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Nondestructive measurement of surface tritium by β-ray induced X-ray spectrometry (BIXS)

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Abstract

Applicability of a newly developed β-ray induced X-ray spectrometry (BIXS) has been examined to measure nondestructively tritium retained on/in the graphite samples. Examination was carried out by using the graphite plates irradiated with tritium ions and an ALT-II limiter tile exposed to D-plasmas in TEXTOR. For the former samples, a sharp intense peak and a broad weak peak appeared clearly in the spectra; the former peak was attributed to the characteristic X-rays from argon used as a working gas, and the latter peak was assigned to the bremsstrahlung X-rays from sub-surface layers of graphite. On the other hand, for the latter sample, a rather weak characteristic X-ray peak was observed along with a diminutive bremsstrahlung X-ray peak. Although the intensities of those X-rays differed from spot to spot, the tritium levels retained on the limiter tile were determined to be 58–132 Bq/cm^2. It was concluded, therefore, that valuable information on the amount and the distribution of tritium retained on/in the wall materials can be nondestructively obtained by using the BIXS. © 2001 Elsevier Science B.V. All rights reserved.

Keywords: Surface tritium measurement; β-ray induced X-ray spectrometry (BIXS); Tritium retention; Graphite materials

1. Introduction

The inner walls of a thermonuclear fusion reactor are exposed to high level of atomic and ionic tritium, causing tritium accumulation in the wall materials. The tritium accumulation will affect the fuel particle balance in the fusion reactor. Further, such tritium retention can bring about a serious problem for safe maintenance work and waste processing of the wall materials. From these viewpoints, the tritium assay will be one of the most important subjects to estimate the precise amount and distribution of tritium on/in the wall materials in a thermonuclear fusion reactor [1,2].

A variety of measurement techniques have been proposed so far for this purpose. Recently, for example, quantitative depth profiling of deuterium and tritium in samples cut from graphite protection tiles at the vessel walls of the fusion experiment ASDEX-Upgrade was carried out using the accelerator mass spectrometry (AMS) by Sun et al. [3]. This method represents basically an extension of a negative secondary ion mass spectrometer with a magnetic mass separator of low mass resolution. In addition, three kinds of techniques were applied to evaluate the surface tritium activity for the bumper limiter tiles in TFTR [4]: the first is an open wall ion chamber, the second ultra thin thermoluminescence dosimeters, and the last a pin diode to detect the charged particles. On the other hand, the surface

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distribution of tritium retained on both the plasma-facing and rear surfaces of ALT-II limiter tiles in TEXTOR was measured using a imaging plate [5]. However, these techniques cannot nondestructively measure tritium existed in a deeper region than the escape depth of β-rays from tritium.

A new nondestructive measurement technique has been proposed by Matsuyama et al. [6,7] for the purpose of in-situ measurements of tritium concentration distribution in bulk of the metallic materials. This technique is based on utilization of the phenomena that emission of β-rays from tritium absorbed in a material gives rise to X-rays, i.e., bremsstrahlung X-rays and characteristic X-rays. The former X-rays form a continuous spectrum. Shape of the spectrum is dependent on a depth profile of tritium, but independent on the total amount of tritium in the bulk. On the other hand, the latter X-rays depend on both the amount and the depth profile of tritium. It is most likely that this technique is principally applicable to measure tritium retained in a surface region of the carbon materials. However, it is hard to employ the characteristic X-rays from carbon atoms to this technique because the characteristic X-rays are low energy (0.277 keV). In addition, since the stopping power of the carbon materials for the β-rays is small, the β-rays can penetrate through the carbon materials of about 1 μm in thickness. Such the escaped β-rays cannot be detected by the present technique.

Because of a precise measurement of surface tritium, we made an important improvement of the technique mentioned above on detection of the β-rays escaped from a surface region of the carbon materials. The improvement is a point that the carbon materials adsorbed/absorbed tritium are kept in an argon atmosphere during the measurement. In this case, the characteristic X-rays of argon will be simultaneously observed along with bremsstrahlung X-rays emitted from bulk of a carbon material. Namely, the amount of surface tritium will be determined from intensity of the characteristic X-rays of argon.

In the present study, applicability of a newly developed β-ray induced X-ray spectrometry (BIXS) was examined using small isotropic graphite plates irradiated with tritium ions, and the BIXS was applied to measure one of ALT-II limiter tiles in TEXTOR.

2. Experimental

2.1. Tritium retention samples

To examine the X-ray spectra induced by the β-rays from tritium retained on the surface of graphite, two kinds of graphite samples containing tritium were prepared: one was made by the implantation of tritium ions into a graphite plate, and the other was one of ALT-II limiter tiles exposed to D-plasmas in TEXTOR. The former sample was prepared by using a specially designed tritium implantation device constructed in the Hydrogen Isotope Research Center, Toyama University, while the latter one was the tile used for D–D experiments for a long period from March 1997 to July 1998. The location of the tile was G8 from blade 4 in TEXTOR. This limiter tile has been exposed to D-plasma for about 5×10^3 s.

The conditions of tritium implantation into a graphite plate were as follows. Small graphite plates were used for the tritium implantation as delivered from Toyo Tanso; the type and the size of them were IG-430U and $15 \times 15 \times 0.5$ mm^3, respectively. Details about a tritium implantation device are given in [8]. The irradiation procedures are briefly noted below. After one of the graphite plates was loaded with a sample holder in an irradiation chamber, it was heated at 673 K and evacuated below 1.3×10^{-4} Pa. The graphite plate was cooled down to room temperature, and then tritium ions were implanted into the graphite plate for a given time. The diameter of an irradiation spot was controlled to be 6 mm. During the irradiation, working pressure in the irradiation chamber was kept at 3 Pa, and the acceleration energy of tritium ions was set 1 keV. Under these conditions, the current observed was around 0.2 μA. Tritium used for the irradiation was diluted with deuterium, and the tritium concentration was determined as 0.7 at.% by using a bremsstrahlung counting method [9]. To minimize the tritium release followed the opening of a sample entrance port, the evacuation of tritium in the chamber was continued for three days and then the inner wall of the chamber was thoroughly decontaminated using a specially designed tritium removal device. After the implantation sample was taken out, it was kept at ambient temperature and atmosphere until measurements of X-ray spectra are conducted.

2.2. Measurements of X-ray spectra

Fig. 1 shows a schematic diagram of the device used to assay a graphite plate irradiated with tritium ions. The device consisted of a X-ray detector, a cryostat, rectangular lead blocks drilled a semi-cylindrical hole and a sample holder. The X-ray detector was equipped with a high pure thin germanium crystal, which is capable of measuring ultra-low energy X-rays. During a measurement of an X-ray spectrum, the distance between a sample and the detector was kept at 5.0 mm, and the space between them was filled with a constant flow rate of argon. In addition, to diminish the effect of natural radioactivity as low as possible, both the sample and the detector were perfectly sandwiched in between two thick lead blocks except for the sample entrance port. In measurements of ALT-II limiter tile, perfect

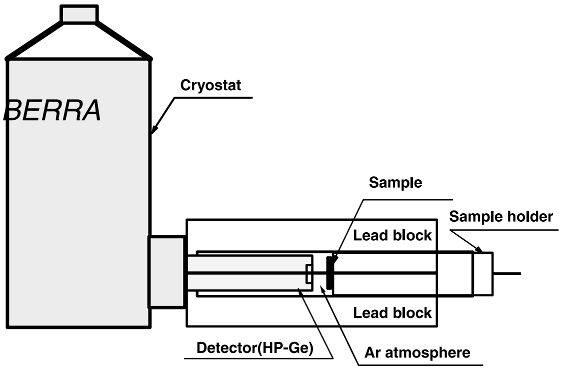

Fig. 1. Schematic diagram of the device used for measurements of X-ray spectra.

shield needed a lot of lead blocks because of large size (114×155 mm^2). Under such configuration, background level of the X-ray detector was reduced as low as 1.71×10^{-2} count/s. Energy resolution (full width at half maximum) of the present detector was below 130 eV at 5.9 keV.

3. Results and discussion

3.1. Tritium ion implantation samples

Fig. 2 shows an example of X-ray spectra observed for graphite plates in an argon atmosphere. Irradiation for this graphite plate was carried out for 25 min. A remarkably intense peak appeared around 3 keV, and it was partly overlapped with a broad peak. The former peak was attributed to the characteristic X-rays of Ar(K_α : 2.96 keV). In addition to this peak, although a peak intensity was very weak, Ar(K_β : 3.19 keV) peak could be also observed closely to the Ar(K_α) peak. Total intensity of both characteristic X-rays amounted to 6.11×10^{-1} counts/s in the energy range of 2.8 to 3.2 keV. On the other hand, the latter broad peak as shown in the inset was attributed to bremsstrahlung X-rays produced mainly by interactions between β-rays and carbon atoms. The total intensity of the bremsstrahlung X-rays amounted to 4.25×10^{-1} counts/s in the energy range of 1.0 to 15 keV, while the background in the same energy region was the intensity of 1.71×10^{-2} count/s, which was much smaller than that of the bremsstrahlung X-rays observed.

By using a conventional combustion method, the total amount of tritium contained in this graphite plate was evaluated to be 1.5×10^5 Bq, which corresponds to 8.4×10^{13} tritium atoms. Such a tritium activity was well reproduced in each irradiation. The acceleration energy of tritium ions was controlled to be 1 keV as mentioned previously. It appears, therefore, that most of tritium implanted in the graphite plates would stay within subsurface layers below about 20 nm in depth. Compared to the range of tritium ions, the escape depth of the β-rays in graphite, about 1 μm, is considerably large because of the fairy small stopping power of β-rays in carbon atoms. This indicates that most of the β-rays can play a major role for generation of the characteristic X-rays of argon, penetrating through subsurface layers of the graphite. Namely, it was seen that the amount of tritium in the sub-surface layers can be evaluated from the intensity of characteristic X-rays observed.

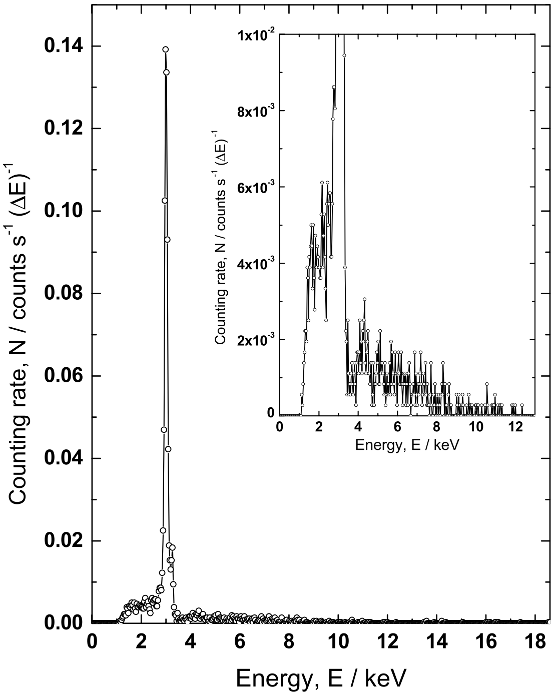

Fig. 2. A typical spectrum observed for graphite plates irradiated with tritium ions.

The broad peak which overlapped partly with the characteristic X-ray peak was assigned to the bremsstrahlung X-rays from sub-surface layers of graphite. Similar X-ray peak profile was observed for samples irradiated with different amounts of tritium. The peak intensities and profiles will give important information to estimate not only the amount but also the depth profile of tritium retained in a graphite plate. Analysis of a tritium depth profile in the materials is described in [6].

3.2. ALT-II limiter tiles exposed to D–D shots

Fig. 3 shows a photograph and the spots of the plasma-facing surface of the limiter tile where the

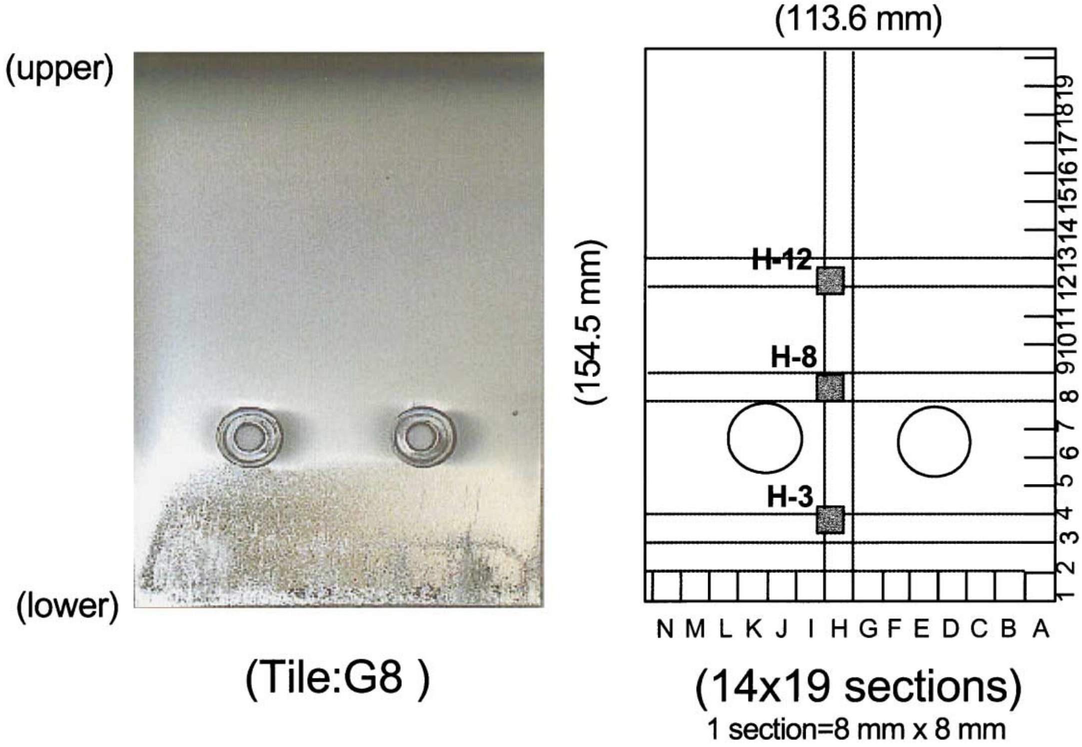

Fig. 3. Photograph (plasma-facing surface) of an ALT-II limiter tile and measuring spots.

surface tritium was measured. Tritium assay was carried out for three spots (H-3, H-8, and H-12) of the plasma-facing surface. It can be seen from the photograph that deposition layers exist on the surface of a part lower than two holes to fix the tiles.

Three X-ray spectra observed are shown in Fig. 4. Each spectrum was obtained by measuring for about 100 h to minimize the statistic error because the considerably small amount of tritium was expected. There appeared a weak characteristic X-ray peak of Ar and a diminutive bremsstrahlung X-ray peak in the spectra. Although the intensities of those X-rays differed from spot to spot, this is a clear indication that a rather small amount of tritium is captured on the surface and it is not distributed uniformly on the surface. Namely, it is suggested that the present method is similarly applicable to measure tritium species trapped on the wall materials in other fusion experimental devices.

The X-ray intensities in each spot are summarized in Table 1. It should be noticed here that the intensity of bremsstrahlung X-rays does not similarly correspond to that of characteristic X-rays because the former X-rays depend strongly on a tritium depth profile and conversion efficiency to X-rays is different in each X-rays. Further investigation needs to solve this problem. Therefore, although the total amount of tritium in the tile cannot be determined quantitatively from the intensities of the bremsstrahlung and characteristic X-rays, it is possible to estimate qualitative difference of a tritium depth profile from the bremsstrahlung X-ray spectra. The intensity of bremsstrahlung X-rays was the highest at the spot H-3, which corresponds to the highest tritium concentration in the bulk. The deposition layers were clearly observed at this spot. On the contrary, the spot H-8 was lowest, where the surface was eroded. Namely, these indicate that such difference in tritium distribution is strongly related to phenomena of erosion and deposition on the surface of the limiter tiles. The highest tritium concentration at the H-3 is in good agreement with the results of deuterium distribution determined by a nuclear reaction [10].

On the other hand, an evaluation of the amount of tritium in a shallow sub-surface layer from the intensity of characteristic X-rays was performed. The evaluation needs several correction factors such as the conversion efficiency from β-rays to characteristic X-rays of Ar, the geometric factor, the absorption factors for the protection materials of the detector, and so on. Thus the detection efficiency including those correction factors was previously determined by using a standard tritium source whose surface radioactivity was evaluated precisely. As a result, it was evaluated to be

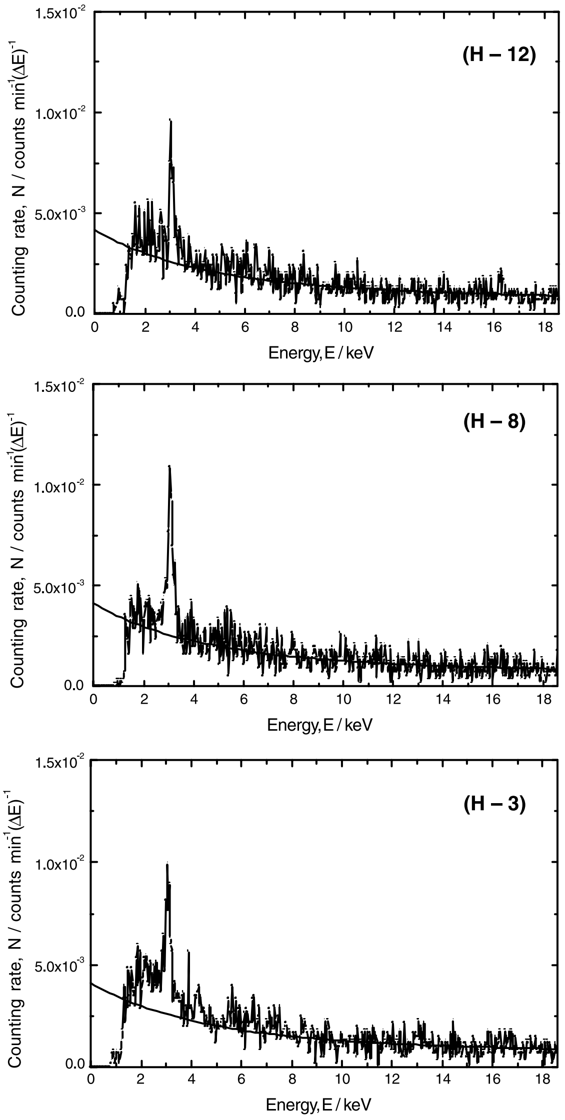

Fig. 4. The X-ray spectra for each measuring spot. Solid lines represent a background level measured without a limiter tile.

2.6×10^{-5} (photons/β-particle). Taking into account this efficiency, for example, the amount of tritium at the H-8 spot was estimated to be 132 Bq/cm^2, which corresponds to 7.4×10^{10} T-atoms/cm^2. Tritium concentration of other spots can be also determined in the same way. This is the first quantitative measurement of tritium formed by D–D experiments in TEXTOR. In addition, the detection limit of the present device was estimated as low as 30 Bq/cm^2, depending on the background level of the detector. Accordingly, it was concluded that the present method is thoroughly applicable to nondestructive measurements relating to the amount and the distribution of tritium retained on/in the wall materials.

4. Conclusions

Applicability of a newly developed BIXS was examined using small isotropic graphite plates irradiated with tritium ions, and the BIXS was applied to measure one of ALT-II limiter tiles in TEXTOR. As a result, the following conclusions were obtained.

For the graphite samples irradiated with tritium ions, a sharp intense peak attributed to the characteristic X-rays from Ar, which was used as a working gas to convert effectively β-rays from sample surfaces to X-rays, appeared around 3 keV. In addition to this peak, a broad weak peak was observed. The latter peak was assigned to the bremsstrahlung X-rays from sub-surface layers of graphite induced by tritium β-rays in the sample. The peak intensities and profiles give important information to estimate not only the amount of tritium on the surface but also the distribution of tritium in the sample.

For the ALT-II limiter tiles exposed to D-plasmas in TEXTOR, there appeared a rather weak characteristic X-ray peak of Ar and a diminutive bremsstrahlung X-ray peak in the spectra. Although the intensities of those X-rays differed from spot to spot, the tritium levels retained on the surface were determined to be 58–132 Bq/cm^2. This is the first quantitative nondestructive measurement of tritium formed by D-D experiments in TEXTOR. Taking into account the background level of the detector, the detection limit of tritium concentration was estimated as low as 30 Bq/cm^2. It is concluded, therefore, that the present method is quite useful for nondestructive measurements of tritium retained on/in plasma-facing materials.

Table 1
Summary of the intensities of both X-ray peaks with measuring spots[a]

X-rays	Spot no.		
	H-3	H-8	H-12
Characteristic	3.74×10^{-4}	8.57×10^{-4}	5.74×10^{-4}
Bremsstrahlung	2.03×10^{-3}	0.68×10^{-3}	0.92×10^{-3}

[a] (UNIT: counts/s).

References

[1] W.R. Wampler, B.L. Doyle, A.E. Pontau, J. Nucl. Mater. 145&147 (1987) 353.

[2] W.R. Wampler, B.L. Doyle, S.R. Lee et al., J. Vac. Sci. Technol. A 6 (1988) 2111.

[3] G.Y. Sun, M. Friedrich, R. Grotzschel et al., J. Nucl. Mater. 246 (1997) 9.

[4] C.A. Gentile, C.A, Skinner et al., in: Fifth International Symposium on Fusion Nuclear Technology, Rome, 19–24 Sept. 1999, P5-8-26, Fus. Eng. Design, in press.

[5] T. Tababe, private communication.

[6] M. Matsuyama, K. Watanabe, K. Hasegawa, Fus. Eng. Design 39&40 (1998) 929.

[7] M. Matsuyama, S. Ueda, K. Watanabe, in: Fifth International Symposium on Fusion Nuclear Technology, Rome, 19–24 Sept. 1999, P3-3-30, Fus. Eng. Design, in press.

[8] M. Matsuyama, S. Ueda et al., Ann. Rep. Hydrogen Isotope Res. Center, Toyama University, 18 (1998) 69.

[9] M. Matsuyama, K. Watanabe, Fus. Technol. 28 (1995) 1045.

[10] M. Rubel, private communication.