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論　文

制動X線計測法によるトリチウムの非破壊分析（Ⅰ）有機材料への適用性

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Nondestructive Tritium Analysis with Bremsstrahlung Counting Method (Ⅰ) Applicability to Organic Polymers

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Abstract

Applicability of the bremsstrahlung counting method for estimating the depth profile of tritium captured in materials has been examined. During the first step, the commercial tritium source (1 mm in thickness) made of a poly-[^{3}H]-methyl-methacrylate disk was employed as the emitter of the bremsstrahlung x-rays, because it was suited for numerical analysis of bremsstrahlung x-ray spectra owing to the homogeneous distribution of the tritium atoms in the emitter. The bremsstrahlung x-ray spectra observed for the tritium source showed a single broad peak having a maximum intensity of around 8 keV. Simulation of the bremsstrahlung x-ray apectra was carried out for various thicknesses of the emitter by using a calculation program mainly consisting of a tritium β-ray spectrum, energy conversion of β-rays to the bremsstrahlung x-rays, and the attenuation of them in the tritium source, taking into account the range of the β-rays determined experimentally as 2 μm. It was suggested from the computational simulation that the present method can measure tritium existing up to a few millimeters in thickness from surface. In addition, the peak position and shape of the bremsstrahlung x-ray spectra obtained from the computational simulations agreed very well with those of the spectra observed by the experiments. This indicates that the peak profile of a bremsstrahlung x-ray spectrum

obtained from materials such as organic polymers can be reproduced by numerical analysis. Accordingly, it was revealed that the present method was thoroughly applicable to nondestructive measuremens of tritium existing in significantly deeper regions than the depth measurable by the conventional β-ray counting method

1. Introduction

Estimation of depth profiles of tritium captured in materials for construction, the first wall, tritium breeder and tubing in a thermonuclear fusion reactor is indispensable from view points of safe confinement and a best profitable use of tritium. It is also important for precise evaluation of the recycling efficiency of the fuel in the rector system as well as the waste management of materials containing tritium[1, 2]

Several methods have been developed so far to measure the depth profiles of tritium captured in materials; for example, chemical or electrochemical etching and thermal desorption analyses. The former is a conventional and most fundamental technique. However, it has problems such as scraping of the surface layers to leave tritium waste, tedious procedures to assure the homogeneity of etching, limited applicability to metals and so on. On the other hand, the latter is applicable to measure the total amount of tritium captured in materials, desorption kinetics and trapping energy of tritium. But it is not effective to obtain a depth profile of tritium.

A conventional β-ray counting method will be also applicable to the above purposes. This method has been generally used for measurement of contaminant tritium on a surface. Owing to the shallow escape depth of tritium β-rays, however, the detectable depth will be limited to a surface region within $1\,\mu$m for organic and $0.1\,\mu$m for metallic materials. Namely, the β-ray couting method is worthwhile for only thin sub-surface layers. The depth of tritium diffusing into a material under high temperature conditions will be usually much deeper than $1\,\mu$m. The evalution of tritium existing beneath sub-surface layers is of great importance for the above purposes. From this view point, it is required to develop a new method applicable to depth-profiling beyond $1\,\mu$m from the surface.

On the other hand, electromagnetic waves have much larger escape depth than tritium β-rays. Bremsstrahlung x-rays are radiated by the interactions between β-rays and constituent atoms of a material containing tritium. Bremsstrahlung x-rays form a continuous spectrum, and its maximum energy should become lower than that of the tritium β-rays. Despite this effect, the escape depth of high energy bremsstrahlung x-rays radiated by the interactions should be significantly greater than the tritium

β-rays. Thus, utilization of the bremsstrahlung x-rays will make it possible to measure tritium existing in a deep region of materials.

To investigate this applicability, an organic polymer sample labeled with tritium was taken up first as a model material, although the conversion efficiency of β-rays to bremsstrahlung x-rays in organic materials is low in comparson with high-Z materials. The selection of the organic polymer sample is due that tritium distribution is homogeneous throughout the material.

The present paper describes the spectra of bremsstrahlung x-rays emitted from the polymer sample labeled with tritium and the comparison between observed and simulation spectra obtained from numerical analyses. The applicability of bremsstrahlung counting method will be discussed as well.

2. Experimental

2. 1. An emitter of bremsstrahlung x-rays

The bremsstrahlung x-ray emitter used in the present study was a disk made of poly-[^{3}H] methy methacrylate (described hereafter as PMM), which had been supplied as a standard tritium source. It was purchased from Amersham Japan Ltd. The thickness and diameter were 1 and 25 mm, respectively. The weight was 5.84×10^{-1}g, and the specific activity was 2.61×10^8 Bq/g. The number of β-rays emitted from the sample surface was measured by a specially designed windowless G-M counter, whose characterization is described elsewhere[3].

2. 2. Apparatus for measurement of bremsstrahlung x-rays

Figure 1 shows a block diagram of the experimental apparatus used to measure bremsstrahlung x-rays emitted from the PMM disk. The apparatus consisted of a silicon avalanche photodiode (Si-APD) detector, amplifiers, a power supply unit and a multichannel analyzer. Data of an energy spectrum temporarily taken

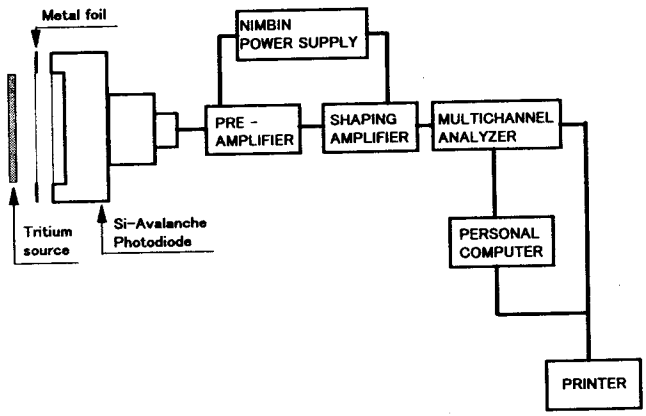

Fig.1. A block diagram of the experimental apparatus used for measurement of bremsstrahlung x-rays.

in the multichannel analyzer were saved by a personal computer. The Si-APD used in the present study was operated at room temperature without any special cooling device, although its energy resolution was not better than a radiation detector consisting of semiconductor elements such as litium-drifted silicon and high pure germanium crystals cooled at a temperature of liquid nitrogen. The effective diameter of the Si-APD for incident radiations was 25 mm, and its incident window was made of a thin polymer film evaporated aluminum to shield from the light. Other basic performance of the Si-APD is described elsewhere[4].

To prevent the contamination of detector due to tritium and the direct incidence of β-rays, thin aluminum foil was used when bremsstrahlung x-ray spectra were measured. The aluminum foil was inserted into the space between the PMM disk and the Si-APD detector as shown in Fig.1. Its thickness was 15 mg/cm^2.

All of measurements were performed at room temperature. The energy calibration of a bremsstrahlung x-ray spectrum was carried out by using a radiation source of ^{57}Co, which emits low energy characteristic x-rays and γ-rays (6.4, 7.0 and 14.4 keV).

2. 3. Computer simulation of a bremsstrahlung x-ray spectrum

To evaluate the observed bremsstrahlung x-ray spectra by means of numerical analysis, a simulation program to reproduce the observed spectra has been established. This program mainly consisted of three parts; that is, reproduction of the tritium β-ray spectrum, energy conversion of β-rays to bremsstrahlung x-rays, and their attenuation behavior in the PMM disk.

2. 3. 1. β-ray spectrum of tritium

It is represented by Sours[5] that the energy spectrum of β-rays emitted from tritium nuclei can be approximated by the following equations:

$$N(E) = \{\alpha/[1-\exp(-\alpha)]\} \cdot P_e(tot) \cdot E_e(tot) \cdot (E_{max}-Ee)^2 \qquad (1)$$

and

$$\alpha = (4\pi/137C)\ [E_e(tot)/P_e(tot)], \qquad (2)$$

where E_{max} represents the maximum energy of β-rays, C the speed of light, $P_e(tot)$ the total momentum and $E_e(tot)$ the total energy of an electron. Total electron energy includes the rest mass energy ($E(rest)$) as well as the kinetic energy (E_e). Namely, these terms are described by the following equations:

$$E_e(tot) = E(rest) + E_e \tag{3}$$

and

$$P_e(tot) = [(E_e)^2 + 2E(rest) \cdot E_e]^{0.5} / C \tag{4}$$

2. 3. 2. Conversion of β-rays to bremsstrahlung x-rays

Wyard gives an equation describing the energy conversion of high energy electrons to bremsstrahlung x-rays[6]. This equation, however, describes basically a bremsstrahlung x-ray spectrum from single energy electrons. On the assumption that the Wyard's equation is applicable to the tritium β-rays having successive energy distribution, the bremsstrahlung x-ray spectrum can be given by the following equation :

$$N(k)/dk = (\omega/1.25)\ [4(1-k/E_e) + 3(k/E_e)\ln(k/E_e)]/k, \tag{5}$$

where $k\ (k<E_e)$ is the energy of bremsstrahlung x-rays, and ω the radiation yield. The term of ω can be approximated as

$$\omega = Z \cdot E_e/(ZE_e + 800), \tag{6}$$

where Z is the atomic number of material containing tritium. In the present calculation, an averaged value of Z was employed since PMM, $(C_5H_8O_2)_n$ consists of carbon, hydrogen, and oxygen atoms. The factor of ω affects the total intensity of a spectrum, but it does not change the spectrum shape as seen clearly from Eq. (5). Therefore, in the present calulation, unity was assumed for ω in Eq. (5).

2. 3. 3. Attenuation of bremsstrahlung x-ray intensity

Attenuation of the intensity of bremsstrahlung x-rays in a material can be expressed by the following equation :

$$I(k) = I_0(k) \exp\ [-\mu(k) \cdot d], \tag{7}$$

where $I_0(k)$ represents the intensity of bremsstrahlung x-rays generated at a given depth in the material, $I(k)$ the intensity of bremsstrahlung x-rays penetrated through the material, $\mu(k)$ the absorption coefficient[7] depending on the photon energy as shown in Fig.2, and d the penetrating distance through the material. The value of d should be different according to the depth of tritium : hence, it was determined as the distance subtracted the range of β-rays from the depth of each β-ray emission layer. In addition, to gain a total intensity for a given energy photon, the PMM disk was divided into the layers by the range of β-rays, and the contributions from

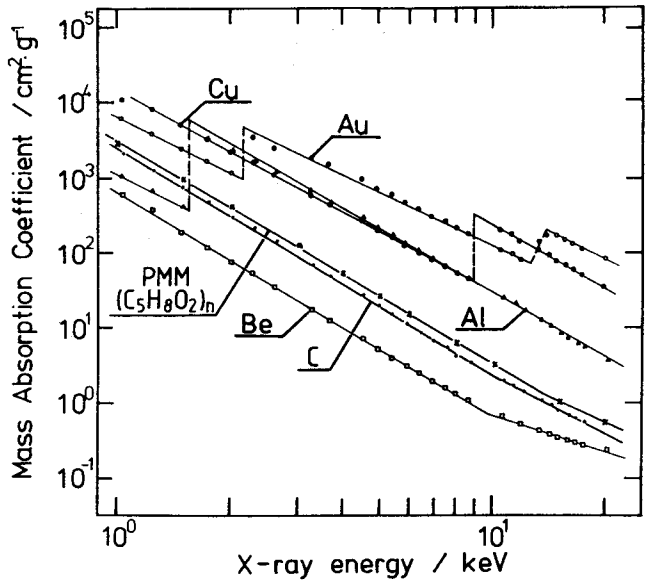

Fig.2. Dependence of mass absorption coefficients for various materials on photon energy.

each division was summed up.

For this calculation process, the range of β-rays in the PMM disk was assumed to be 2μm. This value was estimated from β-ray measurements as described below in detail.

3. Results and discussion

3. 1. Estimation of the range of β-rays in PMM disk

To carry out an approximate calculation of the range of tritium β-rays, the self-absorption coefficient of the tritium β-rays for the PMM disk was examined at first. In general, the attenuation of β-rays due to self-absorption for a thin material whose thickness is much thinner than range of β-rays can be approximately represented by the following equation :

$$A = a_0 \{1 - \exp(-\mu x)\} / \mu , \tag{8}$$

where A is the observed intensity of β-rays, a_0 the intensity per unit thickness, μ the self-absorption coefficient, and x the thickness of the material.

On the other hand, the thickness of the PMM disk used in the present study is expected to be fairly thicker than the range of tritium β-rays. In this case, Eq. (8) can be simplified as

$$A = a_0 / \mu . \qquad (9)$$

From the radioactivity of the PMM disk measured by the windowless G-M counter, the values of A and a_0 were determined as 3.11×10^4 Bq and 1.52×10^9 Bq/cm, respectively. Therefore, the absorption coefficient of tritium β-rays for the PMM was evaluated as $4.89 \times 10^4 cm^{-1}$. By substituting this value into Eq.(8), variation of the radioactivity depending on the thickness of the PMM disk could be estimated. As a result of the calculation, the radioactivity asymptotically increased with increasing the thickness, and was saturated around 2μ m. This indicates the range of tritium β-rays in the PMM disk to be 2μ m.

3. 2. Simulation of a bremsstrahlung x-ray spectrum from PMM disk

Figure 3 shows the bremsstrahlung x-ray spectra obtained from the computational calculation based on the Eqs. (1) - (7). In this calculation, the thickness of the PMM disk was varied in the range from 0.2 to 1.0 mm. All spectra showed a single broad peak, and the intensity asymptotically increased with thickness up to about a few millimeters. This indicates that the bremsstrahlung x-rays could penetrate through about 1.0 mm thickness. In addition to the spectral intensity, the position and shape of the peak varied with thickness: namely, the intensity of higher energy side of each peak increased preferentially with the increase in thickness, and a peak position shifted to higher energy side. Such tendencies are reasonable, because the absorption coefficient of lower energy x-rays is greater than that of higher energy x-rays as clearly seen in Fig.2. It was understood, therefore, that the peak position and shape of a bremsstrahlung x-ray spectrum give important information on the depth distribution of tritium.

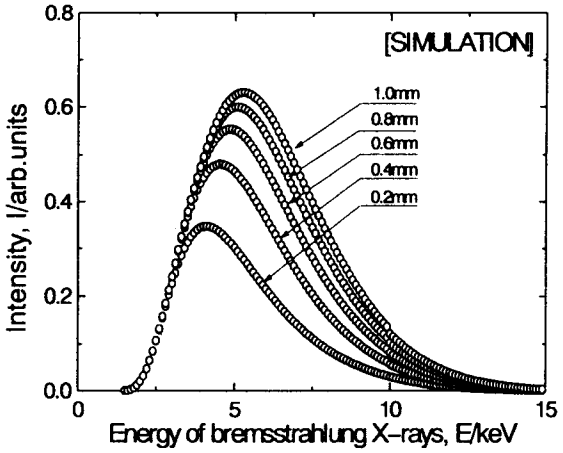

Fig.3. Bremsstrahlung x-ray spectra obtained from computational simulation.

3. 3. Comparison between observed and simulated spectra

Figure 4 shows an example of the observed spectra. In this measurement, a sheet of thin aluminum foil was inserted between the PMM disk and the detector in order to prevent detector contamination, and to prevent the direct incidence of β-rays to the detector. The thickness of the aluminum foil was 55 μ m. The single broad peak observed is due to bremsstrahlung x-rays, where the maximum intensity appeared around 8 keV. The spectral intensity was 19 counts/s. The peak position was observed at higher energy side than that seen in Fig.3. This may be due to the insertion of the aluminum foil.

To examine the effect of the aluminum foil on the peak position and shape, computational calculations were performed, where the aluminum insertion was taken into account. The results are shown in Fig.5. Every peak began to rise about 4.0 keV : it was 2 keV higher than the case calculated without aluminum foil as shown in Fig.3. Furthermore, the peak position also shifted to higher energy side. In general, the absorption yield of photons in a material increases accelerately with decreasing photon energy. Such behavior can be understood by the change in mass absorption coefficients as shown in Fig.2. Namely, the aluminum foil causes additional attenuation effect on low energy photons, resulting in the shift of peak position to higher energy side. Therefore, it was revealed that the peak shift shown in Fig.4 results from the inser-

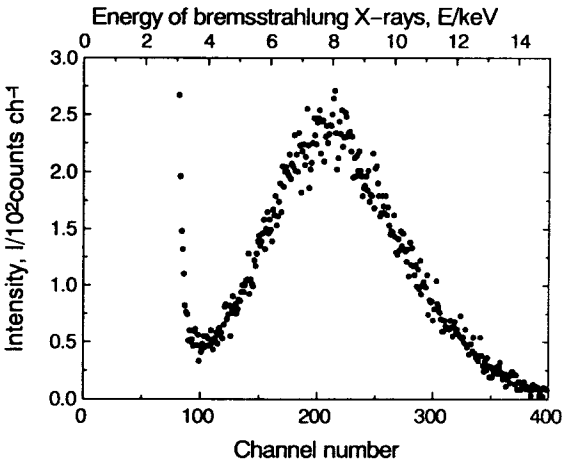

Fig.4. An example of the bremsstrahlung x-ray spectra observed for the tritium source.

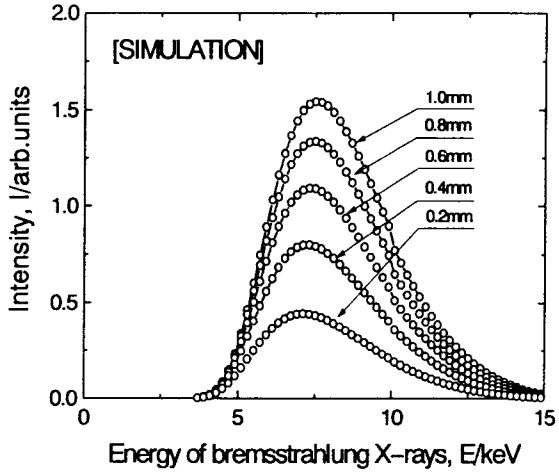

Fig.5. Effect of aluminum insertion on the peak position and shape in calculated spectra.

tion of an aluminum foil.

Figure 6 shows the comparison between the spectra obtained from experiments and simulations. The simulation spectrum in the figure was obtained for the thickness of 1.0 mm. Both spectra agreed quite well except a low energy region. This indicates that the bremsstrahlung x-ray spectrum can be simulated with the computational calculation based on the approximate equations. This means that it is possible to evaluate the depth distribution of tritium in a material mainly consisting of carbon atoms. The discrepancy in peak shape in the low energy region may be mainly due to the electric noise of the x-ray detector system. This would be clarified by application of low noise detectors equipped with semiconductor elements such as highly pure germanium or lithium-drifted silicon crystal cooled cryogenically by liquid nitrogen.

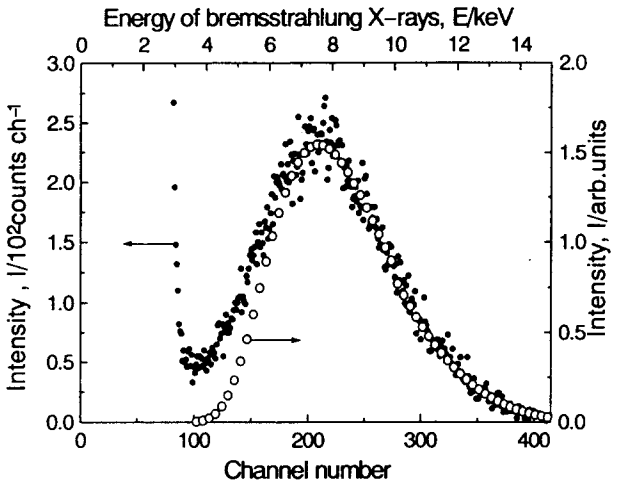

Fig.6. Comparison of observed(●) and calculated (○) spectra.

Figure 7 shows a comparison of detectable depth of tritium between β-ray and bremsstrahlung counting methods as a function of the thick ness of PMM disk. The former shows saturation around 2 μm as mentioned above, while the latter gives further increase in the intensity, indicating that it is applicable to measure tritium existing up to a few millimeters beneath the surface. Namely, the present bremsstrahlung counting method has a high potential for nondestructive measurements of carbonaceous materials con-

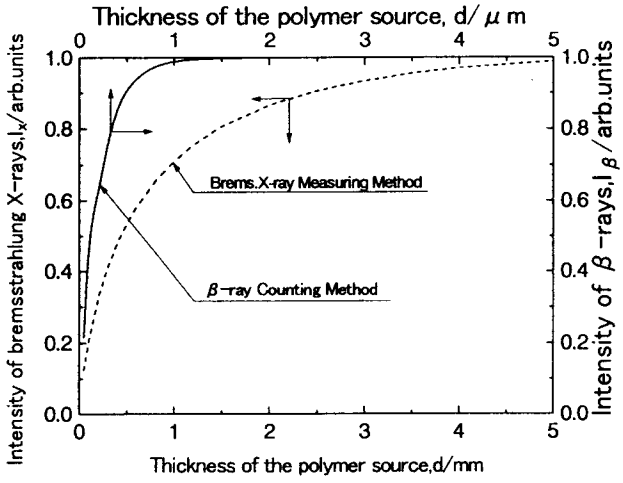

Fig.7. Comparison of detectable depth of tritium between β-ray and bremsstrahlung counting methods.

taining tritium in the region deeper than 2μm. This method would be also applicable to metallic materials although the detection depth should be shallower than the carbonaceous materials because of large absorption coefficients of metallic materials.

4. Conclusions

To estimate the depth profiles of tritium captured in various kinds of materials for thermonuclear fusion reactors, the applicability of the bremsstrahlung counting method was examined. As a first step, a commercial tritium source made of organic polymer was employed as a model, which is convenient for numerical analysis of the bremsstrahlung x-ray spectrum because of homogeneous distribution of tritium. A simulation program was also developed for numerical analysis of the bremsstrahlung x-ray spectrum by considering generation of the tritium β-ray spectrum, energy conversion of β-rays to bremsstrahlung x-rays, and their attenuation in the tritium source.

The bremsstrahlung spectra measured by a low energy x-ray detector system, which was equipped with a silicon avalanche photodiode, was observed for all of a single broad peak having maximum intensity around 8 keV. The feature could be reproduced quite well with the simulation, which showed systematic changes in the peak position, the shape and the intensity. The spectral intensity saturated at a few millimeter thickness. It is, however, about 1000 times greater than that of a β-ray counting method. Namely, the present bremsstrahlung counting method is expected to detect tritium existing in the bulky region. This method is applicable to not only carbonaceous materials but also metals and alloys. Furthermore, it was revealed that the peak position and shape of the observed spectra agreed quite well with those of simulation spectra. This indicates that it is possible to analyze the depth profile of tritium distribution in materials.

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