Measurement of 1-MeV C-ion beam induced X-ray production cross sections of Fe, Nb, Ru, Ce and Ta

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Abstract. While the cross sections of heavy ion beams at energy of a few MeV and above have been measured and reported, experimental data, especially of those minor lines from uncommon materials, for lower-energy heavy ion beams around 1 MeV are lack. In this study, we used 1-MeV C-ion (83 keV/amu) PIXE (particle induced X-ray emission) to measure the X-ray production cross sections of Fe K-line, Nb, Ru and Ce L-line, and Ta M-line from thin films of the materials. The literature-reported experimentally measured Fe Kα cross section for standard 2-MeV proton beam was used as a reference. The measured data were compared with those PWBA- and ECPSSR-theoretically calculated by program ISICS11 and found their deviations from the theoretical predictions in acceptable ranges.

1. Introduction

Particle induced X-ray emission (PIXE) is an important ion beam analysis technique used in the quantitative determining of the elemental make-up of a material or sample particularly in the near surface region. PIXE was developed initially with using MeV light proton beam but MeV heavy-ion PIXE has later come into the researchers' vision. Compared with light-ion PIXE, heavy-ion PIXE has advantages such as larger scattering cross section and larger stopping cross section [1] and hence higher sensitivity. The technique has been applied to analyze not only solid metals [2] but also biological soft materials [3]. Compared with the rich knowledge on standard proton PIXE, knowledge on heavy-ion PIXE has not yet been sufficient. The X-ray production cross section is such a basic one. Recently there have been some efforts in the measuring of high-energy (> MeV) heavy-ion induced X-ray production cross sections in various materials and relevant data have been reported (e.g. [4-8]). The International Atomic Energy Agency (IAEA) has recently launched a Coordinated Research Project (CRP) entitled Development of molecular concentration mapping techniques using MeV focused ion beams [9] with one of two basic tasks to measure the X-ray production cross sections for MeV heavy ion beams. In this CRP, standard samples of thin films of various materials were made and then distributed to the CRP members for measurements. Participating in this IAEA CRP, we applied PIXE to measure the cross sections for 1-MeV C-ion beam based on our accelerator capability as well as some physical interests. The reported cross section data are generally for high-energy heavy ion beams at energy above several MeV up to tens to hundreds MeV and also normally for the X-ray K lines which are the most prominent and thus easy to measure. Our interests were in the ion beam energy around 1 MeV (83 keV/amu) which is almost the lowest ion energy limit for PIXE availability, minor X-ray lines and some uncommon materials, as the X-ray production cross sections for 1-MeV heavy-ion induced minor lines in uncommon materials have rarely been studied and reported. Our work was then aimed at attempting to contribute an important part to the database of the relevant cross sections from experimental measurement for comparison to theoretically calculated data.

2. Experiment

The calculation principle is based on a ratio of the X-ray yields between a reference material and the material to be determined. The X-ray yield is (slightly modified from [10])

$$Y \propto N\,(It/eA)\,A\,\Omega\,\varepsilon\,\sigma(E, S_i, S_t, X)\,\theta(\theta_i, \theta_d), \quad (1)$$

where N is the atomic number density of the target material in cm^{-3}, I is the ion beam current in Amp, t is the measurement time in second, e is the charge constant, i.e. 1.6×10^{-19} C, A is the beam analyzing area in cm^2, i.e. It/eA is the beam fluence (F) or intensity in cm^{-2} and $(It/eA)A$ is the number of incident ions, Ω is the detector solid angle, ε is the detector efficiency, $\sigma(E, S_i, S_t, X)$ is the X-ray production cross section depending on ion energy E, ion species S_i, target species S_t, and X-ray emitted electron shell X, and $\theta(\theta_i, \theta_d)$ is the yield dependence on the beam incident angle (θ_i) and the detecting angle (θ_d). Note that Eq. (1) is for thin film target and thus the ion penetration dependence is ruled out. A unknown cross section could be measured and calculated by a ratio Y_1/Y_2 where 1 represents the known and 2 represents the unknown. From Eq. (1), this ratio is

$$Y_1/Y_2 = (N_1/N_2)\,(F_1/F_2)\,(\sigma_1/\sigma_2). \quad (2)$$

In our measurements, the PIXE spectral acquisition time was kept the same, e.g. 1000 sec, and as different ion beams certainly had different currents, the beam fluence ratio (F_1/F_2) becomes a beam current ratio (I_1/I_2). Therefore, the unknown cross section is

$$\sigma_2 = (N_1/N_2)\,(I_1/I_2)\,(Y_2/Y_1)\,\sigma_1. \quad (3)$$

The measurement was carried out utilizing the 1.7-MV Tandetron tandem accelerator with its ion beam analysis beamline. The ion species was singly charged carbon (C^+, in the later text it is designated as C-ion). The C-ion beam energy available at our system was then tested from the ion beam current measurement. Limited by the current technical capabilities of both switching magnet and mass analyzing magnet, heavy ion energy applicable at the endstation had a limited range at the system. The testing result showed that the C-ion energy was ranged from 0.8 MeV to 1.4 MeV, only where the C-ion beam current could be measured. The ion beam currents were measured from the electrically isolated sample stage.

The IAEA standard samples that we used in the experiment were thin films of Fe_2O_3 on Si, NbO on sigradure, RuO_2 on sigradure, CeO_2 on Si, and Ta_2O_5 on sigradure in a disk shape of about 1.5 cm in diameter and 1 mm in thickness. The film thicknesses were determined and reported elsewhere [7], as shown in Table 1. Beams of 2-MeV proton and 1-MeV C-ion were applied for the PIXE spectrum measurements. Characteristic X-rays were detected by a Si(Li) detector kept at an angle of 120^o relative to the beam direction. A mylar foil (74 µm thickness with 0.38% relative hole area) was placed in front of the Si(Li) detector in order to protect the detector from scatterd high-energy-ions damaging and high-rate X-ray counting. The Si(Li) detector calibration was performed using the X-rays of a ^{109}Cd radioactive source. The energy resolution of the detection system was estimated from the FWHM of the Fe K_α peak at 6.4 keV to be 180 ± 10 eV. In the PIXE measurement calibration, SRMs (standard reference materials) SRM 610 Trace Elements in Glass Matrix [12] were analyzed as a quality control. The measurement demonstrated the accuracy of the experimental setup to be within ±10%. Before we decided to use 1-MeV C-ion beam for the measurement, the workability of the MeV C-ion beam for PIXE was checked, demonstrating the heavy-ion PIXE at the lower energy around 1 MeV workable. All of the PIXE spectral peaks under investigation were first fitted to Gaussian peaks and the mean values of the Gaussians were taken as the X-ray yields.

Table 1. All relevant parameters of the film materials. *N* = (mass-density/atomic-mass) N_A, N_A = 6.022 × 10^{23}/mol.

^{Z}Element (Material film/substrate)	Atomic mass	Film mass density (g/cm^3)*	Film thickness (nm)	Bulk mass density (g/cm^3) (as reference)	Film number density (*N*) ($10^{22}/cm^3$)
^{26}Fe (Fe_2O_3/Si)	56	5.17	160	7.87	5.56
^{41}Nb (NbO/sigradure)	93	4.6	50	8.57	2.98
^{44}Ru (RuO_2/sigradure)	101	6.97	22	12.2	4.16
^{58}Ce (CeO_2/Si)	140	3.5	30	6.77	1.5
^{73}Ta (Ta_2O_5/sigradure)	181	8.18	24	16.65	2.72

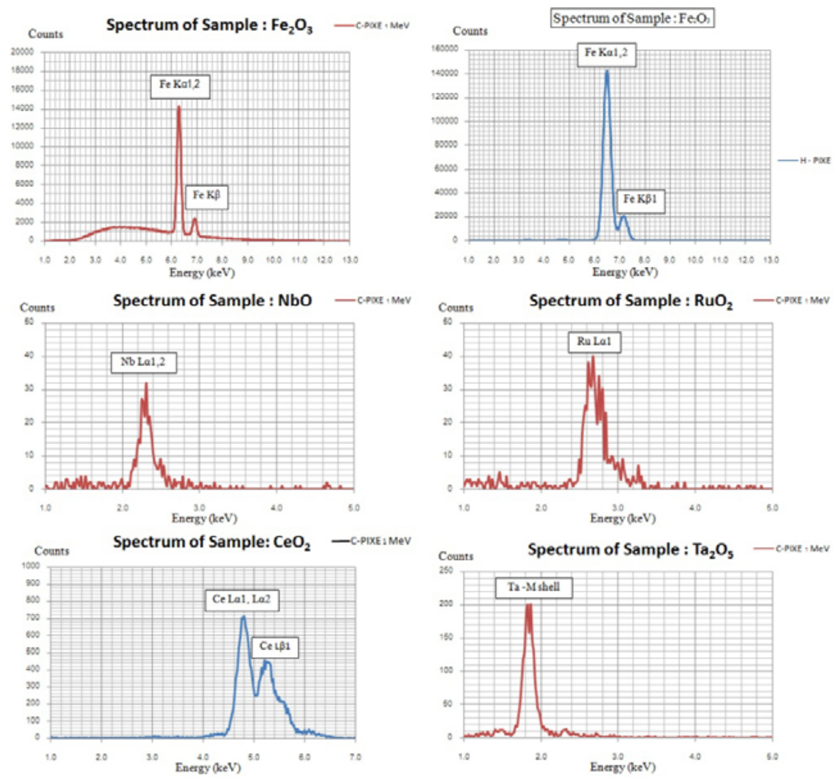

Figure 1. All PIXE spectra measured. H-PIXE: 2-MeV proton beam; others: 1-MeV C-ion beam.

3. Results and discussion

The 2-MeV proton PIXE spectrum was used as the reference which was actually based on the X-ray Kα line production cross section data published in the literatures. There have been a number of data of the experimentally measured 2-MeV proton induced Fe Kα X-ray production cross section reported from various publications, and also theoretical calculation predicted some results. The data were certainly scattered but not significantly and hence showed a clear trend line. We only relied on reported experimental data and fitted the data to find a mean for using as the final reference. It was shown from the fitting that the mean was 74.4 barn, which was used as σ_1. Note that the theoretical PWBA and ECPSSR calculations gave 95 and 83, respectively, for this cross section [13]. Hence, the experimentally measured and theoretically calculated results were fairly close. From the beam currents measured, the beam current ratio between 2-MeV proton and 1-MeV C-ion beams was 7/2 = 3.5, and thus (F_2/F_3) or (I_1/I_2) = 3.5. Then, Eq. (3) could simply be

$$\sigma_2 = 3.5 \times 74.4\ (N_1/N_2)\ (Y_2/Y_1) = 260.4\ (N_1/N_2)\ (Y_2/Y_1). \quad (4)$$

The number densities (N) of the material films were calculated, as shown in Table 1. The X-ray line yields were determined from the PIXE spectral peak heights, as shown in Fig. 1.

To calculate the cross sections, it was first determined that Y_1 = 142,000, as measured from the spectrum of 2-MeV proton in Fe, and N_1 = 5.56, as shown in Table 1. Then, all of the cross sections of relevant X-ray lines of the materials due to 1-MeV C-ion beam are calculated from using Eq. (4), as shown in Table 2. In the table, the measured data are also compared with the theoretically calculated results from the ISICS program [13] with two approaches, namely, PWBA - Plane Wave Born Approximation, a quantum perturbation method [14,15], and ECPSSR - accounts for the energy loss (E) and Coulomb deflection (C) of the projectile and the perturbation of the targets atomic stationary states (PSS) by the projectile as well as the relativistic nature (R) of inner shell target electron [16,17]. It is seen that for Fe, Ce and Ta, the measured results are closer to the PWBA prediction, while for Nb and Ru the measured data are very close to the ECPSSR prediction. The measurement certainly involved not negligible errors from a number of sources, such as ion energy resolution, beam current stability and measurement, spectral determination of the X-ray yield, measurement of the number density of the thin film, X-ray detection efficiency, the reference data, and so on. But, a trend seems to be that for lower Z and higher Z materials, the measured data are closer to the PWBA prediction, while for the medium Z materials, the measured data are closer to the ECPSSR prediction. The differences in the comparison should be due to the special properties of the 1-MeV C-ion. For relatively heavy C-ion at relatively low energy 1 MeV, the ion velocity is fairly lower than those of higher energy (> n-10^2 MeV) ions, plus its single-charge character, the ion interaction with electrons of the target atoms is relatively limited compared with high-energy multiply-charged ions. We speculated three different processes in 1-MeV C-ion interaction with atoms of lower-Z, medium-Z and higher-Z materials, respectively. With the lower-Z materials, the ion fully interacts with the electrons in all shells and is little deflected by the nucleus to pass by due to much energy remaining; with higher-Z materials, the ion partially interacts with the electrons in some shells and is then mildly deflected by the nucleus due to most energy lost but being farther away from the nucleus; while for the medium-Z materials, the ion fully interacts with the all electron shells to lose its most of energy at the most inner shell and is then strongly deflected due to a high closeness to the nucleus. Thus, in the medium-Z case, strong deflection and greatest energy loss of the ion simultaneously take place to cause strong perturbation to the target atom, whereas in the lower-Z and higher-Z cases, the ion deflection is weak or mild, and the ion energy loss is less for the lower-Z case while the ion does not interact with very inner electrons for the higher-Z case, and hence the perturbation is lower. Therefore, for the cases of lower-Z and higher-Z cases, the simpler PWBA theory might be enough to describe the process, whereas for the medium-Z case, the ECPSSR could better describe the process.

Table 2. Calculation of the cross sections of relevant X-ray lines for experimented materials, compared with the theoretically calculated results from the ISICS program.

Material (Z)	X-ray line	Y_2, measured from the spectral peak height	N_1/N_2	Y_2/Y_1	σ_2 (barn)	ISICS result on σ_2 (X-ray production)
Fe (26)	Kα1,2	12,500 (- background)	1	0.088	23	0.25 (PWBA)
	Kβ1	600 (- background)		0.004	1.0	1.1e-3 (ECPSSR)
Nb (41)	Lα1,2	30	1.87	2.1e-4	0.1	26 (PWBA)
	Lβ1,2	15		1e-4	0.05	0.49 (ECPSSR)
Ru (44)	Lα1	39		2.75e-4	0.1	12.6 (PWBA)
	Lα2	4	1.34	2.8e-5	0.01	0.23 (ECPSSR)
	Lβ1	20		1.4e-4	0.05	
Ce (58)	Lα1,2	700		0.005	4.8	0.87 (PWBA)
	Lβ1	420	3.71	0.003	2.9	9e-3 (ECPSSR)
	Lβ2	150		0.001	1.0	
Ta (73)	Mα1	190	2.04	1.34e-3	0.71	0.4 (PWBA) 0.03 (ECPSSR)

4. Conclusion

The X-ray production cross sections for relatively lower energy (1-MeV), singly charged and relatively heavier C-ions in Fe, Nb, Ru, Ce and Ta, most of which are uncommon materials, mostly for minor X-ray lines (except Fe K line) were measured using PIXE from standard IAEA thin film samples. The measured results were compared with the theoretical predictions and the differences were in the acceptable range which was explainable.

Acknowledgements

The work has been supported by the Coordinated Research Projects (CRP) of International Atomic Energy Agency (IAEA), the Chiang Mai University, and the Thailand Center of Excellence in Physics.

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