Development of the pressure driven AGMD process applicable to a multi-stage oxygen isotope separation system

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1. Introduction

Membrane distillation (MD) has been developed especially for desalination of seawater since 1960's. MD process appears to be more useful for removal of salt compared to the conventional reverse osmosis (RO) process. However, it is still under the development stage due to its high energy expenditure for generating thermal gradient to membrane interface compared to the RO process. Nevertheless, its compactness, high separation factors and durability of materials are still attractive to the researchers and engineers searching for the advanced separation process. In addition to MD application to desalination, isotope separation of the light isotopes such as oxygen and hydrogen isotopes contained in water has been investigated for a decade since Chmielewski firstly showed its usefulness for isotope separation in early 90's.^{1,2} AGMD (Air Gap Membrane Distillation) of a single permeation cell showed separation of oxygen isotope with the degree of $1.01 \sim 1.03$. In practice, it is necessary to build a multi-stage MD system to enrich isotopes. Permeation fluxes and the degree of oxygen isotope separation of AGMD and VEMD (Vacuum Enhanced Membrane Distillation) were explored.³⁻⁴ VEMD shows slightly higher isotopic separation degree with higher permeation flux compared to AGMD. It is however virtually impossible to build a multi-stage system due to its system complexity. Although AGMD is suitable for constructing a multi-stage cascade system, permeation flux for AGMD is still too low to be applied to an operational production system. In this investigation, we increased permeation flux using the pressure driven AGMD process, which is different from VEMD, while isotope selectivity was also increased. Permeation flux and the degree of isotope separation of the pressure driven AGMD process were measured by using a multi-stage system.

2. Methods and Results

2.1 Pressure Driven AGMD process

Fig. 1 shows the fundamental scheme of the pressure driven AGMD process. Water vapor is permeated by driving force generated by the temperature gradient and pressure gradient applied to the membrane interface in this scheme. In general, VEMD uses vacuum pump to permeate the vapor without cold heat exchange plate as shown in Fig. 1. For the pressure driven AGMD process, however, air in the gap is evacuated to ~ 20 torr by the peristaltic pump, and cold surface condensed water is sucked out to the outlet port and is fed to the previous stage directly by the peristaltic pump for the feed reflux when a multi-stage system is used. The hydrophobic PTFE porous membrane with the effective diameter 12.6 cm (effective area ~ 125 cm²) was used. Permeation flux was increased about two-folds and $\sim 40\%$ compared to the AGMD process only and AGMD-DCMD combined process respectively as shown in Table 1.



Figure 1. Conceptual diagram of pressure driven AGMD process and VEMD process.

Table I.	Perme	ation flux	comparise	on be	etween A	AGMD,
AGMD-D	CMD	combined	process,	and	Presure	driven
AGMD pi	rocess.					

Operational conditions: $\Delta T = 35 \ ^{\circ}C (45 \ ^{\circ}C - 10 \ ^{\circ}C) / 10 \ ^{\circ}C$					
feed flow rate = 10 mL/min					
AGMD process	$\sim 1.7 \text{ L/hr m}^2$				
(1 mm air gap)					
AGMD-DCMD					
combined process	$\sim 2.7 \text{ L/hr m}^2$				
(1 mm air gap)					
Pressure Driven AGMD	2.9 J/hrm^2				
(1 mm air gap)	~ 3.8 L/III III				

Fig. 2 shows O-18 isotope selectivity expressed by the isotope enrichment factor, $\beta = (x/(1-x))_{\text{product}}/(1/(1-x))_{\text{feed}} = 1.0127$, which was increased about 30% compared to AGMD process.⁴



Figure 2. O-18 isotope enrichment for the pressure driven AGMD process.

2.2 Multi-stage membrane cascade system

Using the pressure driven AGMD process, a multi-stage cascade system was designed and built as shown in Fig. 3. To maintain temperature of the hot feed water, a ceramic heater with ~ 45 W power was installed in each cell and the feed temperature was then controlled by using the PID control system. Permeation fluxes were measured by weighing the collected membrane-permeated water. Isotopic selectivity was analyzed by a Tunable Diode Laser Absorption Spectroscopy.⁵



Figure 3. Schematic design of a multi-stage pressure driven AGMD system.

2.3 Stage cut and stage enrichment coefficient

The system operational conditions and the results are shown in the Table 2. Degree of isotope separation was expressed by enrichment factor which is the isotopic ratio ¹⁸O/¹⁶O of the retentate and the initial feed. The product cut was maintained at 85% during the experiment.

Table 2. Operational conditions and experimental results for the pressure driven AGMD multi-stage stage cascade system

Experimental conditions	Parameters			
Feed temperature	$40^{\circ}C$			
Temperature gradient	$\Delta T = 30 \ ^{o}C$			
Feed flow rate	5 mL/min			
Product flow rate	4.25 mL/miin			
Tail flow rate	0.75 mL/min			
Enrichment factor	1.0127			
Product cut (θ)	85%			

3. Conclusion

The pressure driven AGMD multi-stage system was designed and constructed for oxygen isotope separation. Both permeation flux and isotope selectivity were increased dramatically. In this investigation, we found that the pressure driven AGMD process might be highly applicable to oxygen isotope production pilot module with economic viability.

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