Tritium separation using proton exchange membrane water electrolysis method

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1. Introduction

Recently, many activities related to tritium, including separation, recovery, or removal of tritium, have attracted attention. For example, tritium from wastewater generated by reactors must be removed to meet emission standards, and thousands of tons of diluted tritium from accidents such as the Fukushima Daiichi nuclear power plant are pending [1-3]. Tritium produced in heavy water reactors should be continuously removed to maintain neutron moderation performance and safety. Therefore, research on hydrogen isotope separation technology is significant and in high demand. In this study, a proton exchange membrane (PEM) water electrolyzer with the graphene/Nafion composite membrane was applied for the separation of various concentrations of tritium.

2. Methods and Results

2.1 Preparation of PEM water electrolyzer

Recently, single-layer graphene has been reported as a highly efficient hydrogen isotope separation membrane in electrochemical environments. Bukola et al. fabricated a membrane structure with commercial CVD graphene sandwiched between two proton exchange membranes (Nafion) and found a relatively high proton to deuterium conductivity ration of 14 [4]. In this work, we prepared a Nafion composite membrane with graphene for PEM water electrolysis application as shown in Fig. 1. And the performance of the PEM water electrolysis cell with the graphene/Nafion composite membrane was investigated using liquid water containing tritium.

2.2 Tritium separation using PEM electrolysis

The tritium separation factor was determined by the tritium ratio distributed in liquid water and generated gas during the PEM water electrolysis process. A fixed current of 7 A was supplied to the PEM cell through the power supply, and the voltage was 3.07~3.24 V. Also, 180 mL of the tritiated water as supplied through a peristatic pump at a rate of 300 rpm. During the PEM water electrolysis, produced HT gas and H2 gas was delivered to the sensing device and the concentration of tritium in the gas was analyzed through a gaseous tritium monitor. The activity of tritium in liquid water was measured using a liquid scintillation counter.

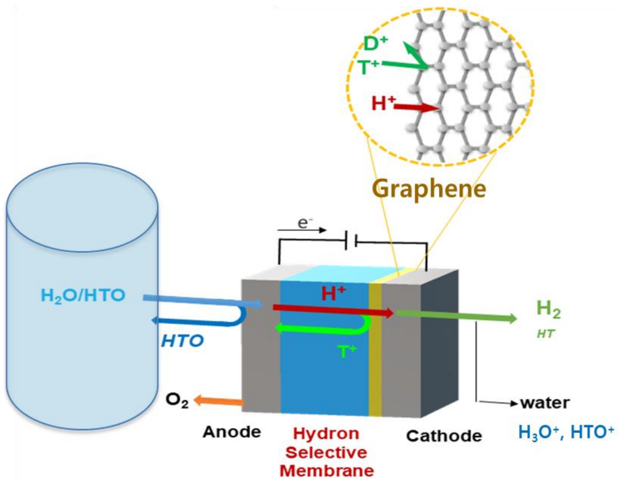

Fig. 1. Conceptual design scheme of PEM cell electrolyzer with the membrane containing graphene and electrode assembly for water electrolysis.

2.3 Results and discussion

Firstly, we compared the performance of a cell with a single Nafion membrane and a cell with a graphene/Nafion composite membrane. The cell performance was studied through a current-voltage curve, and the results found that the composite membrane cell had higher cell resistance. This finding is consistent with Bukola's work, in which the graphene layer was found to have little resistance on the HER process [4].

In the tritium separation experiment using water electrolysis, the separation factor was higher in the cell with the graphene composite membrane. In addition, experiments comparing separation factors by initial tritium concentration confirmed that the concentration of tritium in the gas phase increased after water electrolysis in proportion to the initial concentration in liquid water as shown in Table 1. It also confirmed that the tendency to maintain a relatively high H/T separation factor even at various initial tritium concentration in liquid water.

Table 1. Results of tritium separation using water electrolysis results according to initial tritium concentration.

Initial *tritium conc.* (kBq/L)	**Final *tritium conc.* (kBq/L)**	**Cathode *tritium conc.* (kBq/L)**	***Tritium conc. in g*as (kBq/m^3)**	**SF(H/T)**
3524.8	3573.7	3712.3	80.3	32.3
8841.6	8835.8	9013.4	354.7	18.4
31963.1	32361.7	33519.8	1,488	15.8

3. Conclusions

In this study, a PEM cell with a graphene/Nafion membrane was applied for the electrolytic separation of tritium. The PEM cell with the composite membrane showed a slightly higher cell resistance than that with a single Nafion membrane, but had a higher separation factor. Also, the relatively high separation factor at various initial tritium concentration confirmed the potential of graphene-based PEM cells for tritium separation.

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