Oxygen Isotope Separation using a Multi-stage Membrane Permeation Cells

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1. Introduction

The demand for and applications of stable isotopes in medicine, industry, and science in the modern era has increased and expanded significantly. Especially, ^{18}O-enriched water (> 90%) is used as a target in a cyclotron for the production of the β-emitting radioisotope ^{18}F, which is essential for PET (Positron Emission Tomography) pharmaceutical [^{18}F]-labeled 2-deoxyglucose (FDG) synthesis. Since the isotope separation processes are technically complicated and costly so as to limit the production of ^{18}O. It is essential to re-use the used target water as much as possible. In order to recycle the used target water, it is necessary to purify the organic and inorganic impurities contaminated during the ^{18}F-FDG production loop and to re-enrich the ^{18}O isotope in the target water diluted during the purification process. For the development of a compact target water ^{18}O re-enrichment system, the ^{18}O isotope separation characteristics of AGMD (Air Gap Membrane Distillation) and DCMD (Direct Contact Membrane Distillation) were investigated, and the combined process that uses advantages of AGMD and DCMD was developed. The ^{18}O isotopic water permeation and isotope separation characteristics of a hydrophobic PTFE membrane for each process were evaluated.

2. Methods and Results

Permeation fluxes were measured by weighing the collected membrane-permeated water vapor. And isotopic ratio $^{18}O/^{16}O$ of each water sample was analyzed by a Tunable Diode Laser Absorption Spectroscopy (TDLAS). We observed the effects of the temperature gradient applied to the membrane surfaces on the vapor permeation flux and the oxygen isotope separation. For all MD processes, the permeation flux and the degree of ^{18}O separation increased as the membrane interfacial temperature gradient increased. Since the oxygen isotope separation and the permeation flux for the DCMD is usually 10 times higher than the AGMD with same isotope separation characteristics, the AGMD and DCMD combined process should be more efficient from the system's operational point of view. For a continuous enrichment of the ^{18}O isotopic water, multi-stage membrane cell system was tested.

2.1 TDLAS system for Isotope Analysis

Littman external cavity tunable diode laser (Sacher, Model TEC500-1380) producing wavelengths centered at 1.392 μm with a power of ~ 3 mW was used. Two multi-pass cells with 10 m path length for the reference and measured samples were used for analysis. Two lock-in-amplifiers (Stanford Research Systems, Model SR850) were employed to increase the S/N ratio (~ 1000) of the 1st harmonic absorption signals. Peak-to-peak ratios of each isotopic 1st harmonic signal in the samples were compared to determine the change of the ^{18}O concentrations.

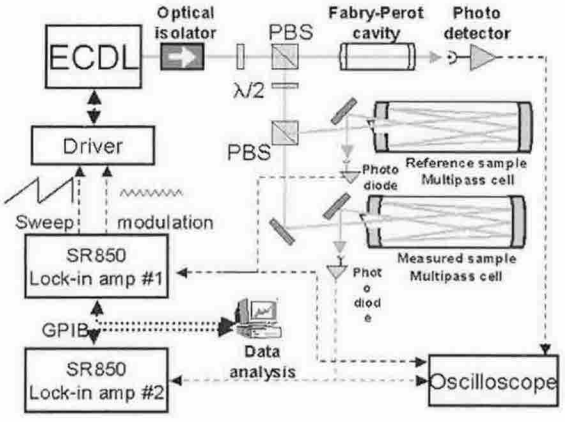

Fig 1. TDLAS system for analysis of the oxygen isotopic concentration.

2.2 Multi-stage Permeation Cell System

Fig. 2 and Fig. 3 show a design of the AGMD and DCMA combined permeation cell and the multi-stage permeation cell system. The hydrophobic PTFE porous membrane with the diameter 14.2 cm in the permeation cell was supported by a stainless steel grid. A membrane with the average pore diameter of 0.2 μm, thickness ~ 150 μm, porosity ~ 70%, and tortuosity factor ~ 2 was used. To maintain the temperature of the process water (50°C) in the permeation cell, a 2.0 mm thick and ~ 20 W Kapton heater (maximum heating capacity is ~ 150°C) was installed in the upper part of the cell. Temperature was controlled by using the PID control system. As shown in the Fig. 3, 6 membrane permeation cells were connected in series. To apply the membrane interfacial temperature gradient to the membrane surface, cold water was circulated through the lower part of the cell using a cooler. Temperature difference between the retentate and the cooling fluid (water) was maintained at $\Delta T \sim 30^{\circ}C$ except the cell #2 in the system.

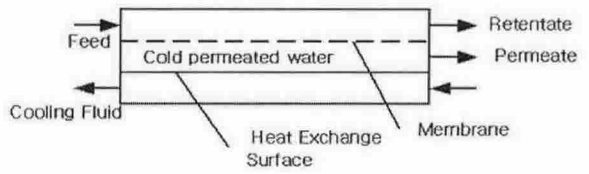

Fig. 2. A diagram of the AGMD and DCMD combined process.

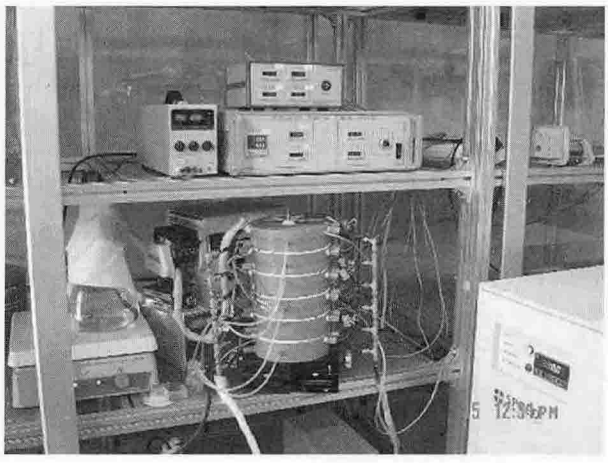

Fig. 3. Multi-stage permeation cell system for oxygen isotope separation.

The feed water was 3 liter at the beginning and flowed with 16 mL/min flow rate through the system using a peristaltic pump. The product cut was determined by weighing the mass of the retentate and was 85%. The separation coefficient was measured for the permeated water of each cell. To compare the effect of temperature gradient applied to the membrane surface, cold water was not circulated for cell #2.

2.3 Enrichment coefficients β for the system

The separation coefficient α for each cell was determined by comparing the isotopic ratio $^{18}O/^{16}O$ in the initial feed and the permeated water in the permeated cell. As shown in the Table 1, the separation coefficients decreased as the cell number increased due to the increase of ^{18}O concentration in the retentate as the cell number increased. Overall enrichment factor for the system was 1.006.

Cell number	1	2	3	4	5	6
α	1.016	1.004	1.004	1.016	1.012	1.007

Table 1. Oxygen isotope separation coefficients for the permeation cell in the multi-stage system.

3. Conclusion

We developed the much higher permeation flux system using the AGMD and DCMD combined process without losing the isotope separation characteristics. There might be some other factors developed to build an efficient system for oxygen isotope re-enrichment of the used cyclotron water target using this design. These include the efficient water reflux and energy expenditure, and temperature control.

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