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

Momoshima, N. (Fac. Sci., Kumamoto Univ.)
Takamura, M. (Grad. Sch., Kumamoto Univ.)
Okai, T. (Fac. Eng., Kyushu Univ.)
Uda, T.

There are several chemical forms of tritium in the atmosphere of the general environment; those are water vapor (HTO), hydrogen (HT), and hydrocarbons (CH_3T). Measurements of tritium in these different chemical forms in the atmosphere have conducted by Okai from 1984 at Fukuoka, Japan. The observed annual average concentrations (Bq m^{-3}) are in the order of HT > HTO > CH_3T. Specific activities on these chemical species have showed quite different levels, which are highest on HT with 10^6 TU, then CH_3T with 10^4 TU and HTO with 10^1 TU. The TU is defined as atom ratio of one tritium atom to 10^{18} hydrogen atoms. The quite different specific activities speculate us different sources of these tritium species in the atmosphere.

In this study, we have designed and constructed an atmospheric tritium sampling apparatus, which is usable for environmental monitoring around feature nuclear fusion facility as well as general environment.

The schematic diagram of the developed tritium sampling apparatus is shown in Fig. 1. Atmospheric tritium is collected successively in the order of HTO, HT and finally CH_3T. All of the tritium species are adsorbed on molecular sieve 3A(MS 3A, 500g) as water molecule after converting their chemical forms.

The water content existing as water vapor in the atmosphere varies with temperature and relative humidity, in summer it sometimes exceeds the MS adsorption capacity packed in the HTO column. Then a cold trap cooled at 2 °C is located before the HTO column. Hydrogen is oxidized to water by metal catalyst. We prepared Pt honeycomb catalyst, in which 0.12 g of Pt is dispersed on surface of honeycomb base metal (25 mm in diameter, 40 mm in length). The Pt honeycomb catalysts can oxidize 4000 ppm H_2 to water completely at 100 °C, tritium free H_2 generated by electrolytic decomposition of tritium free water is added to the sampling air to ensure recovery of HT. Oxidation of CH_4 by the Pt honeycomb catalyst was examined, showing no conversion of CH_4 to water at this operation condition.

Hydrocarbons are oxidized to water by Pd catalyst (DASH-220D, 0.24wt%Pd, 3mm in diameter, 150 cm^3, NECHEMCAT) at 300°C. Tritium free bomb CH_4 is added to the sampling air as to become 2000 ppm using a mass flow controller before the Pd catalyst column.

Teflon tube and metal conjunction (Swagelock) is used for connecting each unit. Total of 5000 liter air is sampled at flow rate of 1.2 L/min (2.89 days) or 4.8 L/min (17.3 h). The performance of the catalysts is confirmed to be satisfactory for the both flow rates and no leakage is observed on the apparatus during above sampling periods, suggesting good tightness of the lines. Water adsorbed on MS is recovered by introduction of N_2 gas heated at 400 °C to the HTO, HT and CH_3T columns. About 15 ml of water is expected to recover from the HT and CH_3T column, respectively. 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.

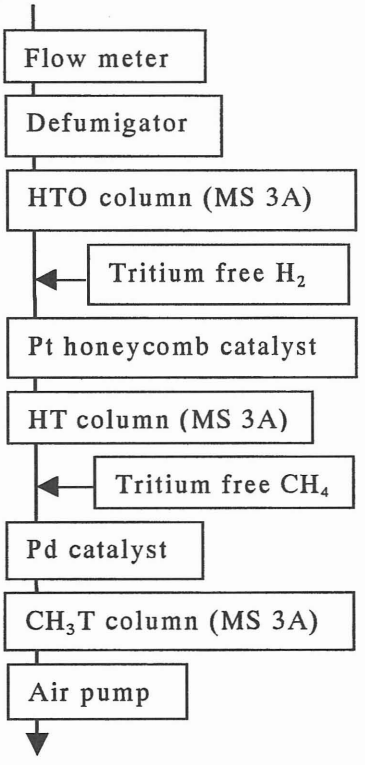

Fig. 1 Schematic diagram of atmospheric tritium sampling apparatus