Nuclear Instruments and Methods in Physics Research B 273 (2012) 33–35

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

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

Non-linearity of prompt nuclear satellites relative intensities observed from high energy protons induced X-ray emission

C.A. Pineda-Vargas [a,b,*], M. Msimanga [a], R.A. Bark [c], D. Gihwala [b]

[a] iThemba LABS, Materials Research Department, Somerset West 7129, South Africa
[b] Faculty of Health and Wellness Sciences, CPUT, Cape Town, South Africa
[c] iThemba LABS, Department of Nuclear Physics, Somerset West 7129, South Africa

ARTICLE INFO

Article history:
Available online 27 July 2011

Keywords:
High-energy PIXE
Nuclear satellites
Rare earths
Cyclotron

ABSTRACT

A study of the relative intensity of K-shell X-ray nuclear satellites as a function of atomic number, *Z* is presented. These satellites, which are generated during bombardment with high energetic protons of 66 MeV from targets composed of pure fluoride compounds for the rare earths and heavier metals including the platinum (Pt, Au) and fissionable metals (Th, U) are of particular interest in the quantitative elemental analysis of rare earth and heavy metals by energetic protons. This paper will describe aspects that may account for the production cross section of nuclear satellites for atomic numbers $57 \leqslant Z \leqslant 92$. Previous results that showed a linear relationship between relative intensities and atomic number will be discussed.

1. Introduction

High-energy protons with energy of the order of 66 MeV can generate K X-rays with sufficient intensity for sensitive analysis [1] with the advantage that the extent of mutual interference is considerably reduced and, in most cases, eliminated. A further advantage is that the energy region of interest for K X-ray spectrometry lies on a relatively low, flat background, thus further facilitating the off-line deconvolution procedure. Since the intensity of the background will obviously depend on the amount of material in the target, the most advantageous conditions for analysis will be the use of thin targets.

Previous studies have shown the potential advantages of High Energy PIXE (HE-PIXE) using energetic H^{1+} – ions for the determination of rare earths and heavier metals using the high ionizations cross sections available for the K-shell X-rays in the energy range of 20–90 keV [1–3]. Over the years only a few authors have measured experimental values for the ionization cross sections of rare earths and heavier metals using protons in the energy range 10–100 MeV. Particularly in the regime of 66–68 MeV experimental data for HE-PIXE has been collected only by Denker and Pineda [4,1]. As pointed out by Lapicki [5] these cross sections, measured by Pineda and Denker bifurcate, particularly for $Z > 56$ with differences greater than 10%. It would be useful to resolve and understand the source of the discrepancy which eventually will contribute to the solidity of the model and to the accuracy in the quantification of elemental concentrations. On the other hand, previous determinations of the contribution of nuclear effects to the total K-shell lines [2] in heavy metals showed that such contributions are related to the excitation of the nuclear satellite signals which in most of the cases appear to be interfering with the original K-shell lines.

The main objective of this paper is to assess and understand the nature of the interference in terms of nuclear effects.

2. $Z + 1$ Nuclear satellites

Rare earths and heavier metals of mass A and atomic number Z subjected to bombardment with energetic protons exhibit nuclear reactions of the type (p, xn), where secondary nuclei in excited states and with masses $A' < A$ are created. These excited nuclear states may decay by internal conversion (I.C.) by giving the transition energy to an orbital electron, which is expelled from the atom (usually an S-electron, as these penetrate the nucleus). The resulting vacancy is filled by outer electrons dropping into fill the shell, thereby emitting X-rays. This process is shown in Eq. (1) for the case of ^{166}Er in which the excited compound nucleus ^{161}Tm* is created by neutron evaporation, and decays to the ground state by either γ-ray emission or I.C.

$$^{166}\mathrm{Er}(\mathrm{p},\,6\mathrm{n})^{161}\mathrm{Tm}^{*} \xrightarrow{\gamma,\mathrm{I.C.}} {}^{161}\mathrm{Tm}_{\mathrm{g.s.}} \quad (1)$$

After series of nuclear decays, the $Z + 1$ nucleus is left in the ground state (or a metastable state) that may decay by electron capture (E.C.) to a Z nucleus, with half-lives that are variable, from

* Corresponding author at: iThemba LABS, Materials Research Department, Somerset West 7129, South Africa.
E-mail address: pineda@tlabs.ac.za (C.A. Pineda-Vargas).

0168-583X/$ - see front matter © 2011 Elsevier B.V. All rights reserved.
doi:10.1016/j.nimb.2011.07.031

hours to μs. Following the above example based on the initial ^{166}Er target nucleus, this process is illustrated in Eq. (2) for the case of $^{161}Tm_{g.s.}$:

$$^{161}Tm_{g.s.} \xrightarrow{E.C.} {}^{161}Er^{*} \xrightarrow{\gamma, I.C.} {}^{161}Er_{g.s.} \quad (2)$$

where $^{161}Tm_{g.s.}$ decays to an $^{161}Er^{*}$ excited state by E.C. and subsequently $^{161}Er^{*}$ decays to the ground state, $^{161}Er_{g.s.}$ by γ-ray emission or I.C.

3. $Z-1$ Nuclear satellites

Often the most intense satellite in the rare earths and heavier elements under bombardment with energetic protons ($E_p > 40$ - MeV) is that emitted from the $Z+1$ compound nucleus. However if the neutron evaporation reaches the 6n or 7n level, satellite contributions from $Z-1$ may also occur. Following the case for $Z+1$ satellite the typical chain of reactions and electron capture decays generating a $Z-1$ nucleus is given by Eq. (3):

$$(A_1, Z_1) \rightarrow (A_2, Z_1+1) \rightarrow (A_2, Z_1) \rightarrow (A_2, Z_1-1) \quad (3)$$

where the decay of all products will in general pass through excited nuclear states. The most important condition for the Z_1-1 nucleus to be populated, is that the reaction should evaporate a sufficient number of neutrons such that the stable elements in the isobaric chain have atomic number values less than Z_1. In general for elements with $Z \geqslant 56$, the probability of occurrence of the $Z-1$ satellites is much greater than for $Z < 56$. This applies to the case of ^{166}Er where the 6n evaporation gives the possibility of successive E.C. decay to the $Z-1$ unstable nucleus $^{161}Ho^{*}$ which subsequently decays to ^{161}Dy. Such a sequence is illustrated in Eq. (4):

$$^{161}Er_{g.s.} \xrightarrow{E.C.} {}^{161}Ho^{*} \xrightarrow{\gamma, I.C.} {}^{161}Ho_{g.s.} \xrightarrow{E.C.} {}^{161}Dy^{*} \quad (4)$$

4. Intermediate Z – excited state contribution to the nuclear satellite

Nuclear satellites for $Z \pm 1$ have been reported and studied previously [6], however little attention has been paid to the contribution to the K – lines of the (Z, A) nucleus from the decay of excited states in the $(Z, A-n)$ nucleus. To illustrate this processes Fig. 1 shows a typical X-ray spectrum obtained from irradiation of Er and Ho with 66 MeV protons obtained previously at the HE-PIXE set-up at the 200 MeV SSC Cyclotron of iThemba LABS [1] by bombardment of thin deposits of Er and Ho fluorides over mylar obtained from Messrs Micrometer Co®. The average thickness of the deposits was of the order of 45 μg/cm^2. Targets were irradiated with a $_1H^{1+}$ beam of 2 mm diameter and with currents of ~ 1 nA. The decay scheme has already being presented for the case of the stable nucleus ^{165}Er in Eqs. (1), (2) and (4).

The cross section for the (p, 6n) reaction is ~1500 mb, and both the $^{161}Tm^{*}$ and $^{161}Er^{*}$ decay by E.C. having, respectively, half-lifes of 30.2 min and 3.21 h. At the same time the ^{161}Ho decays to the stable nucleus ^{161}Dy. All these decay happen on the same time scale as the measurement. The $Z+1$ nuclear satellites in the X-ray energy spectrum of Ho and Er (i.e. Er $K_{\alpha1}$ and Tm $K_{\alpha1}$) are indicated in Fig. 1. Although in this particular example the intensity of the $Z-1$ satellites is relatively small (peaks on the left of the main $K_{\alpha2}$ for Ho and Er), there should be a contribution to the primary target $K_{\alpha1}$ arising from the intermediate state $^{161}Tm^{*}$ which decays to excited states in $^{161}Er^{*}$. The intensity of each satellite $K_{\alpha1}$ X-ray vis-a-vis the primary $K_{\alpha1}$ will be governed by the I.C. mechanism as well as by the relative cross section of the respective reaction. The same rationale may apply for the other K-shell lines.

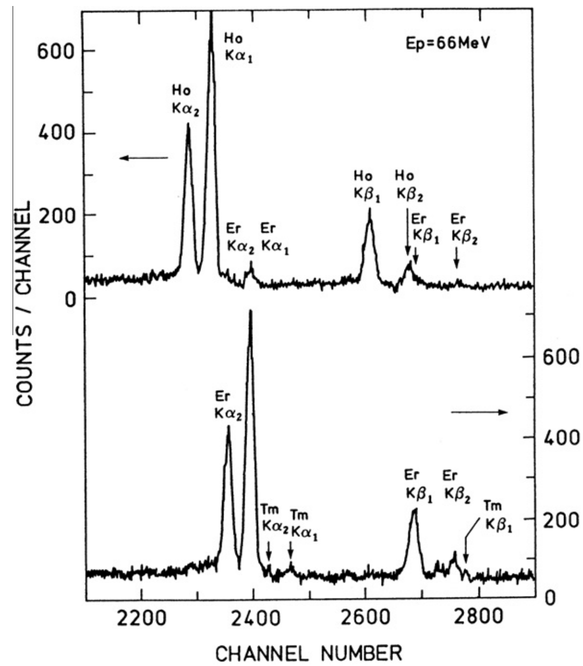

Fig. 1. The high energy portion of the X-ray spectra obtained from the bombardment with 66 MeV protons of the fluorides of holmium (upper spectrum) and erbium. Note the presence of satellite peaks.

5. Discussion: non-linearity of satellites ratios

The relative intensities of the $K_{\alpha1}$ X-ray line of the $(Z+1)$ nuclear satellite to that of the target element Z, under bombardment with 66 MeV protons are plotted in Fig. 2, as a function of the atomic number of the target element.

As it is evident from Fig. 2 the contribution of the nuclear satellite apparently increases with atomic number of target element. For elements below $Z = 56$ the effect is negligible. However for elements with $Z > 56$ this effect contributes to significant interfering problems if a good estimation of the true elemental concentration for the primary element is required. Consequently it is of interest to evaluate and quantify the relationship between the relative yield of the nuclear satellites and the primary atomic species. An inspection of Fig. 2, indicates a general increase of this ratio as a function of Z, with a large scatter in the data points. The general upward trend in the data can be understood qualitatively by considering the dependence of both K-shell ionization cross-sections and internal conversion coefficients with atomic number. The former decreases with increasing Z [3] while the latter increases with Z [7], implying that the ratio should increase. A scatter in the data is expected on the grounds that internal conversion is also a strong function of the nuclear transition energy and multipolarity [7]. The intensity of the satellite peaks will depend on the details of the nuclear levels populated both in the initial nuclear reaction and subsequent electron capture decays. Since nuclear level structure does not depend in a smooth way on Z, strong fluctuations in the intensity of the satellite peaks as a function of Z could be expected. Lastly, one must also consider the details of nuclear reaction cross-sections as a function of Z. While fusion-evaporation cross-sections would be expected to be approximately constant throughout the rare-earth region, evaporation would give way to fission in the actinide region. Clearly a quantitative estimate of nuclear satellite intensities must be made on a case by case basis.

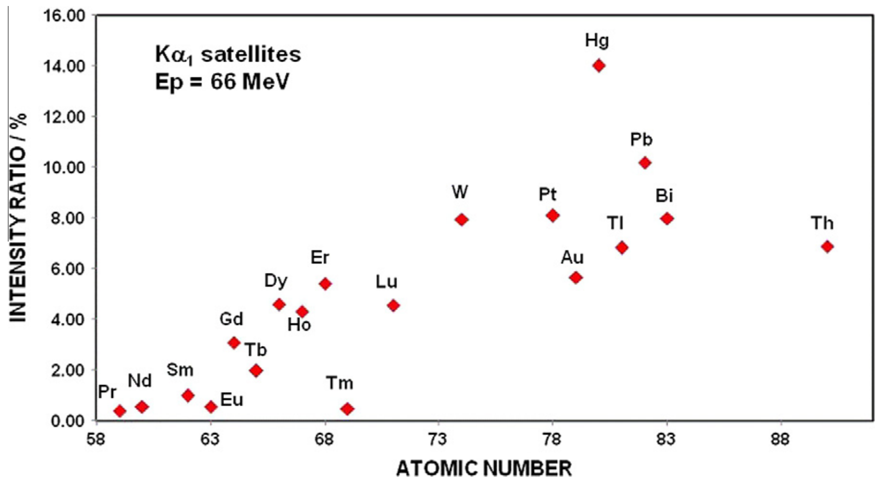

Fig. 2. Plot of the $K_{\alpha 1}$ nuclear satellites intensity ratio (as a % of the $K_{\alpha 1}$ of the primary element) for elements with $58 \leqslant Z \leqslant 90$. The ratios were obtained from irradiation with 66 MeV protons. A beam size of ~2 mm was used with an average intensity of 1 nA.

6. Conclusions

The production of several nuclear species during irradiation highlights the need to correct the primary K-shell X-ray yield from contributions from internal conversion in nuclear species having the same Z-value but populated by electron capture decay of nuclear reaction products. This also applies to other stable nuclei being analyzed in a particular specimen where other satellites from different parent nuclei may interfere.

The contribution of each of the nuclear satellites present in the energy spectrum therefore should be estimated according to the relative cross section of each related nuclear reaction. It will be relevant to investigate in the future the relationship of the beam current intensity and the extent of the nuclear satellite contribution.

In a more realistic case as for example the characterization of a geological material where many elements may be presents in the target matrix a good understanding of the nuclear satellites relative intensities is necessary. The contribution of each of the satellites present in the energy spectrum therefore should be estimated according to the relative cross section of each related nuclear reaction.

References

[1] M. Peisach, C.A. Pineda, Nucl. Instr. Meth. B 49 (1990) 10.
[2] C.A. Pineda, M. Peisach, Nucl. Instr. Meth. A 299 (1990) 618.
[3] C.A. Pineda, M. Peisach, J. Radioanal. Nucl. Chem., Articles 151 (1991) 387–396.
[4] A. Denker, W. Bohne, J.L. Campbell, P. Heide, T. Hopman, J.A. Maxwell, J. Optiz-Coutureau, J. Rauschenberg, J. Röhrich, E. Sraub, X-Ray Spectrom. 34 (2005) 376.
[5] .G. Lapicki, J. Phys. B: At. Mol. Opt. Phys. 41 (2008) 115201 (13pp).
[6] J.S.C. McKee, Int. J. PIXE 3 (1993) 187–214.
[7] F. Rösel, H.M. Fries, K. Alder, H.C. Paul, At. Nucl. Data Tables 21 (1978) 91.