

§6. Study on Measurement Technique of Atmospheric Tritium for Different Chemical Forms

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We have developed the atmospheric tritium sampling apparatus applicable to tritium levels in the general environment. It collects successively 3 different chemical forms in air that are water vapor (HTO), hydrogen (HT) and hydrocarbon (CH₃T). In this paper we report an improved atmospheric tritium sampling apparatus and recent tritium level at Kumamoto obtained by the developed apparatus.

Water vapor is at first collected on molecular sieve 3A (MS-3A) in a HTO column, which is made of glass, 50 cm in length and 5 cm in diameter and 500 g of MS-3A is packed. The water vapor content in air varies with season; in summer it sometimes exceeds the MS-3A adsorption capacity packed in the HTO column. Then a HTO cold trap, being made of glass and cooled at 2°C, is located just before the HTO column to avoid breakthrough of the HTO column. Hydrogen is converted to water by catalytic oxidation and collected as water on MS-3A in a HT column. To increase the amount of water subjected to activity measurement, tritium free H₂ that was generated by electrolytic decomposition of deep well water is added to the sampling air after the HTO column. The H₂ generation rate is controlled to obtain about 15 ml of water with oxidation of H₂. We prepared Pt honeycomb catalysts for H₂ oxidation, in which 0.12 g of Pt is dispersed on the surface of the honeycomb base metal (25 mm in diameter, 40 mm in length). The apparatus uses 3 pieces set of the Pt honeycomb catalyst at 100°C. Hydrocarbons are oxidized to water by Pd catalyst (DASH-220D, 0.24 wt% Pd, 3mm in diameter, NECHEMCAT Co. Lt.) at 300°C. Tritium free bomb CH₄ is added to the sampling air through a mass flow controller before the Pd catalyst column. The Pd catalyst column is made of quartz and 150 cm³ Pd catalyst is packed. The flow rate of CH₄ is controlled to obtain about total 15 ml of water by the sampling.

Waters adsorbed on the MS are desorbed by flowing N₂ gas at 450°C and recovered on a cold trap. Tritium activity is measured by low background liquid scintillation counting after mixing the recovered water with liquid scintillation cocktail (Pico Flow LLT, PerkinElmer) in Teflon vial.

The oxidation of H₂ and CH₄ by the Pt honeycomb catalyst is presented in Fig. 1, showing satisfactory oxidation yields for 4000-ppm H₂ in the temperature range from 30°C to 100°C both the sampling air rate of 1.2 L min⁻¹ and 4.8 L min⁻¹. The oxidation yields of CH₄ by the Pt honeycomb catalyst are below the detection limit of the gas chromatography in that temperature range, suggesting selective oxidation of H₂ and HT in the sampling air. The oxidation yields of 2000-ppm CH₄ by the Pd catalyst are shown in Fig. 2. A sharp increase in oxidation yield above 200°C and almost 100% oxidation yield is achieved at 300°C. Based on above results the Pt honeycomb catalyst is

decided to operate at 100°C and the Pd catalyst at 300°C, respectively.

To confirm the performance and applicability of the present system to atmospheric tritium of the general environment, air was collected by the developed tritium sampling apparatus at the campus of the Kumamoto University, Kumamoto, Japan. The results are shown in Table 1.

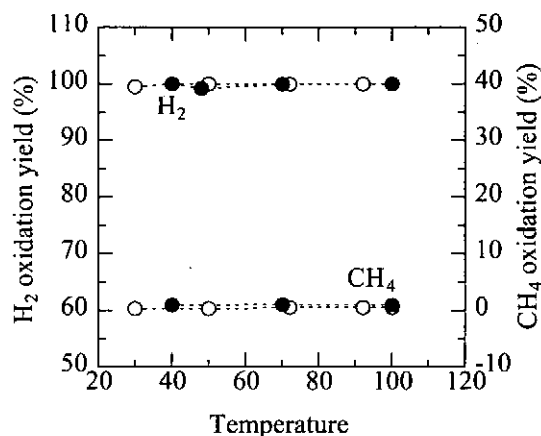


Fig. 1 Oxidation yields of 4000ppm H₂ and 2000ppm CH₄ with temperature by the Pt honeycomb catalyst. Air flow rates are 1.2 L min⁻¹(○) and 4.8 L min⁻¹(●).

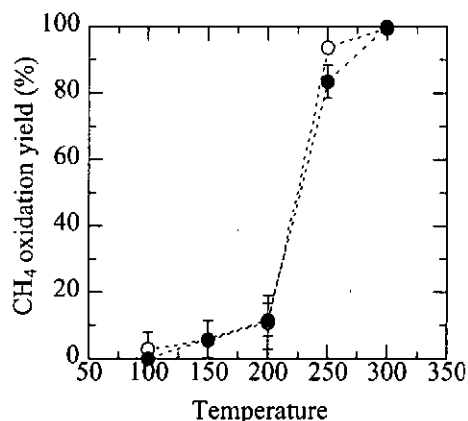


Fig. 2 Change in the oxidation yields with temperature by the Pd catalyst for 2000ppm CH₄. Air flow rates are 1.2 L min⁻¹(○) and 4.8 L min⁻¹(●).

Table 1 Atmospheric tritium concentrations of three different chemical forms at Kumamoto University.

Date (2003)	HTO (mBq m ⁻³)	HT (mBq m ⁻³)	CH ₃ T (mBq m ⁻³)
3-6 June	*ND	18.0 ± 1.6	32.3 ± 3.7
10-13 June	ND	25.4 ± 1.7	5.5 ± 3.3

*ND: below the detection limit