Nuclear Instruments and Methods in Physics Research B 410 (2017) 193–199

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb

H^+, O^{2+}, O^{3+} and high resolution PIXE spectra of Yb_2O_3

P.C. Chaves [a,*], M.A. Reis [a,b]

[a] Centro de Ciências e Tecnologias Nucleares (C^2TN), Instituto Superior Técnico, Universidade de Lisboa, Campus Tecnológico e Nuclear, EN10 km 139.7, 2695-066 Bobadela, Portugal
[b] IEQUALTECS, Lda, R. Dr. Francisco Sá Carneiro, 36 2500-065 S. Gregório CLD, Portugal

ARTICLE INFO

Article history:
Received 17 May 2017
Received in revised form 19 July 2017
Accepted 19 August 2017

Keyword:
XMS
HRHE-PIXE
DT2
Multiple-ionizations
Rare Earth Elements

ABSTRACT

The number of X-ray spectrometry systems having energy resolution of the order of 10 eV, or less, has increasing recently, included already energy dispersive systems (EDS). Access to previous unseen spectra details and enhanced information including speciation, becomes more common and available. Analysis of high resolution EDS PIXE spectra is, nevertheless a complex task due to the need to carefully account for contributions from minor and satellite transitions. In this work, a pure Yb_2O_3 sample was irradiated at the HRHE-PIXE setup of C^2TN, and simultaneous CdTe and X-ray Microcalorimeter Spectrometer (XMS) spectra were collected. The L-shell spectrum of Yb emitted during irradiations using H^+, O^{2+} and O^{3+} ions in the energy range from 1.0 to 6.5 MeV was studied. Measured L X-ray spectra were analysed taking into account the effects of the multiple ionization in the L and M shells. All spectra were analysed using the DT2 code, which allows to include in the fitting model diagram lines as well as multi-ionization satellites and any other contributions. In this communication we present the results and discuss details and problems related to the transition energies, intensity, line width data, and multiple ionization satellites.

1. Introduction

Particle induced X-ray emission (PIXE) is a powerful technique for quantitative analysis because it is non-destructive, multi-elemental (from Na to U), highly sensitive and requires no special sample preparation. Usually proton beams with an energy around 2 MeV are used in PIXE offering high sensitivity. Since the sensitivity of PIXE strongly depends the ionization cross-sections, which are proportional to the square of the projectile atomic number, it can be expected that the use of heavy ion beams improves the sensitivity of the analysis considerably. However, the X-rays emitted from atoms ionized by heavy ions exhibit, apart from the X-ray diagram lines, a significant satellite structure corresponding to different multi-vacancy configurations. Heavy ions PIXE is still far from being an established analytical method and various issues such as the determination of fluorescence yield, line shifts and peak broadening due to multiple ionization, still need to be properly established. In Fig. 1 the overlap of three spectra of thick Yb_2O_3 targets irradiated using different ion beams is shown. In this case, spectra obtained in the lowest standard stable energy conditions of the used accelerator are presented. The complexity of the analysis becomes clear from the differences put in evidence. In fact, even if the differences in L_β to L_α groups could be explained by matrix effects, the different shapes of the L_β group cannot. Furthermore, since the physics of oxygen beam ionizations is more complex then that of proton beams the result is not unexpected. The differences in shape may be explain by differences in L2 and L3 ionization cross sections since the L_β group is composed by transition to both sub-shells. Nevertheless, high resolution studies in this area can enhance the gathering of information about the structure of multi-vacancy configurations, helping to extract information present in data collected by other detector systems. In this work the characteristics of the C^2TN High Resolution High X-ray Energy (HRHE)-PIXE [1,2] and the DT2 [3] code versatility will be shown to be a major support to the study of these spectra.

2. Materials and methods

2.1. Experimental chamber and ion beams

Experiments were carried out using the High Resolution High X-ray Energy (HRHE)-PIXE setup [1] endstation of the CTN 3.0 MV Tandetron. X-rays were collected using an Amptek Peltier Cooled $3 \times 3 \times 1$ mm^3 CdTe detector placed at about 25 mm from target and at 145° relative to the beam direction. Detector window is a 250 μm Beryllium window. EDS high resolution L-shell spectra were collected simultaneous to the CdTe ones, using a Vericold Tech. GmbH Polaris XMS, referred here as the C^2TN XMS, set at 90° to beam direction. More details about the HRHE-PIXE system

* Corresponding author.
E-mail address: cchaves@ctn.tecnico.ulisboa.pt (P.C. Chaves).

http://dx.doi.org/10.1016/j.nimb.2017.08.026
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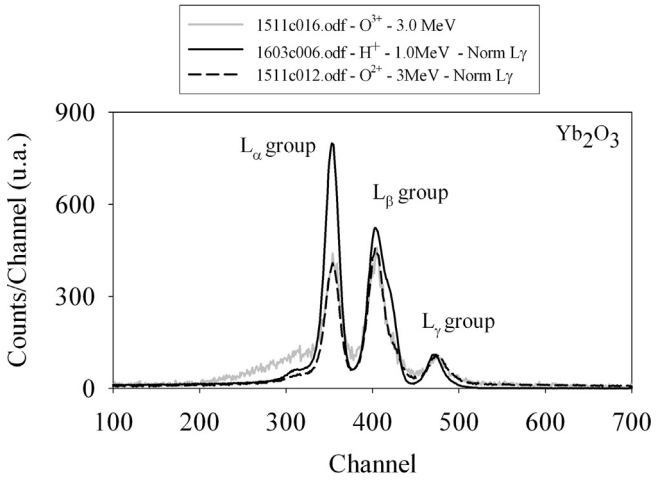

Fig. 1. Overlap of spectra obtained for Yb_2O_3 when irradiated the sample with 3.0 MeV O^{2+} and O^{3+} ion beams with the H^+ spectrum collected when sample was irradiated with 1.0 MeV H^+ ion beams. Spectra were normalized to L_γ peak. It can be seen an effect in the low energy region of O^{3+} spectrum not observed in the O^{2+} spectrum.

can be found in references [1,2]. A thick pellet of pure Yb_2O_3 (99.998%) target was prepared from powder by pressing to 5.6 tons/cm^2 and studied using ion beams of H^+, O^{2+} and O^{3+} having energies from 1.0 to 6.5 MeV. Experimental condition details are presented in Table 1. Ion beam spot in the target was set to a circus of 3 mm in diameter by using a dual collimator providing an elliptical shaped beam cross-section [1]. An electron gun was used to avoid target charging up. In total, 8 spectra were collected. In the case of the irradiation using H^+ ions at 3.8 MeV, a funny filter of a 1 mm Al thick foil having a hole of 1 mm in diameter in the center, was used in front of the detector to level the detector efficiency over its wide dynamical range (5–120 keV). Beam currents were adjusted to eliminate the possibility of exist charge effects during the irradiation.

2.2. Spectra lines Gaussian standard deviation energy dependence

The semiconductor detector response function is modeled as [4]:

$$F(E) = G(E) + Ca(E) + G_e(E) \quad (1)$$

where

$$G(E) = \frac{1}{\sqrt{2\pi}} \cdot \sigma_l(E_0) e^{-\left[\frac{E-E_0}{\sqrt{2\pi}\cdot\sigma_l(E_0)}\right]^2} \quad (2)$$

$\sigma_l(E_0)$ being the spectra line Gaussian standard deviation. The other two terms are relative to tail and escape peaks also dependent on the $\sigma_l(E_0)$. The spectra line Gaussian standard deviation is therefore, one of the most important parameters to take into account for a detailed fitting process. Fine adjusting its energy dependence for the present case, was made considering the Yb_2O_3 spectrum collected by the CdTe detector when the sample was irradiated using 3.8 MeV H^+ ions.

As shown in Fig. 2 (top), in this conditions K and L lines are present in spectrum so that an energy range from 7 to 61 keV can be used. The spectrum was fitted in two regions considering separately K and L X-ray lines. Based on the values for $\sigma_l(E_0)$ of main lines, $L_{\alpha 1}$, $L_{\beta 1}$, $L_{\gamma 1}$, $K_{\alpha 1}$ and $K_{\beta 1}$ it was possible to establish the energy dependence of the spectra line Gaussian standard deviation, modeled as:

$$\sigma_l(E_0) = \sigma_{coef} \cdot (asig + bsig \cdot E_0 + csig \cdot E_0^2) \quad (3)$$

where the *asig*, *bsig* and *csig* parameters must be carefully determined. The parameter σ_{coef} is adjustable during each spectrum fitting procedure.

In Fig. 2 (bottom) the experimental data and fit for the "Sigma", $\sigma_l(E_0)$ energy dependence are presented.

2.3. XMS analysis of Yb transition energies, rates and widths

Once set the $\sigma_l(E_0)$ parameter variation in energy, the second major issue is knowing the exact energy of all transitions present in the spectrum. Usually, the ratio of intensities is also required, but the DT2 code overcomes this difficulty when used in free line mode [3]. As mention above, in heavy ion PIXE, multiple ionization components are significant. Knowing their location is thus a major issue.

In Fig. 3, the L_α energy region of the XMS high resolution spectrum of the Yb_2O_3 target irradiated using a 1.0 MeV H^+ beam is shown. Fit and partial contributions to this, namely $L_{\alpha 1}$, $L_{\alpha 2}$ and $L_{\alpha 1}$ multi ionizations are also presented. Transition energies for diagram lines were obtained from Bearden [5] and multiple ionizations transitions were calculated from these based on the work of Uchai et al. [6].

As can be seen, in this L X-ray energy range the background is close to zero and X-ray lines are well separated allowing the identification of L_α multi ionizations and making it possible to obtain a good fit.

In Fig. 4 the L_β energy region is shown. In this region it is possible to identified the $L_{\beta 4}$, $L_{\beta 1}$, $L_{\beta 6}$, $L_{\beta 3}$ and $L_{\beta 2}$ X-ray lines. Single and double spectator vacancies were considered and fitted for $L_{\beta 1}$. In this fit the $L_{\beta 6}$ line was excluded due to the energy proximity (8.6 eV) to the double spectator vacancy satellite of $L_{\beta 1}$, **L2M4m^2**, combined with the fact that $L_{\beta 6}$ has low intensity.

High resolution spectra lines are modeled by DT2 using a true Voigt function. Based on the fit of the XMS spectrum presented in Figs. 3 and 4 spectrum, it was verified that the natural width values reported by Perkins and co-workers [7] for $L_{\beta 1}$ and $L_{\beta 2}$ are not

Table 1
Description of all experimental conditions used in the present work. XMS and CdTe specra collected at 1.0 MeV using proton beams were collected using the detectors simultaneously. Yb_2O_3 spectrum collected at 3.8 MeV was used to study the sigma energy dependence.

Detector	Particle	Beam energy (MeV)	Diffuser	Filter	Collimator (mm)	Charge (μC)
XMS	H^+	1.0	Yes	No	3	90.6
CdTe	H^+	1.0	Yes	No	3	90.6
	H^+	3.8	Yes	BN + Al funny	3	60
	O^{2+}	2.0	No	No	3	771
	O^{2+}	3.0	No	No	3	203
	O^{3+}	3.0	No	No	3	42
	O^{3+}	4.5	No	No	3	240
	O^{3+}	6.5	No	No	3	32