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ノート

BIXS 法によるトリチウム分圧の測定（Ⅱ）
－混合気体の影響—

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Measurement of Tritium Partial Pressure by BIXS (II)
- Effects of Mixture Gases -

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Abstract

The bremsstrahlung X-ray counting method is one of the promising methods for in-situ measurements of high-level tritium, and the applicability of this method to a tritium handling system has been examined. In this study, pressure dependence of X-ray intensity in a high pressure region above 1 kPa was mainly examined using mixtures of H_2-, D_2-, He- and Ar-1%T_2. In a low pressure region below 1 kPa, the X-ray intensity was proportional to the partial pressure of tritium in all of the mixtures, while above that pressure it deviated downward from the proportional tendency. The deviation behavior in the mixtures of H_2-, D_2-, He-1%T_2 was similar, but that in the mixture of Ar-1%T_2 was different from that in the other three mixtures: namely, the downward deviation in the three mixtures is mainly caused by the self-absorption of β-rays in the hydrogen isotope and helium gases, whereas in the mixture of Ar-1%T_2 the emission of characteristic X-rays from argon atoms plays a major role in the deviation.

In order to safely and precisely control tritium in a fuel circulation system of the thermonuclear fusion devices, it is indispensable to measure *in-situ* the concentration of tritium in circulation gases. For this purpose, it is widely recognized that an ionization chamber is convenient as a conventional method. However, there are some disadvantageous points: for example, since the ionization current depends on the total pressure and the composition of a mixture gas containing tritium, different ionization currents appear even if the number of tritium atoms in an ionization chamber is the same. In addition to this, a small quantity of impurity such as helium strongly affects the ionization current, which is so-called Jesse's effect [1]. Helium is a main product in the D-T fusion reactions. This effect is caused by energy transfer from an excited state of the helium to a hydrogen isotope molecule, causing it to ionize, since ionization potential of a hydrogen isotope molecule is less than the excitation potential of the helium. Because the helium is the main products in the D-T fusion reactions, it is necessary to develop a new measuring method to evaluate precisely the partial pressure of the tritium species in a mixture gas.

Recently, a bremsstrahlung X-ray counting method has been proposed by Matsuyama et al. [2-3], and it is known as one of the promising methods for *in-situ* measurements of gaseous tritium. This method is based on utilization of the bremsstrahlung and characteristic X-rays emitted by the interactions between β-rays and substances. A schematic drawing of the proposed measuring cell is shown in Fig. 1. The present measuring cell was specially designed and manufactured to measure *in-situ* high-level tritium. The conversion from tritium β-rays to X-rays mainly takes place on the surface of an Au/Be plate equipped with the measuring cell as a radiation window.

Inner surfaces of the measuring cell except two Cu gaskets are coated with a thin gold film. The X-rays are mainly generated on the gold surfaces bombarded with β-rays. A part of X-rays generated penetrates through a radiation window material which was made of a thin beryllium disk (150 μm) coated with a thin gold film (described as Au/Be). The penetrated X-rays impinge on the head of an X-ray detector and then those are detected. Namely, the structure of the measuring cell is very simple and the detection processes of the X-rays are not complicated. These are advantageous points of the present method. The X-rays produced on the surface of the Au/Be disk are major part of the detected X-rays.

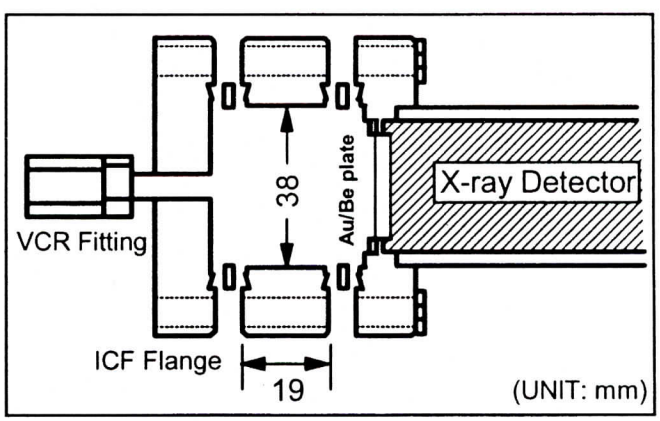

Fig. 1 Cross-sectional view of the measuring cell for gaseous tritium.

In the previous report [4], total pressure dependence of X-ray intensity was examined using tritium species such as elemental T_2 and the mixtures

of D_2-T_2,and H_2-T_2. As a result, it was found that the X-ray intensity was proportional to the total pressure below a few kPa for all of the gases, while above this pressure downward deviation from the linear relation appeared. Quite similar phenomenon was also observed for the mixture of He-T_2 [3]. The deviation from the linear relation was attributed to the self-absorption of β-rays in a gas phase of the measuring cell. The energy loss of tritium β-rays in various gases strongly depends not only on the total pressure but also on atomic number of gaseous species. The absorption coefficients [5-6] of β-rays for several gases are shown along with the average *W*-values [7] in Table 1. It is considered that the similar phenomenon to the deviation mentioned above is due to a reason that the absorption coefficient for He is almost equivalent to that for H_2.

Table 1. Absorption coefficient (μ) of β-rays and average *W*-value for various gases.

Gas	μ (cm^{-1})	*W*-value (eV/ion pair)
H_2	1.8	36.6 ± 0.3
He	1.7	41.5 ± 0.4
Ne	7.6	36.2 ± 0.4
N_2	11.0	34.6 ± 0.3
Ar	12.9	26.2 ± 0.2
CH_4	----	27.3 ± 0.3
H_2O	8.0	30.1 ± 0.3

To examine in detail the effects of a mixed gas in the tritium mixtures on the bremsstrahlung X-ray counting, a mixture gas of Ar-T_2 was prepared and total pressure dependence of the mixture was measured at an ambient temperature. Tritium concentration in the prepared mixture was nominally 1 atomic percent (hereafter described as Ar-1%T_2 mixture). Total pressure of the mixture was measured using a capacitance manometer equipped with two sensors of 1.3 kPa and 0.13 MPa, and the X-ray intensity was measured using a specially designed NaI(Tl) scintillation counter. The X-ray intensity was repeatedly measured 5 times for 1 minute in each pressure and the average value was calculated. Background level was 443 counts/s, which was mainly due to the adsorption of tritiated water vapor on the surface of the Au/Be disk.

Figure 2 shows the total pressure dependence of X-ray intensity for the Ar-1%T_2 mixture. The solid lines in the figure are an eye guide. Two kinds of linear relation were observed below/above about 2 kPa. In

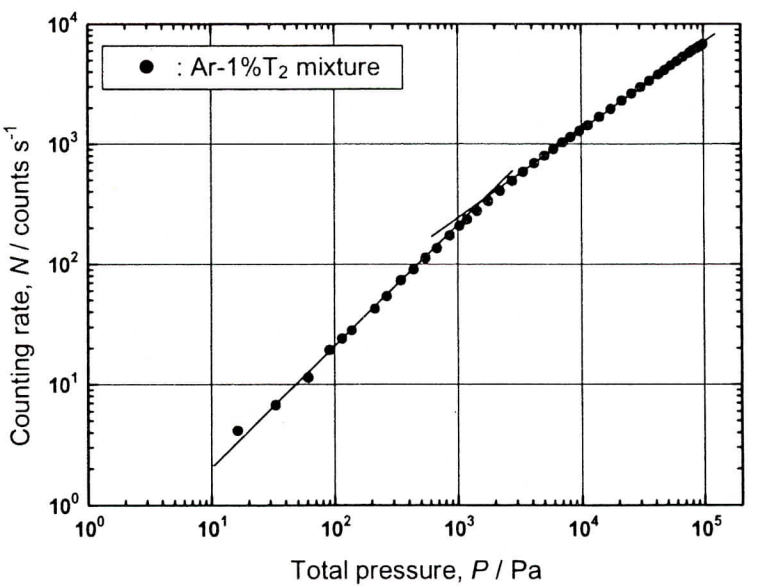

Fig. 2 Pressure dependence of X-ray intensity for the Ar-1%T_2 mixture.

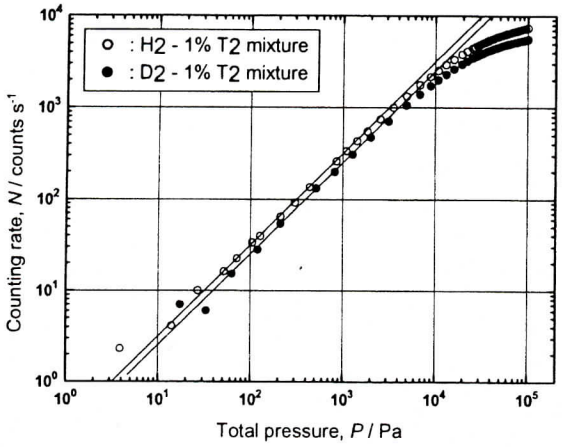

Fig. 3 Pressure dependence of X-ray intensity for the H_2- and D_2-1%T_2 mixtures.

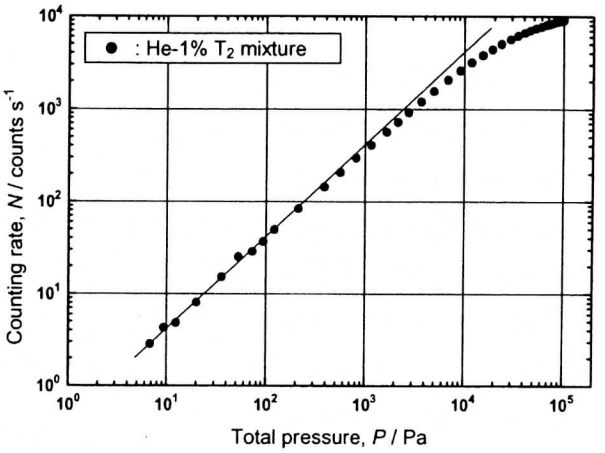

Fig. 4 Pressure dependence of X-ray intensity for the He-1%T_2 mixture.

the lower pressure region, ramp of the line was unity, while it was 0.73 in the higher pressure region. Namely, such a different pressure dependence was also observed for the mixtures of H_2- and D_2-1%T_2 and He-1%T_2 as shown in Figs. 3 and 4, respectively.

However, there was a large difference in the higher pressure region between the Ar-1%T_2 mixture and other mixtures. Above a pressure of about 2 kPa, a linear relation appeared in the Ar-1%T_2 mixture, although the ramp of the line was different from that in the lower pressure region. On the other hand, the convex changes were observed for pure tritium gas and the mixtures of H_2- and D_2-1%T_2 and He-1%T_2 as shown in Figs. 3 and 4. Namely, this indicates that the linear relation observed in the higher pressure region of Ar-1%T_2 can not attribute to only the self-absorption of β-rays in the gas phase. If the linear relation at the higher pressure region is mainly due to the self-absorption of β-rays like other mixtures, a convex deviation should be observed since the absorption coefficient of β-rays for argon is seven times greater than that for hydrogen and helium as shown in Table 1. Therefore, we have to consider contribution of other interactions in the Ar-1%T_2 mixture.

It is well known that the characteristic X-rays are emitted by the interactions between β-rays and substances. In the case of argon atoms as a substance, three characteristic X-rays ($K_{\alpha 1}$=2.958, $K_{\alpha 2}$=2.956 and $K_{\beta 1,3}$=3.190 keV) can be produced by colliding with β-rays emitted from tritium atoms, because the maximum energy of the β-rays is 18.6 keV. For example, the total number of characteristic X-ray photons produced in the measuring cell is 7.68×10^6 s^{-1}, when the total pressure of the Ar-1%T_2 mixture is 0.1 MPa. Off course, this depends on the total pressure of the mixture. The conversion coefficient of β-rays to the X-rays was assumed to be 3.2×10^{-4} photons/β-particle [8]. About 10% of the produced characteristic X-rays hit on the surface of the Au/Be disk and penetrate through it. Intensity of the characteristic X-rays is reduced to 15% after penetrating through it. Finally, 1×10^5 of photons shall enter the detector, which are large enough in comparison with the

bremsstrahlung X-rays. In this calculation, effects of the scattering and absorption of X-rays in the measuring cell are not taken into account of the calculation.

It is clear, however, that the intensity of characteristic X-rays increases with increasing a partial pressure of argon. If generation of the characteristic X-rays could be negligibly small, the observed X-ray intensity should be largely deviated downward. That is, the deviation based on self-absorption of β-rays as shown in Figs. 3 and 4 will be similarly observed. It is suggested, therefore, that the effects of generation of the characteristic X-rays appeared in the mixture of Ar-1%T_2, whereas the self-absorption in cases of the mixtures of hydrogen isotopes and helium was mainly caused by excitation and/or ionization of gases without emission of X-rays. Accordingly, it is necessary to correct previously the effects of X-ray emission by the impurity molecules if the total pressure in the measuring cell is above about 1 kPa. In addition to this, further detailed analyses of the pressure dependence will be required to clarify the deviation behavior in a pressure region around 2 kPa.

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