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論　文

BIXS 法によるトリチウム分圧の測定（Ⅰ）
－水素同位体の全圧依存性－

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Measurement of Tritium Partial Pressure by BIXS (I)
- Total Pressure Dependence of Hydrogen Isotopes -

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ABSTRACT

The correlation between the intensity of X-rays induced by β-rays of tritium and the total pressure of hydrogen isotope mixtures was examined in a pressure range from 10^{-1} to 10^{5} Pa by using three different tritium gases, i.e., pure T_2, D_2-1%T_2 and H_2-1%T_2 mixtures. It was found that linear pressure dependence was obtained up to a few kPa for all of the measured gases by taking dead time of the X-ray detector into account. Above this pressure, however, downward deviation from the linear relation appeared. It was revealed that the effect of self-absorption of β-rays on the pressure dependence was not negligibly small, because the ratios of the counting rates corrected by dead time to total pressure, i.e., specific counting rates, could not be reproduced by a simple exponential function using the absorption coefficients of hydrogen isotopes as variables. The results suggested that an additional factor such as contribution of an X-ray formation on the surfaces of a measuring cell must be taken into consideration in changing in the specific counting rates.

1. Introduction

From a viewpoint of tritium safety and economy, quantitative measurements of high level tritium are indispensable for the tritium plant of a large D-T fusion facility such as ITER and future fusion devices. For this purpose, a variety of the measuring methods based on different principles have been developed so far: for example, a small ionization chamber [1], laser Raman spectrometer [2], mass spectrometer [3], and so on. Although these techniques are promising, there are some problems. The ionization current is influenced by changes in the total pressure and composition of the measuring gases, the lower detection limit of laser Raman spectrometer is not sufficient for the required detection limit, and the mass spectrometer consumes a part of the measuring gas during measurement.

On the other hand, Matsuyama et al. recently proposed β-ray-induced X-ray spectrometry (BIXS) for measurements of tritium partial pressure in tritium-containing gases [4]. This technique is based on detection of X-rays induced by interactions between materials and β-rays emitted from gaseous tritium in a specially designed cell. Intensity of the observed X-rays was proportional to the partial pressure of tritium in the cell, when the total pressure was lower than about 2 kPa. Above this pressure, the downward deviation from the linear relation increased with increasing total pressure because of self-absorption of β-rays in measuring gases. To apply the present technique in a wide range of tritium partial pressure, it is necessary to examine the pressure dependence of the self-absorption in a pressure region above a few kPa. In addition, effects of hydrogen isotope species in a measuring gas on the self-absorption are also important, because of the difference in the ionization energy among the isotope species.

From these viewpoints, correlation between the intensity of β-ray-induced X-rays and the total pressure of hydrogen isotopes has been examined using three different tritium gases i.e., pure tritium, deuterium-diluted tritium, and hydrogen-diluted tritium.

2. Experimental

Figure 1 shows a schematic diagram (A) and a photograph (B) of the measuring cell used in this study. The measuring cell consisted of a double-sided flange, two Conflat flanges (ICF with 70 mm in diameter), a radiation window made of thin beryllium plate, and a metal bellows valve. All the flanges were made of stainless steel, and they were connected using copper gaskets. The inner surface of the radiation window as

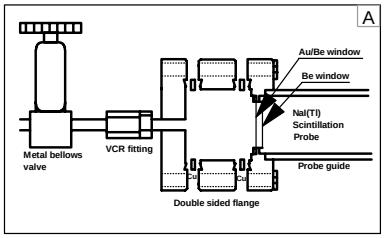

Fig. 1 Cross-sectional view (A) and photograph (B) of the measuring cell.

well as flanges was coated with thin gold film in order to enhance the conversion of β-rays to X-rays and to impede ad/absorption of gaseous tritium on/in construction materials of the measuring cell.

Internal volume of the measuring cell was about 35 cm^3. One of the two Conflat flanges was connected to a metal bellows valve for supply and recovery of tritium, and the other was used to measure X-rays by a conventional NaI(Tl) scintillation probe. Total pressure of hydrogen isotopes in the measuring cell was measured using two capacitance manometers with different sensitivities.

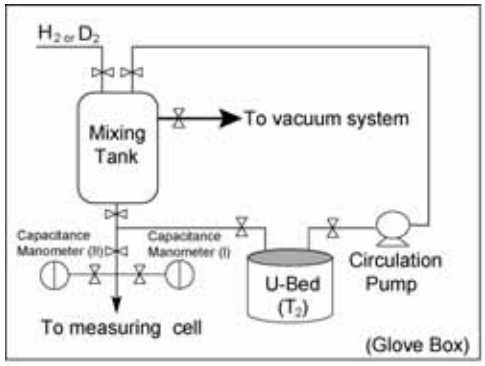

Fig. 2 Schematic diagram of the device used for preparation of tritium mixtures.

Fig. 2 shows the preparation device of different tritium gases, which consists of a mixing tank, U-bed, circulation pump and was connected with the metal bellows valve of the measuring cell. Both the measuring cell and preparation device except a radiation counter (electronics such as high voltage, scaler, and timer) were installed in a glove box to ensure safety. The glove box is constructed at Tritium Engineering Laboratory at Japan Atomic Energy Research Institute.

In the present study three different tritium gases were used: high purity tritium (the tritium concentration was determined as 99.2% by gas chromatography) and about 1% tritium gases diluted with deuterium or hydrogen. These hydrogen isotope mixtures were prepared by supplying a given amount of high purity tritium from the uranium bed to the mixing tank. In the measurements of high purity tritium, it was directly introduced into the measuring cell. Pressure dependence of the X-ray intensity was measured by reducing stepwise the total pressure of T_2 and mixture gasses from about 100 kPa to 0.1 Pa. Measuring time was varied depending on the counting rate of X-rays at each pressure.

3. Results and discussion

It has been already reported that a linear relation between the intensity of X-rays induced by β-rays and the total pressure of tritium-containing gas appears in a pressure region below about 2 kPa [5]. The results obtained by the present examination are illustrated by three symbols in Fig. 3. Although three solid lines having a unit ramp

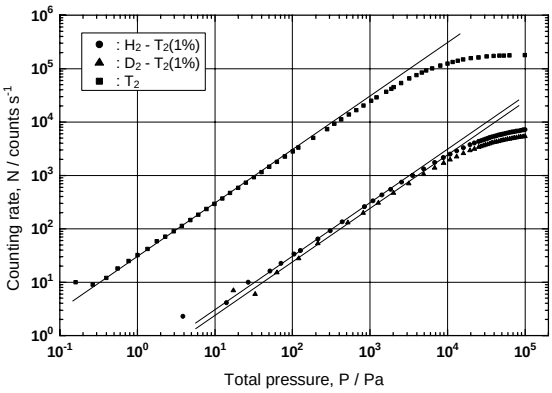

Fig. 3 Total pressure dependence of the counting rates for T_2, D_2-1%T_2 and H_2-1%T_2 mixtures.

are shown in the figure, they are only eye guides. As clearly seen, the counting rates for the high purity tritium were about 100 times greater than those for H_2-T_2 and D_2-T_2 mixtures. This difference is mainly based on a large difference in the partial pressures of tritium among those gases. On the other hand, the counting rates for H_2-T_2 and D_2-T_2 mixtures were proportional well to their total pressures up to about 4 kPa, whereas for the high purity tritium lower deviation from the linear relation appeared above a pressure of about 0.5 kPa. There are two plausible reasons for the downward deviation: one is the effect of dead time of the present detector, and the other is the effect of self-absorption of β-rays in the measuring gases. On the other hand, below 10 counts/s the counting rates scattered around the line. It is considered that this is mainly due to significant increase in the background level caused by adsorption of tritium oxides on the inner surface of the measuring cell.

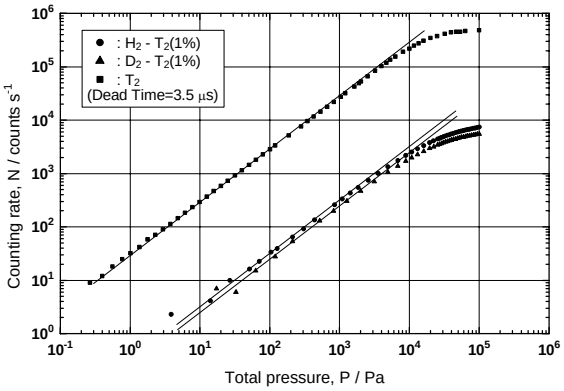

Fig. 4 Total pressure dependence of the counting rates corrected by dead time of the X-ray detector.

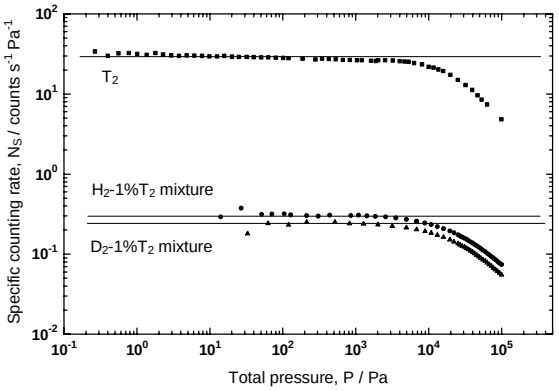

Fig. 5 Total pressure dependence of the specific counting rates for T_2, D_2-1%T_2 and H_2-1%T_2 mixtures.

The dead time of the present radiation counter was 3.5 μs. When τ is the dead time of the counting system and N(obs) the observed counting rate, the unconcerned fraction in N(obs) counts is τN(obs). If N(true) is the true counting rate, the number of counts lost is τN(obs) N(true). Therefore,

$$N(true)=N(obs) + \tau N(obs)\, N(true) . \qquad \text{———— (1)}$$

Figure 4 shows the results calculated by Eq. (1). The dead-time correction appeared above 10^4 counts/s. Namely, the linearity of the counting rates for the high purity tritium was improved in a pressure region higher than a few kPa.

During the present examinations, each tritium concentration in three measured gases was kept constant at a whole pressure region. Specific counting rate (Ns), i.e., the ratio of the corrected counting rate by Eq. (1) to total pressure, should be constant, if the self-absorption of β-rays in a measuring gas is negligibly small. To confirm this effect, the specific counting rates (closed symbols) were plotted against the total pressure as shown in Fig. 5. At a pressure region above around a few kPa, the specific counting rate for each gas became lower than the respective constant value with increasing total pressure. This indicates clearly that the

self-absorption of β-rays in the gas phase is not negligible.

Supposing that the effect of self-absorption can be represented by a simple exponential function, the specific counting rate can be described by the following equation as a first approximation:

$$Ns = a_0 \{1 - \exp(-\mu P)\} / \mu P, \quad \text{----- (2)}$$

where a_0 is the true counting rate per unit pressure, μ the absorption coefficient of tritium β-rays for each gas, and P the total pressure. Figure 6 compares the observed specific counting rate with the estimated values by Eq. (2) for the H_2-1%T_2 mixture as an example (broken line). In this calculation, the absorption coefficient for pure hydrogen gas (1.8 cm^{-1} at 101.3 kPa [Ref]) was employed, because the tritium concentration was quite low. The broken line can be represented by the following equation:

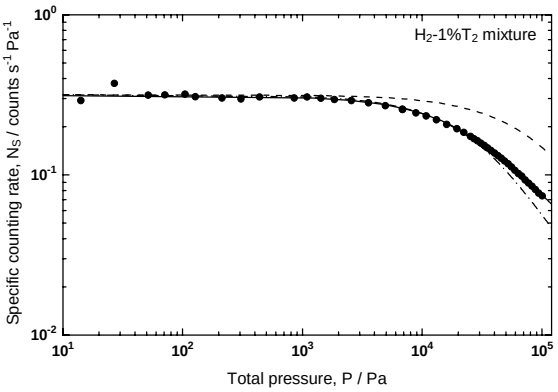

Fig. 6 Comparison between the specific counting rates and those calculated by Eq. (2) for the H_2-1%T_2 mixture. Broken line: μ=1.8 cm^{-1}, dotted and broken line: μ=5.5 cm^{-1}.

$$Ns = 0.317\{1-\exp(-1.8P/101300)\}/(1.8P/101300) \quad \text{------(3)}$$

Disagreement between the observed specific counting rate and the broken line appeared in a pressure region above 1 kPa. Even though the different absorption coefficients were applied to Eq. (3), as shown by the dotted and broken line (μ=5.5 cm^{-1}), they did not agree well in a high-pressure region. This indicates that the major change in the specific counting rates does not solely arise the self-absorption of β-rays in the gas phase.

A certain fraction of β-rays emitted in the measuring cell strikes on the gold surfaces on both the beryllium window and stainless steel constructing the measuring cell, and then contributes X-ray emission. A part of such X-rays can penetrate through the beryllium plate. Namely, it is considered that the observed counting rate include contribution of X-rays produced at the gold surfaces of stainless steel as well as beryllium. The intensity of the former X-rays may also increase with increasing total pressure, but a linear increase in the X-ray intensity can not be expected owing to the self-absorption of β-rays. It is hard to evaluate quantitatively the contribution of such X-rays to the observed counting rate, because a complex structural factor has to be taken into account.

The solid line in the Fig. 6 shows a fitting curve of the specific counting rate. The difference (A) between the broken and the solid lines could be described as

$$A = 0.496 + 0.489\exp(-3.84P/101300). \quad \text{----- (4)}$$

Namely, the pressure dependence of the counting rate for H_2-1%T_2 mixture gas can be written by the following equation.

(1) H_2-1%T_2 mixture gas:

Ns=0.317{1-exp(-1.8P/101300)}/(1.8P/101300){0.496+0.489exp(-3.84P/101300)}

In a similar manner, the pressure dependence of specific counting rates for the D_2-1%T_2 mixture gas and the T_2 gas were described by the following equations:

(2) D_2-1%T_2 mixture gas;

Ns=0.248{1-exp(-2.1P/101300)}/(2.1P/101300){0.524+0.471exp(-3.85P/101300)} ,

(3) T_2 gas;

Ns=29.6{1-exp(-2.2P/101300)}/(2.2P/101300){0.344+0.626exp(-2.67P/101300)} .

The specific counting rates for three hydrogen isotopes were finally reproduced well using above equations as shown in Fig. 7. The first numerical values in the three equations mentioned above depend on the tritium concentration in the measuring gases. Since the chemical purity of T_2 gas determined by gas chromatography was 99.2%, those of the H_2-1%T_2 and D_2-1%T_2 mixtures used in this study were evaluated as 1.06 and 0.83%, respectively. In addition, the absorption coefficients for pure D_2 and T_2 gases estimated to be 2.1 and 2.2, respectively.

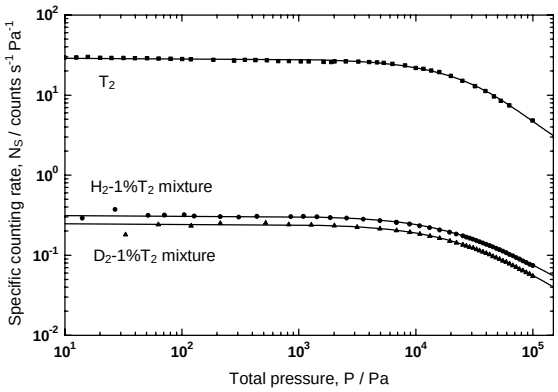

Fig. 7 Comparison between the specific counting rates and the reproducing equations.

Absorption coefficient of β-rays, in general, depends on some factors such as electron density in a given volume, energy of β-rays, and average excitation energy of each hydrogen isotope species. The average excitation energy includes excitation and ionization energies of each hydrogen isotope species. In the present case, the former two factors are the same, while the last one may be different among H_2, D_2, and T_2 molecules, because there is a little difference in ionization energy among them, i.e., H_2: 15.43, D_2: 15.47 and T_2: 15.49 eV [6].

4. Conclusions

Total pressure dependence of the X-ray intensities induced by β-rays of three different tritium gases such as pure T_2, D_2-1%T_2 and H_2-1%T_2 mixtures was examined. The results obtained in this study were as follows:

(1) Linear relations between the intensity of X-rays induced by β-rays of tritium and the total pressure of hydrogen isotope mixtures were obtained up to a few kPa for all the measured gases by taking the dead time of the X-ray counting into account.

(2) Above this pressure, however, downward deviation from the linear relation still appeared

even though the observed counting rates were corrected by self-absorption of β-rays in gas phase.

(3) Changes in the observed counting rates with total pressure were finally reproduced by taking a structural factor in addition to two factors of the dead time and self-absorption into account.

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