Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Jaeger, Marc J.(1996) 'The Enrichment of the Lighter of Two Gases by Enhanced Diffusion (example He/N_2)', Separation Science and Technology, 31: 7, 987 — 992
To link to this Article: DOI: 10.1080/01496399608002499
URL: http://dx.doi.org/10.1080/01496399608002499

SEPARATION SCIENCE AND TECHNOLOGY, **31**(7), pp. 987–992, 1996

The Enrichment of the Lighter of Two Gases by Enhanced Diffusion (example He/N_2)

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ABSTRACT

Molecular diffusion is greatly enhanced when it takes place in a carrier that is moving in a reciprocating fashion within a pipe. The enhancement reaches three to four orders in gases if the oscillation frequency and amplitude are tuned to the physical properties of the diffusing gases and of the carrier gas. Unexpectedly, the heavier gas shows a greater enhancement than the lighter one and therefore diffuses faster. The enrichment of the lighter gas, which we describe in this paper, is therefore more complex than the enrichment of the heavier one. It can only be achieved productively if the standard diffusion protocol is modified and includes diffusion in a continuous, upstream flowing carrier and if the carrier is condensed in the container that receives the feed. Experimental results confirm the theory. This theory predicts unusual findings: thus, separation ratio and yield may be freely set by adjusting the numerical value of independent variables such as the rate of inflow of the feed. Moreover, the yield is found to rise proportionately with the separation ratio! The technique may therefore be practically useful in addition of being quite interesting. Cost estimates for the purification of He are given.

A number of researchers have shown that diffusion is appreciably enhanced if it takes place in an oscillating fluid. The enhancement is all the higher, the lower the molecular diffusion coefficient. Thus, in liquids, an enhancement of six orders is achieved; in gases, the maximal enhancement is three to four orders. The larger of two gas molecules, i.e., the one with the lower molecular diffusivity, is transported at a higher rate. We discuss in a companion paper [*Sep. Sci. Technol.*, *31*, 811–827 (1996)] how this observation can be used in a practical application to enrich deuterium. The references are found in that paper.

Any time the heavier component of a gas mixture is enriched, the lighter components are enriched, too, in what is usually waste product. This paper focuses on ways and means to improve the enrichment of the lighter molecule. It would seem at first glance that this process should not be very efficient since the lighter components has a lower effective rate of diffusion, but the results we show are encouraging. For brevity, the theory is not repeated and the description of the setup is kept to a minimum. The enrichment of an He/N_2 mixture is used as an example with H_2O vapor as the carrier gas. The theory of enhanced diffusion predicts values for the standardized effective diffusion coefficient $\lambda = D_{\text{eff}}/\omega\Delta x^2$ for He and N_2 (Table 1). The ratio $\Gamma = \lambda_{N2}/\lambda_{He}$ approaches at low frequency the inverse of the molecular diffusion coefficients of these gases in H_2O. We choose in the following a frequency of 3.81 s^{-1} in order to optimize the separation Γ which is then 2.44. At higher frequencies, Γ falls, even though the predicted diffusional flux in a setup of given geometry (G_L) rises remarkably (Table 1). G_L is defined below. A glossary in the companion paper may clarify some definitions common to both papers.

For the sake of brevity, only what is pertinent for the enrichment of the lighter species is discussed here. The experimental schematic in Fig. 1 shows that the feed is injected at a rate Q_0 into reservoir (1) and the carrier is injected at a rate Q_3 into reservoir (2). The controlled outflows of the two reservoirs are Q_1 and Q_2, respectively. By application of the conservation of mass principle, we derive from equations in the companion paper

$$f_{i1} = 1 - f_{i2} = \frac{c_{i1}Q_1}{c_{i0}Q_0} = \frac{Q_0}{Q_0 + G_iQ_2/(G_i + Q_2)} \qquad (1)$$

where "i" stands either for the heavier (H) or the lighter gas (L). G_i is defined as

$$G_i = AD_{\text{effi}}/\text{L} \qquad (2)$$

TABLE 1

α	f (second)	λ_{N2}	λ_{He}	G_L (mL/min)
1	0.95	2.59 − 03	1.00 − 03	136.5
2	3.81	9.56 − 03	3.913 − 03	213.7
3	8.57	1.59 − 02	7.93 − 03	979.4
4	15.20	1.20 − 02	1.177 − 02	3207.0
10	95.2	5.39 − 03	5.88 − 03	8089.0

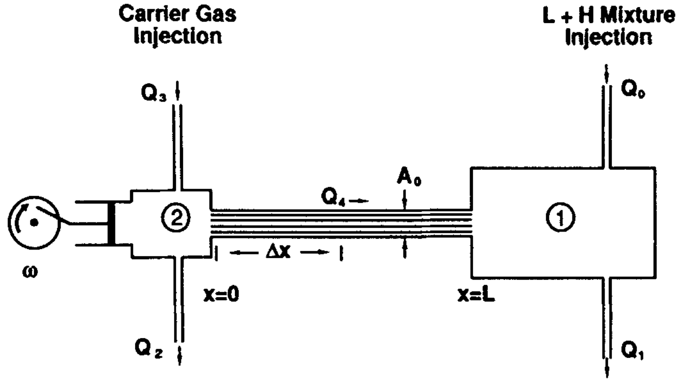

FIG. 1 The setup consists of two small reservoirs connected by a bundle of capillaries with a radius ($a = 0.19$ cm) chosen in order to optimize the transport of the gases considered. The capillaries are 120 cm long; their total cross section is 8.63 cm^2. The feed is injected at a rate Q_0 into reservoir (1); the outflow of that reservoir, Q_1, is controlled. The carrier gas is injected into reservoir (2) at a rate of Q_3; the outflow is kept at Q_2. Inflow and outflow of each reservoir are sometimes the same ($Q_2 = Q_3$; $Q_1 = Q_0$). Q_4 is a continuous, small velocity current from (2) to (1), which is termed counterflow since it is directed against the concentration gradients for H and L.

with D_{effi} being the effective or enhanced diffusion coefficient. The separation ratio for the gas exiting reservoir (1) follows as

$$s_{\mathrm{L1}} = f_{\mathrm{L1}}/f_{\mathrm{H1}} \tag{3}$$

Numerical evaluation of this equation yields a very much lower separation than the analogous equation for the enrichment of the heavier compound s_{H2}. For example, applying the equations to the separation of He/N$_2$, the enrichment of the heavier component s_{H2} is 1.95; by comparison, with the same settings, $s_{\mathrm{L1}} = 1.10$ ($Q_0 = Q_2 = 2000$ mL/min). A separation ratio of only 1.10 precludes any attempt at a practical enrichment, since it is about 15 times lower than the ratio of the molecular diffusion coefficients ($D_{\mathrm{mHe}}/D_{\mathrm{mN2}} = 2.59$).

The use of a continuous, low velocity current of the carrier directed against the concentration gradient is quite successful in raising the separation ratio when the heavier component is to be enriched. One might therefore wonder if it is also of use for the problem at hand, the enrichment of the lighter molecule. To test this hypothesis, we derive the transfer fractions and the separation ratio in counterflow from similar equations in the companion paper:

$$f_{\mathrm{L1}} = \frac{Q_1Q_3[(e^P - 1) + Q_4]}{Q_1Q_3[(e^P - 1) + Q_4] + Q_2Q_4} \tag{4}$$

$$f_{H1} = \frac{Q_1Q_3[(e^{P/\Gamma} - 1) + Q_4]}{Q_1Q_3[(e^{P/\Gamma}) + Q_4] + Q_2Q_4} \tag{5}$$

with $s_{L1} = f_{L1}/f_{H1}$ as before. Numerical solution shows that counterflow is disastrous in our present application since s_{L1} falls asymptotically to 1.0 when the rate of counterflow is raised!

The separation ratio s_{L1}, however, is improved when the carrier flowing from reservoir (2) to reservoir (1) is condensed or absorbed upon arrival in reservoir (1). This modification of the diffusion conditions is discussed in some detail in the companion paper; it raises the concentrations of the feed in reservoir (1). The effect on s_{L1} is illustrated in Fig. 2: s_{L1} is plotted against the rate of counterflow P and is seen to reach maxima. These maxima depend on P, Q_1, and Q_2 as indicated in the legend of the figure. Highest separation ($s_{L1} = 4.42$) is reached for low values of the rate of feed Q_1. This value is remarkably higher than the figure of 1.10 predicted when there is no countercurrent.

The finding that s_{L1} rises to reach a maximum when the upstream flowing carrier is condensed in reservoir (1) is perhaps unexpected, but certainly fortunate. The next observation is even more surprising: The yield $Y_1 = Q_1(c_{L1} + c_{H1})$, i.e., the outflow of reservoir (1) enriched in L and freed of any carrier, also rises with P. The rise is curvilinear and approaches Q_0 at high levels of counterflow. Figure 3 illustrates this relation-

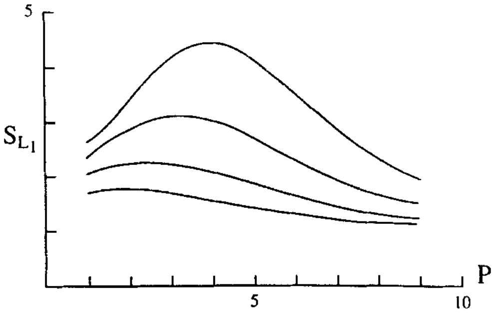

FIG. 2 The separation ratio s_{L1} is plotted against the rate of counterflow expressed as $P = Q_4/G_L$. It depends on P, Q_1, and Q_2 in an interactive fashion. Four curves were generated; from top to bottom Q_1 varied from 25 to 50, 100, and 200 mL/min. When the rate of inflow of the carrier, Q_2, is set at 2000 mL/min, the coordinates of the peak values of s_{L1} are:

$Q_1 = 25$ mL/min; $P = 4.0$; $s_{L1} = 4.42$
$Q_1 = 50$ mL/min; $P = 3.2$; $s_{L1} = 3.11$
$Q_1 = 100$ mL/min; $P = 2.6$; $s_{L1} = 2.27$
$Q_1 = 200$ mL/min; $P = 2.0$; $s_{L1} = 1.75$

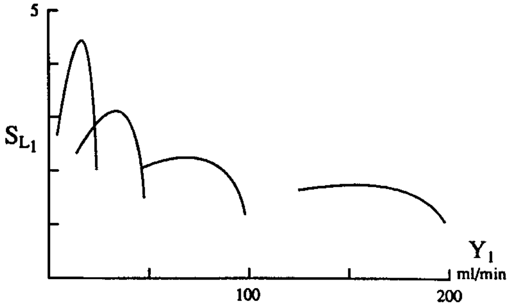

FIG. 3 The separation ratio s_{L1} is plotted against the yield, i.e., the outflow of reservoir (1) freed of carrier gas. Each curve is generated by varying $1 < P < 10$. $Q_1 = 25, 50, 100, 200$ mL/min for the four curves from left to right, respectively. $Q_2 = 2000$ mL/min and is kept constant. The coordinates of the peak values as set for each curve are:

P	s_{L1}	Y_1 (mL/min)
4.0	4.42	15.6
3.2	3.11	32.6
2.6	2.27	70.3
2.0	1.75	150.0

ship by plotting Y_1 and s_{L1} for different values of Q_1 and P. Each dome-shaped curve is derived from the above equations by keeping Q_1 constant while P is varied from zero to nine. It is seen that s_{L1} and Y_1 increase in concert up to the maximal value of s_{L1}. This surprises, since separation and yield are usually inversely related in separation processes. The peak value of s_{L1} and the corresponding Y_1 depend on P, Q_1, and Q_2. Numerical examples in the legend of Fig. 3 illustrate this relationship which offers rich possibilities of optimization.

Experimental verification was done using the same gases and the same setup as described in detail in the companion paper. CO_2 was used as the carrier gas. This affects the gas viscosity and the molecular diffusion coefficients. The theory predicts new optimal settings than for He/N_2 diffusing in H_2O vapor. The experimental verification gave good agreement between experiment and theory. Thus, for $Q_0 = Q_1 = 20$ mL/min and $Q_2 = 1000$ mL/min, s_{L1} is predicted to be 7.58 at $P = 4.67$. The experiment gave $s_{L1} = 9.00$.

DISCUSSION

Since enhanced diffusion, in contrast to simple molecular diffusion, furthers the transport of the heavier of two molecules, it may appear that it is uniquely applicable to the enrichment of the heavier components in a mixture. However, the process can be turned around to enrich the lighter component, too. The proposed setup makes this reversal rather simple; the variables are readily adjusted to maximize the new purpose.

The possible use of enhanced diffusion to enrich He may be evaluated by modeling a putative cascade. It is assumed that a 1% He/99% N_2 gas mixture is to be enriched in He using water vapor as the carrier. It is further assumed that the recycling of water vapor is the main fraction of the energy cost; indeed, the frictional losses of oscillating the gas mixture in the capillaries are minimal, if the losses in the reciprocating pump are neglected. The cascade requires five stages, the first stage having an overall cross-sectional area about 600 times larger than the last one. Taking as the last stage a unit identical to the one used in the our own experiments, the cascade would produce 11.5 mL of 95% He/5% N_2 per minute at a recycling cost of 60 L of water vapor. If the cascade is enlarged to seven stages, it would produce 11.2 mL/min 99.8% He at a cost of 177.5 L of water vapor. The energy cost would then be about 8.8 kWh/L of 99.8% He. This evaluation does not account for any recovery of heat in the recycling of water vapor. Such a recovery is usually achieved at a rate that increases with the size of the operation. It would greatly reduce the operating costs.

As mentioned in the companion paper [*Sep. Sci. Technol.*, *31*, 811–827 (1996)], the method is in the early stages of development. Improvements of both theory and experiments are planned. Especially easy would be to increase Δx and therefore the yield; Δx may be raised until turbulence occurs. We have not tested this possibility to the limit. The increase of operating pressure is another avenue to higher yield. It would increase the molar yield while keeping the operating costs per mol essentially the same. In brief, the method may get very close to becoming competitive with only small improvements.

Received by editor July 17, 1995