Trapping of heavy gases in stationary ultrasonic fields

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Abstract

We have observed heavy-gas trapping in stationary ultrasonic fields. This effect as well as the recently observed cold-gas traps have been investigated. Both can be explained by the difference in mass density between the trapped gas and ambient media. A first theoretical approach is given. © 2002 Elsevier Science B.V. All rights reserved.

Acoustic levitation [1,2] has been applied as a technique for suspending small liquid and solid samples with diameters in the range of micrometers to millimeters in the pressure nodes of a stationary ultrasonic field (SUSF). In the past, acoustic levitation has been applied in different research areas, e.g., in fluid dynamics [3,4], material science [5] or analytical chemistry [6,7]. In earlier work [8,9] we have observed a trapping effect of cold gases and aerosols in the SUSF and – closely connected – the formation of ice particles from ice aerosol in the pressure node regions of the SUSF. The latter phenomenon is not discussed in this Letter. Here we focus on the trapping effect: at suitable experimental conditions cold gases or aerosols gather in stable rotational ellipsoidal systems around the pressure nodes of the SUSF. These systems are well separated in temperature from the ambient medium, and therefore we call them cold-gas traps. However, the effect still remains unexplained.

In order to get more experimental data we studied the effect more precisely. The temperature-field of the cold-gas traps within the SUSF has been measured, and the dependence of the effect on the intensity of the SUSF and on the gas flux into the SUSF has been studied. Furthermore, to answer the question whether the trapping effect basically depends on the temperature or on the mass density of the trapped gas we varied the mass density of the sample gas at constant temperature. These experiments clearly show that heavy gases can be trapped as well as cold gases, and that the effect depends on the mass density of the sample gas as explained in detail below. Based on the

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experimental results and according to a model of acoustic levitation of solid and liquid samples we give a first theoretical explanation of the effect.

The experiments of this investigation have been performed in an axially tuned levitator with an ultrasonic frequency of 20 kHz which has been described in detail elsewhere [2,8]. The sound pressure level (SPL) of the SUSF was estimated by drop-out condition for suspended drops [10], i.e., balance between gravitational and levitation forces, to cover the range of 160–180 dB with an accuracy of approximately ± 2 dB. As sample gases we used helium, nitrogen, synthetic air, carbon dioxide, bromine, crypton, and octafluoro cyclobutane ($C_4F_8$). The sample gases were either precooled by liquid nitrogen or heated up before flowing into the SUSF. Gas fluxes were in the range of 0–1 l/min, and temperatures of the sample gases were between 173 and 373 K. The temperature within the SUSF was monitored by a 0.25-mm-diameter mantle thermocouple which was adjustable in the vertical $z$- and radial $r$-axes. In order to reduce convection of ambient air the whole acoustic levitator was enclosed in an acrylic glass tube.

As described earlier by some of the authors [8,9] the cold gas trapping effect is visible in case of ice aerosol, i.e., cold gas with suspended small ice particles of a few μm in size. In Fig. 1a, zones of cold gas in the SUSF of the 20-kHz levitator are marked by ice aerosol which has been generated by cooling ambient gas mixtures. In the cold-gas traps, temperatures range from 173 to 273 K. The vertical diameter of the zones corresponds to half of the ultrasonic wavelength, and the horizontal diameter depends on the radial extension of the SUSF, i.e., roughly the diameter of the ultrasound transducer which is 30 mm in our experiment. The temperature within the cold-gas traps is mainly determined by the cold-gas source. The temperature field of the cold-gas traps shows a strong gradient of 10–20 K/mm at its boundaries. A typical temperature field of cold-gas traps in the SUSF of a 20-kHz levitator is given in Fig. 1b.

Stable conditions for cold-gas traps require a continuous cold-gas flux into the SUSF. Because of acoustic streaming in the ultrasonic field, thermal diffusion along the strong temperature gradient at the boundaries, and convective heat exchange between cold gas and ambient air the effect disappears immediately (in the range of a second) without continuous supply from a cold-gas source. The cold gas is sucked into the SUSF in the planes of the pressure nodes. Because of

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**Fig. 1.** (a) Rotational ellipsoidal zones of cold-gas traps visualized by ice aerosol in the stationary ultrasonic field (SUSF) of a 20-kHz levitator. The ice aerosol which was generated from ambient air using a metal rod precooled with liquid nitrogen is sucked into the SUSF in the planes of the pressure nodes. There is a sharp boundary between the cold-gas traps and the ambient host medium. (b) Typical temperature field of cold-gas traps. The temperature, in °C, is plotted as a function of the radius $r$ and the height $z$ in the levitator axis of the SUSF. As cold-gas source gaseous nitrogen of approximately 173 K and with a flow rate of 10 ml/s was used. (c) Trapped gaseous bromine in a SUSF of a horizontal positioned 20-kHz levitator. The heavy bromine is sucked into the SUSF against gravity and forms similar rotational ellipsoidal zones similar as shown in (a).
continuity of the whole gas flux the regions of pressure antinodes act as gas outlets.

The trapping effect strongly depends on the SPL and the flow rates from the cold-gas source. Measurements of the minimum temperature with a 0.25-mm-diameter mantle thermocouple at the pressure nodes of the SUSF at a constant flow rate from the cold-gas source show that the effect appears above SPLs of approximately 130–150 dB and slightly diminishes at higher SPLs. The decline is probably caused by a stronger acoustic streaming and by other non-linear effects of the SUSF in higher SPL levels [11]. The dependence of the effect on the gas-flow rates into the SUSF at a constant SPL seems to be linear: minimum temperatures of cold-gas traps decrease linearly with increasing flow rate over a wide range.

To study the gas trapping effect in general, sample gases with different mass densities were used at different temperatures. In these experiments, the temperature of the sample gas was used as a marker as well to indicate the spatial distribution of the gas in the SUSF. For example, gaseous bromine at ambient temperature (see Fig. 1c) as well as warm (353 K) octafluoro cyclobutane, krypton or carbon dioxide in a host medium of ambient air are sucked into the SUSF, forming stable rotational ellipsoidal systems around the pressure nodes similar to the cold-gas traps. This indicates that the temperature of the gas is not responsible for the gas trapping in SUSFs. Otherwise, the warmer gas would have to be displaced out of the SUSF. The opposite case was also investigated. Cold helium (173 K) with a low mass density (about a seventh compared to ambient air) is displaced out of the SUSF (see Fig. 2e). Moreover, as seen as in Fig. 2, gas trapping increases with increasing mass density. In the presented series, all sample gases were injected into the SUSF at approximately 170 dB and at the same experimental conditions, i.e., with the same temperature (353 K) and flux rate (10 ml/s). The trapping effect is most remarkable in the case of octafluoro cyclobutane indicated by the highest maximum temperature inside the SUSF at the pressure nodes, Fig. 2a. With decreasing mass density of the sample gas the effect is reduced and totally disappears when sample gases with a lower mass density than the host medium are used (see Fig. 2d). To summarize, there is strong evidence that the difference in mass density but not in temperature of the sample gas and the host medium are governing the effect.

To our knowledge the heavy gas trapping effect in SUSFs has not been described before, and
therefore no theoretical model explaining the phenomenon exists. However, the acoustic levitation of liquid and solid samples has been modelled by Barmatz and Collas [12]. This model is based on two assumptions: (i) the radius of the suspended sample has to be much smaller than the wavelength of the ultrasonic field, and (ii) there has to be a sharp boundary between the surface of the sample and the host medium which leads to scattering effects of the acoustic wave. Scattering is caused by different speed of sound in the sample and the host medium. According to this model we make a first attempt to give an explanation of gas-trapping in SUSFs. To meet the two model conditions we assume small well-separated volume elements of a suspended high density sample gas probe which enters the SUSF. Any kind of gas mixing processes like diffusion and convection is neglected. These assumptions are supported by the facts that binary gas diffusion is a slow process and that turbulent convection can be suppressed at low SPLs.

In a SUSF of an axially tuned levitator with cylindrical symmetry, the normalised sound pressure $p$ and components of velocity $v_z$ and $v_r$ are approximately given by

$$\frac{p(r,z)}{p_{\text{max}}} = \sin(k_z z) \cdot J_0(k_r r),$$  \hspace{1cm} (1a)

$$\frac{v_z(r,z)}{v_{\text{max}}} = \frac{k_z}{k_0} \cos(k_z z) \cdot J_0(k_r r) \quad \text{and}$$

$$\frac{v_r(r,z)}{v_{\text{max}}} = \frac{k_r}{k_0} \sin(k_z z) \cdot J_1(k_r r),$$  \hspace{1cm} (1b)

with the Bessel function $J_i$ of order $i$. The origin of the cylindrical coordinates $z$, $r$ is chosen to be at one of the pressure nodes. The axial and radial wave numbers, $k_z$ and $k_r$, are components of the wave number vector according to

$$k_0^2 = \frac{\omega}{c_0} = k_z^2 + k_r^2.$$  \hspace{1cm} (2)

Sound pressure and velocity of the ultrasonic field lead to (normalised) potential and kinetic energy densities which can be written as

$$E_{\text{pot}} = \sin^2(k_z z) \cdot J_0^2(k_r r),$$  \hspace{1cm} (3)

and

$$E_{\text{kin}} = \left(\frac{k_z}{k_0}\right)^2 \cos^2(k_z z) \cdot J_0^2(k_r r)$$

$$+ \left(\frac{k_r}{k_0}\right)^2 \sin^2(k_z z) \cdot J_1^2(k_r r).$$  \hspace{1cm} (4)

In a first order assumption, the potential energy density is proportional to the square of the normalised sound pressure and the kinetic energy density to the square of the normalised acoustic velocity. As described by Barmatz and Collas [12] the acoustic levitation potential $U$ for small spherical samples in a SUSF, with a mass density $\rho_s$ and a speed of sound $c_s$, in a fluid host medium, with a mass density $\rho_h$ and a speed of sound $c_h$, follows from the energy densities by

$$U = \frac{f_1}{3} E_{\text{pot}} - \frac{f_2}{2} E_{\text{kin}}$$  \hspace{1cm} (5)

with the factors

$$f_1 = 1 - \frac{\rho_0 c_0^2}{\rho_s c_s^2} \quad \text{and} \quad f_2 = \frac{2(\rho_s - \rho_0)}{2\rho_s + \rho_0}.$$  \hspace{1cm} (6)

In case of gaseous test volumes and gaseous host media the factors $f_1$ and $f_2$ can be simplified by use of the relation for ideal gases

$$\rho = \frac{p M}{RT}$$  \hspace{1cm} (6)

and the relation of the speed of sound in gases

$$c = \sqrt{\frac{k \cdot p}{\rho}} = \sqrt{\frac{\kappa RT}{M}},$$  \hspace{1cm} (7)

where $p$ is the main pressure, $T$ the temperature, $M$ the molecular mass and $\kappa$ the adiabatic coefficient of the gas. $R$ represents the universal gas constant. In this way, the equations for the factors $f_1$ and $f_2$ result in

$$f_1 = 1 - \frac{\kappa_0}{\kappa_s} \quad \text{and} \quad f_2 = \frac{2(x - 1)}{2x + 1}$$  \hspace{1cm} (8)

with

$$x = \frac{\rho_s}{\rho_0} = \frac{M_s T_0}{M_0 T_s}.$$  \hspace{1cm} (8)

The factor $f_1$ is zero for gases with the same adiabatic coefficient $\kappa_s = \kappa_0$. In this case the levitation potential is only governed by the kinetic energy of the stationary ultrasonic field (see Eq. (5)).
factor $f_2$ is a function of the relative density $x = \rho_s/\rho_0$, or of the temperatures and the molecular masses of the gases as plotted in Fig. 3. In case of $x > 1$, the factor $f_2$ is positive and leads to an attractive potential while in case of $x < 1$ the factor $f_2$ becomes negative and results in a repulsive potential.

The levitation potential $U$ is illustrated in Fig. 4. The values of the parameters have been adapted to our experimental conditions, i.e., an ultrasonic frequency of 20 kHz, a wave number ratio $k_r/k_z$ of 0.7, $f_1 = 0$ and $f_2 = 0.32$, corresponding to the case of precooled nitrogen as sample gas. In this case, Eq. (5) leads to an attractive potential for small test volumes. At the pressure nodes of the SUSF, potential wells appear which are filled up with the sample gas of higher mass density and form the described zones of trapped gas around the pressure nodes. A repulsive potential appears from Eq. (5) if $f_1$ is approximately zero and $f_2$ is negative. The factor $f_2$ is always negative for sample gases with a lower mass density than the host medium. These gases are displaced from the SUSF.

From the levitation potential follow the axial and radial levitation forces $F_z$ and $F_r$, as components of the vectorial levitation force

$$\mathbf{F} = -\nabla U.$$  \hspace{1cm} (9)

In case of $f_1 = 0$, Eq. (8) results in

$$F_z = \frac{f_2}{2} \frac{k_r}{k_z} \sin(2k_zz) \left[2k_r^2J_0^2 - 2k_z^2J_1^2 \right]$$ \hspace{1cm} (10)

and

$$F_r = \frac{f_2}{2} \frac{k_r}{k_z} \left[k_r^2J_1(j_0 - J_2) \cos^2(k_z) - 2k_r^2J_0J_1 \sin^2(k_z) \right].$$ \hspace{1cm} (11)

Another more qualitative explanation for the reported trapping effect is given by the following. Assuming two nonmiscible fluids with different speeds of sound and with a plain common phase boundary a vertically propagating ultrasonic wave pushes the fluid with the lower speed of sound through the boundary layer into the other fluid independently of the direction of the wave [13]. Transforming this example to two gases with different densities in a SUSF one can expect a similar
result: according to Eq. (7), the gas with the higher mass density has a lower speed of sound and is sucked into the SUSF, then displaces the gas with the lower mass density and forms zones of trapped gas.

In summary, we have presented new experimental results describing the phenomena of cold-gas traps in SUSFs in more detail. The temperature field and its dependence on the strength of the SUSF and the cold-gas source have been measured. Moreover, it is clearly shown that the observed trapping phenomenon also occurs for heavy gases and is predominantly an effect of mass density and not of temperature. A first theoretical description of the effect is given by explaining the experimental results with the theory of acoustic levitation of small liquid and solid samples. In future work trapping conditions have to be optimized. First results have been obtained with stationary sonic fields of 4 kHz. The advantage of lower frequencies is an increasing size of heavy-gas or cold-gas traps. Finally, we will focus on the development of advanced techniques using the phenomenon. There may be possibilities concerning gas separation processes or the development of miniature laboratories for gas or plasma reactions without wall contact.

References