

## Thermoacoustic Separation of a He-Ar Mixture

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We report observation of a new mixture-separation process: an insonified mixture of helium and argon in a narrow duct spatially separates along the acoustic-propagation axis. We measure mole-fraction differences across the ends of the duct as large as 7%. We measure initial separation flux densities as high as  $10^{-3} M^2 c$ , where  $M$  is the acoustic Mach number and  $c$  is the sound speed. This initial separation flux, as a function of both the amplitudes and the relative phasing of the pressure and velocity oscillations in the duct, agrees well with a recent theory involving oscillating thermal diffusion in the acoustic boundary layer.

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Mixture separation is one of the great technological challenges, permeating nearly every area of science and engineering. It is so pervasive that the motto of the Institution of Chemical Engineers is *findendo fingere disco* ("I learn to make by separating") [1]. Here we demonstrate a new mixture-separation phenomenon: acoustic separation of a gas mixture, using simple hardware, through a complex interaction of boundary-layer processes similar to the pumping of heat in a thermoacoustic refrigerator [2,3]. While these experimental data are consistent with our theory [4], the theory is very incomplete and much work remains to be done to understand this phenomenon, its consequences, and its possible practical uses.

Our apparatus is shown in Fig. 1. Two reservoirs of volume  $V = 125 \text{ cm}^3$  are connected by a narrow duct in which the separation occurs; the whole is filled with an initially uniform 50/50 molar mixture of He and Ar at mean pressure  $p_m = 80 \text{ kPa}$  [5]. Each reservoir has a bellows-sealed piston driven by a linear motor, and a side branch on each reservoir leads to an acoustic gas analyzer. Independent phase and amplitude control of the two motor-driven pistons allows great variety in the resulting waves produced in the separation duct. The frequency  $f$  of the piston oscillations is low enough ( $< 20 \text{ Hz}$ ) that the length of the separation duct is much shorter than a wavelength, so the gas oscillations can at first be thought of as essentially uniform along the length of the duct. Sensors in each reservoir detect the amplitude and phase of the oscillating pressure in each reservoir, which are used with the gas density and viscosity and the dimensions of the separation duct to calculate the amplitudes of pressure  $p_1$  and volume flow rate  $U_1$ , and the phase difference  $\theta$  by which  $p_1$  leads  $U_1$ , in the middle of the duct. Each acoustic analyzer [6] is an acoustically excited cylindrical cavity used to detect the local proportions of the He and Ar. The lowest resonance frequency ( $\sim 4 \text{ kHz}$ ) in the cylindrical cavity is  $c/2\ell$ , where  $c$  is the sound speed and  $\ell$  is the cavity length. The sound speed is in turn related to the molar weight  $m$  of the mixture by  $c^2 = \gamma RT/m$ , where  $\gamma$  is the ratio of isobaric to isochoric specific heats,  $R$  is the universal gas constant, and  $T$  is the mean absolute temperature. Mole

fraction can thus be easily obtained from measurement of the analyzer's resonance frequency and gas temperature (not shown in the figure are thermocouples mounted in the center of each acoustic analyzer.)

Figure 2 shows the difference in He mole fraction between the reservoirs as a function of time, for a separation duct with a 5-mm inner diameter and a length of one meter, with the pistons driven at 10 Hz to produce a pressure oscillation in the separation duct of 2 kPa. Here, the pistons are driven so that the wave in the separation duct has nearly traveling-wave phasing ( $p_1$  leading  $U_1$  by only  $15^\circ$ ) but a ratio of pressure to velocity  $6\times$  higher than that of a pure traveling wave. The calculated power dissipation in the duct is 5 mW. An easily measurable mole-fraction

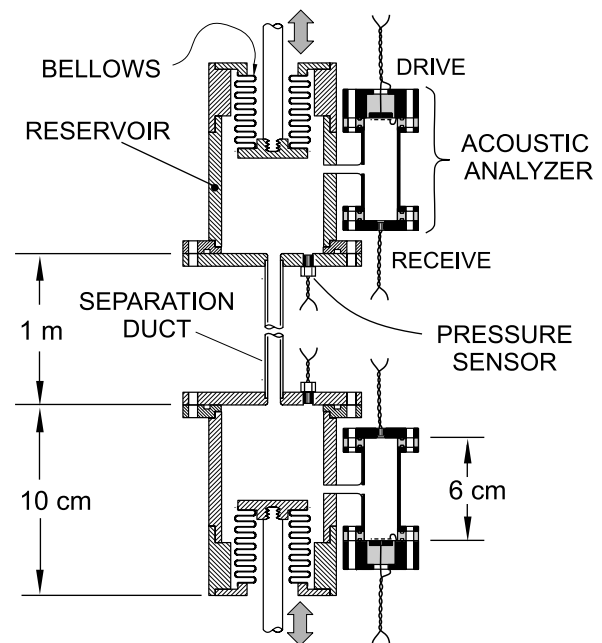


FIG. 1. A schematic of our apparatus. Two  $125\text{-cm}^3$  reservoirs, driven by bellows-sealed pistons, are connected by a narrow duct in which the separation process occurs. Each reservoir has a dynamic-pressure transducer and a side branch to an acoustic gas analyzer that detects the mole fraction of He.

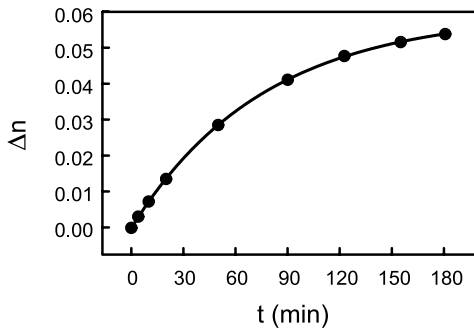


FIG. 2. The difference between the mole fractions of He in the two reservoirs of Fig. 1 vs time, when the experiment is filled initially with a 50/50 molar mixture of He-Ar at 80 kPa and the pistons are driven at 10 Hz to produce  $p_1 = 2$  kPa,  $p_1/U_1 = 6\rho c/A$  (where  $A$  is the duct area), and  $U_1$  temporally lagging  $p_1$  by  $15^\circ$ . The points represent the measurements, and the line is an exponential fit to the points.

difference of 1% arises in only a few minutes; eventually the separation process saturates, leveling off near 6% under these conditions (and as high as 7% under other conditions). The direction of the He and Ar fluxes can be reversed by interchanging the phases and amplitudes of the two pistons. Thus a simple acoustic device may indeed separate the components of a gas mixture.

With an approach similar to that used to develop basic thermoacoustic heat-transport theory [7], we have begun [4] to understand how a binary mixture of gases such as He and Ar in an acoustic field near a solid boundary should experience separation along the axis of acoustic propagation. The idea is shown in Fig. 3, where one cycle of a standing wave in a binary gas mixture, near a solid boundary, is crudely represented by four idealized discrete steps. Near times  $t = \tau/8$  and  $5\tau/8$ , the pressure wave is at an extremum, and the particle velocity is zero. The gas far from the solid boundary experiences adiabatic compression or rarefaction, and is therefore heated or cooled relative to the high-heat-capacity isothermal boundary. A temperature gradient extends over a distance  $\delta_\kappa = \sqrt{k/\pi f \rho c_p}$  in the gas, where  $k$  is the thermal conductivity,  $\rho$  is the density, and  $c_p$  is the isobaric specific heat. Thermal diffusion [8] causes the heavy component to diffuse toward the wall and the light component to diffuse away from the wall near  $\tau/8$  and vice versa near  $5\tau/8$  (directions may be reversed for some gas mixtures), with the components diffusing a distance of the order of  $\delta_D = \sqrt{D_{12}/\pi f}$ , where  $D_{12}$  is the binary mass diffusion coefficient. Near  $3\tau/8$  and  $7\tau/8$ , the pressure is near its mean value and the temperature is spatially uniform, but the velocity is nonzero, with a gradient near the wall extending over a distance of order  $\delta_\nu = \sqrt{\mu/\pi f \rho}$  where  $\mu$  is the viscosity. In the cycle illustrated, the net effect is that light-enriched gas outside the viscous boundary layer moves downward near  $3\tau/8$  while heavy-enriched gas is trapped inside the viscous

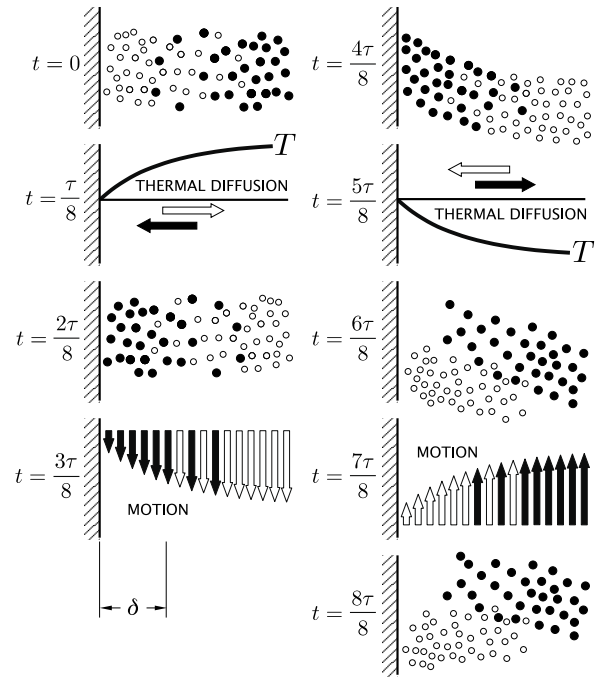


FIG. 3. A simplified view of the interplay between thermal diffusion and viscosity in the acoustic boundary layer, producing spatial separation of the components of a gas mixture. The small, open circles represent light molecules, and the larger, filled ones represent heavy molecules. The figure shows the history of such molecules near one location through a full cycle of an acoustic standing wave, comprising a temporal sequence of high pressure and temperature, downward motion, low pressure and temperature, and upward motion. The period of the oscillation is  $\tau$ , and the temperature is  $T$ . The net result of a full cycle is that heavy molecules move up and light molecules move down.

boundary layer, and vice versa near  $7\tau/8$ , resulting in net fluxes of the heavy component upward and the light component downward. For our He-Ar mixture at 10 Hz,  $\delta_\kappa = 1.6$  mm,  $\delta_D = 1.7$  mm, and  $\delta_\nu = 1.0$  mm. Under the conditions of Fig. 2, the adiabatic temperature oscillation amplitude is  $3^\circ\text{C}$  and the average gas displacement amplitude along the duct is 2 cm.

The full mathematical treatment of this process is challenging, but we have already predicted the initial separation flux [4], before a significant mole-fraction gradient is established, in the boundary-layer limit:

$$\dot{N}_2 = -\frac{\delta_\kappa}{d} \frac{\gamma - 1}{\gamma} \frac{k_T}{RT} [F_{\text{trav}} p_1 U_1 \cos\theta + F_{\text{stand}} p_1 U_1 \sin\theta], \quad (1)$$

where  $\dot{N}_2$  is the second-order molar separation flux of the lighter of the two gases,  $k_T$  is the thermal diffusion ratio,  $d$  is the duct diameter, and  $F_{\text{trav}}$  and  $F_{\text{stand}}$ , which indicate the traveling-wave and standing-wave contributions to the flux, are complicated expressions [4] involving thermo-physical properties of the mixture including  $\mu$ ,  $k$ , and  $D_{12}$ .

Although the idea shown in Fig. 3 is very naive, it captures some features of this quantitative theory: The separation takes place in the boundary layer, due to acoustically driven thermal diffusion perpendicular to the separation direction, with the net separation flux proportional to the product of oscillating pressure and oscillating velocity.

Acoustic enhancement of binary separation in mixtures of three gases (the third gas is a carrier) using the differences between the ordinary mass diffusivities of two of the components through the third component is also under vigorous development [9–11]. This mechanism works independently of the boundary-layer temperature gradients and thermal diffusion of our process. Separation of binary gas mixtures using a sound wave was observed earlier [12] and was attributed both to barodiffusion and thermal diffusion [13], but with the latter involving the interaction of the second-order, time-averaged boundary-layer temperature gradient with the ordinary second-order Rayleigh streaming [14]. We suspect that this fourth-order separation mechanism is generally weaker than our second-order process.

The data in Fig. 2 were taken in a duct with  $d/\delta_\kappa = 3$ , which we find produces a relatively large ultimate separation. However, the theory in [4], in particular Eq. (1), should be tested in the boundary-layer limit, where  $d/\delta_\kappa \gg 1$ . Hence, for subsequent experiments we used a 15-mm-diam duct at 15 Hz, yielding  $d/\delta_\kappa = 11$ . Under these conditions, Eq. (1) reduces to

$$\dot{N}_2 = \left(14 \frac{\mu\text{mol}}{\text{Pa m}^3}\right) (0.03 \cos\theta + 0.01 \sin\theta) p_1 U_1. \quad (2)$$

To measure the flux described by Eqs. (1) and (2) in our apparatus, we take data in the manner of Fig. 2, and find the initial slope by fitting to the data a function of the form  $\Delta n = a(1 - e^{-bt})$ . We easily obtain

$$\dot{N}_2(t=0) = \frac{d(\Delta n)}{dt} \Big|_0 \left( \frac{p_m V}{2RT} \right). \quad (3)$$

Figure 4 shows the experimental dependence of this initial separation flux on  $p_1 U_1$ , where the latter spans nearly a factor of 10, for  $\theta = 1^\circ$ . Evidently the separation flux is linear in this product, strong evidence that this is indeed a second-order acoustic process as indicated in Eqs. (1) and (2). The highest flux recorded,  $6.2 \times 10^{-8}$  mol/s, corresponds to about 100 mm<sup>3</sup>/min of He flowing from one reservoir to the other (with an equal amount of Ar flowing in the opposite direction), while consuming 10 mW (calculated) of acoustic power in the duct. This can also be expressed as a separation flux density of order  $10^{-3} \text{ M}^2 c$ , where  $\text{M} \sim p_1/p_m \sim U_1/Ac$ . Inspired by the high powers of large thermoacoustic engines and refrigerators [3], straightforward scaling suggests that a 100-W mixture-separation device operating at higher amplitude and/or frequency, perhaps around 10 cm in diameter, with a porous

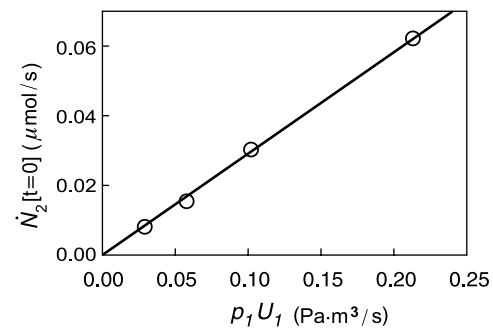


FIG. 4. The initial separation flux as a function of  $p_1 U_1$ . Here  $p_1$  and  $U_1$  are in phase, and  $p_1/U_1 = 5\rho c/A$ . Circles are data, and the line is a linear fit.

element consisting of many parallel, narrow separation ducts filling the entire cross section, could easily achieve separation fluxes on the order of 1 mole/h.

Equation (2) implies that a traveling wave ( $\theta = 0$  or  $180^\circ$ ) should produce a separation flux  $3\times$  larger than a pure standing wave ( $\theta = \pm 90^\circ$ ). This is not obvious from the crude treatment in Fig. 3. The data in Fig. 4 were taken near traveling-wave phasing, to get large separation fluxes; but for a more thorough comparison of experiment to theory, we keep  $p_1 U_1$  essentially constant, and measure  $\dot{N}_2(t=0)$  as a function of  $\theta$ . The results are compared directly to Eq. (2) in Fig. 5. Qualitatively, the agreement is excellent, with the separation flux varying sinusoidally with phase angle as suggested by theory. Quantitatively, the data are shifted only slightly in phase relative to the theory, and are only slightly lower in overall amplitude.

While the data strongly indicate that a separation process such as we describe is occurring, our understanding of the phenomenon is far from complete. For example, one possible cause of the small disagreement between theory and experiment in Fig. 5 is that a duct with  $d/\delta_\kappa = 11$  is too small to be accurately in the boundary-layer limit; the theory developed in [4] does not include realistic duct cross sections. This theory also fails to predict the separation flux with nonzero mole-fraction gradients along the separation duct and the limiting mole-fraction difference

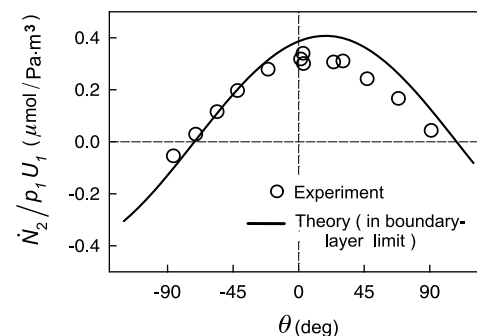


FIG. 5. The initial separation flux, scaled by  $p_1 U_1$ , as a function of the phase between  $p_1$  and  $U_1$ . The circles are data, and the line is Eq. (2).

achievable. Effects tending to remix the separated gases, especially second-order acoustic streaming [14], must also be investigated and understood; note that second-order streaming cannot be minimized relative to the second-order mixture separation by simply choosing low or high acoustic amplitudes. Such improvements to the theory will certainly be necessary for optimal design of arrays of many ducts in parallel that might be used in a large-scale acoustic separation device.

It seems unlikely that this thermoacoustic mixture-separation method can ever reach the high efficiencies obtained in distillation separation plants [15] (e.g., industrial air-separation plants), the largest of which exceed half the efficiency limit imposed by the laws of thermodynamics. Relying as it does on the irreversible processes of mass diffusion and viscous flow, this thermoacoustic separation process is intrinsically irreversible. However, other low-efficiency separation methods, such as concentric-tube thermogravimetric separation [1,15], are in widespread use where convenience and low cost are more important than the highest efficiency. Hence, we are hopeful that thermoacoustic mixture separation, integrated with compatible new concepts such as the thermoacoustic-Stirling heat engine [16] and open-flow thermoacoustic refrigerators [17], can ultimately provide gas-stream processing with useful efficiency and with the high reliability and low capital cost associated with no moving parts.

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- [1] H. R. C. Pratt, *Countercurrent Separation Processes* (Elsevier, New York, 1967).
- [2] T. J. Hofler, Ph.D. thesis, University of California, San Diego, 1986.
- [3] G. W. Swift, *Phys. Today* **48**, No. 7, 22–28 (1995).
- [4] G. W. Swift and P. S. Spoor, *J. Acoust. Soc. Am.* **106**, 1794–1800 (1999); **107**, 2299(E) (2000).
- [5] Atmospheric pressure in Los Alamos.
- [6] E. Polturak, S. L. Garrett, and S. G. Lipson, *Rev. Sci. Instrum.* **57**, 2837–2841 (1986).
- [7] N. Rott, *Z. Angew. Math. Phys.* **26**, 43–49 (1975).
- [8] L. D. Landau and E. M. Lifshitz, *Fluid Mechanics* (Pergamon, New York, 1982).
- [9] U. H. Kurzweg and M. J. Jaeger, *Phys. Fluids* **30**, 1023–1025 (1987).
- [10] G. W. Howell, *Phys. Fluids* **31**, 1803–1805 (1988).
- [11] M. J. Jaeger, *Chem. Eng. Sci.* **53**, 3613–3621 (1998).
- [12] A. M. Dykhne, A. F. Pal', V. D. Pis'mennyi, V. V. Starostin, and M. D. Taran, *Zh. Eksp. Teor. Fiz.* **88**, 1976–1983 (1985) [*Sov. Phys. JETP* **61**, 1171–1175 (1985)].
- [13] V. B. Bozhdankevich, A. M. Dykhne, A. F. Pal', V. D. Pis'mennyi, V. V. Pichugin, A. N. Starostin, and M. D. Taran, *Dokl. Akad. Nauk SSSR* **288**, 605–608 (1986) [*Sov. Phys. Dokl.* **31**, 428–430 (1986)].
- [14] W. L. M. Nyborg, in *Physical Acoustics*, edited by W. P. Mason (Academic Press, New York, 1965), Vol. IIB, pp. 265–331.
- [15] *Encyclopedia of Separation Technology*, edited by D. M. Ruthven (Wiley, New York, 1997).
- [16] S. Backhaus and G. W. Swift, *Nature (London)* **399**, 335–338 (1999).
- [17] R. S. Reid, W. C. Ward, and G. W. Swift, *Phys. Rev. Lett.* **80**, 4617–4620 (1998).