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# New technique for non-destructive measurements of tritium in future fusion reactors

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## Abstract

To establish a new technique for monitoring and control of high-level tritium in a fuel circulation system of the fusion reactors, the applicability of a new technique to elemental tritium and tritiated water was discussed. The new technique is based on the utilization of x-rays induced by  $\beta$ -rays from tritium, and it is called  $\beta$ -ray-induced x-ray spectrometry. The applicability of the same technique to the tritium species retained in solid materials was also discussed. It was concluded from applications to gaseous and aqueous tritium under the static state that the new technique is applicable as an in-line monitor in the fuel circulation system. In addition, it was also shown that the new technique plays an important role in non-destructive evaluation of tritium retained in solid materials such as plasma-facing materials.

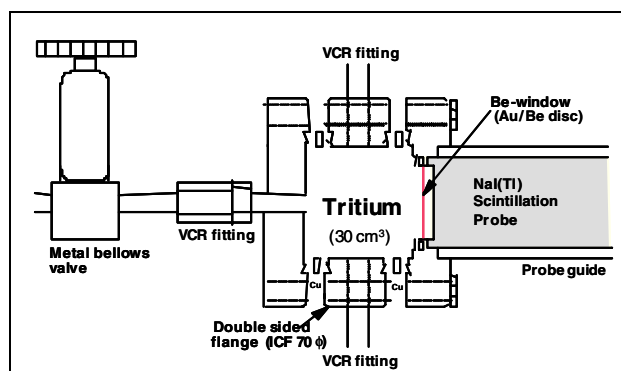
## 1. Introduction

From the viewpoints of safe handling of high-level tritium and tritium economy in fusion reactors, the monitoring and control of gaseous and aqueous tritium in a fuel circulation system of the fusion reactors is indispensable. In addition, quantitative evaluation of the amount and depth profile of tritium retained in the solid materials such as plasma-facing materials is also required. For these purposes, a variety of techniques to measure gaseous and aqueous tritium have been proposed and established so far, but most of them are mainly for relatively low-level or tracer-level tritium and for static measurements. Namely, precise evaluation of elemental tritium and tritiated water in a flowing system needs a new measuring technique, which is able to measure non-destructively high-level tritium and/or a huge amount of tritium. In addition to this, no reliable methods for non-destructive evaluation of the amount and the depth profile of tritium retained in solid materials have been developed so far except a calorimetric method [1].

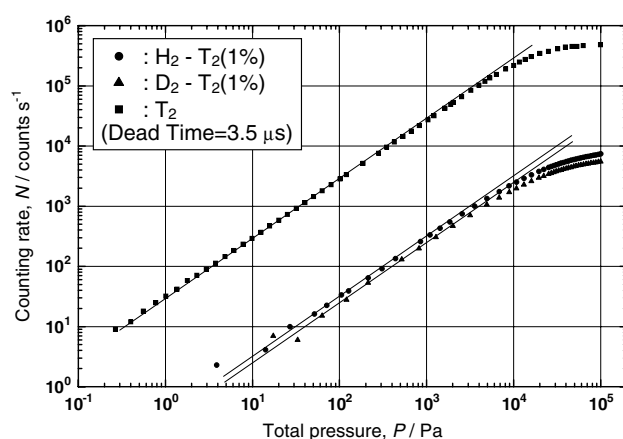
Conventional techniques established so far are mainly based on utilization of an ionization process and a scintillation process by interactions with  $\beta$ -rays emitted from tritium species. For example, a small ionization chamber developed for measurements of high-level tritium [2] is applicable to a flow system, 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 the ionization chamber, even if the number of tritium atoms in the chamber is equal. 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 an upper detection limit and much generation of high-level organic wastes. Inorganic [3] and plastic scintillation counters are also easily contaminated by adsorption and/or absorption of tritium. It is also possible to utilize spectroscopic methods such as infrared [4] and laser Raman [5] to measure tritiated water vapour, hydrocarbon labelled with tritium and elemental tritium. However, it is hard to apply these methods to a practical tritium system in fusion reactors although they are suitable for a laboratory scale experiment of tritium.

A large amount of pure tritium gas can be measured by techniques such as volumetric, gravimetric and calorimetric methods. These methods are suitable for absolute measurement of high-level tritium, but not in-line measurements of tritium. From this viewpoint, a new technique for non-destructive measurements of high-level tritium is required for establishment of a safe and efficient tritium handling system. In this paper, the feasibility of a new technique in the fusion reactors, which is based on utilization of bremsstrahlung and characteristic x-rays produced by interactions between  $\beta$ -rays and substances, will be discussed.



**Figure 1.** Sectional view of the confinement vessel of tritium gas.  
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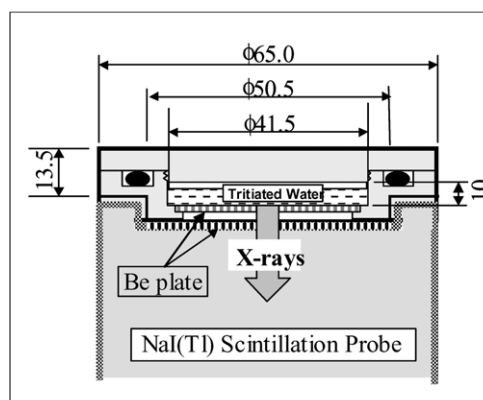


**Figure 2.** Total pressure dependence of the x-ray intensity in gaseous tritium.

## 2. Application of a new technique for 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 figure 1. The confinement vessel of tritium consists of the connection parts employed in the ultra-high vacuum systems to prevent a tritium leak. The volume of the confinement vessel was about 30 cm<sup>3</sup>. X-rays induced by  $\beta$ -rays mainly generate on the surface of a gold film coated on a thin beryllium disc. A part of the produced x-rays penetrates through the thin beryllium disc and 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 that penetrated through the gold-coated beryllium disc.

Changes in the intensity of x-rays with tritium concentration were examined using three kinds of tritium gases [6]. Nominal tritium concentration of T<sub>2</sub>, D<sub>2</sub>-T<sub>2</sub> and H<sub>2</sub>-T<sub>2</sub> mixtures was 99%, 1% and 1% tritium, respectively. Figure 2 shows the correlation between the x-ray intensity and the total pressure of pure tritium and tritium mixtures. It is seen that the x-ray intensity is quite proportional to the total pressure below about 2 kPa in all tritium gases, whereas above 2 kPa the deviation from the linearity gradually increased with the total



**Figure 3.** Sectional view of the metallic vial and x-ray detector for measurements of tritiated water.

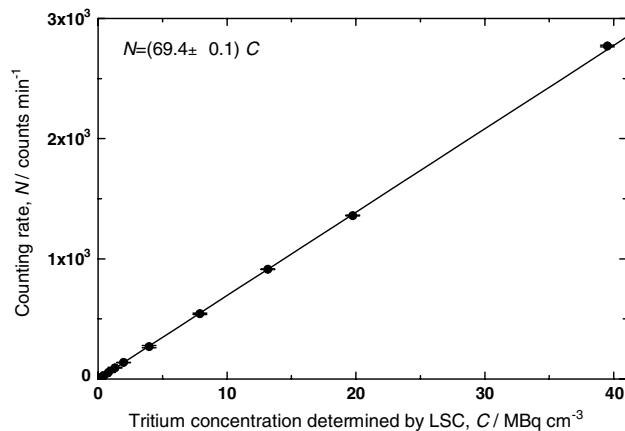
pressure. At a pressure region below 2 kPa, the intensity ratio of x-rays gave a relative concentration of tritium used in the examination. In addition, no memory effects were observed even after measurements of pure tritium.

Considerable deviation from the linearity of x-ray intensity above 2 kPa is basically due to the self-absorption of  $\beta$ -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 derived. Such a calibration curve was also applicable to other tritium mixtures such as He-T<sub>2</sub> and N<sub>2</sub>-T<sub>2</sub> [7].

## 3. Application of a new technique for evaluation of aqueous tritium

A liquid scintillation counter is commonly used to measure the 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 the measurements. To solve such problems, the feasibility of the new technique based on the same principle as that mentioned above was examined [8]. For this purpose, a metallic vial shown in figure 3 was specially designed and manufactured. The metallic vial was basically made of stainless steel and only the bottom was made of a thin beryllium plate. The inner surface of the beryllium plate was coated with gold and the maximum volume was 13 cm<sup>3</sup>. The size of the metallic vial was designed to just fit an entrance window of a NaI(Tl) scintillation probe.

First of all, dependence of x-ray intensity on the 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<sup>3</sup>. This indicates that it is not necessary to load the metallic vial with tritiated water above 7 cm<sup>3</sup>. Thus, 5 cm<sup>3</sup> of tritiated water was selected as a sampling volume, taking the sampling process into account. Subsequently, the dependence of tritium concentration on the x-ray intensity was examined. The result is shown in figure 4. The metallic vial and the x-ray detector were installed in a lead-shielded box to lower the effects of natural radiations. The background level in the present measuring system was  $7.30 \pm 0.12$  cpm. The concentration of



**Figure 4.** Calibration curve of tritium concentration for tritiated water.

tritiated water used for examination was previously determined using a liquid scintillation counter calibrated using standard tritiated water samples. As is shown in figure 4, a quite good linear relation between tritium concentration and x-ray intensity was observed.

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. When we apply this technique to a flow system, some issues have to be examined: for example, durability of gold plating and beryllium disc for a long time, effects of radiations from activated impurities if exist in a flow system, and so on.

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

Quantitative evaluation of tritium retained on/in various materials plays a very important role in future fusion devices as well as ITER from the viewpoints of reduction of the wastes contaminated with tritium and of exposure of workers to tritium. Although surface tritium monitors have been proposed so far [9], they are not enough for the present purpose. Therefore, we examined the applicability of a new technique for non-destructive measurements of tritium retained on/in materials. A new technique applied to the present measurements is basically based on the same principles as that applied to measurements of gaseous and liquid tritium. In this measurement, surfaces of a tritium-containing sample were exposed to argon. The  $\beta$ -rays from tritium species trapped on the surface and 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 intensity of the characteristic x-rays 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 a material is partly converted to bremsstrahlung and characteristic x-rays, and those are emitted

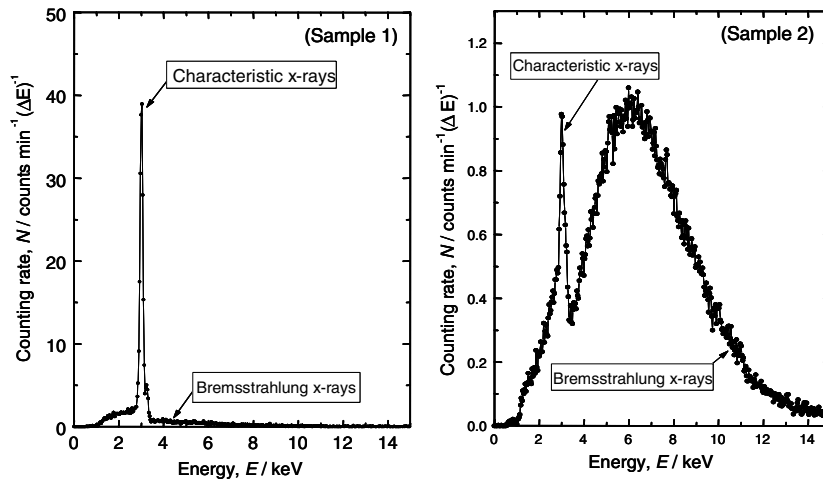
in an argon atmosphere. X-ray spectra are measured using a high purity Ge detector. Since the shape of a bremsstrahlung x-ray spectrum induced by  $\beta$ -rays depends on the depth profile of tritium in a material, analysis of the shape gives information about tritium distribution in the material.

Typical x-ray spectra obtained from the quite distinct depth profile of tritium are shown in figure 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) labelled uniformly with tritium [11]. Most of the 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 sample 1 characteristic x-rays of argon appeared strongly in the spectrum, whereas for sample 2, bremsstrahlung x-ray intensity was relatively strong. Namely, the shape of the x-ray spectrum induced by  $\beta$ -rays reflects tritium distribution in a material.

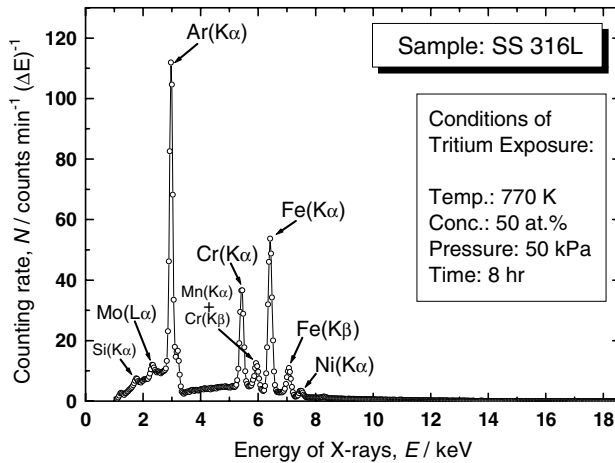
This technique has been successfully applied to the tritium evaluation of ALT-II carbon limiter tile exposed to D-D plasmas in TEXTOR [12]. In this measurement, residual tritium of around  $100 \text{ Bq cm}^{-2}$  was confirmed. Similar non-destructive tritium measurements have been carried out at the 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 not only in a sub-surface layer but also in the bulk and the retention drastically differed from tile to tile.

This technique was applied to evaluate tritium not only in low-Z materials such as boron [14] and carbon but also in high-Z materials such as Pd, Zr, W [15] and stainless steel [16]. One of the many examples is shown in figure 6. This is a sample in which a large amount of tritium was thermally introduced into stainless steel type 316L (SS-316L) as an example. The experiments have been carried out as 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 the 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 analysing procedures are described in detail elsewhere [17].

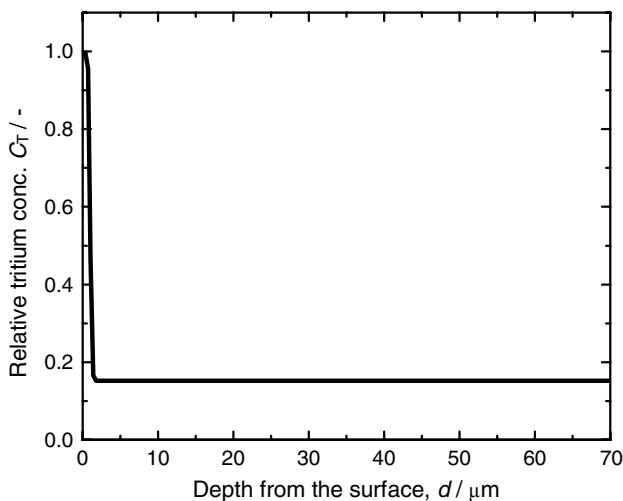
Figure 7 shows the result of the tritium depth profile obtained by computer simulation of the spectrum shown in figure 6. The simulation x-ray spectrum based on the depth profile agreed quite well with the observed one. The tritium depth profile indicates that there is a high-level region in a sub-surface 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 surface layers but also the tritium depth profile in the bulk. This technique can be used for non-destructive evaluation of the plasma-facing materials in fusion reactors by employing a remote control system.



**Figure 5.** X-ray spectra induced by  $\beta$ -rays from tritium species retained on/in the materials: sample 1 is a carbon plate irradiated with tritium ions, and sample 2 is a polymer source labelled with tritium.



**Figure 6.** X-ray spectrum induced by  $\beta$ -rays from tritium atoms dissolved in a SS-316L sample.



**Figure 7.** Tritium depth profile obtained by analysing the x-ray spectrum observed for the SS-316L sample.

## 5. Summary

To establish a new technique for the monitoring and control of high-level tritium in the fuel circulation system of fusion reactors, the feasibility of a new technique based on utilization of x-rays induced by  $\beta$ -rays emitted from tritium atoms was discussed. The new technique is called  $\beta$ -ray induced x-ray spectrometry (BIXS), which has some advantages: namely, it is possible to measure non-destructively without sampling of tritium gases and tritiated water from the fuel circulation system, the working range of the detector is fairly wide, the x-ray conversion part is small and compact and the maintenance of the detector is easy. On the other hand, the basic principles of the new technique are also applicable to evaluate non-destructively the amount and/or depth profile of tritium retained in the solid materials such as the plasma-facing materials. Namely, BIXS is one of the promising techniques for monitoring and control of high-level tritium in the fuel circulation system and the reactor core of the fusion reactors.

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