富山大学水素同位体機能研究センター研究報告13：51−61，1993.

論　文

制動X線計測法による高濃度トリチウムのIn-Situ測定
（Ⅱ）ベリリウム製窓の特性

松山　政夫・荒井　寛幸*・山崎登志成*
池田　長康*・渡辺　国昭

富山大学水素同位体機能研究センター
*富山大学工学部電子情報工学科
富山市五福3190

In-Situ Measurement of High Level Tritium by Bremsstrahlung Counting Method
(Ⅱ) Characteristics of a beryllium window

Masao MATSUYAMA, Hiroyuki ARAI*,
Toshinari YAMAZAKI*, Nagayasu IKEDA*,
and Kuniaki WATANABE

Hydrogen Isotope Research Center,
*Faculty of Engineering, Toyama University
Gofuku 3190, Toyama 930, Japan
(Received August 31, 1993 ; accepted November 7, 1993)

Abstract

Improvements of the bremsstrahlung counting method proposed for in-situ measurements of high level tritium were performed from the viewpoint of increase in sensitivity. The major point of the improvements was made in regard to the window material which acts as a generator and extractor of the bremsstrahlung X-rays. It was expected from computer simulation that a thin beryllium foil would be the most favorable window material for the present purposes.

On the basis of the simulation, characteristics of the 0.25 mm thick beryllium foil was experimentally investigated by using tritium gas diluted with deuterium gas. The bremsstrahlung counting rate was proportional to the concentration of tritium above 1×10^{-5} Ci/cm^3, and also sensitivity increased above 500 times in comparison with a case of former glass tube.

On the other hand, it was seen that the counting rate was affected by the total pressure of the gas containing tritium. Namely, above about 50 Torr, the counting rate decreased gradually with the total pressure. Such behavior was interpreted in terms of the self-absorption of β-rays in the gas phase. It was revealed that the change in the counting rate can be fully reproduced by a exponential function taking into account the self-absorption.

1. Introduction

Precise measurements of high level tritium are indispensble for fuel processing such as supply, recovery, storage, purification, enrichment, and waste treatment in D-T burning fusion devices[1, 2]. For this purpose, various methods to measure the tritium have been developed so far[3-9]. We have proposed the bremsstrahlung counting method as one of the most promising methods[10].

The bremsstrahlung counting method is based on detection of the bremsstrahlung X-rays induced by interactions between material and β-rays emitted from tritium. This method does not require to sample tritium gas for measurements in contrast to the conventional liquid scintillation counting, mass spectrometry[9] and gas chromatography[11]. Namely, it is an in-situ and non-destructive method. Other advantages are independence of the sensitivity on chemical forms of tritium and coexistence of other gas species, and that the detector system is free from contamination due to adsorption of tritium.

We have previously reported the validity of this method, where a specially designed proportional counter was employed to measure the bremsstrahlung X-rays[10]. It has been revealed that the bremsstrahlung counting method has high potential for in-situ and non-destructive measurements of high level tritium. The proportional counter, however, had a large volume, needed counting gas (mixture gas of argon and methane (10%)) and showed low detection efficiency (33%).

The improvement of those points has been done with use of a silicon avalanche photodiode (denoted as Si-APD), which is small in size and does not need cooling with liquid nitrogen[12]. It has been confirmed that the Si-APD effectively detects photons in the energy range from 3 to 20 keV at about 20℃. Much more improvement should be possible by increasing the conversion efficiency of β-rays to bremsstrahlung X-rays through suitable selection of a favorable window material which acts as a generator and extractor of the bremsstrahlung X-rays.

From this viewpoint, we explored window materials through a computer simulation and examined experimentally characteristics of the most favorable one. In this report, we will represent the validity of the window made of beryllium.

2. Selection of a window material

Two major processes are concerned with the bremsstrahlung counting method : namely, generation of bremsstrahlung X-rays at the inner surface of a window material and subsequent absorption in it.

2. 1. Generation of bremsstrahlung X-rays

Generation of bremsstrahlung X-rays by monoenergetic electrons can be represented by the following approximate equation proposed by Wyard[13] :

$$dI(K) = C[4(1-K/E) + 3(K/E)\ln(K/E)]dK \cdots\cdots\cdots\cdots\cdots\cdots (1)$$

where I(K) represents the energy of electrons consumed by generation of photons having energy K, E the kinetic energy of electrons and C the constant. By the Integration of Eq. (1), the total energy of electron consumed by generation of photons in the energy range from 0 to E is $I(K)=1.25CE$, when the initial energy of electrons is E. Since the radiation yield (ω), which corresponds to the ratio of I(K) to E, is equivalent to 1.25C, the value of C in Eq. (1) is 0.8ω. Namely, an energy spectrum of the bremsstrahlung X-rays generating by the electrons of energy E can be written as follows :

$$N(K)/dK = 0.8\omega[4(1-K/E) + 3(K/E)\ln(K/E)]/K \cdots\cdots\cdots (2)$$

The value of ω can be evaluated from the following equation proposed by Wu[14].

$$\omega = 1.98\times10^{-4}(1.96E+2)Z/[1+0.152\ln(82/Z)] \cdots\cdots\cdots\cdots\cdots\cdots (3)$$

where Z is the atomic number of the element constituting the window. Those equations simulate a spectrum of the bremsstrahlung X-rays arising from the monoenergetic electrons of energy E. However, the β-rays from tritium has a continuous spectrum. The bremsstrahlung X-ray spectrum corresponding to the continuous β-ray spectrum would be obtained by introducing an equation[15] reproducing the β-ray spectrum into E of Eqs. 2 and 3 and then integrating in the energy range from 0 to 18.6 keV.

2. 2. Absorption of bremsstrahlung X-rays in a window material

The second process mentioned above, i.e., the depletion of photons in a window material, is evaluated from the equation describing attenuation of electromagnetic wave. For the bremsstrahlung X-ray having energy K, it should also obey the following equation,

$$N_{10}(K) = N_0(K)\exp(-\mu d), \cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots\cdots (4)$$

where $N_{10}(K)$ represents the intensity of the bremsstrahlung X-rays transmitted through the window material, $N_0(K)$ the intensity of the bremsstrahlung X-rays generated at the surface, μ the mass absorption coefficient and d the thickness of the window material.

3. Experimental

3. 1. Apparatus

Figure 1 shows a cross sectional view of the bremsstrahlung X-rays converter (denoted hereafter as BXC) designed in the present study where the β-rays are converted to the bremsstrahlung X-rays. Following the computer simulation, a thin beryllium foil was employed as window material, which was purchased from NGK Co. The thickness of the beryllium foil was 0.25 mm, and the diameter 30 mm. To make BXC shown in Fig. 1 , the beryllium window fabricated by brazing was welded to a conventional ultrahigh vacuum (UHV) flange, which was attached to another UHV flange with a high vacuum fitting. BXC was connected to a tritium handling apparatus. The inner volume of BXC surrounding with two flanges was 10.4cm^3(STP).

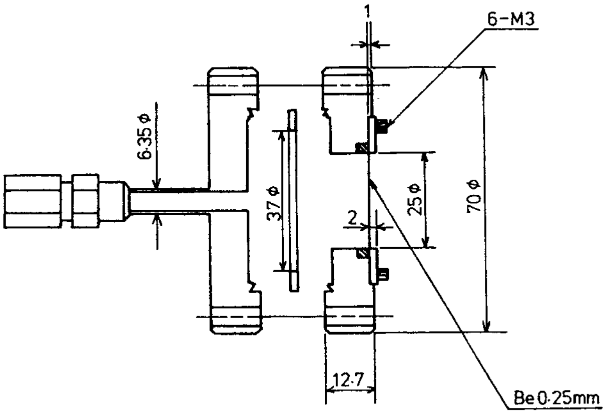

Fig. 1 A detailed cross sectional view of BXC.

Figure 2 shows a schematic diagram of the tritium handling apparatus used for storage, supply and recovery of tritium gas. The apparatus which consisted of metal tubes and tube fittings was connected to a conventional vacuum system with a liquid nitrogen trap evacuated by an oil diffusion pump backed with an oil-sealed rotary pump. The

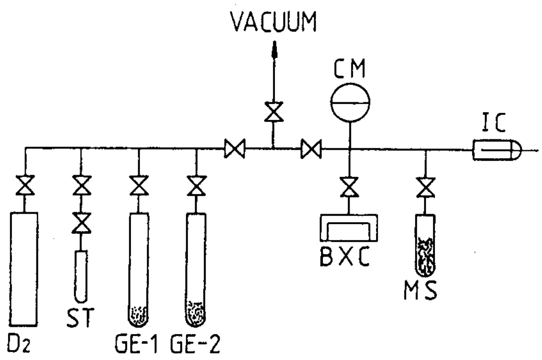

Fig. 2 A schematic diagram of the experimental apparatus used for the supply and recovery of tritium gas.

residual pressure was routinely below 5×10^{-6} Torr ($=6.6\times10^{-4}$ Pa). A small ionization chamber (IC)[3] and a capacitance manometer (CM) were used in each run for the measurements of tritium concentration and total pressure, respectively. The bremsstrahlung X-rays emitted from BXC was measured by using the Si-APD proposed previously[12].

3. 2. Materials

Tritium gas used in the present study was diluted with deuterium. It was stored in a ZrNi getter, which is set in GE-1 shown in Fig. 2 . The details of characteristics of the ZrNi getter have been described elsewhere[16]. The tritium gas was supplied to BXC by heating the ZrNi getter at 400℃. It was recovered from the BXC by cooling it to room temperature. In the supply process, a small amount of molecular sieves, which is denoted as MS in Fig. 2 , was used as a temporary storage material to introduce a large amount of tritium into BXC. Concentration of the tritium gas released from GE-1 was 0.457%, which was determined using the small ionization chamber and a quadrupole mass spectrometer to another UHV system[17].

3. 3. Procedures

Two series of experiments were carried out in the present study : one is partial pressure dependence of the bremsstrahlung counting rate, and another is total pressure dependence. All of the experiments were performed at room temperature.

For the former experiments, the bremsstrahlung counting rate was measured by varying the partial pressure of tritium. The pressure of tritium gas was changed in the range from 0.5 to 60 Torr by heating GE-1 at a given temperature. The partial pressure of tritium employed in the present study corresponds to the concentration in the range from 10^{-5} to 10^{-3} Ci/cm^3.

For the latter experiments, the amount of tritium in BXC was kept constant at 1.12×10^{-3} Ci(equivalent to 7.4 Torr) and the total pressure was varied in the range from 7.4 to 717 Torr by successive addition of a given amount of deuterium gas. After the measurements, the tritium gas used was recovered with GE-2 loaded with the same getter material as GE-1.

4. Results and discussion

4. 1. Selection of a window material

Figure 3 shows spectra of the bremsstrahlung X-rays generated at inner surface

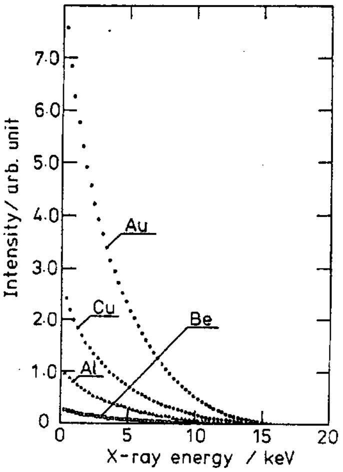

Fig. 3 Bremsstrahlung spectra generated at the inner surface of the window (simulation).

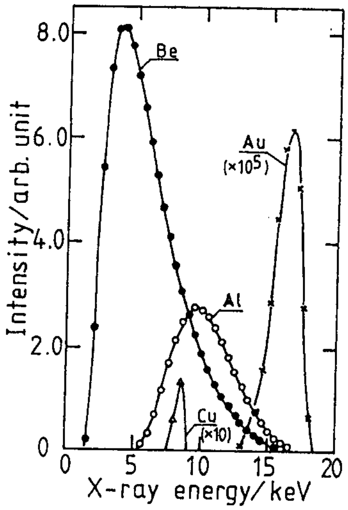

Fig. 4 Bremsstrahlung spectra transmitted through each materials (simulation).

of several window materials. They were calculated from Eqs. 2 and 3. To examine the effect of atomic number on the intensity of the bremsstrahlung X-rays, materials chosen for calculations were Be(Z=4), Al(Z=13), Cu(Z=29) and Au(Z=79) as shown in the figure. These materials are advantageous for the window from viewpoint that hydrogen could be adsorbed little. It was seen that the intensities were 3.7, 9.1 and 29 for Al, Cu, and Au, respectively, when they were normalized to that for Be. Namely, the higher the atomic number of the material is, the larger the intensity. It suggests that gold is most favorable as the X-ray generator.

Figure 4 represents spectra of the bremsstrahlung X-rays transmitted through the window materials. For the calculation of attenuation in each window material based on Eq. (4), values of two parameters are required : that is thickness and mass absorption coefficient of the window materials. Hence, the thickness of each material was constant at 0.13 mm and a mass absorption coefficient was cited from literature in the present calculations[18]. Since a mass absorption coefficient depends on the energy of photons as shown in Fig. 5 , the energy dependence was represented by the following equations to calculated intensity of the bremsstrahlung X-rays transmitted through each window material :

$$\mu(\mathrm{Be}) = 620K^{-2.925} \quad (1<K<10)$$
$$= 50K^{-1.825} \quad (10<K<20)$$

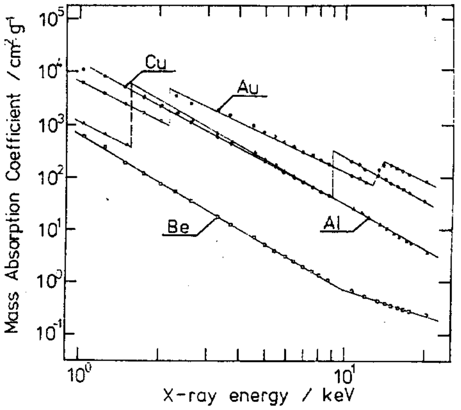

Fig. 5 Energy dependence of the mass absorption coefficients for materials.

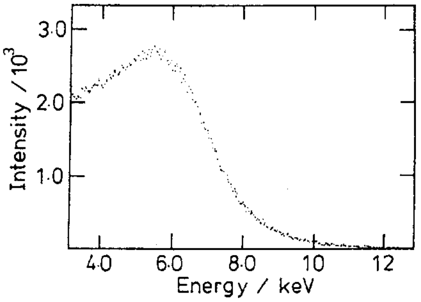

Fig. 6 An example of the bremsstrahlung spectrum observed by the present BXC.

$$\begin{aligned}\mu(\mathrm{Al}) &= 1200\mathrm{K}^{-2.575} \quad (1<\mathrm{K}<1.5)\\ &= 23100\mathrm{K}^{-2.925} \quad (1.5<\mathrm{K}<20)\\ \mu(\mathrm{Cu}) &= 15800\mathrm{K}^{-2.725} \quad (1<\mathrm{K}<8.9)\\ &= 177800\mathrm{K}^{-2.610} \quad (8.9<\mathrm{K}<20)\\ \mu(\mathrm{Au}) &= 6700\mathrm{K}^{-2.525} \quad (1<\mathrm{K}<2.1)\\ &= 28200\mathrm{K}^{-2.360} \quad (2.1<\mathrm{K}<12)\\ &= 111000\mathrm{K}^{-2.415} \quad (12<\mathrm{K}<20)\end{aligned}$$

where units of μ and K for photons were cm^2/g and keV, respectively. It was seen that the order of intensity of the bremsstrahlung X-rays is $\mathrm{Be} > \mathrm{Al} \gg \mathrm{Cu} \gg \mathrm{Au}$, and the peak of spectrum was shifted to higher energy side with increasing atomic number. The intensities normalized to Be for Al, Cu and Au were 0.4, 3.0×10^{-3} and 4.3×10^{-6}, respectively. This indicates that the attenuation in copper and gold is remarkably greater than that in beryllium. It is suggested, therefore, that the most promising material should be beryllium on account of the X-ray generator as well as the extractor of the X-rays.

Figure 6 shows an example of the bremsstrahlung X-ray spectrum observed for tritium gas. In this run, the amount of tritium enclosed in BXC was 1.03 mCi, and the measuring time with Si-APD was 1800 s. The spectrum showed a single peak at 5.5keV. No other peaks corresponding to characteristic X-rays were observed, although a part of BXC was made of stainless steel. The total intensity of the spectrum was 2.92×10^5 counts and the background counting rate was 6×10^2 counts : that is, bremsstrahlung counting rate was equivalent to 162 s^{-1}. The peak position observed was slightly higher than that of calculated spectrum for Be in Fig. 4 . One plausible reason for this discrepancy is the difference in thickness of beryllium : the beryllium used for experiments was two times thicker to ensure the safety of BXC than that for calculation.

The relations between the bremsstrahlung counting rate and the concentration/amount of tritium are shown in Fig. 7 . It is clear that the counting rate is proportional to the tritium concentration. This indicates that the total pressure of tritium gas does not affect the counting rate below the total pressure of 60 Torr and the effect of tritium adsorption on inner wall of BXC is negligible. As reported previously[10], similar linearity has been observed for the device constituting of the glass tube (10 mm in diameter and 140 mm in length) and a specially designed proportional counter in the range from 3×10^{-4} to 3×10^{-1} Ci/cm^3. It is concluded, therefore, that the linearity of the counting rate is insensitive to the geometric of BXC.

The sensitivity of the present detector system was 1.5×10^3 cps/(mCi/cm^3), as seen clearly from figure, while the previous one was 2 cps/(mCi/cm^3)[10]. Namely, a considerable improvement of the sensitivity was established by the present system. It is about 500 times of the previous device, taking account that the volume of the present BXC was 1.5 times greater than that of the previous one. In addition, the background counting rate in the present device was below 1 cps as mentioned above. It is expected, therefore, that the present device will be applicable to in-situ and real-time

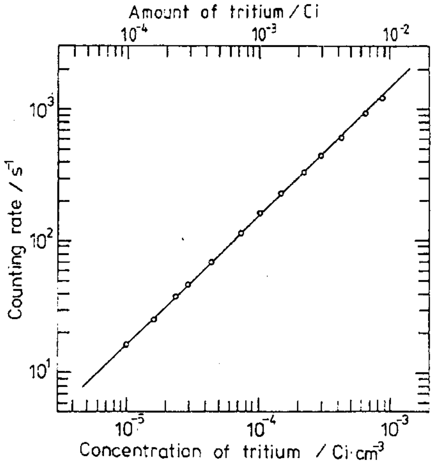

Fig. 7 Correlation between the bremsstrahlung counting rate and the concentration/amount of tritium.

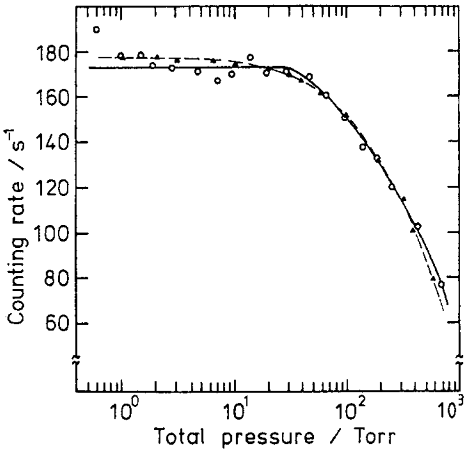

Fig. 8 Total pressure dependence of the bremsstrahlung counting rate.

measurements of tritium concentration in the wide range from 10^{-6} to 2.6 Ci/cm^3.

Figure 8 shows the dependence of the bremsstrahlung counting rate on the total pressure of tritium gas diluted with deuterium gas. It is clear from the figure that the counting rate is almost constant below about 50 Torr. This feature agrees well with Fig. 7 . Above 50 Torr, however, the counting rate decreases gradually with the total pressure. This behavior could be interpreted in terms of self-absorption of the β-rays in gas phase. Namely, a fraction of the kinetic energy consuming by collision with deuterium and tritium molecules should increase with the total pressure. Therefore, the kinetic energy of β-rays striking the beryllium window becomes small. This may result in the increase in the fraction of low energy bremsstrahlung X-rays. However, since the mass absorption coefficients for low energy photons below about 3keV are significantly large, the low energy photons should be almost consumed in the beryllium window and hence they could not contribute to the counting rate.

The number of β-particles striking the beryllium window, A, could be approximated as follows by considering the self-absorption of β-rays in gas phase :

$$A=A_0(1-\exp(-\mu d))/\mu, \quad \cdots\cdots (5)$$

where A_0 is the number of β-rays emitted in a unit thickness of gas phase, μ the absorption coefficient of β-rays in hydrogen isotopes, and d the thickness of gas phase. The counting rate of the bremsstrahlung X-rays is proportional to the number of beta-particles striking the beryllium window as seen in Fig. 7 . Hence, the counting rate shown in Fig. 8 should be described as follows :

$$I=\alpha A=\alpha A_0(1-\exp(-\mu d))/\mu, \quad \cdots\cdots (6)$$

where α is the ratio of the bremsstrahlung counting rate to the number of β-particles ; namely, it corresponds to the conversion coefficient of β-rays to the bremsstrahlung X-rays.

Parameter fittings based on Eq. (6) were carried out using the following values : $A_0=1.963\times10^7\text{cm}^{-1}\text{s}^{-1}$, $\mu=1.81\text{P cm}^{-1}$ (P is the total pressure in atm), and $d=1.365$ cm. The broken line (filled triangle) in Fig. 8 shows the fitting result. As seen in the figure, the observed counting rate can be well reproduced by Eq. (6), where α was evaluated as 6.6×10^{-6}. This value agrees well with one obtained from the relation shown in Fig. 7 . From those results, it was revealed that the total pressure dependence can be estimated by Eq. 6. More precise calculations for the total pressure dependence would be established by using the absorption coefficient for pure tritium, because in the present calculations the absorption coefficient for hydrogen was used as a first approximation with making no account of the isotope effect.

5. Conclusions

To improve the sensitivity of the bremsstrahlung counting method, the promising window materials which act as a generator and extractor of the bremsstrahlung X-rays have been explored. It was predicted from a computer simulation that a thin beryllium foil is the most favorable material among beryllium, aluminum, copper and gold.

Basing on the results of computer simulation, BXC attached the beryllium window was fabricated and then its characteristics were experimentally examined by introducing a given amount of tritium into BXC. As a result, it was revealed that the bremsstrahlung counting rate is proportional quite well to the tritium concentration in the wide range from 10^{-5} to 10^{-3} Ci/cm^3. Such linearity agreed thoroughly with the previous results and the sensitivity of the present BXC increased 500 times than that of the previous one. This indicates that the linearity is obtained irrespective of the structure and size of BXC and that the sensitivity depends significantly on the atomic number of a window material.

On the other hand, the total pressure dependence of the bremsstrahlung counting rate was examined in the range from 7.4 to 717 Torr. The counting rates were almost constant below 50 Torr, while above that pressure a gradual decrease was observed with increase in the total pressure of mixture gas. This was interpreted in terms of the self-absorption of β-rays in gas phase. It was revealed that the bremsstrahlung counting rate observed at higher pressure can be fully reproduced by taking account of the self-absorption of β-rays.

From these results, it was clarified that BXC developed in the present study shows considerable performance and it is applicable to in-situ and real-time measurements in a wide range of the tritium concentration in fusion fuel processing.

Acknowledgement

This work was partly supported by the Grant-in-Aid for Scientific Research from the Ministry of Education, Science and Culture of Japan.

References

1) P. Dinner, M. Chazalon, D. Evans, F. Fauser, M. Iseli and C.H. Wu, Fusion Technol., **14** (1988) 178.

2) H.F. Dylla, J. Vac. Sci. Technol., **20** (1982) 119.

3) M. Matsuyama and K. Watanabe, Fusion Eng. Design, **18** (1991) 91.

4) K. Okuno, T. Uda, S. O'hira and Y. Naruse, J. Nucl. Sci. Technol., **28** (1991) 509.

5) H. Miyake, M. Matsuyama and K. Watanabe, J. Vac. Sci. Technol., **A6** (1988) 158.

6) W.O. Hofer and J.V. Seggern, J. Nucl. Mater., **128&129** (1984) 943.

7) R. Lässer, Nucl. Instrum. Methods, **215** (1983) 467.

8) R.E. Ellefson, J. Vac. Sci. Technol., **A1** (1983) 865.

9) R.E. Ellefson and W.E. Moddeman, J. Vac. Sci. Technol., **18** (1981) 1062.

10) M. Matsuyama, H. Nakatani and K. Watanabe, Fusion Eng. Design, **10** (1989) 405.

11) T. Schober and C. Dieker, Rev. Sci. Instrum., **58** (1987) 1116.

12) M. Matsuyama, H. Arai, T. Yamazaki and K. Watanabe, Ann. Rept. Hydrogen Isot. Res. Center, Toyama Univ., **10&11** (1991) 69.

13) S.J. Wyard, Proc. Roy. Soc., **A65** (1952) 377.

14) C.S. Wu, Phys. Rev., 59 (1941) 481.

15) M. Matsuyama, H. Nakatani and K. Watanabe, Ann. Rept. Tritium Res. Center, Toyama Univ., **8** (1988) 51.

16) K. Watanabe, K. Tanaka, M. Matsuyama and K. Hasegawa, Fusion Eng. Design, **18** (1991) 27.

17) K. Watanabe, H. Miyake and M. Matsuyama, J. Vac. Sci. Technol., **A5** (1987) 237.

18) "Handbook of Chemistry-Fundamentals (II)", ed. Japan Chemical Society, (Maruzen, Tokyo, 1975), p.1556. (in Japanese)