cengiz et al. (2010) have determined chemical effects on K and L x-ray intensity ratios and production cross-sections for gold compounds. Tıraşoğlu and Tekbiyik (2005) have studied K_β/K_α intensity ratios for some Ca and K compounds. Raj et al. (2000) have measured K_β to K_α intensity ratios of Cr, Mn and Co in pure metals and in CrSe, MnSe, MnS and CoS compounds.

In this study we have investigated chemical effects on K_β/K_α x-ray intensity ratios, w_K fluorescence yields and η_{KL} vacancy transfer probabilities for some Ce compounds at 59.54 keV.

2. Experimental procedure and theoretical basis

The schematic arrangement of the experimental setup used in the present study is shown in Fig. 1. We have used a sample chamber shown in Fig. 2. The samples were excited at 59.54 keV γ-rays emitted from 5Ci ^{241}Am annular source and fluorescent x-rays were detected by an HPGe detector. The HPGe detector is a DSG planar high purity germanium crystal with a diameter of 16 mm, a length of 10 mm, a beryllium window of 0.12 mm and active area of 200 mm^2. A bias voltage of -1500 V is applied to the detector with a resolution of 182 eV at 5.9 keV. The samples of thickness ranging from 0.125 g/cm^2 to 0.426 g/cm^2 were prepared in the form 13 mm dia cylindrical pellets by being pressed in a hand operated hydraulic press at a pressure of 10 t. In this work, the studied compounds were CeCl$_3$, CeF$_3$, Ce (NO$_3$)$_3$, Ce$_2$ (SO$_4$)$_3$ and Ce$_2$O$_3$ for Ce. The spectra were recorded using a Canberra (AccuSpec) PC-based multichannel analyzer card. The time constant of the Ortec model 472 amplifier was set to 6 µs, ensuring optimum detector performance as specified by manufacturers. Operating parameters of the system are governed and controlled by the computer program Genie-2000. The pulse height spectra of x-rays were acquired for a period of 7200–14400 s. The data were collected into 1024 channels of the MCA. Data were analyzed by the Origin 7.5 software program. The mean of 10 channels of each side of the peaks was used to calculate the background and to define the peak. A linear background function was selected since the background is constant in this region. The background count rate was subtracted from measurements. X-ray peaks have been fitted into one Gaussian. A typical x-ray spectrum of CeCl$_3$ target is shown in Fig. 3.

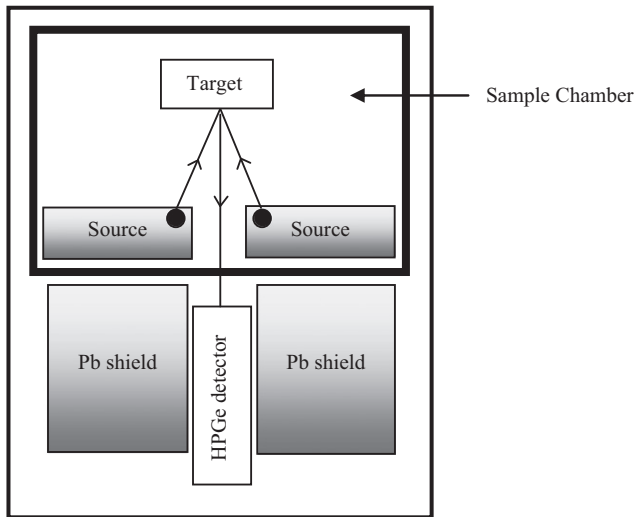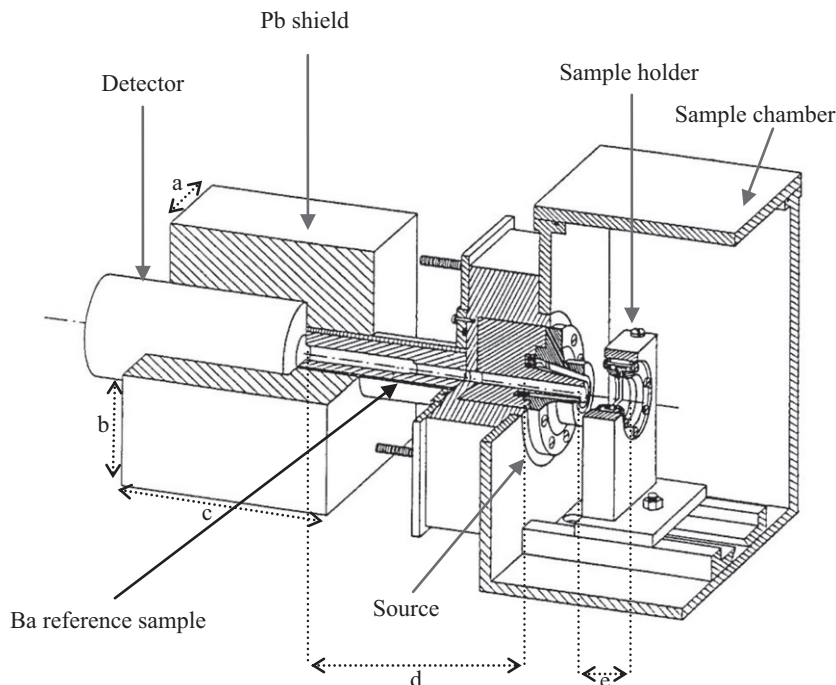**Fig. 1.** Experimental setup.

Fig. 2. Sample chamber (a=6.5 cm, b=6.3 cm, c=13.5 cm, d=11 cm, e=5 cm).