X-ray Emission Measurement from Organic and Insulating Materials with Low Energy Ga Ion Beams

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We observed a phenomenon of X-ray emission from various types of organic and insulating materials under irradiation of a low energy (30 kV) gallium ion beam. Energy dispersive X-ray spectrometer combined with a focused ion beam instrument could detect light elements with low K-shell electron excitation energy such as carbon, oxygen, fluorine, sodium and silicon. Effect of the current intensity of gallium ion beam on the normalized X-ray yield was investigated. It was found that a strong irradiation beam current could effectively enhance the normalized X-ray yield. [DOI: 10.1380/ejssnt.2006.365]

Keywords: low energy ion beam; X-ray emission; organic and insulating materials; the normalized X-ray yield

I. INTRODUCTION

Organic and insulating materials are important constituents of the natural world. To study the elemental composition information of them, X-ray emission spectroscopy has been established as a powerful analysis technique. From X-ray emission spectra, the compositions are identified by the characteristic X-ray energy position and the concentrations are deduced from their integral intensities. Among various types of detection methods based on X-ray emission, ions or particles induced X-ray emission has its unique advantages. The dominant merit of particle induced X-ray emission (PIXE) technique is that it is a highly sensitive detection method, so that this technique has been established as a routine analytical technique to characterize elemental composition [1–10]. It is already confirmed that PIXE analysis has important applications in various materials such as medieval stained glass [11], magnesium aluminate spinel [12], human skin sections [13], mineral assemblages and base-metal ores [14], proteins [15], trace elements inside plants [16], and so on.

However, conventional PIXE techniques need ions with primary energy of several millions of voltages to bombard the surface of the target samples, especially in case of use of light element gas ions such as proton and helium [17–19]. Compared to high energy ion irradiation, low energy ion irradiation technique has its unique characteristics. It was expected that ion bombardment with low kinetic energy only cause slight damage at the surface of specimen [22–25]. The low kinetic energy of the projectile limit the total path and region where the energetic ion penetrate a solid surface and therefore show different movement cascade from the case of high energy ion irradiation [26–28].

Recently, it was found that heavy ions enable to induce characteristic X-ray from the metal samples even with low primary energy [20]. Furthermore, it has been reported that the signals of the characteristic X-ray with energy less than about 2 keV could be enhanced if the samples are of insulating [21]. This enhancement was deduced to be due to the charge up effect, but the systematic studies

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FIG. 1: (a) Experimental set-up and (b) schematic diagram of JEM9310-FIB.
and detailed analysis have not been carried out yet. In the present work, characteristic X-ray emission from organic and insulating materials was measured using a low energy Ga ion beam with 30 kV primary energy to investigate the features of X-ray signals of light elements with K-shell electron excitation energy, and the influence of irradiation beam current on the X-ray yield was systematically examined.

II. EXPERIMENTAL

The ion beam irradiation experiments were carried out using a JEM-9310 focused ion beam (FIB) system equipped with an energy dispersive X-ray spectrometer (EDS) detector (EX-94013JNU analysis system with an energy resolution of 0.138 keV). X-ray emission measurements were carried out in the main chamber under a base pressure of $1 \times 10^{-4}$ Pa. The gallium ion beam of the FIB, with working voltages ranging from 5 kV to 30 kV and emission currents ranging from $0.8 \times 10^{-12}$ A to $6 \times 10^{-9}$ A, was utilized to irradite different types of organic and insulating samples. Intensity counts of each X-ray spectrum were normalized by the dose of gallium ion (beam current multiplies irradiation time).

We chose a series of organic and insulating specimens for X-ray analysis. Leaf and wood was picked up from a fir tree and an ilex tree respectively and then dried at 100°C for 12 hours. Teflon, SiO$_2$, Al$_2$O$_3$ and glass specimens were commercially bought. The glass specimen consists of silicon, oxygen and sodium elements. Each sample was cut into a rectangle shape using a slice saw. Organic samples and glass have a dimension of 2 mm (wide) $\times$ 2 mm (length) $\times$ 1 mm (thick). SiO$_2$ and Al$_2$O$_3$ specimens have a dimension of 2 mm (wide) $\times$ 2 mm (length) $\times$ 0.2 mm (thick).

Figures 1(a) and 1(b) show the experimental setup of JEM-9310 FIB system and the schematic diagram of its basic working principle, respectively. The gallium emitter is composed of a Ga liquid-metal ion source (LMIS). All of the ion beam induced X-ray emission measurements shown here were performed using this FIB-based system.
TABLE I: Detection results of organic and insulating samples as shown in Figs. 2a-2f. Symbol meaning: X — successfully detected; o—appear due to contamination, not main constitutes of samples.

| Fig. sequence | Fig. 2a | Fig. 2b | Fig. 2c | Fig. 2d | Fig. 2e | Fig. 2f |
|---------------|--------|--------|--------|--------|--------|--------|
| Sample       | Teflon | Wood   | Leaf   | SiO$_2$ | Al$_2$O$_3$ | Glass  |
| C-K, 0.27 KeV| X      | X      | X      | X      | o      | o      |
| O-K, 0.52 KeV| X      | X      | X      | X      | X      | X      |
| F-K, 0.67 KeV| X      |        |        |        |        |        |
| Al-K, 1.48 KeV|       |        |        |        | X      |        |
| Na-K, 1.04 keV|       |        |        |        |        | X      |
| Si-K, 1.74 KeV|       |        |        |        |        | X      |

The working distance used here is 40 mm and the angle between specimen surface and EDS detector is 40°.

III. RESULTS AND DISCUSSION

Figures 2(a) to 2(f) show the results of X-ray measurement from teflon, wood, leaf, SiO$_2$, Al$_2$O$_3$ and glass using a 30 kV Ga ion beam. Each X-ray spectrum consists of several characteristic X-ray peaks superimposed on a background due to various atomic bremsstrahlung processes. The beam current used here was 1.185 x 10$^{-9}$ A and the acquisition time was 50 s (live time) for each sample. Thus, the X-ray counts of each spectrum could be normalized by dividing a factor of 1.185 x 50 = 59.25 x 10$^{-9}$ C (dose of gallium ion).

As shown in Figs. 2(a)-2(f), carbon-K peak at 0.27 keV, oxygen-K peak at 0.53 keV and fluorine-K peak at 0.67 keV could be found from Teflon; carbon-K peak at 0.27 keV and oxygen-K peak at 0.53 keV could be found from wood and leaf; oxygen-K peak at 0.53 keV and silicon-K peak at 1.74 keV could be found from SiO$_2$ sample; oxygen-K peak at 0.53 keV and aluminum-K peak at 1.49 keV could be found from Al$_2$O$_3$ sample; oxygen-K peak at 0.53 keV, sodium-K peak at 1.04 keV and silicon-K peak at 1.74 keV could be found from glass sample. These data were summarized in Table I in detail. The weak carbon-K signals showed in the EDS spectra of SiO$_2$, Al$_2$O$_3$ and glass might come from the possible contamination.

Hence, X-ray emission phenomenon could be observed from various types of organic and insulating materials under irradiation of a low energy (30 kV) gallium ion beam. These EDS results clearly show that light elements from organic and insulating samples, especially for those elements with low K-shell electron excitation energy such as carbon, oxygen, fluorine, sodium and silicon could be detected.

To obtain a satisfied EDS spectra with high ratio of signal to background, important factor such as beam current, which would affect the X-ray yield, was investigated and the results were shown below.

During the process of gallium ion beam bombarding the surface of insulating samples, current intensity of irradiation ion beam is an important parameter determining the X-ray yield. To study the relationship between beam current and X-ray yield, we applied three different beam currents. The values of beam currents used were 0.402 x 10$^{-9}$ A, 1.185 x 10$^{-9}$ A and 3.09 x 10$^{-9}$ A, respectively. We kept the irradiation time (50 seconds) and the size of the scanned area to be constant, while only the irradiation current was changed. The X-ray counts of each set of data was divided by its corresponding ion dose, i.e., 0.402 x 50 = 20.1 x 10$^{-9}$ C, 1.185 x 50 = 59.25 x 10$^{-9}$ C and 3.09 x 50 = 154.5 x 10$^{-9}$ C, respectively. Hence, the normalized X-ray yield of the three spectra could be compared reasonably and the effect of current intensity on normalized X-ray yield will be known.

Figure 3(a) is a combined X-ray spectra with normalized X-ray yield, which shows the detection results under all the three irradiation beam currents as listed above. Based on Fig. 3(a), the integral intensities of carbon-K...
peak and fluorine-$K$ peak of each spectrum were calculated and the results were illustrated in Fig. 3(b). Briefly speaking, Fig. 3(b) shows X-ray yield as a function of ion beam current. The integral arithmetic is, in fact, an accumulation sum of intensity counts in the range of from 0.18 to 0.36 keV for C-$K$ and from 0.58 to 0.76 keV for F-$K$ after subtracting an average background, respectively. An average background is used for background subtraction when we calculate the integral X-ray yield of one characteristic peak such as C-$K$ and F-$K$, because the characteristic peak shown in the EDS spectrum is a superimposed one, together with the background. For example, to compute an average background, at first, we accumulate the intensity values from 0.09 eV to 0.13 eV and from 0.41 eV to 0.45 eV and get an average value by dividing the total channel number (10, in this case). Second, after summing the intensity values from 0.14 eV to 0.40 eV for C-$K$ peak, a background value resulting from the average background multiplying channel number (27, in the case) is subtracted. Thus, an actual integral area over the characteristic peak could be obtained, in which the background contribution was not included.

It is found from Figs. 3(a) and 3(b) that with the irradiation current increasing, the X-ray yields of both carbon-$K$ peak and fluorine-$K$ peak increase correspondingly. For example, while the beam current increases from $0.402 \times 10^{-9}$ A to $1.185 \times 10^{-9}$ A and $3.09 \times 10^{-9}$ A, the integral counts of C-$K$ peak increases from $\sim 658$ to $\sim 1645$ and $\sim 2070$; the integral counts of F-$K$ peak increases from $\sim 825$ to $\sim 1690$ and $\sim 2525$, respectively. Thus, based on the above experimental data, it was concluded that current intensities of Ga ion beam have effects on the X-ray yield and intense ion beam can induce strong X-ray from specimen. The possible mechanism behind the current effect of gallium ion beam on the X-ray yield will be addressed in the future.

IV. CONCLUSIONS

In summary, X-ray emission was observed from the surface of various types of organic and insulating materials under irradiation of a low energy (30 kV) gallium ion beam. Light elements with low $K$-shell electron excitation energy such as carbon, oxygen, fluorine, sodium and silicon could be detected by an energy dispersive X-ray spectrometer together with a focused ion beam instrument. The influence of irradiation beam current on the normalized X-ray yield was studied. It was concluded that a strong beam current could evidently enhance the normalized X-ray yield from organic and insulating specimens.

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