## §2. Development of Tritium-in-Air Monitor Attached with Proton Conductor as Membrane Separator

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To account for and control of tritium, the detection of low level tritium is one of the key issues for tritium safety management in tritium handling facilities. Various type tritium monitors such as an ionizing chamber, a proportional counter and scintillation type detectors have been developed and utilized for radiation protection and emission control. However, tritiated water vapor in air exits with the presence of other airborne radioactive species such as Rn. Thus they disturb the detection of low level tritium in an ionization chamber. On the other hands, a proportional counter has higher sensitivity than an ionization chamber because tritium can be detected separately from other airborne radioactive species. However, the efficiency would be reduced for the detection of tritium when air is introduced, since primary electrons produced by a  $\beta$ -ray are lost by electron attachment to oxygen molecules in air. Thus, to detect such a low level tritium, the separation of disturbance species and the enrichment of tritium are an effective measure. The application of an electrochemical hydrogen pump using a proton conductor that can recover only hydrogen isotopes under elevated temperatures have been proposed as a membrane separator. We have reported the results of the system integration test under the conditions without oxygen gas. In this study, the integration test under the practical condition using room air was conducted at Isotope Separation Laboratory in Nagoya University.

The proposal tritium monitor system is described in elsewhere.<sup>1)</sup> The system mainly consists of proton conductor, metal bellows pump, commercial proportional counter LB110 (Berthold Technology) and data logger LB5320. A proton conductor  $CaZr_{0.9}In_{0.1}O_{3-\alpha}$  tube, closed at one end, was supplied by TYK Co. Ltd. The electrode area of the proton conductor was  $62 \text{ cm}^2$ . The test sample was heated to 973 K with a dual electric furnace. The electrochemical hydrogen pump was examined by applying a direct current using a galvanostat. To supply tritiated water vapor into the anode compartment, room air was passed through a water bubbler filled with tritiated water, which was immersed in a controlled temperature bath. Under these conditions, the radioactive concentration of the tritiated water vapor was approximately 1–4 MBq/m<sup>3</sup>. Dry argon gas was fed into the cathode. The flow rates were 300 cm<sup>3</sup>/min for the anode compartment and 10 cm<sup>3</sup>/min for the cathode compartment. The extracted tritium from tritium water vapor in argon gas is carried by metal bellows pump and mixed with room air and P-10 gas. Then, the tritium concentration is measured by the proportional counter. The conversion to the supplied tritium concentration from the tritium concentration measured by the proportional counter is described in Ref 1.

Figure 1 shows the evolution rate of hydrogen with and without oxygen gas in the anode compartment as a

function of applied current at 973K. The experimental conditions are shown in the figure. Although the hydrogen extraction increased with increase of the applied current in the presence of oxygen gas, the hydrogen extraction rate was low under the air atmosphere. Under the atmosphere containing oxygen, the electron-hole migrates through the proton conductor with a proton at elevated temperatures. Thus, the water vapor electrolysis efficiency under the air atmosphere is lower than that under the atmosphere without oxygen gas. Figure 2 shows the correlation between the tritium concentration introduced and the tritium concentration measured by this system. The estimated tritium concentration in this system shows good agreement with the introduced tritium concentration regardless of the atmosphere. Technically, the proposal system has reached a level at which it can be put into practical use.

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Fig. 1. Evolution rate of hydrogen with and without oxygen gas in the anode as a function of applied current at 973K.



Fig. 2. Correlation between the introduced tritium concentration and the estimated tritium concentration by the system.