

Separation of Gas Mixtures by Thermoacoustic Waves

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Imposing sound on a binary gas mixture in a duct separates the two gases along the acoustic-propagation axis. Mole-fraction differences as large as 10% and separation fluxes as high as $0.001 M^2 c$, where M is Mach number and c is sound speed, are easily observed. We describe the accidental discovery of this phenomenon in a helium-xenon mixture, subsequent experiments with a helium-argon mixture, and theoretical developments. The phenomenon occurs because a thin layer of the gas adjacent to the wall is immobilized by viscosity while the rest of the gas moves back and forth with the wave, and the heat capacity of the wall holds this thin layer of the gas at constant temperature while the rest of the gas experiences temperature oscillations due to the wave's oscillating pressure. The oscillating temperature gradient causes the light and heavy atoms in the gas to take turns diffusing into and out of the immobilized layer, so that the oscillating motion of the wave outside the immobilized layer tends to carry light-enriched gas in one direction and heavy-enriched gas in the opposite direction. Experiment and theory are in very good agreement for the initial separation fluxes and the saturation mole-fraction differences.

INTRODUCTION

A few years ago, we were experimenting [1] with acoustically coupled acoustic resonators, using two identical half-wavelength resonators, each of which was driven at its natural resonance frequency by a thermoacoustic engine. Under some circumstances, these experiments demonstrated mode locking when the two resonators were connected through a smaller-diameter half-wavelength coupling tube. When those measurements were made with a helium-xenon mixture, we sometimes noticed a totally unexpected near-perfect equality in the resonance frequencies of the two resonators while we were trying to force unequal resonance frequencies by imposing a temperature difference (as large as 15°C) between the resonators. This behavior could not be explained by any obvious candidates such as temperature uncertainty or geometry mismatch between the resonators. We concluded that the sound wave in the small-diameter acoustic coupling tube was somehow separating the helium and xenon, thereby enriching one resonator with helium and the other with xenon.

We realized that this mass separation could be due to a combination of three effects in the boundary layers adjacent to the inside surface of a tube: oscillating temperature gradients in the thermal boundary layer, thermal diffusion, and oscillating velocity gradients in the viscous boundary layer. In a typical mixture of helium and xenon, the Prandtl number is about $1/4$, so the viscous penetration depth is about half of the thermal penetration depth. For

standing-wave phasing in a channel whose diameter is much larger than these penetration depths, we might think of the wave as consisting of four sequential steps equally spaced in time, as illustrated in Fig. 1. In the first step, while the pressure is high, the time-dependent part of the temperature has a steep gradient within a thermal penetration depth of the wall due to the adiabatic temperature rise in the gas far from the wall and the large solid heat capacity of the wall itself. During this time, thermal diffusion drives the heavy component down the temperature gradient toward the

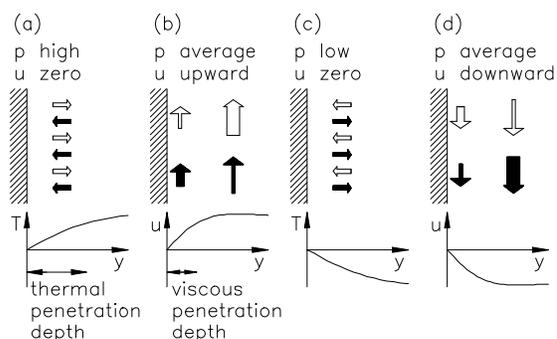


FIGURE 1. Illustration of the separation process occurring near a solid wall (hatched) in a standing wave in a gas mixture with Prandtl number near $1/4$. Solid arrows show motion of the heavy component, and light arrows show motion of the light component. The arrow lengths represent velocity, and widths represent local concentrations. (a) through (d) show processes occurring at time intervals separated by $1/4$ of the period of the wave, as described in the text.

wall and the light component up the temperature gradient away from the wall. Hence, at the end of this time the gas near the solid wall is enriched in the heavy component and depleted of the light component, while the gas approximately a thermal penetration depth from the wall is enriched in the light component and depleted of the heavy component. In the second step, the gas moves upward, with a steep gradient of velocity within a viscous penetration depth of the wall due to viscosity.

During this time, the heavy-enriched gas is relatively immobilized in the viscous boundary layer, while the light-enriched gas, just outside of the viscous boundary layer, moves easily upward. In the third step, low pressure reverses the sign of the temperature gradient, so the thermal diffusion reverses direction, forcing the heavy component away from the boundary and the light component toward the boundary. Thus, in the fourth step, light-enriched gas is relatively immobilized while heavy-enriched gas moves easily downward. The net, time-averaged effect of these four steps is that some of the heavy component moves downward while some of the light component moves upward.

RESULTS TO DATE

Thus far, the agreement between our theory [2] of this process and our measurements [3,4] of it is very good.

The theory is based on a monofrequency, steady-state acoustic approximation to the equations of momentum, continuity, heat transfer, and mass diffusion, assuming that the viscous penetration depth, thermal penetration depth, mass-diffusion penetration depth and the tube radius are all small compared to the tube length. The three penetration depths themselves are of comparable magnitudes. Results of the theory include the time-averaged mole fluxes of each component along the tube axis. This result is expressed as a function of pressure amplitude, volume-velocity amplitude, phase between pressure and volume velocity, axial concentration gradient, and gas properties (including mean pressure and temperature, thermal diffusion ratio, viscosity, thermal conductivity, and mass diffusivity). Setting the time-averaged mole fluxes equal to zero yields the axial concentration gradient at which the effect saturates.

The experiments begin with a mixture of 50% helium (mole fraction) and 50% argon near atmospheric pressure. The separation tubes are 1 meter long and either 5 mm or 15 mm in diameter, filled with the mixture and driven by bellows-sealed pistons. When a tube is insonified at 10 Hz, helium separates toward

one end and argon toward the other end. The concentrations at the ends, which reach extremes of 45% and 55%, are detected as functions of time by exciting acoustic resonance (near 3 kHz) in small cavities at each end and inferring the concentrations there via the dependence of sound speed on average molar mass [5].

Measurements include the initial rates of mole flux toward each end, before a significant concentration gradient has had time to develop, and the saturation concentration difference reached in steady state. These are measured as functions of pressure amplitude, volume-velocity amplitude, and phase between pressure and volume velocity.

In these experiments, tube length and frequency have been chosen so that the tubes are "short," much shorter than a wavelength and also short enough that the resulting concentration differences from one end to the other do not cause large differences in thermophysical properties. This eases comparison of the experimental results with the theory. We are confident that longer tubes and/or higher frequencies would result in larger separations. The larger tube diameter is chosen to be ten times the largest of the three penetration depths, so that the boundary-layer approximation is useable in the theory. The smaller of the two tubes is chosen to test the more challenging Bessel-function aspects of the theory.

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