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Thermally enhanced membrane gas separation

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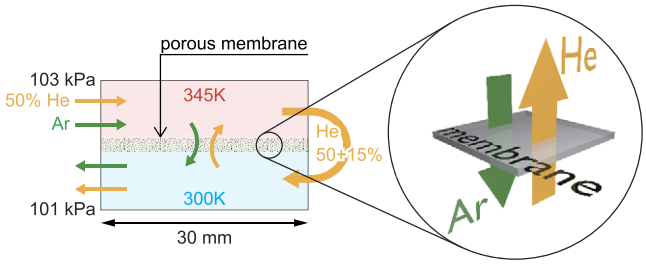
HIGHLIGHTS

- Novel gas separation with temperature and pressure differences across the membrane.
- Molecular exchange flow by the thermal transpiration in the porous membrane.
- Low cost membrane is available for gas separation.
- 15% gain of mole percentage by a membrane of 3 × 3 cm size with 45 K temperature difference.

GRAPHICAL ABSTRACT



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ABSTRACT

An enhancement of membrane gas separation, which makes use of the temperature difference in addition to the pressure difference across the porous membrane, is proposed. The thermal transpiration flow through the membrane induced by the temperature difference enables control of two physical quantities of the permeate flow, since the temperature difference is controllable separately from the pressure difference. It is possible to establish the molecular exchange state of the permeate flow, where a component gas of binary mixture flows in the opposite direction from the flows of the other component. A model gas separation device, which makes use of the counter flow arrangement to accumulate the molecular exchange of the micro-channels in the membrane, is devised using a single mixed cellulose ester membrane of 110 μm thickness and 3 × 3 cm size. The device induces 15% variation of mole percentage of a helium–argon mixture by a temperature difference of 45 K when the speed of the counter flow is around 50 cm/s. The performance of the device is well represented by numerical simulations that use the results of molecular gas dynamics for mixture flow in micro-channels.

1. Introduction

Gas separation by membranes is an attractive method, because of its simple device structure, easy operation, and high energy efficiency. A great number of research efforts have been carried out for the fabrication of separation units and membranes with appropriate macroscopic and microscopic structures, including the design of molecules of the membrane [1–4]. These various implementations of membrane separation have a common, simple principle. The feed is supplied to one side of the membrane at higher pressure relative to the other side, and the gas molecules permeate according to the pressure difference. The separation occurs with the dependency of the permeation speed on molecular species. The dependency comes from the fact that the mechanism of molecular transport in the membrane is quite different from that in bulk gas: various diffusion effects such as Knudsen diffusion, molecular sieving, and solution diffusion play important roles. In these

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Nomenclature

α = A, B	mixture component
F (R)	channel at front (rear) side of membranes
H(C)	heated (unheated) plate
subscript 1, 2	1: inlet and 2: outlet at H and C
ε	effective porosity of the membrane
$\theta\,\Delta T$	effective temperature difference
κ	Boltzmann constant
χ	mole percentage of A-gas
χ_{I}	χ of the feed mixture
$\Delta\chi$	increment of χ by separation device
ℓ_{C}	mean free path of A-gas at p_{C}, T_{C}
ℓ_{R}	mean free path of A-gas at p_{R}, T_{R}
ℓ^{BGK}	mean free path in the BGK model
ℓ^{S}	mean free path in the S-model
c_{R}	sound speed of A-gas at T_{R}
D	width of channel F and R
d	pore size of the membrane
d_{MCE}	effective pore size of the membrane
d_m^{α}	diameter of α-molecule
Err_{Q}	$= 100\,(Q_{\mathrm{I}} - Q_{\mathrm{L}})\,/Q$
Kn	Knudsen number in micro-channel
$L = 2dm$	length of channel F and R
L_{-} (L_{+})	length of inlet (outlet) of F and R
M_1, M_2	mass flow controller
m	number of simulated micro-channels
m^{BGK}	mass of a molecule in the BGK model
m^{S}	mass of a molecule in the S-model
m^{α}	mass of α-molecule
N_p (N_T)	nondimensional flow rate of pressure (temperature) induced flow
n^{α}	number density of α-gas
p_{C}	pressure at C_2
p_{D}	pressure drop between F and R
p_{F}	pressure at the inlet of channel F
p_{R}	pressure at the outlet of R
p_i	pressure at gauge P_i ($i = 1,\ 2,\ 3$)
p^{α}	partial pressure of α-gas
Δp	$= p_2 - p_3$
Δp_{I}	pressure difference between $\mathrm{H}_1 - \mathrm{C}_2$
Δp_{L}	pressure difference between $\mathrm{H}_2 - \mathrm{C}_1$
Q	$= (Q_{\mathrm{I}} + Q_{\mathrm{L}})\,/2$
Q_{I}	volume flux of the feed mixture
Q_{L}	volume flux at U-bend part
Q^{α}	net volume flux of α-gas through the membrane
Q^{*}	$= Q^{\mathrm{A}} + Q^{\mathrm{B}}$
S	area of the membrane
$T_{\mathrm{F}}(T_{\mathrm{R}})$	temperature at the front (rear) channel
$T_{\mathrm{H}}(T_{\mathrm{C}})$	temperature of heated (unheated) plate
ΔT	$= T_{\mathrm{H}} - T_{\mathrm{C}}$
t	thickness of the membrane
V	openings of valve V_1 at U-bend part
v	flow velocity of mixture
v_{F}	flow velocity of feed mixture

implementations, we can control only one driving parameter for the molecular transport—the pressure difference across the membrane. It means that we can control only one physical quantity of permeate flow, which may be the total molecular flux of the mixture through the membrane. As a result, we cannot control the ratio of molecular fluxes of component gases; the performance of gas separation is mostly determined by the physical and chemical properties of the membrane.

The above use of membranes, however, utilizes only a part of the ability of membranes. That is, the pressure difference is just one of the driving forces of molecular transport in the membrane. According to the results of molecular gas dynamics, which deals with the behavior of the gas with a finite mean free path of gas molecules, the temperature field may induce various steady gas flows other than natural convection [5]. These flows vanish in the continuum (i.e., small mean free path) limit, but they remain appreciable in the rarefied gas, where the size of mean free path of gas molecules is comparable to the scale of the system. The rarefied condition applies to the gas in micro-channels of porous membrane if the pore diameter is comparable to the mean free path. For these porous membranes, we have two driving forces for gas flow in the membrane, which can be controlled by two different physical parameters—pressure and temperature differences across the membrane. In this case, we can control two physical quantities of permeate flow. In the case of binary mixtures, we can control molecular flux of each component independently [6]. Thus we can change the selectivity of a given membrane. It is even possible that one component flows in the opposite direction of the flow of the other component. This is one of the keys of the thermal enhancement of membrane gas separation presented in this paper.

Although there are various kinds of temperature induced flows, only a few of them can be used owing to the limitations in fabrication. The local flow speed of thermal edge flow [7,8] is proportional to the square root of the mean free path, but the requirement for the shape of micro-channel is not simple. As for the flows whose speed is proportional to the mean free path, two types of flow are investigated: the nonlinear thermal stress flow [9] and the thermal transpiration (or thermal creep) flow [10,11]. The former requires a larger variation of temperature, which is difficult to achieve in the membrane. The latter is well known linear effect and is extensively studied by many researchers, including the case for gas mixtures [10–21]. (Please refer reviews [22–24] for extensive references.) The studies lead the development of the pump with no moving parts—Knudsen pump [25]. In 2011, Gupta and Gianchandani [26] succeeded to induce the thermal transpiration flow under atmospheric pressure by using a mixed cellulose ester (MCE) membrane. Their dime-sized pump induces appreciable gaseous flux around 1 sccm and a pressure difference around 1 kPa with a temperature difference of 30 K. The pump is also stable to work continuously over 6 months. It is one crucial step for membrane technology to utilize the thermal transpiration flow. In the present paper, we make use of the design of the pump by Gupta and Gianchandani to devise our gas separation device.

The purpose of our device is to demonstrate the enhancement of membrane separation by a temperature field. The native gas separation effect of small portion of MCE membrane with temperature difference around 30 K is, of course, expected so small for engineering purposes. Some improvements are required to amplify the effect: e.g., the use of long tube [27] or a series connection of Knudsen pump [28]. In the present paper, we apply one of the existing methods of membrane gas separation. That is, we use the counter flow setup similar to the hollow fiber membrane gas separation unit [1]. This is the second key of the present work, which enables the detection of the thermal enhancement of membrane separation. First we will carry out numerical simulations on the gas flow in the test device to confirm the effect of the combination of the thermal enhancement and the counter flow setup. Second we will carry out experiments with binary mixtures of noble gases to demonstrate the new method of membrane gas separation.

2. Structure of gas separator

2.1. Concept

The schematic of our gas separator [29,30] is shown in Fig. 1. The device consists of a porous membrane with micro-channels and