§5. Recovery of Tritium from Tritiated Water Vapor Using a Proton Conductor


From the view points of the public acceptance and the safety management, it is necessary to monitor the low level of tritium in the air in a fusion test facility. In order to measure lower concentrations of tritium, a number of tritium monitors combining an ionization chamber with semipermeable polymer membranes have been researched and developed. The sensitivity of certain tritium monitors equipped with semipermeable membrane improved by an order of magnitude over that of the tritium monitor without membrane.

To further improve the sensitivity of the tritium monitor, a proportional counter with a proton conductor as the hydrogen isotope gas separator has been proposed. The proposed system would be useful not only for the elimination of unwanted interference in the proportional counter, but also for the concentration of tritium, because the proton conductor has a function as an electrochemical hydrogen isotope pump. The monitor system would offer the following advantages over the liquid scintillation counting method: reduction of waste material such as an organic solvent, ease of operation and handling, treatment in the form of gas, continuous monitoring, etc.

From the viewpoint of the practical use, we have examined the hydrogen pump of the one-end-closed tube made of CaZrO$_3$:In$_2$O$_3$ with wide electrode area as candidate materials of the proton conductor. As the results under the water vapor electrolysis condition, a maximum hydrogen evolution rate of 1.8 cm$^3$/min was obtained at 0.5 A and 973 K. The next stage of development is to use tritiated water vapor to demonstrate the electrochemical tritium pump. To use the tritiated water vapor, the experimental apparatus was transferred to ISL (Isotope Separation Laboratory) of tritium handling facility in Nagoya University and was set up as shown in Fig. 1. To supply tritiated water vapor to the anode, argon gas was passed through a water bubbler filled with tritiated water of about 1000 Bq/cm$^3$, which was immersed in a constant temperature water bath adjusted to 10°C. The concentration of tritiated water vapor was approximately 6.6x10$^7$ Bq/cm$^3$. The ratio of T and H corresponds to approximately 10$^{11}$. Dry argon gas was fed to the cathode. These flow rates were 100 cm$^3$/min. The test tube was heated up to 973 K by an electric furnace. Then, the current constant was passed through between the electrodes by a galvanostat. The gas concentrations of hydrogen in the cathode were analyzed by a gas chromatograph (GTR Tech, G2700T). Tritium gas was collected by a water bubbler system equipped with copper oxide heated to 325 °C. The sample water collected tritium gas was mixed with a scintillation solvent. The mixed solution sample was measured five times for ten minutes by a liquid scintillation counter (Aloka Ltd., LSC-5100). The distillation water was used as a background sample.

Figure 2 shows the evolution rate of hydrogen and tritium. The dashed line shows theoretical hydrogen evolution rate calculated by Faraday’s low. Evolution rate of hydrogen and tritium increased with the current. Tritium evolution rate reached 0.28 Bq/min and tritium recovery rate of 0.43 at 124 mA. It indicates that tritium gas can be extracted by the electrochemical hydrogen isotope pump using proton conductor even at very low tritium concentration. The ratio of recovery rate of tritium and hydrogen as hydrogen isotope effect was the range of 0.55 and 0.72. The ratio of recovery rate approached to 1 as the recovery rate of hydrogen increased. For application of proposed tritium monitor, it is necessary to design in consideration of the hydrogen isotope effect of recovery rate.

![Fig. 1: A photograph of hydrogen pump apparatus (left) and gas analyzing system (right) at ISL (Isotope Separation Laboratory) in Nagoya University](image)

![Fig. 2: Evolution rate of hydrogen and tritium as a function of current at 973 K](image)