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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

松山政夫・荒井寛幸*・山崎登志成*・渡辺国昭

富山大学水素同位体機能研究センター，*工学部
〒930 富山市五福3190

In-Situ Measurements of High Level Tritium by Bremsstrahlung Counting Method (I) Feasibility Tests of Si-Avalanche Photodiode

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

Hydrogen Isotope Research Center, Toyama University
*Faculty of Engineering, Toyama University
Gofuku 3190, Toyama 930, Japan
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Abstract

To develop some in-situ measurement techniques of high level tritium, the basic characteristics of a silicon-avalanche photodiode (Si-APD) were examined in detail using low energy X-ray emitters. We also investigated the applicability of the Si-APD to measurements of low energy bremsstrahlung X-rays caused by interactions between materials and the β-rays of tritium. The Si-APD worked well at room temperture without using any particular cooling device, and it proved to be satisfactory for the measurements of low energy X-rays below 20 keV except for significant temperature dependence of the peak energy in observed spectra, which is considered as its inherent characteristic on the one hand and owing to temperature dependence of the leakage current of the Si-APD on the other. In addition, the bremsstrahlung X-ray spectra measured using a polymer tritium source and tritium gas showed a broad single peak in the region below 15 keV. It has been revealed that the Si-APD is valuable for fabricating a compact

detector system capable of detecting low energy bremsstrahlung X-rays and is easy in maintenance.

1. Introduction

Various promising methods have been already proposed for the measurements of high level tritium : for example, utillzation of mass spectrometer[1, 2], laser spectrometer[3], infrared spectrometer[4], calorimeter[5], inorganic scintillator[6], small ionization chamber[7], and so on. They have their own merits, but also have disadvantageous features for some applications. More development would be indispensable to apply them for D-T fuel processing systems of thermonuclear fusion reactors.

We developed an in-situ method using bremsstrahlung counting for measuring high level tritium[8] : namely, detection of the bremsstrahlung X-rays induced by interactions between materials and the β-rays emitted from tritium. This has many advantageous points : it is non-destructive, in-situ, highly durable and unconcerned with chemical forms of tritium and presence of other gas species. To confirm validity of this method, we used a specially designed proportional counter to measure the bremsstrahlung X-rays. It has been revealed that this method has high potential for measurements of high level tritium. This counter, however, has several disadvantageous points such as large volume, need of multiplying and quenching gases and low detection efficiency. These features would limit its applicability to the tritium processing systems of thermonuclear fusion reactors.

From this viewpoint, we examined the applicability of a Si-avalanche photodiode (to be described hereafter as Si-APD), which is a kind of semiconductor detectors and does not need cooling with liquid nitrogen. The semiconductor has the following merits: 1) the detector is small in size and light in weihght, 2) W-value (energy required for generation of one electron-hole pair) is small (e.g., 3.6 eV for silicon[9]), 3) both detection efficiency and energy resolution are high. In this report, we will represent some basic characteristics of the Si-APD and the bremsstrahlung X-ray spectra, which were studied as a first step of potential evaluation of the Si-APD.

2. Experimental

Figure 1 shows a block diagram of the experimental apparatus used for measurements of basic characteristics of the Si-APD and of bremsstrahlung X-rays. The apparatus consisted of a Si-APD (Model SX25R), preamplifier (Model SA-101), BIN power source (Model 401AD : ORTEC Co. Ltd), shaping amplifier (Model 2021 : Canbera

Industries, Inc.), multichannel analyzer (Series 35 plus : Canbera Industries, Inc.) and printer (Model VP-550 : Seiko Epson Co., Ltd.). The X-rays were detected using the Si-APD and the preampifier which were delivered from Radiation Monitoring Devices, Inc. The working area of the Si-APD was a circle of 25mm in diameter. Its front side, facing to X-ray sources, was protected against air with a thin Mylar film coated with aluminum evaporated film.

The Si-APD was calibrated to the energy of bremsstrahlung X-rays using three nuclides as shown in Table 1[10], which emit low energy X-rays as well as γ-rays. ^{57}Co

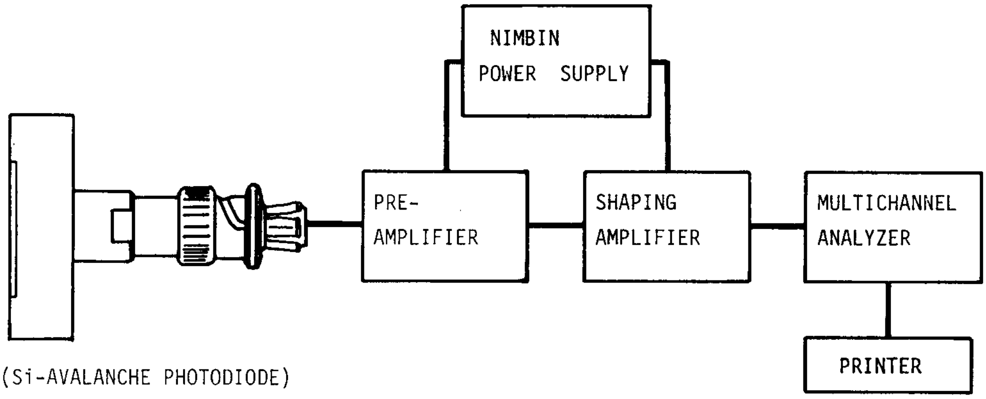

Fig. 1. Block diagram of the system employed for X- and γ-ray measurements with silicon-avalanche photodiode.

Table 1. Radioactive nuclides and photon energies.

Nuclide(half-life)		
^{57}Co(0.744y)	^{133}Ba(10.54y)	^{241}Am(432.7y)
6.391 keV(Fe(Kα_2))/16.49% 6.404 keV(Fe(Kα_1))/32.52% 7.058 keV(Fe(Kβ_1))/ 5.83% 14.41 keV(γ) / 9.54% 122.1 keV(γ) /85.54% 136.5 keV(γ) /10.69%	4.285 keV(Cs(Lα))/6.78% 4.728 keV(Cs(Lβ))/6.59% 30.63 keV(Cs(Kα_2))/35.6% 30.97 keV(Cs(Kα_1))/65.7% 34.97 keV(Cs(Kβ_1))/18.0% 36.01 keV(Cs(Kβ_2))/4.39% 80.99 keV(γ) /34.2% 302.8 keV(γ) /18.4% 356.0 keV(γ) /62.2%	13.93 keV(Np(Lα))/13.0% 17.61 keV(Np(Lβ))/20.2% 21.00 keV(Np(Lγ))/5.27% 26.34 keV(γ) /2.41% 59.54 keV(γ) /35.7%

was also used as a X-ray source to examine basic characteristics of the Si-APD : namely, effects of applied voltage, gain amplification and operation temperature on peaking channel number and total count rate.

Poly-methyl methacrylate labeled by ^{3}H and tritum gas were used as β-sources of tritium. The former was purchased from Amersham Japan Co. Its nominal activity and specific activity were 3.1 mCi and 5.4 mCi/g, respectively. The latter was purchased from New England Nuclear Co., whose nominal activity was 1 Ci. This was loaded in a glass tube.

With respect to the measurements of the bremsstrahlung X-rays, thin aluminum foil was used as a target of the β-rays emitted from the polymer source. The aluminum foil was a commercial one with $15\pm1\mu$m thickness.

3. Results and discussion

3. 1. Basic characteristics of Si-APD

Figure 2 shows a spectrum of low energy X- and γ-rays emitted from ^{57}Co source. Two peaks were observed. It is seen from Table 1 that the lower energy peak is due to the X-rays of Fe(Kα_1) and Fe(Kα_2), the higher one does the γ-rays of 14.4 keV. Hereafter, they will be denoted as peak 1 and peak 2, respectively.

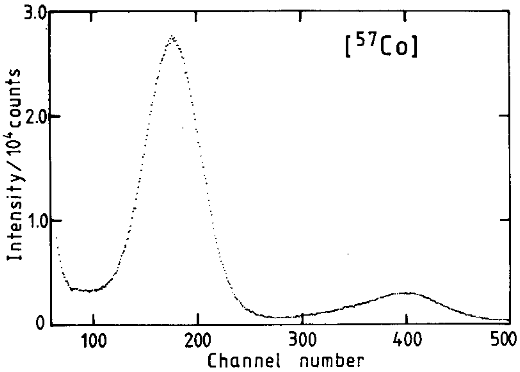

Fig. 2. An example of spectra of the photons emitted from ^{57}Co. The spectrum was obtained under the following conditions: the ambient temperature 19.0℃, applied voltage 1180 V, coarse gain 1000, fine gain 0.6 and integrating time 300 s.

The integral intensities of peak 1 and peak 2 were 1.93×10^6 and 3.51×10^5 counts, respectively. The ratio of them was 5.50, agreeing within 5% error with the intensity ratio (5.75) evaluated from Table 1. The observed ratio, however, was slightly smaller than the table value. This difference is considered partly due to the energy dependence of the transmittance of low energy photons through the window material of the Si-APD. Namely, the lower the energy of photons is, the larger the absorption coefficient does. As a result, the transmittance of peak 1 becomes smaller than that of peak 2. On account of this effect, the accuracy of the counting rate measurements with the Si-APD is quite satisfactory for the present purpose.

The energy resolution of a radiation detector is commonly defined by FWHM (Full

Width at Half Maximum) of a peak in a spectrum. It is the ratio of the peak width at half maximum to the peak position described as energy unit or channel number. A channel number corresponds in general to a photon energy. The observed resolution of peak 1 was 36% and that peak 2 was 25%. As for a scintillation detector, which is usually employed for measurements of γ-ray spectra, it is 5~10%. It is below 1%[11] for a semiconductor detector cooled with liquid nitrogen. Although those values are considerably smaller than those obtained in the present study, the poor resolution does not give rise to any serious problem for the present purpose. This is because the bremsstrahlung X-rays have inherently a continuous spectrum, and in general only a single peak is expected owing to significant absorption of them by material in a low energy below few keV as described later.

Figure 3 shows the shifts of the peaking channel numbers with applied voltage, which was varied in a range from 1135 to 1180 V. This corresponds to "HV control setting" in a range from 7.3 to 7.6. As can be seen from the figure, the channel numbers of both the peaks sifted to higher channel numbers with increasing applied voltage, indicating the increase in the output of Si-APD. This is because the output is directly proportional to the extent of secondary ionization by electrons of electron-hole pairs produced by incident photons.

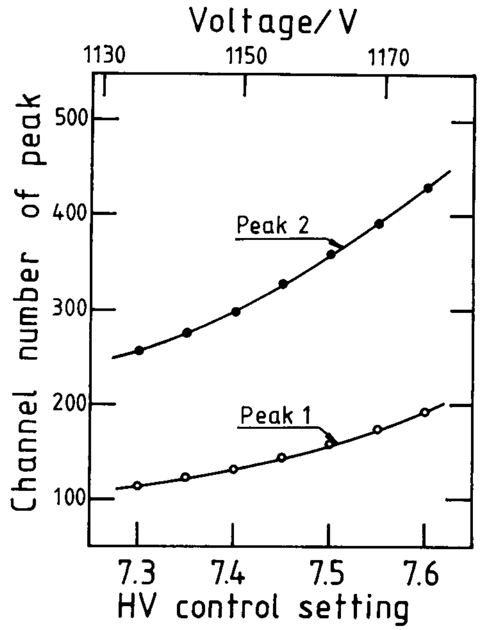

Fig. 3. Variations in peak channel numbers with HV control setting. Conditions for measurements were as follows : the ambient temperature 17.1℃, coarse gain 1000, fine gain 0.6, and integrating time 300 s.

Figure 4 shows the variation in the resolution of the peak 1 accompanied with the peak shifts shown in Fig. 3. The resolution decreased linearly with increasing applied voltage. Similar variation in the peak 2 was also observed : namely, the energy resolution of the Si-APD is improved with increasing applied voltage.

Figure 5 shows the peak shifts with the fine-gain control setting of the shaping amplifier. It is clear that the channel number of each peak varies linearly with fine-gain control setting.

Figure 6 shows the temperature dependence of the peak channel number. Both channel

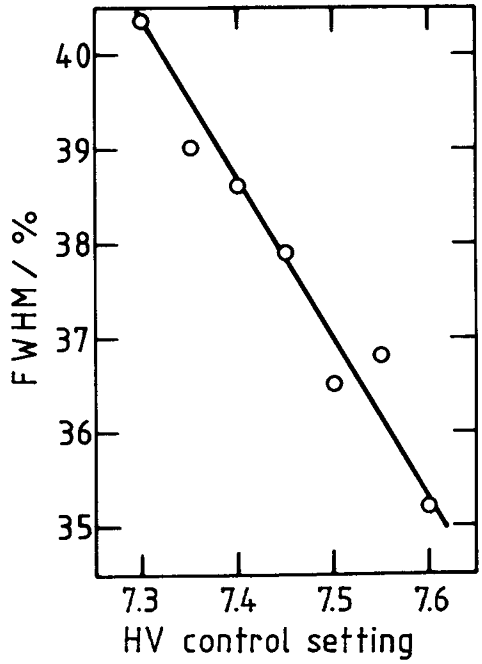

Fig. 4. Variation in FWHM of peak 1 with the HV control setting.

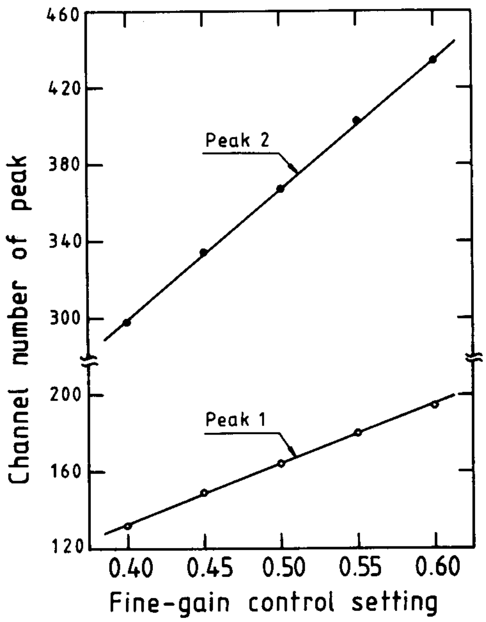

Fig. 5. Variations in peak channel numbers with the fine gain control setting.

numbers of peak 1 and 2 decreased with increasing ambient temperature of the Si-APD. This is considered as follows. According to the data sheet delivered from the manufacturer, the leakage current of Si-APD increases concavely with the ambient temperature. This phenomenon is principally due to the narrow band gap (1.11 eV at 300K[9]) of silicon. Namely, the temperature increase causes the increase in the leakage current, which gives rise to the decrease in the net voltage applied across the Si-APD. Accordingly, the peak channel number shifts to lower side as a consequence of the applied voltage dependence of the peak channel number as described above.

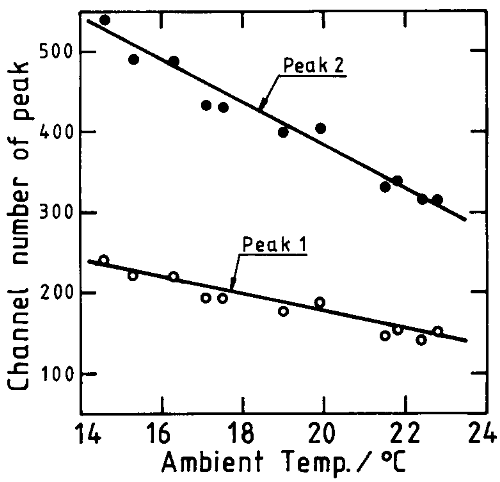

Fig. 6. Temperature dependence of channel number of peaks. Conditions of the measurements were as follows: the applied voltage was 1180 V, coarse gain 1000, and fine gain 0.6.

For the present purpose, the most important factor is constancy of the integrated peak intensity although the peak channel number varied with the applied voltage, gain and ambient temperature.

Figure 7 shows the variation of the total count in each peak with the applied voltage and fine-gain. In these measurements, the HV control setting was varied by keeping the fine-gain control constant at 0.6, whereas the fine-gain control setting was varied by setting the HV control at 7.6 (equivalent to 1180 V). This figure shows that the total count was not affected by the applied potential if the HV control setting was kept over about 7.4 (1150 V). Namely, the recombination of electrons and holes generated by photons does not play an important role at high applied potentials over 1150 V. On the other hand, the fine-gain control setting affected slightly the total count. Based on these observations, we adopted the standard experimental conditions for measurements of the low energy X-rays as HV control setting=7.6, coarse gain setting=1000, and fine-gain control setting=0.6.

Figure 8 shows an example of energy calibration curves, which was measured at 17.1℃. As seen clearly from the figure, a good linear relation was obtained. Slope of the line was determined as 33 eV/channel. As mentioned above, however, the slope depends on the ambient temperature of the Si-APD as well as the applied voltage and gain. For energy analysis, therefore, it is necessary to examine, in each run, the peak channel number at the ambient temperature by using some radiactive nuclides.

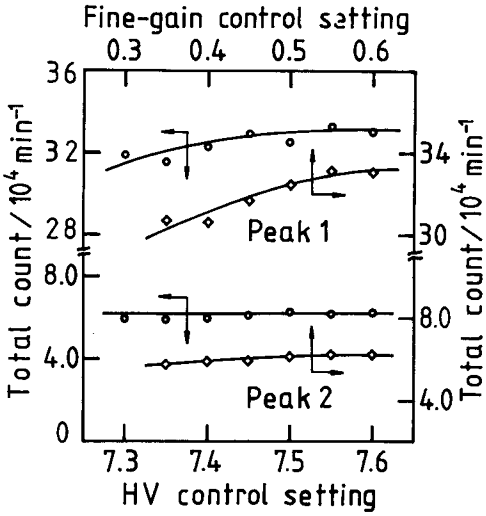

Fig. 7. Variations in total count with the HV control setting and fine gain control setting. This was obtained under the following conditions : the ambient temperature was 17.1℃, coarse gain 1000, integrating time 300 s.

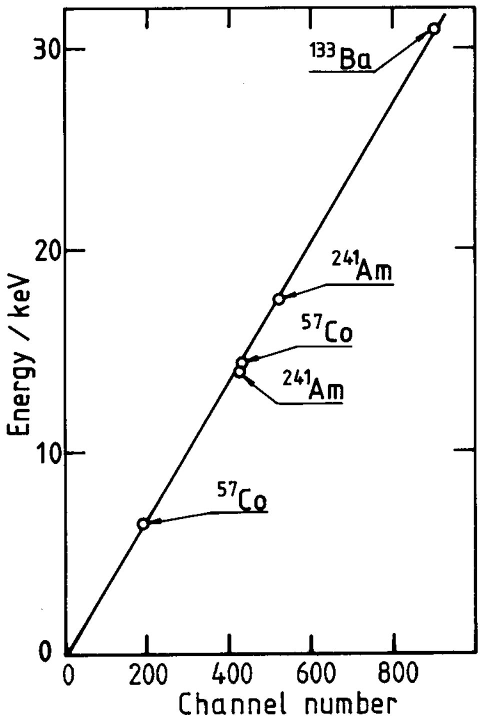

Fig. 8. An example of energy calibration curves.

3. 2. Measurements of bremsstrahlung X-Rays

Figure 9 shows an example of the spectra of bremsstrahlung X-rays generated by the interaction between aluminum foil and β-rays from the polymer source of tritium. The thickness of aluminum foil was 75 μm. This spectrum was measured at 19.0℃ and for an integration time of 1.8 ks. A single broad peak was observed in the spectrum in the region from 4.0 to 14 keV, being peaked at about 7.5 keV. Both the characteristic X-rays and bremsstrahlung X-rays are emitted due to the collisions of β-rays to aluminum atoms. However, since the energy of characteristic X-rays from aluminum is about 1.5 keV, it is not detectable in the present conditions. In addition, it appears hard to detect it owing to the large absorption in aluminum foil itself, even though the noise level of the Si-APD is low enough to detect the characteristic X-rays. Namely, the spectrum observed in the present study consists of the bremsstrahlung X-rays. The maximum energy of the β-rays emitted from tritium is 18.6 keV. Accordingly, the bremsstrahlung X-rays should be in lower energy than the β-rays.

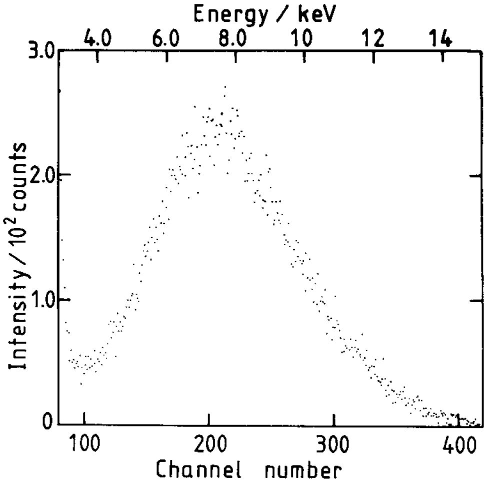

Fig. 9. An example of bremsstrahlung X-ray spectra measured by using a polymer source of tritium.

The absorption of bremsstrahlung X-rays in aluminum foil being taken into account, the total count will depend on the thickness of aluminum foil. Figure 10 shows the effect of thickness of the aluminum foil. The logarithmic plots showed convex decrease with the increase in the thickness. We have previously reported[12] that the variation in the total count obeys −2.38 power of the thickness of the target in the vicinity of 1 mm, and below this thickness it increases convexly with decreasing thickness. This was estimated theoretically by considering energy dependence of the absorption coefficient of continuous X-rays for a target of β-rays. This explana-

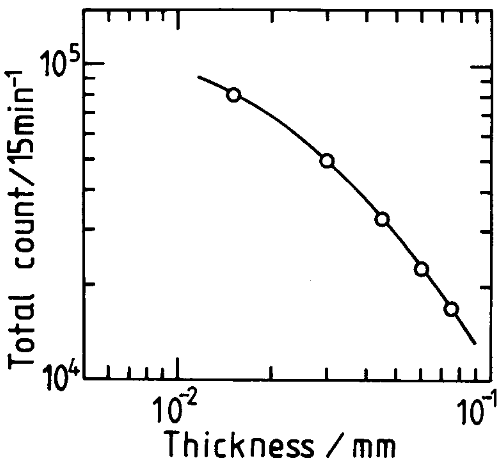

Fig.10. Dependence of the total count on the thickness of aluminum foil.

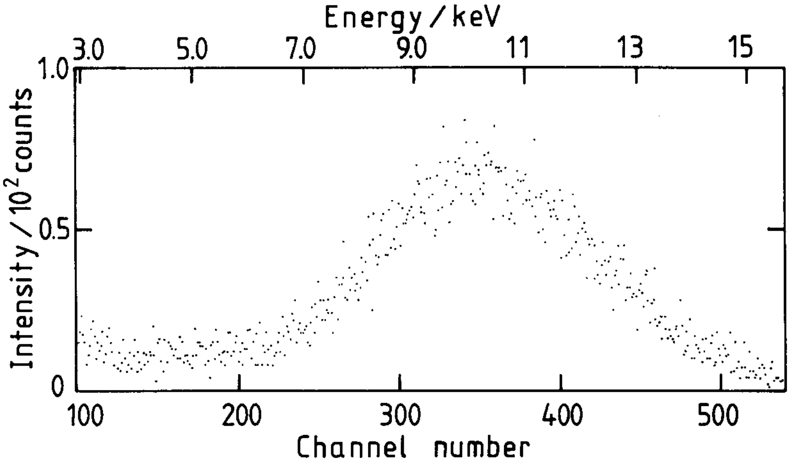

Fig.11. An example of bremsstrahlung X-ray spectra measured by using tritium gas enclosed in glass ampoule.

tion is also valuable for the present observations shown in Fig.10.

Figure 11 shows an spectrum obtained for tritium gas enclosed in a glass ampoule. In this measurement, the glass ampoule was placed directly in front of the Si-APD as it stands : namely, without aluminum foil. The ambient temperature and integrating time were 16.1℃ and 0.9 ks, respectively. Other conditions were the same as the case of Fig.10. It is seen that a peak was observed in about 10 keV and total count was 1.29×10^4. The total count should be proportional to the number of β-rays although it is not easy to evaluate quantitatively the number of β-rays responsible for the generation of the bremsstrahlung X-rays because the length of the glass ampoule used was considerably larger than the diameter of the Si-APD. On the other hand, the peak shifted to higher energy side by 2.5 keV in comparison with the spectrum shown in Fig. 9 . This appears mainly due to the difference in the thickness of glass tube and aluminum foil. The wall thickness of the glass ampoule was about 1.0 mm, which is 13 times greater than that of the aluminum foil. Therefore, since the absorption of the bremsstrahlung X-rays is expected to become larger with decresing energy, the target of β-rays must be fabricated as thin as possible to yield high counting rate.

The preliminary measurements of bremsstrahlung X-rays by the Si-APD revealed that the Si-APD is quite useful for the in-situ and non-destructive measurements of high level tritium. It is possible to fabricate a compact detector system with easy maintenance. Further studies, however, are needed to improve the efficiency of generation of the bremsstrahlung X-rays.

4. Conclusions

To confirm the feasibility of Si-APDs for detection of low energy bremsstrahlung X-rays, we examined basic characteristics of a Si-APD and measured bremsstrahlung X-rays using two different tritium sources. It was confirmed that the Si-APD effectively detect photons in the energy range from 3 to 20 keV at about 20℃, and that the total count of photons was kept almost constant if the applied voltage and fine gain were set over 1150 V and 0.6, respectively, although the channel number of a peak depended on some extrinsic factors such as the applied voltage, gain and ambient temperature.

It was observed that a broad single peak of the bremsstrahlung X-rays was generated in the region below 15 keV by the interactions between a thin aluminum foil and the β-rays of tritium. Total count of the bremsstrahlung X-rays depended on the thickness of aluminum foil. The thickness dependence could be explained very well with theoretical analyses basing on the energy dependence of the absorption coefficient for bremsstrahlung X-rays. On account of those observations, we concluded that the Si-APD is considerably valuable for in-situ measurements of high level tritium.

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