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Diffusional Separation of Gases and Solutes in Oscillatory Flow

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Abstract

We present a method for separating gases and solutes by oscillating the fluid column. The process is based on augmented diffusion and is akin to Taylor dispersion. The augmentation of the flux, as compared to simple molecular diffusion, is 3 orders of magnitude in the gaseous system and 6 orders of magnitude in the liquid system. Proper choice of experimental conditions (capillary radius, frequency, and fluid properties) is crucial for best results. In the gaseous system, the best separation factor achieved was 0.4; the expected separation under the conditions prevailing was 0.3. Highest diffusional flux was 850 mL/min in a system with a cross-sectional area of 8.62 cm^2. The net energy consumption at these conditions was only 14 W. The separation in the liquid system was similar to that in the gaseous system. The method provides good separation at low energy cost.

INTRODUCTION

Taylor (*1*) has shown that the spreading of a substance introduced in small quantity into a fluid flowing in a pipe is greatly enhanced by the fluid velocity. The spreading is the result of the combined action of radial molecular diffusion and the variation of velocity over the cross section. It is especially high in oscillating, laminar flow because of the particular time-dependent velocity profile in this type of flow. Oscillatory flow has the further advantage that there is no net, continuous flow in the pipe, and that the spreading occurs, if averaged over time, from a stationary site. The increased spreading can be expressed as effective diffusion or dispersion (*2–7*) We show in this paper that the effective diffusion coefficient is

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4 to 6 orders of magnitude greater than the molecular diffusion coefficient of the same substance, and that the process can be used in gases and liquids to separate substances with different molecular diffusion coefficients (*8*, *9*).

THEORY

Early work on dispersion in an oscillatory liquid goes back to Dreyer et al. (*10*). They developed their work based on the assumption that the flow pattern in slow oscillations is close to that in laminar steady flow and succeeded in separating solutes using a single capillary. A more complete study was performed by Harris et al. (*11*). The most comprehensive analysis is by Watson (*12*). He showed that the transport of a tracer in oscillatory pipe flow may be expressed as an effective diffusion coefficient, D_{eff}, which is a function of (a) the molecular diffusion coefficient D_m of the tracer; (b) the kinematic viscosity, ν, of the carrier fluid; (c) the radius, a, of the cylindrical pipe; and (d) the angular velocity, ω, of the oscillations. For convenience, the above parameters are combined into two nondimensional numbers: the Schmidt number $\sigma = \nu / D_m$, and the Wormersley number $\alpha = a\sqrt{\omega/\nu}$. The transport equation for a straight, cylindrical pipe of length L and cross-sectional area A may be written, in analogy to the conventional diffusion equation, as

$$G = \dot{q}/(c_1 - c_2) = D_{\mathrm{eff}} A/L \tag{1}$$

where $\dot{q}$ is the rate of diffusional transport, c_1 and c_2 are the concentrations of the diffusing substance at each end of the diffusing path, and D_{eff} is the effective diffusion coefficient. Watson (*12*), and earlier Harris (*11*), found that D_{eff} is proportional to the square of the oscillation amplitude ΔX (ΔX = stroke volume/cross-sectional area). For convenience, D_{eff} is normalized by dividing by the product of the angular velocity times the square of the oscillation amplitude, $\lambda = D_{\mathrm{eff}}/\omega \Delta X^2$ (*3*, *5–7*).

The normalized diffusion coefficient λ is, under certain limiting conditions, a simple function of α and σ. These limiting conditions are different for gases and liquids, and they are tabulated for clarity in Table 1. If the limiting conditions do not apply, a more general formulation must be used:

$$\lambda(\alpha,\sigma) = D_{\mathrm{eff}}/\omega \Delta X^2 = AB/C \tag{2}$$

with

$$A = \sigma/4\alpha(\sigma^2 - 1)$$

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TABLE 1

Gas	$\alpha < 1$	$\alpha > 20$
$0.1 < \sigma < 2.0$	$\lambda = \sigma\alpha^2/384$	$\lambda \approx \dfrac{1}{12\sqrt{2\alpha}}$
Liquid	$\alpha < 0.01$	$\alpha > 1$
$500 < \sigma < 5000$	$\lambda = \sigma\alpha^2/384$	$\lambda \approx \dfrac{1}{4\sqrt{2\sigma\alpha}}$

$$B = F_I(\alpha) - \frac{1}{\sqrt{\sigma}}\left[\frac{|F(\alpha)|}{|F(\alpha\sqrt{\sigma})|}\right]^2 F_I(\alpha\sqrt{\sigma});\ F(\alpha) = F_R(\alpha) + iF_I(\alpha)$$

$$C = |1 + 2F(\alpha)/\alpha|^2$$

$$F_R(\alpha) = [\text{bei}\,\alpha\ \text{ber}'\,\alpha - \text{ber}\,\alpha\ \text{bei}'\,\alpha]/[\text{ber}^2\,\alpha + \text{bei}^2\,\alpha]$$

$$F_I(\alpha) = [\text{ber}\,\alpha\ \text{ber}'\,\alpha + \text{bei}\,\alpha\ \text{bei}'\,\alpha]/[\text{ber}^2\,\alpha + \text{bei}^2\,\alpha]$$

Note that the theory predicts that λ (and therefore D_{eff}) may be either proportional to D_m (in liquids if $\alpha > 1$) or inversely proportional to D_m (in liquids and in gas if α is small). Dispersion is quasi-independent of D_m in gases if $\alpha > 20$ (*13*).

Separation of gases in a gaseous carrier or of solutes in a liquid carrier can be achieved whenever the ratio of the diffusion coefficients D_{eff} for the two substances is not unity. Optimal separation, however, exists only under narrowly defined conditions. This is illustrated for gases in Fig. 1 and for liquids in Fig. 2. In each case there is a heavy molecule (H) that is to be separated from a lighter one (L). We plot λ against α.

We choose as examples in the gaseous phase the dispersion of CO_2 ($\sigma = 1.0$) and of He ($\sigma = 0.2$) in O_2 (Fig. 1). The normalized dispersion has a different maximum for each tracer. The maxima occur at a narrowly defined value of α and fall off rapidly if α is varied above or below that particular tuning point (*6, 14*). It seems appropriate to attempt separation near the maxima, since λ predicts the flux of the tracers. However, best qualitative separation, i.e., the lowest ratio of λ_L/λ_H, is only given at conditions when the flux is relatively low; according to Table 1, one may expect best separation in the gaseous phase for $\alpha < 1$, i.e., at a much lower value of α than the tuning point for either gas. We add in Fig. 1 a dotted curve that expresses a compromise between best qualitative separation and highest flux of the gas one wishes to collect. The relationship is obtained by multiplying the separation ratio λ_H/λ_L by the standardized flux λ_H. This

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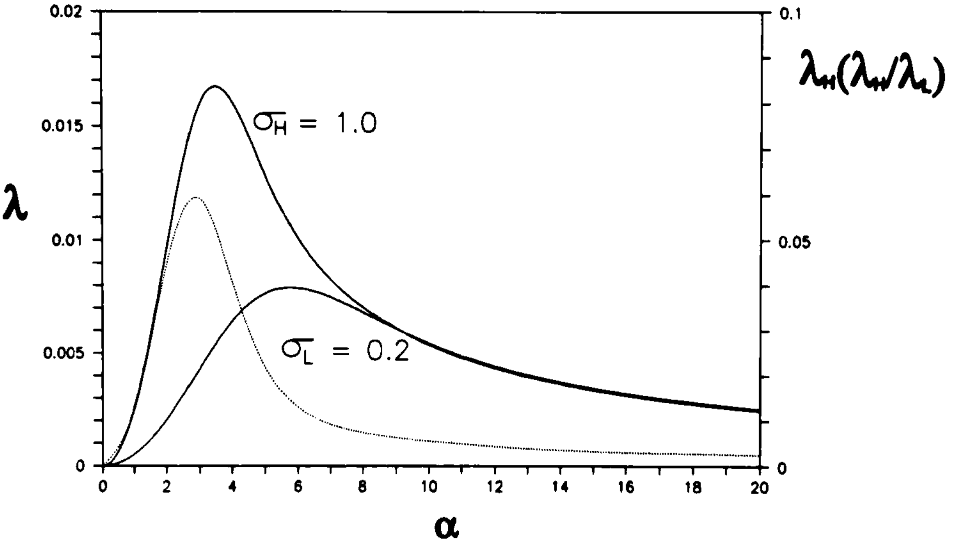

FIG. 1. The normalized dispersion coefficient $\lambda = D_{\mathrm{eff}}/\omega\Delta X^2$ is plotted against the Wormersley coefficient $\alpha = a\sqrt{\omega/\nu}$ for CO_2 diffusing in O_2 ($\sigma = 1.0$) and He diffusing in O_2 ($\sigma = 0.2$); in addition, the factor $\lambda_H(\lambda_H/\lambda_L)$ is shown as a dotted line.

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factor has a maximum at $\alpha = 3.0$; the ratio λ_L/λ_H equals 0.27 under these conditions, and is close to the theoretical best separation of the two gases ($D_{m\mathrm{He}}/D_{m\mathrm{CO_2}} = 0.20$).

Figure 2 shows the solution of Eq. (2) for the separation of two solutes in a liquid. We choose two solutes with $\sigma_L = 2000$ and $\sigma_H = 500$ diffusing in H_2O with $\nu = 0.01$ cm^2/s. Comparison with Fig. 1 illustrates several differences, as well as interesting similarities, between the liquid and the gaseous phase:

1. Maximal normalized dispersion is found at values of α which are much lower in liquids than in gases. To estimate optimal tuning of α, we may write $\alpha_{H_2O}/\alpha_{\mathrm{gas}} = \sqrt{\sigma_{\mathrm{gas}}/\sigma_{H_2O}}$.
2. The achieved maximal dispersion is nearly the same in gases and in liquids; thus, e.g., λ_{CO_2} in O_2 reaches 0.016 while maximal dispersion of a heavy molecule in H_2O is 0.019. The respective molecular diffusion coefficients are nearly 4 orders of magnitude apart.
3. In liquids, λ_H may be either higher or lower than λ_L. In gases, λ_H is always higher.

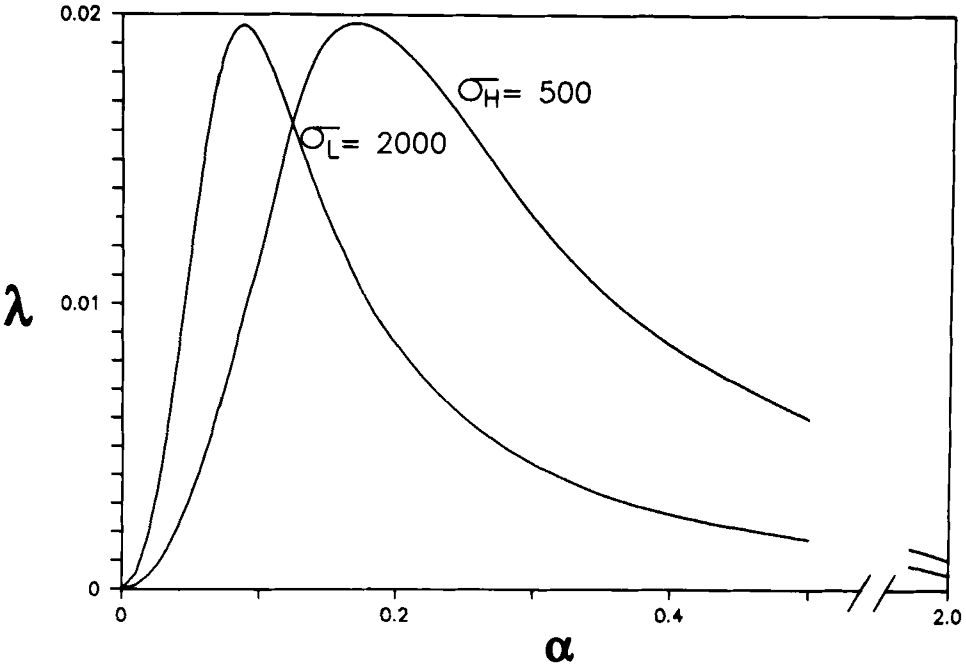

FIG. 2. The normalized dispersion coefficient λ is plotted against α for two solutes in water, one heavy (σ_L = 2000) and one light (σ_H = 500).

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METHODS

The experimental setups for gaseous and liquid separation are similar (Fig. 3). They consist of a reciprocating pump that generates oscillatory flows in capillary tubes. A reservoir at one end of the capillaries holds the fluid (gaseous or liquid) which contains the two tracers that are to be separated. The area at the other end of the capillaries is flushed with pure carrier fluid at a rate Q. This generates a concentration gradient for the diffusing substances H and L, which are transported at rates $\dot{q}_H$ and $\dot{q}_L$ from the container toward the area being flushed. The partially separated substances are collected and analyzed at the outflow of the system. The continuity equation requires the carrier fluid to diffuse in the opposite direction at a rate $\dot{q}_c = \dot{q}_H + \dot{q}_L$.

The specific conditions for gaseous separation were as follows: We chose a bundle of 76 capillaries with radius a = 0.19 cm and length L = 120 cm. The value of α was varied between 2.66 and 4.61. The oscillation amplitude was less than 20% of L. We used a gas mixture of 10% He,

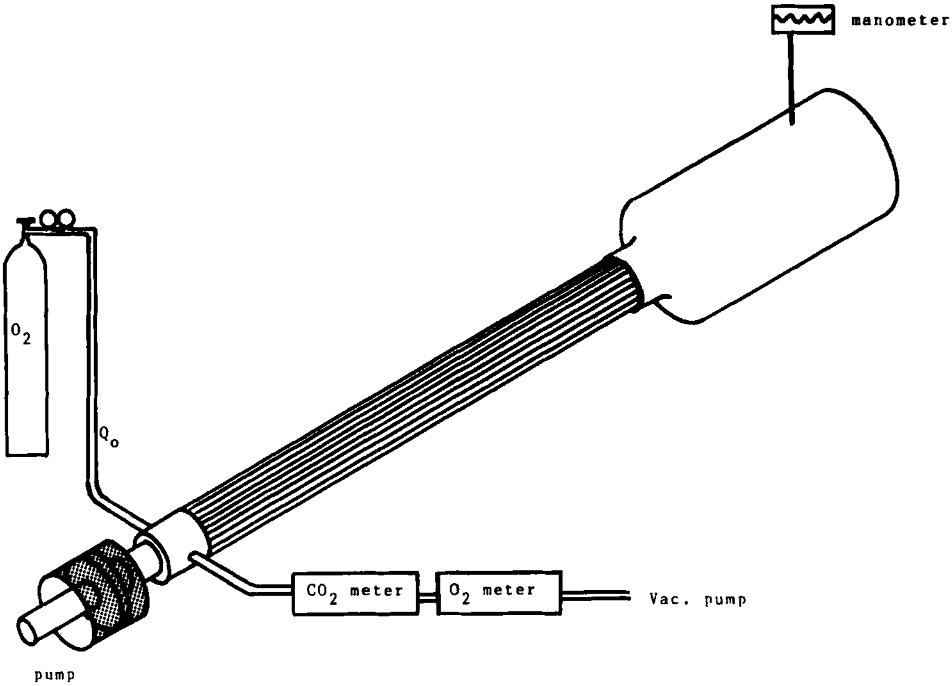

FIG. 3. Experimental setup showing a reservoir with the gases to be separated, the capillaries which serve as the diffusing path, the inflow of carrier fluid (Q), and the pump.

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10% CO_2, and 80% O_2, O_2 being the carrier gas. The reservoir had a volume of 20 L. The pressure in the reservoir was continuously monitored and kept at an average value of $P_{amb} \pm 0.01$ mmHg by adjusting the outflow in order to match the outflow exactly to Q.

The system for liquid separation consisted of a bundle of 91 capillaries with $a = 0.087$ cm and $L = 120$ cm. Alpha was varied between 0.52 and 0.89. The oscillation amplitude was kept below 40% of L. The reservoir had a volume of 0.2 L and had a distensible top. We used the following solutes: KCl, $CuSO_4$, glucose, Methylene Blue.

Experimental separation depends on Q, the rate of flushing with carrier fluid. If Q is high, the available concentration gradient is high and the flux is maximal. This provides for best separation, but the concentration of the diffusing substances in the outflow is low. On the contrary, if Q is reduced, the flux and separation are reduced as well. It may be shown (7) that the separation factor ρ_L equals:

$$\rho_L = C_{L2}C_{H1}/C_{L1}C_{H2} = (G_L/Q + \lambda_L/\lambda_H)/(G_L/Q + 1) \qquad (3)$$

RESULTS

The theory predicts that transport is proportional to $\omega\Delta X^2$ for each given value of α (Eqs. 1 and 2). We therefore plot D_{eff} vs $\omega\Delta X^2$ for each gas at different frequencies. Results of such plots are shown in Table 2, which includes the experimental slopes λ^*, the intercepts, and the correlation coefficients of three sets of experiments conducted at $\alpha = 2.66$, 3.76, and 4.61. Each set includes measurements at four different amplitudes ΔX. The correlation coefficients are high, indicating good agreement with the predicted relationship. The experimental slopes λ^* for He are higher than predicted; the slopes for CO_2, lower; this leads to the ratio $\lambda^*_{\mathrm{He}}/\lambda^*_{\mathrm{CO_2}}$ being higher than predicted. However, the magnitude of the effective diffusion coefficient is close to the theory: $D_{\mathrm{effCO_2}}$ is at maximum amplitude ($\Delta X = 23.2$ cm), 3625 times higher than $D_{m\mathrm{CO_2}}$, close to the theory.

The measured separation factor ρ_{L} is plotted in Fig. 4. The variation is due to different settings of Q (Eq. 3). Best separation is found for high values of Q. The best achieved separation approaches 0.4; the expected separation under those conditions is 0.3.

Results of liquid separation are given in Table 3. The ratios of individual separations of three pairs of substances are plotted in Fig. 5 against the predicted ratio $\lambda_{\mathrm{H}}/\lambda_{\mathrm{L}}$. Open squares represent the averages. A fairly good relationship is found, but as with the gaseous data, the separation is less than predicted. The maximal flux obtained with KCl was equivalent to an augmentation of the diffusion rate by a factor of 840,000 when compared to simple diffusion without oscillations.

DISCUSSION

The discussion deals first with a critique of the method and with a comparison between the results and the theory. The theory makes a number

TABLE 2
Regression Analysis of Gaseous Data

α	λ^*	Intercept	r	N	λ	$\lambda^*_{\mathrm{He}}/\lambda^*_{\mathrm{CO_2}}$	$\lambda_{\mathrm{He}}/\lambda_{\mathrm{CO_2}}$
2.66	$D_{\mathrm{effCO_2}} = 0.0100\omega\Delta X^2$	−7.52	0.97	8	0.0137	0.42	0.26
	$D_{\mathrm{effHe}} = 0.0042\omega\Delta X^2$	−4.18	0.96	8	0.0035		
3.76	$D_{\mathrm{effCO_2}} = 0.0135\omega\Delta X^2$	−37.4	0.99	11	0.0161	0.69	0.37
	$D_{\mathrm{effHe}} = 0.0094\omega\Delta X^2$	−46.1	0.96	11	0.0059		
4.61	$D_{\mathrm{effCO_2}} = 0.0113\omega\Delta X^2$	−3.2	0.98	11	0.0142	0.68	0.50
	$D_{\mathrm{effHe}} = 0.0077\omega\Delta X^2$	−3.1	0.95	11	0.0072		

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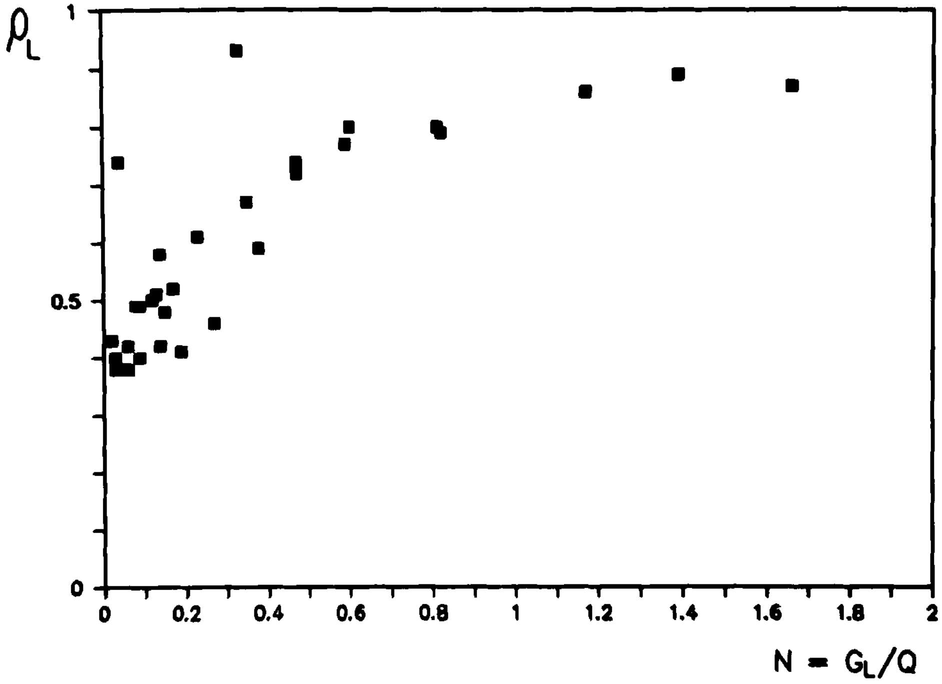

FIG. 4. Effect of dilution by the carrier fluid flow Q on the separation. Squares are experimental values of $G_{\mathrm{He}}/G_{\mathrm{CO_2}}$.

TABLE 3
Results: Liquid Data[a]

	α	σ	λ	λ*	N	r
KCl	0.89	500	0.00212	0.00107	8	0.98
$CuSO_4$	0.89	1136	0.00099	0.00060	8	0.99
Glucose	0.89	1450	0.00078	0.00081	8	0.99
Methylene Blue	0.89	1923	0.00062	0.00057	12	0.91
KCl	0.52	500	0.0059	0.00229	7	0.92
$CuSO_4$	0.52	1136	0.0031	0.00190	6	0.99
Glucose	0.52	1450	0.0024	0.00127	7	0.99
Methylene Blue	0.52	1923	0.0019	0.00231	5	0.99

[a] λ = predicted standardized dispersion coefficient. λ* = measured standardized dispersion coefficient.

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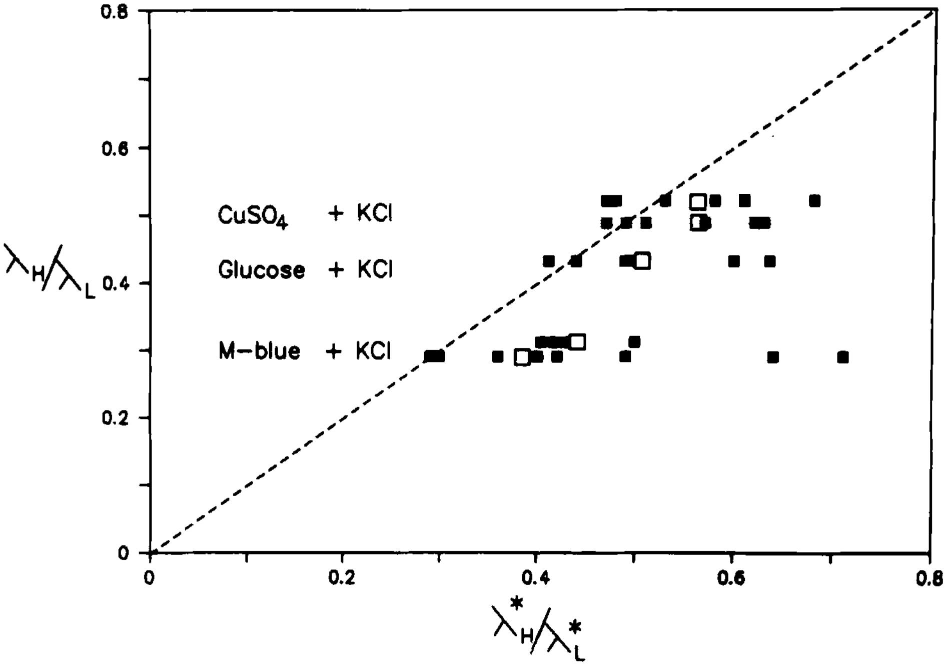

FIG. 5. Separation of 3 pairs of solutes. λ_H/λ_L is the expected separation, λ_H^*/λ_L^* is the measured separation.

of assumptions which are only incompletely satisfied in the experimental setup used. These assumptions include 1) uniform geometry, 2) incompressible fluid, 3) low tracer concentration, and 4) laminar flow. These assumptions are discussed separately.

Uniform geometry implies several notions. It assumes that the diffusion path has the same geometry everywhere and, therefore, does not account for the connecting tubes needed to connect the capillaries with the pump, with the container, and with the measuring equipment. Oscillations occur in all of those conduits. Dispersion takes place in those conduits at values of α that are different from those in the capillaries. This reduces separation. Moreover, there is likely to be an entrance length in each capillary in which the flow is not fully developed. The effect of entrance conditions on dispersion has not been evaluated, neither theoretically nor experimentally. Thus, it is not possible to estimate its effect. The entrance length itself is not as clearly defined in oscillatory flow as it is in steady flow. If one

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assumes the length to equal approximately ΔX, it includes a significant amount of the diffusing length in our experiment and may be a significant factor explaining the descrepancies between experiment and theory.

Another factor is gas compressibility and/or conduit distensibility. It is, presumably, of importance in the gaseous phase only. Our system has a mechanical resonance at >20 Hz. Gas compressibility and inertance combine to result in differences between the stroke volume of the pump and the tidal volume measured at the distal end of the capillaries of up to 33%. The effect of compressibility is difficult to evaluate; it should, however, be the same on the two gases that are separated. It is therefore, presumably, not the cause of the discrepancy between expected and measured separations (Table 4).

The theory further assumes that the properties of the oscillating fluid are constant along the diffusing path; thus, ν, α, and σ are assumed constant along the capillaries (*12, 14*). This is the case if the diffusing substance is introduced at tracer concentration only. Such low concentrations are technically uninteresting. The concentration was therefore raised to 10% for each diffusing gas. This has an appreciable effect on ν, which varies by 60% along the capillaries. The effects of such variation are also not known. To test this effect, we conducted experiments raising the concentration of CO_2 and He in the container to 50% each. The results are preliminary, but encouraging. With $\alpha \simeq 3.76$, the measured ratio $\lambda^*_{He}/\lambda^*_{CO_2}$ was 0.38 ($N = 12$). This equals the separation obtained with low-concentration gas mixtures listed in Table 2.

The theory assumes laminar, oscillatory flow. Indeed, the dispersion is independent of the diffusivity of the tracer in turbulent flow. The conditions for turbulence in oscillatory flow have been described in different ways. Signs of turbulence may be very transient, occurring at particular times of the cycle only (*15–18*). Kamm et al. (*19*) used the nondimensional number Re/α to define the onset of changes of dispersion in oscillatory flow. This ratio is not to exceed 200. It was exceeded by a small amount in some of

TABLE 4
Gas Separation Data

	ρ_L[a]		
α	Experimental	Theory	G_L/Q
2.66	0.40 ± 0.03 ($N = 8$)	0.28	0.06
3.76	0.43 ± 0.08 ($N = 6$)	0.44	0.13
4.61	0.49 ± 0.15 ($N = 5$)	0.56	0.13

[a] See Eq. (3).

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our experiments, indicating that we reached the safe limit for separation and that a further increase of ΔX would presumably be detrimental to separation. This has not been tested, however.

CONCLUSIONS

Oscillations of the fluid column in appropriately sized capillaries greatly increase the diffusional flux of tracer substances. In gases, the enhancement is 3 orders of magnitude; in liquids, 5–6 orders of magnitude. It is assumed that technical improvements would further increase this flux. The highest $\dot{q}_{CO_2}$ measured was 850 mL/min; the simultaneous $\dot{q}_{He}$ was 612 mL/min. The process offers a new approach for separation in either the gaseous or the liquid phase. The process may be used in cascades. It is suitable for continuous operation. We used steady-state operation for the experiments mentioned earlier using 50% CO_2 and 50% He. This was achieved through perfusion of the container (Fig. 3) with the gas mixture. The operational energy cost is low. With a diffusional flux of CO_2 of 850 mL/min and a separation factor $\rho_L = 0.38$, the net energy cost was 14 W. To that cost one has to add the cost of removing the carrier fluid. This fluid can be chosen for its convenience and low processing costs, since optimizing of the diffusional process is independent of the carrier fluid's properties. Our choice of O_2 was made for technical reasons.

Some features shown in Figs. 1 and 2 give an interesting insight into the mechanism of enhanced diffusion. Thus, it may be seen that maximal standardized dispersion (λ) occurs when $\alpha^2\sigma = \pi^2$ (*6*). This means, in effect, that the process is as effective in liquids as in gases when conditions are optimized $[\alpha^2\sigma = (a^2\omega/\nu)(\nu/D_m)]$. Optimal conditions are realized when the boundary layer thickness δ has a fixed relationship to the radius ($\delta/a = \sqrt{\sigma}/\pi$). In gases, for which σ is close to unity, the boundary layer thickness needs to be equal to about 1/3 of the radius. In liquids, however, optimal conditions are met when the boundary layer is, in effect, thicker than the radius.

The oscillation of the fluid column results in large velocity and concentration gradients. Fluid coming alternatively from either end of the capillaries moves into the core of the fluid column. This results in very large concentration gradients and tracer fluxes between core and boundary region. The fluxes change direction during each half-cycle. Optimal net transfer occurs when $t_d/t_e = \pi$, i.e., when the time required for radial diffusion ($t_d = a^2/D_m$) has a simple, fixed relationship to the length of the half-cycle, which is the time effectively available for each radial flux. In liquids this time is relatively long and the diffusion takes place within the very narrow outer region of the boundary layer, where the velocity gradient is highest. A further interesting theoretical prediction is that there are conditions for

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which the dispersion of substances of different sizes is identical (Figs. 1 and 2). This implies that the effective diffusion of substances of different sizes is the same. This prediction may have its own theoretical and practical applications.

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