

## 論 文

### 厚いアクリル樹脂製のトリチウム線源で観測された X線スペクトルの解析及びシミュレーション

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Analysis and Simulation of  $\beta$ -Ray-Induced X-Ray Spectrum Observed  
for a Thick Tritium Source Made of Methylmethacrylate Resin

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#### Abstract

The  $\beta$ -ray-induced X-ray spectra observed for a polymer disk (1 mm thickness), which was made of poly-methylmethacrylate labeled with tritium, were analyzed and they were reproduced by computer simulation based on the uniform distribution of tritium in the polymer disk. The X-ray spectra were measured in both helium and argon atmospheres. An X-ray spectrum observed in the helium atmosphere consisted of only a broad bremsstrahlung X-ray peak, while in the argon atmosphere a characteristic X-ray peak appeared in addition to bremsstrahlung X-rays. From differences in the shape and intensity of X-ray spectra observed in both atmospheres, the conversion fraction of the bremsstrahlung X-rays to the characteristic X-rays in an argon atmosphere was determined to be  $7.1 \times 10^{-2}$  by taking the absorption coefficient of X-rays into account. The spectra observed in both atmospheres could be reproduced quite well by using the equations to estimate generation and attenuation of X-rays in various materials. It was suggested, therefore, that a tritium depth profile in carbonaceous materials can be evaluated by the numerical simulation. Furthermore, the simulation indicated that nitrogen could be also used as a promising alternative gas of helium for evaluation of a tritium depth profile in bulk.

## 1. Introduction

In the International Thermonuclear Experimental Reactor (ITER) and a future advanced reactor, a huge amount of tritium will be required as fuel in comparison with that in the conventional tracer studies. Therefore, quantitative evaluation of tritium activity not only in a gaseous state but also a dissolved state in materials is indispensable from a viewpoint of safe handling in the fusion reactors. Especially, evaluation of the amount and depth profile in the latter state is of a great importance for safe management of tritium wastes. However, the penetration depth of  $\beta$ -rays from tritium is very short because of their low energy. This is a reason why it is difficult to measure the amount of tritium retained in bulk of a material by means of  $\beta$ -ray detection.

Electromagnetic radiation, however, is emitted with  $\beta$ -rays as a result of interactions between  $\beta$ -rays and constituent atoms, when tritium dissolves in a material: characteristic and bremsstrahlung X-rays are emitted. Penetration power of the X-rays is much higher than that of the  $\beta$ -rays, and therefore X-rays give information on the amount of tritium in a deeper region of a material. Intensity and shape of these X-ray spectra depend on the amount and the depth profile of tritium in bulk of the material. In particular, the shape of a bremsstrahlung X-ray spectrum basically reflects the depth profile of tritium. Analyzing the shape of a bremsstrahlung X-ray spectrum by numerical simulation, therefore, the depth profile of tritium can be estimated. The present method has been applied so far to some inorganic materials to examine the amount and changes in the depth profile of tritium [1-4], but in most of cases the depth profile of tritium was only a near surface region shallower than 1  $\mu\text{m}$ .

In the present study, a polymer disk (1 mm in thickness) labeled with tritium was employed for the examination, and the X-ray spectra observed in argon and helium atmospheres were analyzed in detail. In addition, to examine the applicability of the present simulation method to a thicker material, reproduction of the X-rays spectra was carried out by numerical simulation based on a uniform distribution of tritium in the polymer disk.

## 2. Experimental

### 2.1. Materials

A polymer disk used in this study was made of poly-methylmethacrylate that was uniformly

labeled with tritium (hereafter described as T-PMMA), and the size was 25mm in diameter and 1mm in thickness. This was delivered from Amersham Japan Co., and the specific activity was 178.7 MBq/g (15 March, 1999). Weight of the disk was 0.584 g. Purity of argon gas used for measurements of  $\beta$ -ray-induced X-rays was 99.995%, and that of helium was 99.9995%.

## 2.2. Measurements of $\beta$ -ray-induced X-ray spectra

A semiconductor detector equipped with a high purity Ge element was used for measurements of  $\beta$ -ray-induced X-rays. Energy resolution of the present X-ray detector was determined to be 129 eV at 5.9 keV by using a  $^{55}\text{Fe}$  source. The radiation entrance window of the X-ray detector was made of a specially designed thin beryllium plate (8  $\mu\text{m}$  in thickness) to gain an effective transmittance of low energy X-rays. The polymer disk was fixed at location of 5mm from the front of the radiation entrance window of the X-ray detector. The X-ray spectra were measured in both argon and helium atmospheres. During a measurement, these gases were kept at the constant flow rate of 40  $\text{cm}^3/\text{min}$ . In the former atmosphere X-rays induced by tritium of surface and bulk can be detected, while in the latter atmosphere X-rays induced by tritium in bulk can be mainly measured. To reduce the effects of natural radiations on the X-ray measurements, the X-ray detector was surrounded with two lead bricks during a run. The schematic drawing of the present measuring device is shown in Fig. 1.

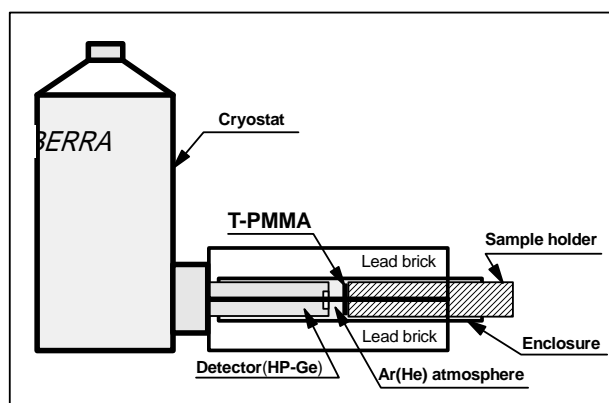


Fig. 1: Cross sectional view of the  $\beta$ -ray-induced X-ray measuring device.

## 2.3. Simulation of the observed X-ray spectra

The simulation of the observed X-ray spectra was carried out by the aid of computer. Basic equations for numerical calculations of the simulated spectra were shown elsewhere [5]. In the calculation process, absorption coefficients of various materials are important parameter to calculate the attenuation of X-rays, which affect the shape and intensity of X-ray spectra. Table 1 shows the summary of energy dependence of mass absorption coefficient for each material in the present measuring condition.

### 3. Results and discussion

#### 3.1. Analyses of the observed X-ray spectra

Figure 2 shows a typical spectrum of the background level for the present measuring system. The X-ray spectrum was obtained by accumulating for 72 hrs. The long accumulation is to obtain a precise background spectrum. The background level was as low as  $(2-5) \times 10^{-3}$  counts/min $\Delta E$  in the interesting region, while a steep increase appeared below 1 keV. It is considered that

this is mainly due to noise of electric circuits of the detector. The value of  $\Delta E$  was set at 34.5 eV under the present measuring conditions. Total intensity of the background was 1.03 counts per minute (hereafter described as cpm) in a range from 1.0 to 18.6 keV.

Figures 3 and 4 show the X-ray spectra observed in helium and argon atmospheres, respectively. A broad X-ray peak was observed as shown in Fig. 3, and this is bremsstrahlung X-rays based on interactions between  $\beta$ -rays and constituent atoms in the polymer disk. Intensity of

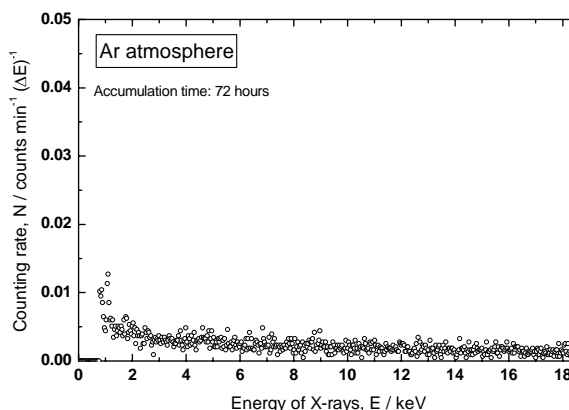


Fig. 2: A typical background spectrum observed in the argon atmosphere.

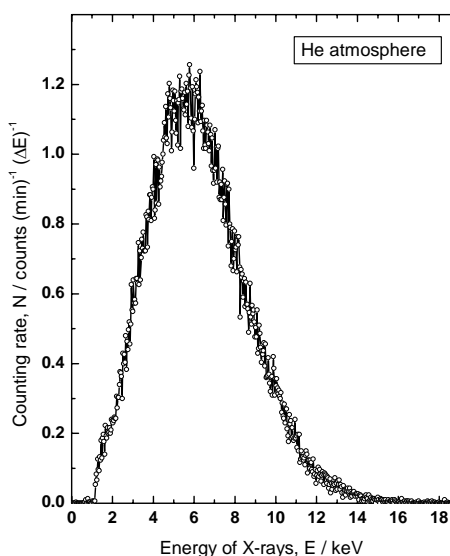


Fig. 3: The X-ray spectrum observed in the helium atmosphere.

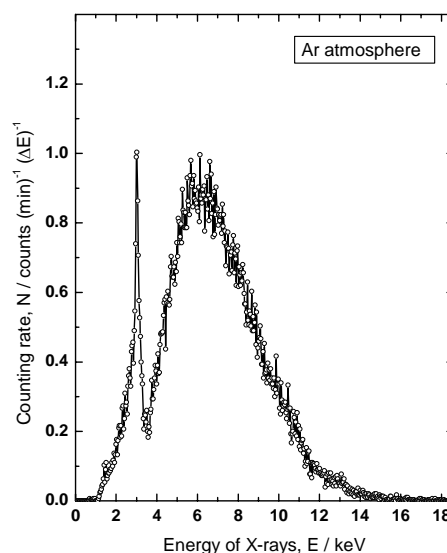


Fig. 4: The X-ray spectrum observed in the argon atmosphere.

Table 1: Summary of energy dependence of a mass absorption coefficient for each material employed in the present measuring device.

Absorber / thickness	K-Absorption Edge (keV)	Mass Absorption Coefficient <sup>#3</sup> (cm <sup>2</sup> / g)	
		$\mu(1)$	$\mu(2)$
T-PMMA (1 mm) #1	0.2838 #2	2835E <sup>-2.8984</sup>	
Ar gas (5 mm)	3.2029	3350E <sup>-2.7104</sup>	40519E <sup>-2.8169</sup>
Be window (8 $\mu$ m)	0.111	620E <sup>-2.9250</sup>	
Al-evaporated film (200nm)	1.5599	1200E <sup>-2.5750</sup>	23100E <sup>-2.9250</sup>
Poly-carbonate film (2 $\mu$ m)	0.2838 #2	1753E <sup>-2.8777</sup>	
Dead layers of Ge crystal (100nm)	11.1036	16015E <sup>-2.6268</sup>	110571E <sup>-2.6268</sup>

#1 T-PMMA: poly-methylmethacrylate labeled with tritium (T-C<sub>4</sub>H<sub>6</sub>O<sub>2</sub>)<sub>n</sub>.

#2 The energy is available for carbon atoms.

#3 Values of  $\mu(1)$  and  $\mu(2)$  correspond to a energy range below and above the K-absorption edge, respectively.

the bremsstrahlung X-rays was 198.8 cpm. As is shown in Fig. 4, on the other hand, a characteristic X-ray peak of argon appeared in addition to the bremsstrahlung X-ray peak, although a total X-ray intensity observed in argon atmosphere was a little lower. Intensities of the bremsstrahlung and the characteristic X-rays were evaluated to be 145.4 and 4.82 cpm, respectively.

Two kinds of generation processes for characteristic X-rays are considered: the excitation of argon atoms is caused by not only collisions with  $\beta$ -rays but also the photoelectric effect of bremsstrahlung X-rays. The former excitation is led by colliding with  $\beta$ -rays from tritium existing within a few  $\mu$ m beneath the surface, whereas the latter excitation is responsible for all photons having energy above 3.203 keV (absorption edge of the K-shell in argon atoms), which are emitted from the bulk of the polymer disk. In this paper, the surface means a shallower depth than a penetration depth of  $\beta$ -rays, and the bulk means a deeper region than the penetration depth.

The absorption fraction of bremsstrahlung X-rays in argon atmosphere can be determined from a difference in the intensity of bremsstrahlung X-rays in both atmospheres, while one can also estimate it by using a mass absorption coefficient of X-rays for argon. The calculated X-ray

spectrum (solid line) is shown in Fig. 5 together with the spectrum in helium atmosphere, where the mass absorption coefficient for argon shown in Table 1 was used. The open circles again represent the X-ray spectrum shown in Fig. 3. Both X-ray spectra agreed quite well except for the part of characteristic X-rays. Namely, this indicates that the absorption of the bremsstrahlung X-rays in helium atmosphere is negligibly small, and that the bremsstrahlung X-ray spectrum emitted from the surface of the polymer disk in argon atmosphere can be reproduced by the calculation. Agreement of the two spectra plays an important role for computer simulation of the observed X-ray spectra. In addition, it was revealed that the difference in the bremsstrahlung X-ray intensity above 3.203 keV in both atmospheres should be mainly responsible for the absorption of X-rays by photoelectric effect, and it was evaluated as 44.28 cpm.

To examine a conversion efficiency (fluorescence yield) of bremsstrahlung X-rays to characteristic X-rays due to the photoelectric effect in argon, an X-ray spectrum was measured in a similar manner by packing the polymer disk with a thin poly-propylene film of 6  $\mu\text{m}$  to shield perfectly the  $\beta$ -rays. As a result, a similar spectrum to that shown in Fig. 4 appeared, but a considerable decrease in the characteristic X-ray intensity was observed, suggesting that the photoelectric effect of bremsstrahlung X-rays is not negligible for the generation of characteristic X-rays of Ar( $K\alpha$ ). The fluorescence yield for argon was finally determined to be  $7.1 \times 10^{-2}$  from the present examination, assuming that absorption of bremsstrahlung X-rays in the poly-propylene film is negligibly small.

The photoelectric effect gives rise to a competitive emission between Auger electrons and characteristic X-rays, and the ratio of both emission processes depends on the atomic number. It is known that a fluorescence yield  $\omega(K)$  for the emission of characteristic X-rays can be approximately estimated by the following equation [6]:

$$\omega(K) = 1/(1+1.12 \times 10^6 Z^{-4}),$$

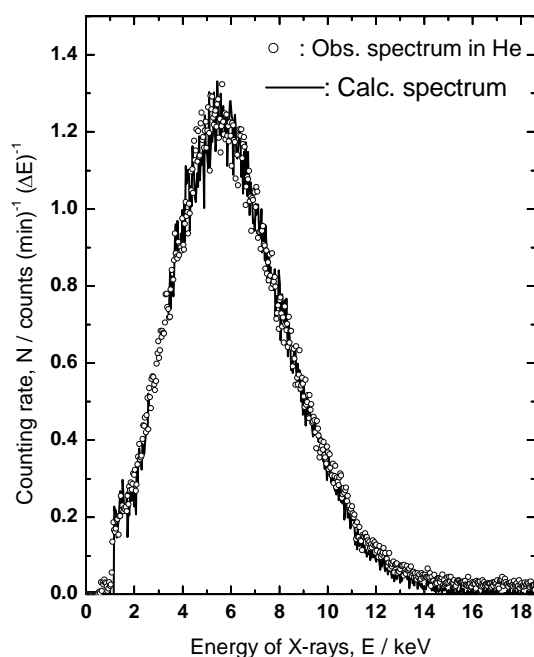


Fig. 5: Comparison of the bremsstrahlung spectrum calculated from the X-ray spectrum observed in argon atmosphere (solid line) with that observed in helium atmosphere (open circles).

where  $Z$  represents the atomic number of a target. For example, applying this equation to the argon system, the value of  $\omega(K)$  is calculated to be  $8.5 \times 10^{-2}$ , which agrees well with the value obtained from the experiment mentioned above. It can be seen, therefore, that the characteristic X-ray intensity described in Fig. 4 consist of the sum of characteristic X-rays induced by  $\beta$ -rays (1.68 cpm) and those induced by bremsstrahlung X-rays (3.14 cpm). A conversion efficiency of  $\beta$ -rays to characteristic X-rays of argon has been already reported to be  $3.1 \times 10^{-4}$  photons/ $\beta$ -particle by Matsuyama et al., which was determined from the irradiation experiments of graphite with tritium ions [3].

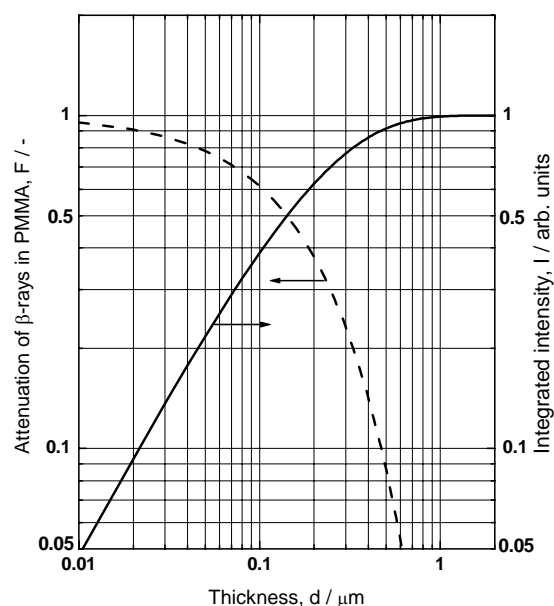


Fig. 6: Attenuation and integral intensity of  $\beta$ -rays as a function of thickness of T-PMMA.

### 3.2. Simulation of the observed X-ray spectra

In order to simulate a  $\beta$ -ray-induced X-ray spectrum by numerical calculation, it is necessary to determine basic two parameters prior to the calculation. The first parameter is the penetration depth of  $\beta$ -rays in T-PMMA. The penetration depth can be estimated by determining the absorption coefficient of  $\beta$ -rays. The absorption coefficient can be calculated from results of total and surface activity measurements for T-PMMA. It was determined to be  $4.89 \times 10^4 \text{ cm}^{-1}$ . The details are described elsewhere [7]. Assuming that the attenuation of  $\beta$ -rays obeys a simple exponential function, as shown in Fig. 6, the average penetration depth is expected to be  $2 \mu\text{m}$  from the above value. Namely, it was assumed in the numerical calculation that the initial kinetic energy of the  $\beta$ -rays is consumed during plowing through T-PMMA of  $2 \mu\text{m}$  in thickness. This penetration depth is remarkably small in comparison with the thickness of T-PMMA.

The second parameter is energy dependence of absorption coefficients of X-rays for T-PMMA, argon and construction materials (Be, Al, Ge, and a thin poly-carbonate film) of the Ge detector. Those values used in the numerical calculation are represented in Table 1.

Figure 7 shows the comparison of the simulation spectra (solid line) with the spectra observed in the argon atmosphere. The detailed numerical calculation process of an X-ray spectrum by

computer simulation is described in the literature [5]. Reduction of the characteristic and/or bremsstrahlung X-rays in the polymer disk and the Ge detector was calculated using parameters summarized in Table 1. The simulation was carried out by basing on the uniform distribution of tritium in the polymer disk. It was seen that the simulation spectra agreed fairly well with the observed spectrum although there appeared to be a little deviation from the observed intensity at a low energy side.

A comparison of simulation spectra calculated in three kinds of atmospheres (He, N<sub>2</sub> and Ar) is shown in Fig. 8. The same calculation parameters were used to calculate for each atmosphere, except that a different parameter is an energy dependence of the mass absorption coefficient. It is clear that the shape of a simulation spectrum for helium atmosphere was quite similar to that of the observed spectrum shown in Fig. 3. This indicates, therefore, that the present simulation of a β-ray-induced X-ray spectrum is applicable to tritium-containing carbonaceous materials as thick as about 1mm.

Furthermore, the simulation spectrum expected for helium atmosphere was perfectly overlapped with that expected for nitrogen atmosphere, although absorption coefficients of X-rays for both atmospheres considerably differed: the mass absorption coefficient (cm<sup>2</sup>/g) for helium is  $56.89E^{-2.8838}$ , and that for nitrogen is  $3354E^{-2.8940}$ , where E describes the X-ray energy (keV). The observation that little difference of X-ray spectra in both atmospheres appeared is due to the following multiplier effects: the first is

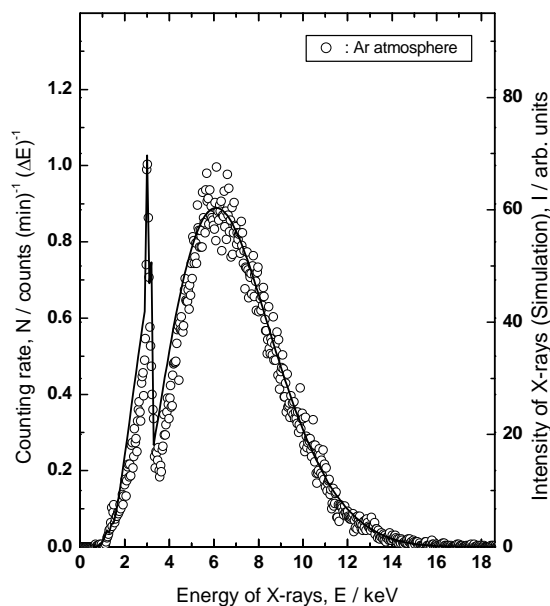


Fig. 7: Comparison between the simulated and the observed spectrum in argon atmosphere.

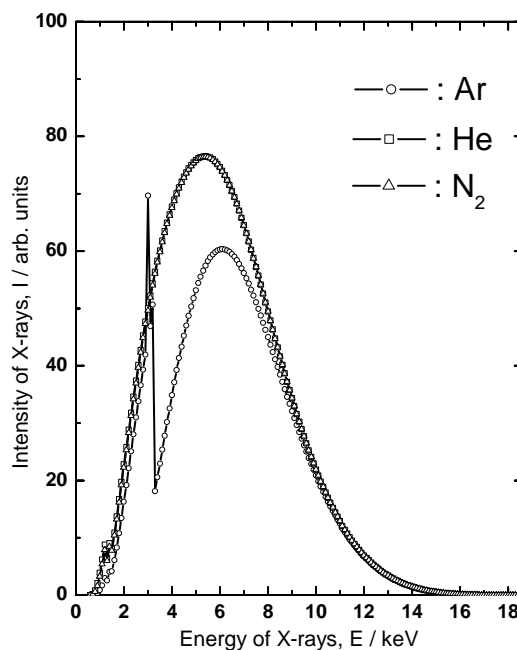


Fig. 8: Simulation spectra for He, Ar, and N<sub>2</sub> atmospheres.



difference in the conversion efficiency to bremsstrahlung X-rays, and the second is the closed distance between the polymer disk and the beryllium window. From viewpoint of practical measurements of a tritium-containing material, the simulation spectra indicate that nitrogen is more useful than helium, because a long time measurement in helium atmosphere is difficult owing to the permeation through a beryllium window of the Ge detector

#### 4. Conclusions

The  $\beta$ -ray-induced X-ray spectra observed for a thick polymer tritium disk, which was made of poly-methylmethacrylate, were analyzed and the reproduction of them was attempted by computer simulation based on the uniform distribution of tritium. The X-ray spectra were measured in both helium and argon atmospheres. An X-ray spectrum observed in the helium atmosphere consisted of only a broad bremsstrahlung X-ray peak, while in the argon atmosphere a characteristic X-ray peak appeared in addition to bremsstrahlung X-rays. Total X-ray intensity observed in the argon atmosphere was about 75% as low as that in the helium. The bremsstrahlung X-ray spectrum calculated using the absorption coefficient of X-rays for argon agreed quite well with that the bremsstrahlung X-ray spectrum observed in helium. The characteristic X-rays of argon were attributed to excitation by both  $\beta$ -rays from tritium in surface layers of the polymer disk and the photoelectric effect of bremsstrahlung X-rays. It was seen that the conversion fraction of the bremsstrahlung X-rays to the characteristic X-rays in an argon atmosphere was  $7.1 \times 10^{-2}$ . The observed spectra in both atmospheres could be reproduced quite well by computer simulation, suggesting that a tritium depth profile in tritium-containing carbonaceous materials can be estimated by simulation analyses. Furthermore, it was seen from the simulation that nitrogen could be used as a promising alternative gas of helium.

#### References

- [1] M. Matsuyama, T. Murai, K. Watanabe, K. Tsuzuki and N. Noda, Ann. Rept. Hydrogen Isotope Research Center, Toyama University, **20** (2000) 53.
- [2] M. Matsuyama, T. Tanabe, N. Noda, V. Philipps, K.H. Finken and K. Watanabe, J. Nucl. Mater., **290-293** (2001) 437.
- [3] M. Matsuyama, T. Murai and K. Watanabe, Fusion Sci. Technol., **41** (2002) 505.

- [4] M. Matsuyama, T. Murai, K. Yoshida, K. Watanabe, H. Iwakiri and N. Yoshida, *J. Nucl. Mater.*, **307-311** (2002) 729.
- [5] M. Matsuyama, K. Watanabe and K. Hasegawa, *Fusion and Eng. Design*, **39-40** (1998) 929.
- [6] “Hyoumenbunseki”, Eds. M. Someno and I. Yasumori, Kodansha Scientific, (1976), p.128.  
( in Japanese)
- [7] M. Matsuyama and K. Watanabe, *Ann. Rept. Hydrogen Isotope Research Center, Toyama University*, **16** (1996) 81.