New Technique for In-Situ Tritium Measurement in Future Fusion Reactors

M. Matsuyama, Y. Torikai, M. Hara, and K. Watanabe
Hydrogen Isotope Research Center, University of Toyama
Gofuku 3190, Toyama 930-8555, Japan
E-mail: masao@ctg.u-toyama.ac.jp

Abstract
To establish a new technique for in-situ measurements of high-level tritium, applicability of the new measuring technique that is based on utilization of X-rays induced by β-rays from tritium was examined for three physical states of tritium such as gaseous tritium, aqueous tritium, and solid tritium retained on the surface and/or in bulk of the tritium-exposed materials in fusion reactors. It was found that the intensity of X-rays induced by β-rays was well proportional to tritium partial pressure in a mixture gas and also to tritium concentration in water in a wide range. In addition, it was seen that this technique is applicable to evaluate the amount of tritium retained in a sub-surface layer of materials and to determine a depth profile of tritium in the bulk of materials. This measuring technique was named β-ray-Induced X-ray Spectrometry (BIXS), and it was concluded that BIXS is widely applicable to in-situ measurements of high-level tritium in future fusion reactors as well as ITER.

1. Introduction

It is indispensable to evaluate tritium distribution and/or inventory not only in the reactor core, but also in the tritium plant of fusion reactors from viewpoints of safe handling of high-level tritium and tritium economy. A variety of techniques to measure tritium have been proposed and established so far, but most of them are mainly for measurements of low-level or tracer-level tritium. Namely, precise evaluation of gaseous tritium, tritiated water, and tritium-contaminated materials in fusion reactors needs a new measuring technique that is able to measure huge amount of tritium and/or high-level tritium, but no reliable methods have been developed so far except a calorimetric method[1].

Conventional techniques established so far are mainly based on utilization of an ionization process and a scintillation process by interactions with β-rays emitted from tritium species. For example, a small ionization chamber was developed for measurements of high-level tritium[2], but contamination of the inner walls of the ionization chamber is not avoidable. In addition, the observed ionization current is apparently variable by the changes in the total pressure of a tritium-mixing gas, even if the number of tritium atoms in the chamber is the same. On the other hand, a liquid scintillation counter is conventionally used for measurements of low-level tritiated water, whereas it is not suitable for measurements of high-level tritiated water due to a lack of upper detection limit and generation of high-level organic wastes. Inorganic[3] and plastic scintillation counters are also easily contaminated by
adsorption and/or absorption of high-level tritium. It is also possible to utilize spectroscopic methods such as infrared\textsuperscript{[4]} and laser Raman\textsuperscript{[5]} to measure tritiated water vapor, hydrocarbon labeled with tritium, and elemental tritium. However, it is hard to apply to a practical tritium system in fusion reactors although they are suitable for experiments in laboratory scale of tritium.

High-level tritium can be measured by techniques such as volumetric, gravimetric and calorimetric methods. These methods are suitable for absolute measurements of high-level tritium, but not in-situ and/or in-line measurements of tritium. From this viewpoint, a new technique for in-situ measurements of high-level tritium is required for establishment of a safe and efficient tritium handling system. In this paper, feasibility of the new technique, which is based on utilization of bremsstrahlung and characteristic X-rays produced by interactions between β-rays and substances, will be discussed.

2. Application of a new technique to evaluation of gaseous tritium

To apply a new measuring technique for non-destructive measurements of high-level gaseous tritium, a specially designed confinement-device of tritium was constructed as shown in Fig. 1. The confinement-vessel of tritium consists of the connection parts employed in the ultra-high vacuum systems to prevent a tritium leak. Volume of the confinement-vessel was about 30 cm\textsuperscript{3}. β-rays strike the surface of a thin beryllium disk coated with a gold film and X-rays are emitted. A part of the produced X-rays penetrates through the thin beryllium disk and it enters into an X-ray detector. A modified NaI(Tl) scintillation detector equipped with a beryllium window was usually used to measure the intensity of X-rays penetrated through the gold-coated beryllium disk.

Changes in the intensity of X-rays with tritium concentration were measured using three kinds of tritium gases\textsuperscript{[6]}. Nominal tritium concentration of T\textsubscript{2}, D\textsubscript{2}-T\textsubscript{2} and H\textsubscript{2}-T\textsubscript{2} mixture was 99, 1, and 1%-tritium, respectively. Figure 2

![Fig. 1 Sectional view of the confinement-vessel of tritium gas.](image)

![Fig. 2 Total pressure dependence of the X-ray intensity in gaseous tritium.](image)
shows the correlation between the X-ray intensity and the total pressure of each tritium gas. It was found that the X-ray intensity is proportional quite well to the total pressure below about 2 kPa in all tritium gases, whereas above 2 kPa the lower deviation from the linearity gradually increased with the total pressure. At a pressure region below 2 kPa, the intensity ratio of X-rays gives a relative concentration of tritium.

Considerable deviation from the linearity of X-ray intensity above 2 kPa is basically due to the self-absorption of β-rays in the tritium gases and dead time of the X-ray detector. Accordingly, it was seen that the deviation was thoroughly improved by considering them, and a linear calibration curve applicable up to a higher pressure was obtained. Such a calibration curve was also applicable to other mixtures such as He-T$_2$ and N$_2$-T$_2$[7].

3. Application of a new technique to evaluation of aqueous tritium

A liquid scintillation counter is commonly used to measure concentration of tritiated water. However, the upper detection limit is not enough for measurements of high-level tritiated water, and flammable organic wastes containing high-level tritium are released after measurements. To solve such problems, feasibility of the new technique depending on the same principle as that mentioned above was examined[8]. For this purpose, a metallic vial as shown in Fig. 3 was specially designed and manufactured. The metallic vial is basically made of stainless steel and only the bottom is made of a thin beryllium plate. Inner surface of the beryllium plate was coated with gold and the maximum volume was 13 cm$^3$. Size of the metallic vial was designed to just fit an entrance window of NaI(Tl) scintillation detector.

First of all, dependence of X-ray intensity on volume of tritiated water was measured. The results showed that the X-ray intensity convexly increased with the volume of tritiated water: the saturation intensity appeared in around 7 cm$^3$. This indicates that it is not necessary to load the metallic vial with tritiated water above 7 cm$^3$. Thus, tritiated water of 5 cm$^3$ was selected as a sampling volume, taking a sampling process into account. Subsequently, dependence of tritium concentration on the X-ray intensity was examined. The result is shown in Fig. 4. The metallic vial and X-ray detector was installed in a lead shielding box to lower the effects of natural radiations. The background level in the present measuring system was 7.30±0.12 cpm. The concentration of tritiated water used for examination was

![Fig. 3 Sectional view of the metallic vial and X-ray detector for measurements of tritiated water.](image-url)
determined with a liquid scintillation counter calibrated previously by using standard tritiated water samples. As is shown in Fig. 4, it is clearly seen that a quite good linear relation between tritium concentration and X-ray intensity can be obtained in a wide concentration range from 4 GBq/m$^3$ to 40 TBq/m$^3$. In this run, a given concentration of tritiated water was measured one by one, but the present technique is also applicable to a flow system of high-level tritiated water. To examine the effects of metallic impurities in tritiated water, tritiated water containing a given amount of nickel was subjected to the measurements as a model. As a result, if the impurity level is below 300 ppm, it was seen that the effects of metallic impurities on X-ray intensity are negligibly small.

4. Application of a new technique to evaluation of tritium retained on/in materials

Quantitative evaluation of high-level tritium retained on/in various materials plays a very important role in future fusion devices as well as ITER from viewpoints of reduction of the wastes contaminated with tritium and of occupational exposure by tritium. Although surface tritium monitors have been proposed so far$^{[9]}$, they are not enough for the present purpose. Therefore, we examined applicability of a new technique for in-situ measurements of tritium retained on/in materials. A new technique applied to the present measurements is basically based on the same principle as that to measurements of gaseous and aqueous tritium. In this measurement, a tritium-containing sample was placed in an argon atmosphere. The $\beta$-rays from tritium species trapped in a sub-surface layer of materials can excite argon atoms, and characteristic X-rays of argon are emitted in an argon atmosphere. It is expected, therefore, that the characteristic X-ray intensity of argon is proportional to the amount of tritium retained in a sub-surface layer of the material. In addition, kinetic energy of the $\beta$-rays emitted from tritium species dissolved in materials is partly converted to bremsstrahlung and characteristic X-rays, and also those are emitted in an argon atmosphere. Measurements of X-ray spectra were carried out using a high purity Ge detector. Since shape of a bremsstrahlung X-ray spectrum depends on a depth profile of tritium in materials, analysis of the shape gives information about tritium distribution in the materials.

Typical X-ray spectra obtained from the quite distinct depth profile of tritium are shown in Fig. 5: one is a graphite sample (sample 1: thickness is 0.5 mm) that was irradiated by tritium ions with kinetic energy of 1 keV$^{[10]}$, and the other is a polymer source (sample 2:
thickness is 1 mm) labeled with tritium$^{[11]}$. Most of tritium in the former sample 1 is retained in a sub-surface layer, while tritium in the latter sample 2 uniformly distributes in the bulk. It can be seen that quite different spectra are observed for both samples: for the sample 1 characteristic X-rays of argon appeared strongly in the spectrum, whereas for the sample 2 bremsstrahlung X-ray intensity was relatively strong. Namely, shape of the X-ray spectrum induced by $\beta$-rays reflects tritium distribution in the materials.

This technique has been successfully applied to the tritium evaluation of ALT-II carbon limiter tiles exposed to D-D plasmas in TEXTOR$^{[12]}$. In this measurement, residual tritium of around 100 Bq/cm$^2$ was confirmed. Similar non-destructive tritium measurements have been carried out at the facility of Tritium Laboratory Karlsruhe to examine a series of divertor tiles exposed to D-T plasmas in JET$^{[13]}$. The observed spectra were quite different from those of ALT-II carbon limiter tile: that is, it was found that a large amount of tritium was retained in not only a sub-surface layer but also the bulk and the retention drastically differed from tile to tile.

This technique was applied to evaluate tritium in not only low-Z materials such as boron$^{[14]}$ and carbon but also high-Z materials such as Pd, Zr, W$^{[15]}$ and stainless steel$^{[16]}$. One of many examples is shown in Fig. 6. This is a sample that a large amount of tritium was thermally introduced into stainless steel type 316L (SS-316L) as an example. The experiments have been carried out under the collaboration work with JET. As clearly seen from the X-ray spectrum, characteristic X-ray peaks attributed to the constituent elements of SS-316L appeared with a bremsstrahlung X-ray peak. From the observed X-ray spectrum, the depth profile of tritium in the SS-316L sample was examined by a
method of numerical analysis. The analyzing procedures are described in detail elsewhere[17].

Figure 7 shows a result of the tritium depth profile obtained by computer simulation of the spectrum shown in Fig. 6. The simulation X-ray spectrum recalculated by basing on the depth profile agreed quite well with the observed one. The tritium depth profile indicates that there is a high-level region in a sub-surface layer of the SS-316L sample. It is considered that enrichment of tritium in a sub-surface layer is due to some oxides on the sample surface of SS-316L. The present depth profile agreed well with that obtained from the other method such as a chemical etching by an acid mixture. Namely, the present technique gives information about not only the amount of tritium in a sub-surface layer but also a tritium depth profile in the bulk.

5. Summary

To establish a new technique for in-situ and non-destructive measurements of high-level tritium, feasibility of a new technique based on utilization of X-rays induced by β-rays emitted from tritium atoms was examined. This technique was named the β-ray-induced X-ray spectrometry (BIXS). From the first examination, it was found that the X-ray intensity was well proportional to the total pressure of tritium gas although deviation from the linearity appeared above 2 kPa owing to the self-absorption of β-rays and dead time of a NaI(Tl) detector. From the second examination, it was found that the observed X-ray intensity showed proportionality in tritium concentration over 4 orders of magnitude above 4 GBq/m³. From the third examination, it was seen that two kinds of X-ray peaks (characteristic and bremsstrahlung X-rays) appeared in the observed X-ray spectra and that the characteristic X-ray intensity of argon gave information about the amount of tritium in a sub-surface layer of the materials. The X-ray intensity of argon was proportional to the amount of tritium retained in a sub-surface layer. On the other hand, it was seen that a peak shape of the bremsstrahlung X-rays depends on a depth profile of tritium in a material. From these findings it was revealed that the present technique is able to evaluate separately tritium retention on/in a material.

Acknowledgements

This work was continuously supported by the Grant-in-Aid for Scientific Research of
the Ministry of Education, Culture, Sports, Science and Technology, and also supported by the TEXTOR program under IEA. The authors are in debt to many investigators of Tritium Laboratory and Institute for Materials Research II in FZK, Institute for Plasma Physics in FZJ, EURATOM/UKAEA Fusion Association in Culham Science Center and Tritium Engineering Laboratory in JAEA about helpful discussion for research and development of the new tritium measuring technique.

References