Dynamics of gas-mixture separation in the field of a sound wave

A. M. Dykhne, A. F. Pal', V. D. Pis'mennyĭ, V. V. Pichugin, A. N. Starostin, and M. D. Taran

Institute of Metal Physics, Ukrainian Academy of Sciences
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An analysis is given of the dynamics of the spatial separation of a binary mixture of a molecular gas and helium in a broad range of concentrations (0.1–0.001) of the heavy component. The acoustic pressure was up to 9×10^4 dyn/cm^2 (bar) and the pressure of the mixture in the resonator was $P_0 = 760$ Torr. It was found that the degree of separation was increased by the radial temperature gradient and by the circulation multiplication effect in acoustic vortex flows. A possible separation mechanism is discussed together with the expected magnitude of the separation effect. A comparison is made with the experimentally observed effect.

1. INTRODUCTION

The possibility of gas-mixture separation in the field of a traveling sound wave was first investigated in Ref. 1. The basic mechanism producing the spatial separation of particles of different mass was assumed in Ref. 1 to be barodiffusion (see, for example, Ref. 2), and the reason for the appearance of a time-averaged pressure gradient in the traveling wave was considered to be the attenuation of sound by dissipative volume processes. The true nature of the separation of the 1:1 mixture of CO_2 and H_2, observed in Ref. 1, has not been entirely clear: the process took hours to settle, and the magnitude and sign of the effect did not fit the theoretical ideas.[1]

The separation of gas mixtures in the field of a standing acoustic wave was investigated in Ref. 3. The separation of mixtures by barodiffusion is conveniently described in terms of an individual particle when the sound is excited in a light gas containing a small heavy-particle impurity.[3] Friction forces exerted on a test particle by vibrating light molecules makes the heavy-particle concentration distributed in space in accordance with the Boltzmann law

$$n=n_0 \exp\,(U/T),$$

where the effective high-frequency potential U is given by

$$U=M\bar{\tilde{v}}^2,$$

in which M is the mass of the test molecule and $\tilde{v}$ is the velocity of light particles in the acoustic wave (the bar represents time-averaging over one period of the acoustic waves).

Precisely the same expression for the spatial separation of a small heavy-particle impurity is obtained from the condition that there is no resultant flow by diffusion and barodiffusion (see Ref. 2 and the expressions given below):

$$x_1/x_0\approx(P_1/P_0)^{M/m}\infty\exp\,(M\Delta\bar{p}/mP_0),$$

where the assumption that the amount of impurity is small implies that the relative volume concentration x of the impurity is $x\ll m/M$ and m is the mass of the light molecules. The subscripts 0 and 1 in the above expression refer to two chosen points in space, P is the time average of the pressure in the standing acoustic wave, and $\Delta\bar{p}=P_1-P_0$. Using the relation $\Delta\bar{p}\simeq-\rho\bar{\tilde{v}}^2$ and $P_0=\rho T/m$ (ρ is the mass density of light particles), we obtain

$$x_1/x_0\infty\exp\,(-M\bar{\tilde{v}}^2/T),$$

as above. Heavy-particle enrichment should be observed at points of higher pressure.

The experiments reported in Ref. 3 were in qualitative agreement with these ideas. The concentration of the components of the mixture was measured by the gas-sampling time-of-flight mass-spectrometer MSKh-4, which was unsuitable for investigating the dynamics of the separation process. In this paper, we present the results of experiments on the separation of a binary mixture of a molecular gas (CO, CO_2, CF_2HCl, SF_6, CF_3I) and helium. The concentration of the heavy component of the mixture was recorded continuously by observing infrared absorption lines.

Our aim was to study experimentally the dynamics of gas-mixture separation in a standing acoustic wave, and to elucidate the mechanism responsible for this separation.

2. EXPERIMENTAL SETUP AND RESULTS

The measurements were carried out in an apparatus consisting of an acoustic resonator, an acoustic-wave radiator, an exhaust system, and a gas-mixture supply system. The apparatus is shown schematically in Fig. 1. The resonator was in the form of a cylindrical tube, 2 cm in diameter and 86 cm long. An electrodynamic radiator was mounted at

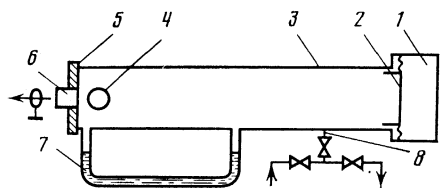

FIG. 1. Schematic diagram of the apparatus: 1—radiator; 2—radiator diaphragm; 3—resonator; 4—NaCl windows; 5—flange; 6—LKh-610 pressure pickup; 7—liquid-filled manometer; 8—exhaust and inlet system for the gas mixture.

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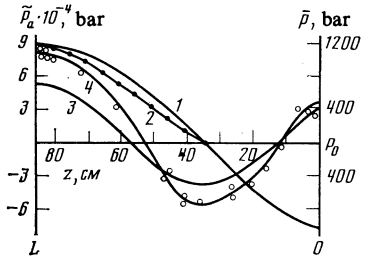

FIG. 2. Distribution of sound pressure $\tilde{P}_a$ and mean acoustic pressure $\overline{P}(z)$ along the resonator. The point marked 0 corresponds to the equilibrium position of the radiator diaphragm. Graphs of $\tilde{P}_a$: calculation—1, experiment—2; $\overline{P}$ curves: calculation—3, experiment—4. Air, $P_0 = 760$ Torr, $f_r = 167$ Hz.

one end of the resonator, whilst the other was covered by an attachment equipped with NaCl windows. An LKh-610 acoustic pickup was mounted on the flange of this attachment. The resonator was equipped with side ports used for exhausting, for introducing gas mixtures, and for attaching liquid-filled manometers. The resonator was first pumped down to about 0.03 Torr by a roughing pump. The radiator was connected to an audio-frequency generator through a 20-W amplifier.

The resonator was excited by the harmonic oscillations of the radiator diaphragm at the first natural frequency $f_r = c_s/2L$, and this produced a standing wave in its interior. The pickup was used to measure the acoustic pressure $\tilde{P}_a$ at the wave, antinode and the resonance frequency was determined from the maximum signal. The pressure distribution $\tilde{P}_a(z)$ along the resonator was also determined with an acoustic pickup. A mobile liquid-filled manometer was used to measure the acoustic pressure distribution $\overline{P}(z)$ along the resonator, and the pressure difference $\Delta\bar{p}$ between the wave node and antinode as a function of the acoustic frequency when a U-manometer was connected at these points. The results of these measurements are shown in Fig. 2. We also measured the function $\Delta\bar{p}(\tilde{P}_a)$ at the resonance frequency. The maximum value of $\tilde{P}_a$ at the antinode was $\tilde{P}_a \simeq (9.9 \pm 0.8) \times 10^4$ bar; the result $\Delta\bar{p} \simeq (1.5\text{–}2) \times 10^3$ bar is in good agreement with calculations made for different mixtures. The experimental result $\Delta\bar{p} \sim \tilde{P}_a^2$ is also in agreement with the theoretical relation.

The dynamics of the separation process was investigated with an infrared spectrophotometer. We used Specord 75 IR and IKS-29 spectrophotometers, working in conjunction with the K-201 null-type strip chart recorder manufactured by Karl Zeiss of Jena. This enables us to expand the transmission scale of the two instruments by a factor of about 10. The time constant of the recording system was about 1 s. The instrument was calibrated by varying the pressure in the resonator within $\pm$ 10% of the working pressure. The validity of this method of calibration was verified by comparing it with calibrations against mixtures differing by 10% in concentration. The resonator end equipped with windows was inserted into the sample section of the spectrophotometer, which was then tuned to the center of the chosen absorption line. The transmission coefficient was measured continuously as the mixture was irradiated with acoustic waves. The transmission coefficient was proportional to the concentration of the heavy component of the mixture.

Mixtures of Co, CO_2, CF_2HCl, SF_6, and CF_3I with helium were prepared in advance with component volume ratio of 1:9. Mixtures of SF_6 and He in ratios of 1:99, 1:332, and 1:999 were also examined.

We measured $\delta = \Delta n_T/n_T^0$ as a function of time during exposure to the acoustic waves (Δn_T is the change in the concentration of the heavy component of the mixture and n_T^0 is the initial concentration) for different acoustic pressure $\tilde{P}_a$, initial mixture concentration, mass of the heavy component, resonator length, and sound frequency. Figure 3 shows typical spectrophotometer traces reflecting the dynamics of the separation process. It is clear from the shape of these curves that separation did, in fact, take place, since the heavy-particle density recovered within a time $\sim$20–25 min, which corresponds to the time necessary for the equalization of disturbances by diffusion.

Our measurements show that:

1. There is an optimum pressure $\tilde{P}_a$ for which the change in the concentration is a maximum. For the 1:9, SF_6-He mixture it was found that $\Delta n_T/n_T^0 \simeq (4.5\text{–}5)\%$ for $\tilde{P}_a \simeq 2.9 \times 10^4$ bar. Figure 4 shows the ratio $\Delta n_T/n_T^0$ as a function of $\tilde{P}_a$.

2. When M/m was varied between 7 and 49 at $\Delta\bar{p} \simeq 280$ bar, the increase in concentration varied from 2.5% for CO-He to 5–6% for CF_3I-He [for equal concentrations

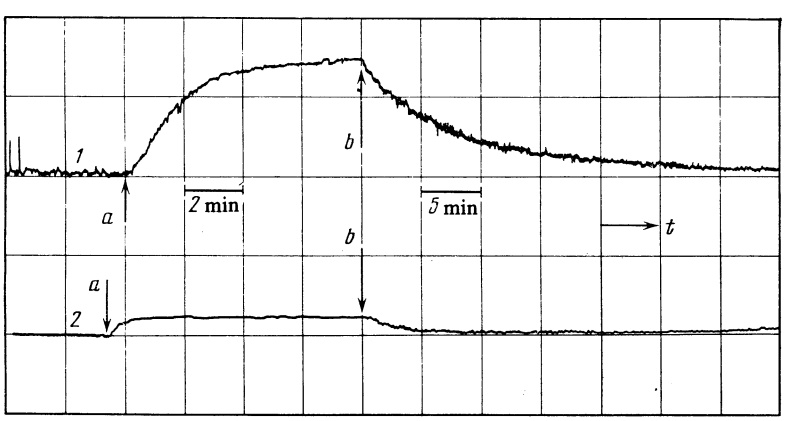

FIG. 3. Spectrophotometer output. Curve 1—resonator without partition, 2—resonator with partition in place. Arrows show: a—sound turned on, b—sound turned off; chart velocity is reduced after b. SF_6-He mixture, 1:9, $P_0 = 760$ Torr, $f_r = 205$ Hz. 1—$\Delta n_T/n_T^0 = 5\%$; 2—$\Delta n_T/n_T^0 = 0.8\%$; $P_a = 2.8 \times 10^4$ bar.

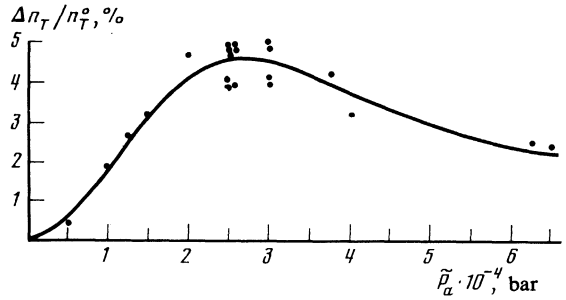

FIG. 4. Degree of separation $\Delta n_T/n_T^0$ as a function of acoustic pressure $\tilde{P}_a$. SF_6–He, 1:9, $P_0 = 760$ Torr.

x:$(1-x) = 1$:9], i.e., there was some increase in the degree of separation with increasing M/m. The results are shown in Fig. 5 (the spread of the points corresponding to CO_2 can be explained by the low precision of the measurements due to absorption in atmospheric CO_2, whereas the spread obtained for CF_2HCl was due to the fact that there was a C_3F_6 impurity of about 2%, and the absorption line used in these measurements overlapped the C_3F_6 absorption line).

3. A reduction in the concentration of the heavy component was found to be accompanied by an increase in the degree of separation at constant $\Delta\bar{p}$ (Fig. 6). When the SF_6 concentration was varied from 0.1 to 0.001, the ratio $\Delta n_T/n_T^0$ was found to increase from 4.8 to $\simeq$13.5%, i.e., by a factor of about 3. For all the mixtures that we have investigated, maximum separation was achieved for the same value $\Delta\bar{p}\simeq$280–290 bar in a time $\tau = 4$–6 min.

4. When the resonator length was reduced to 43 cm, we found that $\tau\simeq 3$ min and $\delta = 4.8\%$, i.e., the same as for $L = 86$ cm.

5. Measurements performed at the center of the resonator (node of the pressure $\tilde{P}_a$) showed that the concentration of the heavy component was lower by 3–5% (the experimental accuracy was lower in this case because the K-201 was not used).

We have also carried out measurements with the resonator divided into two along its entire length (with the exception of the attachment with the windows) by a thin metal partition in the diametral plane. This partition had practically no effect on the properties of the resonator, but had a considerable influence on the structure of acoustic flows.

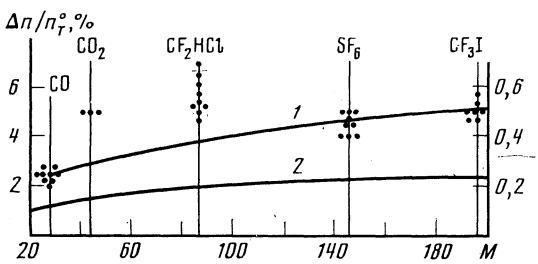

FIG. 5. $\Delta n_T/n_T^0$ as a function of the mass M of the heavy component. M–He concentration 1:9, $P_0 = 760$ Torr, $\tilde{P}_a \simeq 3\times 10^4$ bar. 1—Experiment, left-hand scale, 2—calculation, right-hand scale.

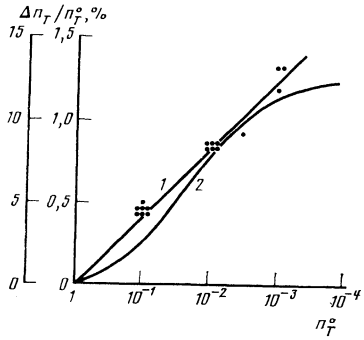

FIG. 6. Dependence of n_T/n_T^0 on the concentration n_T^0 (logarithmic scale) of the SF_6–He mixture, $P_0 = 760$ Torr; 1—experiment, left-hand scale; 2—calculation, right-hand scale.

These experiments showed that the separation was reduced by a factor of about 5–6, and increased linearly with increasing $\Delta\bar{v}$.

3. ANALYSIS OF EXPERIMENTAL RESULTS

Let us consider in greater detail the expected spatial separation of a gas mixture in the field of a standing acoustic wave. A standing wave with velocity amplitude $\tilde{v}$ is established in a tube in which sound is produced by an oscillating cover plate at one end.[4] This amplitude has the following distribution:

$$\tilde{v} = v_0 \cos\omega t \frac{\sin K_0(L-z)}{\sin K_0 L}, \tag{1}$$

where v_0 is the velocity of the oscillating plate, ω is the oscillation frequency, L is the length of the acoustic resonator, and the length z is measured from the equilibrium position of the cover plate. The acoustic wave undergoes slight attenuation in the gas at atmospheris pressure, the main contribution to the attenuation being due to dissipation by viscosity, thermal conduction, and diffusion in boundary layers near the walls.[2]

Standard analysis of the attenuation of acoustic waves near a wall in a two-component gas mixture in the absence of surface flow yields the following dispersion relation for sound in the tube when the wall is held at constant temperature:

$$K = (\omega/c_s) + i\Gamma, \tag{2}$$

where c_s is the velocity of sound in the mixture, given by

$$c_s^2 = \frac{P_0}{\rho_0}\frac{\langle\gamma/(\gamma-1)\rangle}{\langle 1/(\gamma-1)\rangle} \equiv \frac{P_0}{\rho_0}\langle\gamma\rangle, \tag{3}$$

$\langle F\rangle = xF_1 + (1-x)F_2$, x is the relative volume concentration of particles of type 1 (in our case, these are heavy particles of mass M, where the mean mass of the mixture is $\langle m\rangle = xM + (1-x)m$), γ_i is the adiabatic exponent for particles of type i, and

$$\Gamma = \frac{1}{Rc_s}\left\{\left(\frac{\nu\omega}{2}\right)^{1/2} + \frac{\mu_2[1+(\mu_2/\mu_1)^{1/2}]}{\langle 1/(\gamma-1)\rangle[1+(\chi/D\mu_2^{1/2})]}\left(\frac{\chi\omega}{2}\right)^{1/2}\right\}. \tag{4}$$

In this expression, R is the radius of the tube, ν is the kine-

matic viscosity, χ is the temperature diffusivity, and D is the diffusion coefficient. The quantities μ_1 and μ_2 are defined by

$$\mu_{1,2}=\frac{1}{2}\left\{1+\frac{\chi^*}{D}\pm\left[\left(1+\frac{\chi^*}{D}\right)^2-4\frac{\chi}{D}\right]^{1/2}\right\}, \tag{5}$$

$$\chi^*=\chi+D^*; \tag{6}$$

where D^* can be expressed in terms of the thermal diffusion coefficient k_T and the barodiffusion coefficient[2] k_p:

$$D^*=\frac{D(m-M)\langle m\rangle k_T^2}{\langle\gamma/(\gamma-1)\rangle k_p Mm}. \tag{7}$$

The quantity k_p is usually expressed in terms of the mass concentrations c of the particles:

$$c=xM/\langle m\rangle. \tag{8}$$

According to Ref. 2,

$$k_p=(m-M)c(1-c)[(1-c)/m+c/M]. \tag{9}$$

Estimates show that the renormalization of the barodiffusion coefficient by viscous forces[5] was unimportant under our conditions. It will be more convenient to use the barodiffusion coefficient $\bar{k}_p$, which relates the current to the volume concentration gradient:

$$i\propto(\nabla x+\bar{k}_p\nabla P/P).$$

In our case,

$$\bar{k}_p=[(m-M)/\langle m\rangle]x(1-x). \tag{10}$$

Estimates of the attenuation of sound based on (4)–(9) show that the Q-factor of a perfect acoustic resonator is given by

$$Q=\omega/\Gamma c_s\geqslant 30, \tag{11}$$

which is appreciably greater than the value found experimentally from the halfwidth of the resonance pressure curve ($Q_{\text{exp}}\leqslant 3$). The large width of the resonance curve is probably due to absorption in the sound-generating plate. The reactive component of the conductance of the plate assembly produces a reduction in the wave vector[6] of the sound wave as compared with $K_0=\pi/L$, and hence in the corresponding resonance frequency f_r, which amounts to 167 Hz ($f_r^0=c_s/2L=197.7$ Hz for air at $P=1$ atm). Since estimates have shown that the attenuation of sound near the walls was small under our conditions, the time-averaged pressure can be estimated from

$$\rho\bar{v}^2+\bar{P}=\text{const}, \tag{12}$$

which follows from the conservation of momentum. The constant in (12) can be found by considering the nonlinear equation for the standing sound wave (see, for example, Ref. 4). We thus have

$$\bar{P}(z)=P_0+\rho_0\tilde{v}^2[(\langle\gamma\rangle+1)/8-{}^1/_2\sin^2K_0(L-z)], \tag{13}$$

where $\tilde{v}=v_0\sin K_0L$ and $K_0=\omega/c_s$. Neglecting thermal diffusion, it can be shown that the following relations ensues from the condition of zero heavy-particle current:

$$\nabla x+(\bar{k}_p/\bar{P})\nabla\bar{P}=0. \tag{14}$$

Integrating this, we find that, in the steady state,

$$\left[\frac{x(z)}{x(0)}\right]^{m/(M-m)}\left[\frac{1-x(0)}{1-x(z)}\right]^{M/(M-m)}=\frac{\bar{P}(z)}{\bar{P}(0)}. \tag{15}$$

Since the temperature of the gas mixture varied only slightly under the conditions of our experiment, the quantity $x(0)$ can be found from the conservation of the number of heavy particles:

$$\frac{1}{L}\int_0^L x(z)\bar{P}(z)\,dz=x_0P_0, \tag{16}$$

where x_0 is the initial concentration of particles of type 1 in the gas mixture. Figures 5–7 show numerical calculations performed using (15), (16), and (11). They also show the experimental points for the partitioned resonator (see above), used to reduce the influence of circulating currents.

It is important to note that the observed separation effect is greater than that calculated from (13), (15), and (16). Let us therefore estimate the role of thermodiffusive separation due to the radial temperature gradient and the circulation multiplication effect in acoustic vortex flows.[4] Let us compare the velocity amplitude of the Rayleigh vortex $\tilde{v}_R\sim 0.1v^2/c_s\sim 1$ cm/s with the convective flow due to the radial temperature gradient[7] ($\Delta T/T\sim 10^{-3}$ according to thermocouple measurements):

$$v_{\text{conv}}\sim(\nabla P/(2\pi)^2\rho\nu)(\Delta T/T)R^2. \tag{17}$$

Estimates of v_{conv} based on (17) yield ~ 1 cm/s, i.e., $v_R\sim v_{\text{conv}}$. According to Ref. 4, the velocity of the Rayleigh vortex near the wall is directed away from the velocity antinode toward the end. When the temperature on the resonator axis is greater than that at the wall, and the thermal diffusion coefficient is positive (i.e., when heavy-particle enrichment occurs near the wall), the Rayleigh vortex should multiply the transverse separation effect and should lead to an increase in heavy-component enrichment at the end of the tube. For the same sign of the radial temperature gradient, the convective vortex is directed in the opposite direction to that of the acoustic vortex (the equivalent acceleration $\sim\Delta P/\rho$ points away from the end of the tube), and should produce an effect of opposite sign (and incidentally for the opposite sign of $\nabla_\perp T$, as well). Unfortunately, our estimates

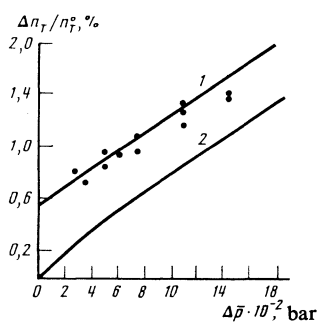

FIG. 7. Dependence of n_T/n_T^0 on $\bar{P}$ for the resonator with partition in place. SF_6–He, 1:9, $P_0=760$ Torr. 1—Experiment, 2—calculation.

are not accurate enough to enable us to draw an unambiguous conclusion about the magnitude and direction of the resultant vortex velocity. However, the fact that the effect exceeds estimates based on barodiffusion suggests that it is more likely that the Rayleigh acoustic flows predominate over convective flows. Estimates of the overall separation effect due to barodiffusion and thermal diffusion yield (cf. Refs. 7 and 8; the subscript 0 corresponds to the end of the tube)

$$\int_{x(0)}^{x(z)} \frac{dx}{x(1-x)\,[H+E_zK_d]} = \int_0^z \frac{dz}{K_c+K_d}, \tag{18}$$

where

$$H=2\pi\alpha \int_0^R \frac{d\ln T}{dr}\psi dr,$$

α is the thermal diffusion constant, $\bar{v}_z$ is the velocity of the vortex along the tube axis,

$$\psi = \int_0^R \rho\bar{v}_z r\,dr, \tag{19}$$

$$K_d = 2\pi \int_0^R \rho D r\,dr, \tag{20}$$

$$K_c = 2\pi \int_0^R \frac{\eta h^2}{\rho D}\,dr, \tag{21}$$

and

$$E_z \approx ((M-m)/\langle m\rangle)\, d\ln P/dz. \tag{22}$$

Estimating the effect for small x and $M \gg m$, we obtain

$$x(z) \sim x(0)\exp\left\{-\frac{M}{m}\left|\int_0^z \frac{d\ln P/dz}{1+(K_c/K_d)}\right| - \left|\int_0^z \frac{Hdz}{K_c+K_d}\right|\right\}. \tag{23}$$

The order-of-magnitude result is $K_c/K_d \sim (Rv_R/D)^2 \sim 1$, although K_c may become greater than K_d as the amplitude of the acoustic wave increases, and this may give rise to a reduction in the separation effect. The second term in the argument of the exponential in (23) gives rise to an increase in the separation effect by thermal diffusion. When $\alpha \sim 0.1$ (cf. Ref. 7),

$$\int \frac{Hdz}{K_c+K_d} \sim \frac{L\alpha\Delta T v_R}{2DT[1+(Rv_R/D)^2]} \sim 10^{-2},$$

i.e., the effect is of the order of 1%, which is comparable with the contribution due to the first term. We note that the dependence on the component mass ratio is determined mainly by the first term in the agreement of the exponential (23). The thermal diffusion effect also passes through a maximum as the amplitude of the second wave increases but, when the partition is introduced, the position of the maximum shifts toward greater amplitudes, which agrees with experiment. Moreover, the time taken for the effect to become established is shorter than the diffusion time due to convection ($\sim L^2/4\pi^2 D$). It is clear that the separation effect in the field of a sound wave can be substantially increased when a forced radial temperature gradient is established (for example, by introducing a hot wire along the tube axis). As noted above, the separation maximum should then be observed on the velocity crests of the sound wave.

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