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論　文

トリチウム汚染のその場測定に対するベータ線誘起X線計測法の適用性

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Feasibility of β-ray Induced x-Ray Counting Method to In-situ Measurement of Tritium Contamination

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Abstract

To conduct in-situ and real-time measurements of contamination due to tritium adsorption/absorption on the surfaces and in sub-surface layers of materials, a new measuring method was established. The method is principally based on measurement of x-rays induced by β-rays from tritium. To assess the applicability of the present method, a graphite sample in which was implanted a given amount of tritium was used as a model. After tritium implantation, measurement was carried out using an ultra-low energy Ge detector under air or argon atmosphere. In the latter atmosphere, a sharp and strong characteristic x-ray peak attributed to K-transition of Ar appeared at 2.96 keV along with a broad bremsstrahlung x-ray spectrum, indicating x-rays more penetrating than the β-rays to be effectively generated by substitution of argon and contamination to be easily evaluated by conventional x-ray detectors without special detectors for β-ray counting. Accordingly, it was revealed that the present method is valuable for in-situ and real-time measurements of surface contamination due to tritium exposure owing to high durability and reliability, simple construction and ease of maintenance.

1. Introduction

The development of techniques related to safe handling and the best use of tritium will be one of important subjects for establishment of a tritium circulation system in the thermonuclear fusion devices. In the circulation system containing a reactor core, surfaces of various materials are exposed to a large number of molecular or ionic tritium with a high specific activity. For this reason, a part of those tritium stays on the surfaces and in sub-surface layers of materials, depending on exposure conditions. Namely, contamination of the material surfaces is unavoidably caused by the adsorption and absorption of tritium. Such tritium-contaminated surfaces also may cause a radiological hazard of workers in tritium handling facilities. Therefore, accurate assessment of the hazard requires a reliable method to measure the contamination level.

Measurement of contamination level of the surfaces has been carried out so far by some methods[1-3]: for example, wipe-assay utilizing a liquid scintillation counter, a windowless gas flow counter, a windowless vacuum scintillation counter, thermal desorption and so on. The first method is the most commonly used method for measurements of removable tritium. The next two methods can be applied to measurements of tritium surface contamination, but there are some disadvantage points in each method: that is, the former gas flow counter has a thin and fragile membrane as a entrance window of β-rays, while the latter counter is sensitive even to feeble light come into the scintillator through a clink between the counter and the surface of a material, and applicable only to large smooth surfaces which permit the necessary vacuum seal. The last method using thermal desorption is applicable to only small samples which are durable against heating at high temperatures. Such weak points in each measuring method are due to inherently low penetration power of tritium β-rays: about 5 mm in the air and a few μm in solids. Therefore, if the x-rays induced by the β-rays from tritium are available for measurements of contamination level, the weak points will be improved because the x-rays are more permeable than the β-rays.

It is possible for the tritium β-rays to excite K x-rays in elements with $Z \leqq 43$. The probability of L- or M-transitions is very lower than that of K-transition. The relative transition probabilities increase as K>L>M by about a factor of 100 in each case. On the other hand, as the energy of characteristic x-rays becomes lower than 2 keV in elements with $Z \leqq 14$, detection of the low energy x-rays becomes difficult because of a large degradation during

a traveling. Namely, promising elements are lying in a region of $15 \leqq Z \leqq 43$, and hence we have selected Ar(Z=18) as a working material, taking account of transition probability, running cost, safety, and so on. According to the reference[4], characteristic x-ray energy due to K-transition of Ar consists of 2.95563($K_{\alpha 2}$) and 2.95770($K_{\alpha 1}$) keV.

In this paper, the feasibility of a new method of β-ray induced x-ray counting (hereafter denoted as BIXC) will be described to evaluate the contamination due to tritium adsorption/absorption on the surface and in sub-surface layers of materials.

2. Experimental

2.1. Apparatus

2.1.1 Implantation of tritium ions

Figure 1 shows the schematic diagram of an apparatus used for the implantation of tritium ions. The apparatus mainly consisted of three parts such as an ultra-high vacuum (UHV) system, a gas supply part, and an ion implantation part. The UHV system was constructed by a turbomolecular pump(TMP) backed with an oil-sealed rotary pump(RP). The residual pressure of the UHV system was routinely below 7×10^{-7} Pa. The gas supply part consisted of two tritium containers, a deuterium storage container, and an argon cylinder. One of two tritium containers was subjected to supply and storage of tritium(GE-1), and the another was used for recovery of tritium during an implantation(GE-2). The double sealed structure was applied to prevent permeation and release of tritium through/from the container, while for a deuterium container a single sealed structure was employed. Each container was loaded with an given amount of Zr_9Ni_{11} alloy powder. The supply of tritium and deuterium was carried out by controlling temperature of the alloy powder with the heater set in the containers.

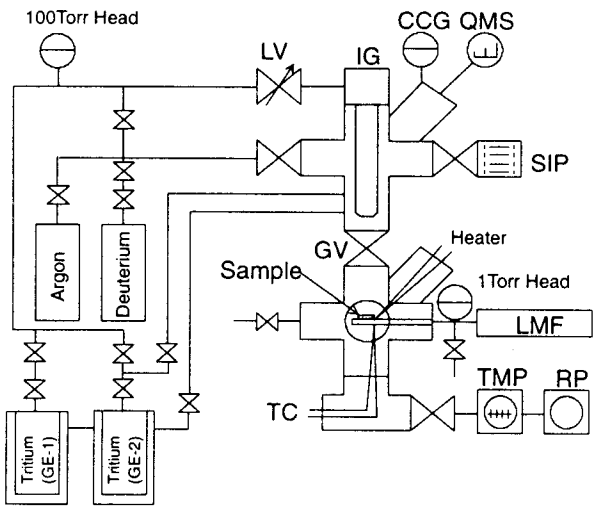

Fig. 1. Schematic diagram of the tritium implantation apparatus.

The ion implantation part consisted of a sample holder fixed by a linear motion feedthrough (LMF) movable horizontally 10cm, an ion gun, pressure measuring and gas analysis equipments. The sample holder was separable from an ion gun(IG) by a gate valve(GV). All of a sample, a K-thermocouple and a heater, which is made by a fine W-Re(26%) wire, were attached with the sample holder. A cold cathode gauge(CCG) and a quadrupole mass spectrometer(QMS) were employed for the residual gas and the purity analysis of tritium. After tritium implantation processing, a sputter ion pump(SIP) was used to minimize the release of residual tritium from the implantation device as much as possible.

2.1.2. Measurements of x-rays induced by tritium β-rays

Figure 2 shows the schematic arrangement of an apparatus used to measure x-ray spectra from the sample. The sample implanted tritium ions was fixed in front of a x-ray detector. An ulta-low energy Ge detector(Model: GUL0055P), which was delivered from Canberra Industries, Inc., was utilized for the present measurements. The energy resolution (Full Width at Half Maximum:FWHM) of the detector was 140eV at 5.9 keV. The distance from the sample to the detector was fixed to 5 mm. To effectively generate characteristic x-rays by β-rays from the implanted tritium, the sample was placed in an argon atmosphere. Flow rate of argon was set about 30 cm^3/min.

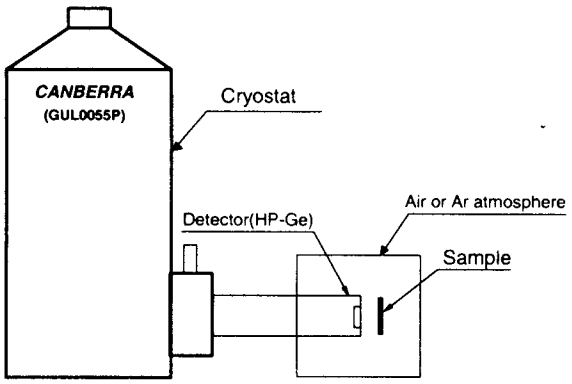

Fig. 2. Schematic arrangement of the apparatus used for measurements of x-rays.

2.2. Materials

The sample of graphite plates used in the present study was purchased from Toyo Tanso Co., and the type and size was IG-430U and 15x15x0.5 mm, respectively. The graphite of IG-430U type will be employed for the first wall material of the Large Helical Device (LHD) constructed in the facilities of National Institute for Fusion Science.

The purity of deuterium and argon was 99.6% and 99.999%, respectively. Those gases were purchased from Nihon Sanso Co. Tritium used for ion implantation was diluted with

deuterium, and the concentration was about 1 %. A given amount of the diluted tritium has been usually stored in the container(GE-2).

2.3 Procedures

The graphite plates were used as received. After the sample of a graphite plate was set at the sample holder in the ion implantation part, evacuation of the part was performed at room temperature. As the residual pressure of the ion implantation part reached below 10^{-5} Pa, the sample was heated stepwise up to 673 K, and the maximum temperature was kept for 1 hour. At this time, if the residual pressure was over 1.3×10^{-4} Pa, additional heating of the sample was performed for 3 hours. After the vacuum heating, the sample was cooled down to room temperature and a given amount of tritium was implanted.

With regard to the implantation of tritium, the procedures are as follows. After the thermal treatment of the sample, tritium gas was prepared at first by heating the GE-1 getter at around 500 K. Released tritium was introduced into the ion implantation part through a leak valve, and during the implantation the GE-2 getter was working to recover residual tritium in gas phase. The tritium pressure in the implantation part was slowly raised by controlling a leak valve until a discharge of the ion gun striked. Filament and discharge currents were previously set at 2.3 A and 27 mA, respectively. Kinetic energy of tritium ions was set 1 keV. After implantation of a given amount of tritium, the discharge was shut off, and then the supply of tritium was stopped by closing a leak valve. The residual tritium in the implantation part was evacuated by using a sputter ion pump for 2 days. Prior to taking out the sample, a sample holder was isolated from a chamber loaded with the ion gun by a gate valve, and then the chamber loaded with the sample holder was exposed to the ambient air. After that, to decontaminate tritium adsorbed on the surface of the chamber wall, a tritium removal system was connected to the chamber. The removal system was constructed by a catalyst reactor, a dryer with molecular sieves, an ionization chamber and a temperature controller. Tritium level in the chamber was followed by the in-line ionization chamber. After the tritium level decreased to a given concentration, the chamber was opened and the implanted sample was taken out.

To measure the tritium activity on the surface of a graphite sample, the sample was set at a distance of 5 mm in front of the ultra-low energy Ge detector. The energy calibration of the detector was previously carried out by using radioactive sources such as ^{57}Co(6.4, 7.0 and

14.4 keV), ^{133}Ba(32.2 and 35.0 keV), ^{137}Cs(36.3 keV) and ^{241}Am(13.9, 17.6 and 59.5 keV). The every measurements of a x-ray spectrum were performed for 1 hour.

3. Results and discussion

3.1. A typical sample bombarded with tritium ions

Figure 3 shows an example of the graphite sample bombarded with a given amount of tritium ions. The shadowy circle in the figure describes the implantation part. The total amount of tritium in the sample was examined by a combustion method. In this measurement, the sample was divided four pieces to examine the distribution of tritium concentration, and then each of them was perfectly burned in pure oxygen flow by the aid of a red-hot platinum coil. Tritiated water vapor formed by the oxidation was trapped by a series of two water bubblers of 100 cm^3.

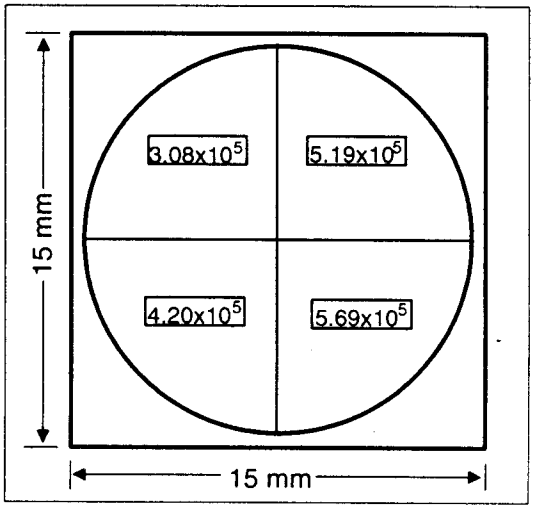

Fig. 3. An example of the graphite sample bombarded with tritium ions.

After the finish of each combustion, water in limited amounts in each bubbler was taken out, and it was measured by using a liquid scintillation counter. The tritium activity for each piece was magnitude of 10^5 Bq as shown in the figure; however, the activities were slightly different from each other because of slight deviation from the vertical arrangement of the ion gun. Total amount of tritium contained in the sample was evaluated as 1.8×10^6 Bq. Such tritium activity was reproduced in each implantation under the present conditions. It was revealed, therefore, that a sample contaminated by a given amount of tritium can be prepared by using the present implantation apparatus.

The kinetic energy of tritium ions implanted into the sample was controlled to be 1 keV as mentioned before. For this reason, most of tritium in the sample would stay within about 20 nm from the surface. The ranges of hydrogen isotopes in graphite are reported by numerous investigators[5, 6]. On the other hand, escape depth of the β-rays emitted from tritium will reach about 1000 nm in graphite. As the range of tritium ions is remarkably shorter than

the escape depth, a part of the β-rays from tritium implantated can penetrate through sub-surface layers of the graphite. Therefore, the escaped β-rays as well as those from the surface tritium will also play a significant role for interactions with constituents in an ambient atmosphere.

3.2. Measurement of x-ray spectra induced by β-rays

Basic performances of the ultra-low energy Ge detector used in the present study were examined at first. Figure 4 shows a background spectrum in the energy range from 0 to 15 keV. In this measurement, the discrimination level of pulse height signals was set at 0.7 keV. A slightly high background was observed below 2 keV, but above that energy the intensity was almost constant as around 0.03 counts/min(ΔE), where ΔE represents the energy per one channel of a multi-channel analyzer used, and it was determined as 34.4 eV/channel by using radiation sources of γ- and x-rays. In addition, the resolution power of the present detector was evaluated to be 125 eV by FWHM of the characteristic x-ray peak(7.04 keV) from a ^{57}Co source.

Figure 5 describes an example of spectra observed in the air. A sharp and small peak appeared around 3 keV, and it overlapped with a broad peak appeared in the range from 1 to 15 keV. Both of those peaks resulted in interactions between β-rays and graphite and/or air components. The total intensity amounted to 143.2 counts/min in the wide energy range of 1 to 15 keV. The intensity of the sharp peak amounted to

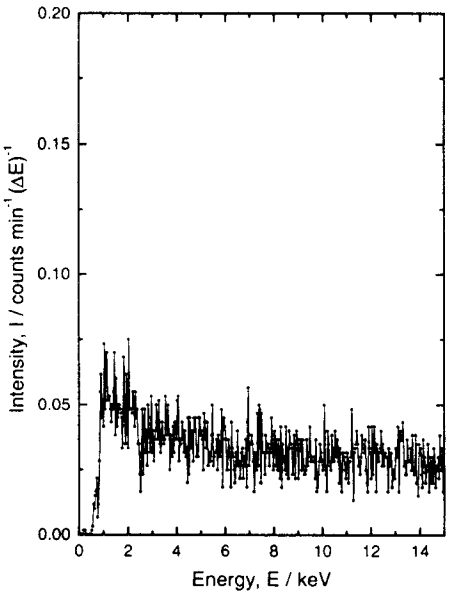

Fig. 4. Background spectrum of the x-ray detector used in the present study.

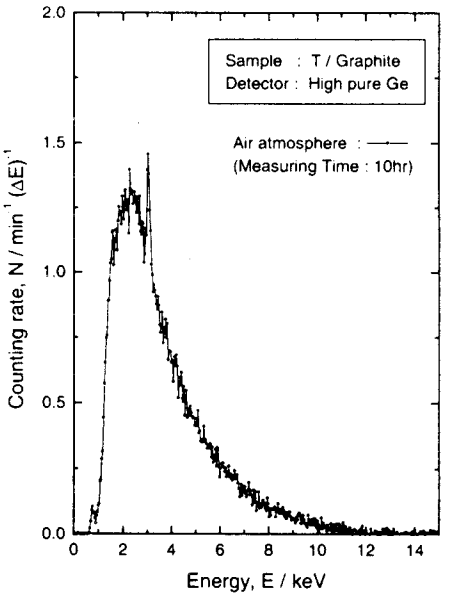

Fig. 5. An example of the spectrum observed in the air atmosphere.

1.7 counts/min in the energy range of 2.8 to 3.2 keV. This peak was attributed to the characteristic x-rays of Ar, taking the peak energy into consideration because argon is contained 0.934 vol% in the atmosphere. On the other hand, the broad peak was attributed to the bremsstrahlung x-rays. Namely, it was seen that a part of β-ray energy was converted to two kinds of electromagnetic wave, characteristic and bremsstrahlung x-rays. Especially, it should be mentioned here that the sharp characteristic x-ray peak coming from K-transition of Ar could be observed easily. Because penetration power of the x-rays is much stronger than the β-rays from tritium, large penetration power will give a wide possibility in the selection of a tritium detector.

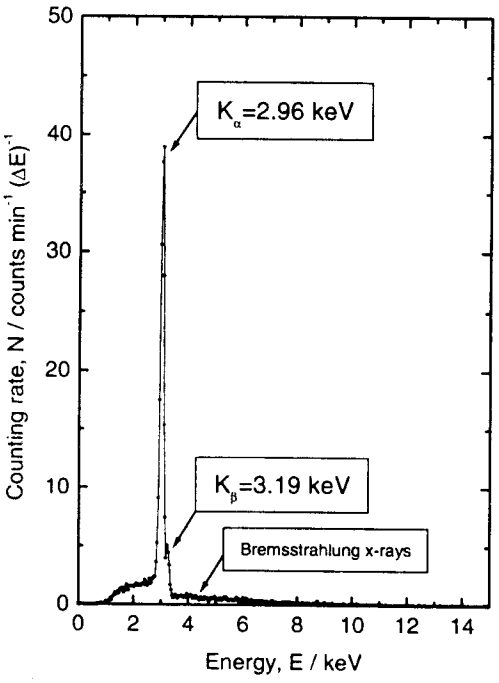

Fig. 6. An example of the spectrum observed in the argon atmosphere.

To confirm effectiveness of argon substitution, similar measurements were performed under the condition of a pure argon atmosphere. An example of the results is shown in Fig. 6. A remarkably intense peak appeared at 2.96 keV, which was attributed to the characteristic x-rays of Ar K_α lines. Intensity of the peak top was 39.0 counts/min and the total intensity in the energy range of 2.8 to 3.2 keV amounted to 158.4 counts/min. From these observations, it should be noticed that the intensity of characteristic x-rays increased about 100 times in comparison with that observed in Fig. 5. On the other hand, back- ground intensity was negligibly as small as 0.5 counts/min in the same energy region.

Table 1. Summary of the peak intensity in each atmosphere.

	Ambient atmosphere	
	Air (counts/min)	Argon (counts/min)
Total intensity (1 - 15 keV)	143.2	354.9
Characteristic x-rays (2.8 - 3.2 keV)	1.7	158.4
Bremsstrahlung x-rays (1 - 15 keV)	127.7	182.7
Background	13.8	

Spectral intensity under the air and the argon atmospheres was

summarized in Table 1. In addition to the characteristic x-ray peak, a broad bremsstrahlung x-ray peak also appeared in an argon atmosphere as described in Fig. 6. The intensity of the bremsstrahlung x-ray peak was evaluated to be 182.7 counts/min, which is comparable to the intensity of the characteristic x-ray peak. The total intensity of the both x-ray peaks amounted to 354.9 counts/min in the energy range of 1 to 15 keV, including the background. The total intensity of the two peaks in the argon atmosphere was mere 2.5 times that in the air. It was revealed from those results, therefore, that the intensity of the characteristic x-rays will be available to evaluate the amount of tritium adsorbed on the surface and absorbed in sub-surface layers.

On the other hand, it can be seen that shape of the bremsstrahlung x-ray spectrum obtained under an argon atmosphere was much different from that observed in the air as shown in Fig. 5. The difference lies in the change of an ambient atmosphere. This is due to a reason that bremsstrahlung x-rays having higher energy than the characteristic x-rays took place an interaction of photoelectric effect during traveling in an argon atmosphere.

3.3 Apparent efficiency for tritium detection

As mentioned before, total tritium activity of the present sample measured by a combustion method was evaluated as 1.8×10^{6} Bq, and geometric area of the sample bombarded with tritium ions was 1.5 cm^2. Namely, average tritium activity per unit area can be calculated as 1.2×10^{6} Bq/cm^2, although the tritium distribution of the sample surface was not uniform. Effective area of a beryllium window of the ultra-low energy Ge detector used was 0.5 cm^2 (0.8 cm in diameter). Taking those values into account, the minimum number of β-rays which can contribute to the generation of characteristic x-rays within the area of the beryllium window will reach about 3.0×10^{5} Bq, provided that a sample is placed near the detector and that the β-rays are emitted isotropically.

The intensity of the characteristic x-ray peak observed in the argon atmosphere was 2.6 counts/s as shown in Table 1. Apparent efficiency will be given by the ratio of tritium activity to the observed intensity. Namely, the apparent efficiency under the present conditions is calculated as about 1×10^{-3} %, provided that only characteristic x-rays is available for measurements of tritium adsorbed on the surface and of tritium absorbed in sub-surface layers of materials. As shown in Fig. 6, however, the broad bremsstrahlung x-ray peak also appeared with the sharp characteristic x-ray peak. The bremsstrahlung x-rays also

will play an important role for measurements of the amount of surface tritium because their intensities were comparable to those of the characteristic x-rays. That is, the apparent efficiency will rise about 2 times.

In addition, a lower detection limit of the detector will be also an important factor for the practical use. A small peak(1.7 counts/min) attributed to the characteristic x-rays of Ar which is contained in the air could be clearly detected along with a broad bremsstrahlung x-ray peak. Basing on this observation and the background level of the present detector, it appears that the present detector is measurable as a weak intensity as about 0.2 counts/min. Therefore, the lower limit of the present measuring system will reach about 1000 Bq/cm^2. If another type of a x-ray detector with a large entrance window can be employed for the measurements, the detection limit can be lowered, and also the apparent efficiency will increase. Although energy resolution is not so good as that of the present x-ray detector, the conventional scintillation detectors attached with NaI(Tl) crystal are convenient for this purpose because a large crystal of NaI(Tl) can be commercially prepared with ease. For example, it is no problem to obtain a NaI(Tl) scintillation detector having a beryllium window of 25mm in diameter, and by employing it the area(5.0 cm^2) of the entrance window is enlarged just 10 times. Namely, it is expected that a x-ray detector having a large beryllium window will be able to cause the increase in feasibility of the present method.

4. Conclusions

To solve some difficulties of the conventional tritium detector for measurements of a contamination level of the material surfaces, feasibility of a new measuring method based on the BIXC has been examined. Prior to the examinations, graphite samples contained a given amount of tritium were prepared by using a tritium implantation apparatus as a model sample of the contaminated surface. It was seen that it is possible to prepare the surface contaminated by various amounts of tritium. A resulted sample was applied to measure the x-rays induced by tritium β-rays, and the measurements were carried out in the argon atmosphere. As a result, a sharp and strong characteristic x-ray peak attributed to the K-transition of Ar appeared at 2.96 keV along with a broad bremsstrahlung x-ray peak. The peak height of the characteristic x-rays was above 1000 times larger than the background intensity of the x-ray detector used in the present study. Applying a conventional large solid scintillation counter, it

is expected that the lower detection limit will reach about 100 Bq/cm^2. Namely, it was reveal that the method of BIXC is promising for in-situ and real time measurements of the contamination due to adsorption of tritium on the surface and to absorption in sub-surface layers of materials.

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