THE FLOW OF A RAREFIED GASEOUS MIXTURE FROM AN ORIFICE

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We investigate the flow of a mixture of gases of markedly different molecular weights through an orifice for a wide range of pressures, including the transitional conditions from continuous to free molecular flow. Quantitative results are given for the flow-rate coefficient of the orifice for the mixtures Ar + He and CO_2 + He and for the change in the concentration following separation of the gases in the transitional conditions.

The investigation of the flow characteristics of nozzles in the transitional conditions from continuous to free molecular flow has been the subject of a number of papers [1-3]. But these investigations were made with single gases and their results cannot determine the flow of a mixture of gases of different molecular weights or of various effective molecular cross sections. When such mixtures expand, diffusion processes in the gradient flow determine the change in the concentrations of the flow components [4].

The aim of this paper is to study the mechanism of the flow of a binary mixture (of a light and a heavy gas) in transitional conditions from viscous to free molecular flow.

To analyze the flow characteristics we introduce the concept of the flow-rate coefficient of the orifice for the separated components as the ratio of the real flow rate G_i of the component to the flow rate $G_{i\text{T}}$ computed from the orifice geometry using inviscid equilibrium flow theory:

$$C_i = \frac{G_i}{G_{i_\text{T}}}. \tag{1}$$

The total flow-rate coefficient is $C_\Sigma = G_\Sigma / G_{\Sigma\text{T}}$. For the continuous inviscid flow of a binary mixture $C_1 = C_2 = C_\Sigma = 1$. In the general case, when the velocity components are equal, $C_1 = C_2 = C_\Sigma \neq 1$

$$C_\Sigma = \frac{N_1 m_1 + N_2 m_2}{N_\text{T} m}, \tag{2}$$

where N_T is the flow of particles in a continuous flow of the gaseous mixture without separation, $m = m_1 f + m_2(1-f)$;

$$C_1 = \frac{N_1}{N_{1\text{T}}}; \qquad C_2 = \frac{N_1}{N_{2\text{T}}}. \tag{3}$$

The inviscid flow of particles of the i-th component is

$$N_{i_\text{T}} = A_* \frac{N_A}{\sqrt{m_i}} \frac{p_{0i}}{\sqrt{RT_0}} \sqrt{\gamma_i \left(\frac{2}{\gamma_i + 1}\right)^{\frac{\gamma_i + 1}{\gamma_i - 1}}}.$$

For a gaseous mixture

$$N_\text{T} = A_* \frac{N_A}{\sqrt{RT_0}} \sqrt{\gamma \left(\frac{2}{\gamma + 1}\right)^{\frac{\gamma + 1}{\gamma - 1}}} \frac{p}{\sqrt{m_1 f + m_2 (1 - f)}}. \tag{4}$$

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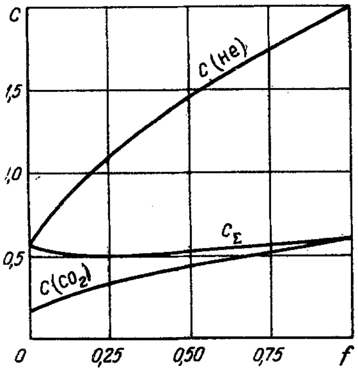

Fig. 1

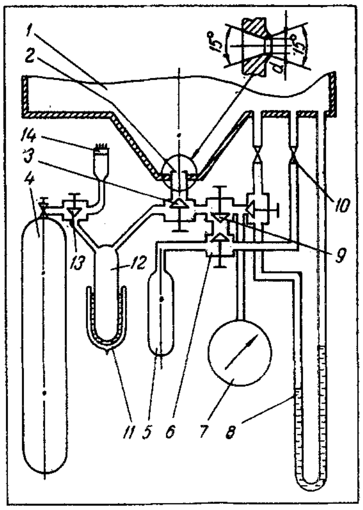

Fig. 2

Fig. 1. Computed flow-rate coefficient for a diaphragm, for the mixture He + CO_2 in molecular conditions.

Fig. 2. Diagram of the working part of the apparatus.

For the separate components

$$N_{1\text{T}} = N_{\text{T}} f; \tag{5}$$

$$N_{2\text{T}} = N_{\text{T}} (1 - f). \tag{6}$$

The flow of the particles of the separate components can be determined from the change in the parameters in the braking chamber:

$$N_1 = -\frac{d}{dt}\left(fp\frac{VN_A}{RT_0}\right) = -\frac{VN_A}{RT_0}\left(p\frac{df}{dt} + f\frac{dp}{dt}\right) \tag{7}$$

(the gas expansion is at constant temperature T_0 in the volume V across the critical cross section of area A_*),

$$N_2 = \frac{VN_A}{RT_0}\left[p\frac{df}{dt} - (1 - f)\frac{dp}{dt}\right]. \tag{8}$$

From this we have

$$C_\Sigma = \frac{V}{A_* \sqrt{RT_0}} \frac{\left[\frac{df}{dt}(m_2 - m_1) - \frac{1}{p}\frac{dp}{dt}m\right]}{\sqrt{\gamma\left(\frac{2}{\gamma+1}\right)^{\frac{\gamma+1}{\gamma-1}}}\sqrt{m}}, \tag{9}$$

$$C_1 = -\frac{V\sqrt{m}}{A_*\sqrt{RT_0}\sqrt{\gamma\left(\frac{2}{\gamma+1}\right)^{\frac{\gamma+1}{\gamma-1}}}}\left[\frac{1}{f}\frac{df}{dt} + \frac{1}{p}\frac{dp}{dt}\right], \tag{10}$$

$$C_2 = \frac{V\sqrt{m}}{A_*\sqrt{RT_0}\sqrt{\gamma\left(\frac{2}{\gamma+1}\right)^{\frac{\gamma+1}{\gamma-1}}}}\left[\frac{1}{1-f}\frac{df}{dt} - \frac{1}{p}\frac{dp}{dt}\right]. \tag{11}$$

Thus, by measuring the pressure and the composition of the mixture in the volume from time to time we can compute the flow-rate coefficient.

The case of the stationary flow of a mixture through an orifice can be computed exactly in molecular conditions. We find the equation for the pressure change in the chamber and for the flow-rate coefficients of a binary mixture.

The flow rate for the various components out of the vessel is given by the equation

$$N_i = \frac{1}{4} u_i A_* n_i. \tag{12}$$

Here u_i is the arithmetic mean of the thermal velocities of the particles of one component. On the other hand,

$$N_i = -V\frac{dn_i}{dt}, \quad \frac{dn_i}{n_i} = -\frac{1}{4}\frac{A_*}{V}u_i dt. \tag{13}$$

Integration of this equation yields

$$n_i = n_{0i}\exp\left[-\frac{1}{4}\frac{A_*}{V}u_j t\right]. \tag{14}$$

Let $(1/4)(A_*/V)u_i = \alpha_i$. Since for a binary mixture,

$$f = \frac{n_1}{n_1 + n_2}, \tag{15}$$

we have

$$f = f_0\frac{\exp\{-(\alpha_1 - \alpha_2)t\}}{1 + f_0[1 - \exp\{-(\alpha_1 - \alpha_2)t\}]}. \tag{16}$$

In accordance with Dalton's law for the flow of a gas out of a vessel at constant temperature

$$p = kT_0[n_{01}\exp(-\alpha_1 t) + n_{02}\exp(-\alpha_2 t)], \tag{17}$$

$$\bar{p} = \frac{p}{p_0} = f_0\exp(-\alpha_1 t) + (1 - f_0)\exp(-\alpha_2 t). \tag{18}$$

In (12) we replace the values of the velocities u_i by their expressions from molecular kinetic theory:

$$u_i = \sqrt{\frac{8kTN_A}{\pi m_i}}.$$

Further, by (2) and (3), we have

$$C_{\Sigma_M} = \frac{1}{\sqrt{2\pi}\sqrt{\gamma\left(\frac{2}{\gamma+1}\right)^{\frac{\gamma+1}{\gamma-1}}}}\,\frac{f\sqrt{m_1} + (1-f)\sqrt{m_2}}{\sqrt{m}}, \tag{19}$$

$$C_{1_M} = \frac{1}{\sqrt{2\pi}\sqrt{\gamma\left(\frac{2}{\gamma+1}\right)^{\frac{\gamma+1}{\gamma-1}}}}\sqrt{\frac{m}{m_1}}. \tag{20}$$

$$C_{2_M} = \frac{1}{\sqrt{2\pi}\sqrt{\gamma\left(\frac{2}{\gamma+1}\right)^{\frac{\gamma+1}{\gamma-1}}}}\sqrt{\frac{m}{m_2}}. \tag{21}$$

Physically, Eqs. (19)-(21) denote the limits to which the real flow-rate coefficients tend as the conditions in the flow of a mixture through an orifice tend to the free molecular.

Figure 1 gives the results of numerical computations of the flow-rate coefficient in molecular conditions for the mixture He + CO_2. In these calculations the ratio of the specific heats for the mixture was taken as

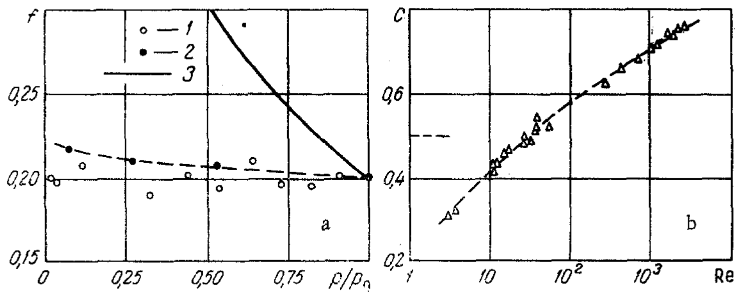

Fig. 3. The flow of a mixture of 20% Ar + 80% He: a) concentration of argon in the cylinder; 1, 2) p_0 = 2190; 42.3 mm Hg, respectively; 3) calculation for molecular conditions; b) flow-rate coefficient.

$$\gamma_\Sigma = \frac{c_{V_1}\gamma_1 f\frac{m_1}{m} + c_{V_2}\gamma_2(1-f)\frac{m_2}{m}}{c_{V_1} f\frac{m_1}{m} + c_{V_2}(1-f)\frac{m_2}{m}},$$

or

$$\gamma_\Sigma = 1 + \frac{1}{\frac{1}{\gamma_1 - 1} f\frac{m_1}{m} + \frac{1}{\gamma_2 - 1}(1-f)\frac{m_2}{m}}. \tag{22}$$

Since the equation for the flow-rate coefficient cannot be given in analytic form in the transitional regime between continuous and free-molecular flow, it is appropriate to determine it experimentally.

In the experiments we used a vacuum system low density wind tunnel with mains supply of the components and apparatus for measuring the gas concentrations in the vessel (Fig. 2).

The mixture flows from the vessel 4 of capacity 30.4 liters through the valve 3 and the nozzle 2 into the vacuum chamber 1 provided with an electron beam system to measure the concentrations of the components. As the mixture flows out the pressure change in the cylinder is recorded from time to time as is the composition of the mixture. The pressure is measured by a manometer 7 or a U-tube manometer 8. Test samples are taken from the cylinder through the valves 6 and 9 using the vessel 5. Then the gas sample is fed to the vacuum chamber. The sample size is chosen so that the pressure in the vacuum chamber is in the range $1\text{-}1.5 \cdot 10^{-2}$ mm Hg.

The composition of the sample is measured by the ratio of the intensity of the radiation of the lines of the components excited by the electron beam with energy 10 kV at a current of 1-3 mA:

$$\frac{n(\mathrm{Ar})}{n(\mathrm{He})} = B\frac{I(\mathrm{Ar})}{I(\mathrm{He})}.$$

To record the radiation of helium the line 5016 Å was used; for argon the region 4200 ± 50 Å was used. The measuring apparatus comprised the spectrograph ISP-51, the photomultiplier FÉU-27 and the recording instrument ÉPPV-60. The coefficient B = 2.89 was found by preliminary calibration in static conditions. The error in measuring the concentration by this method did not exceed ±5% in our experiments.

A mixture of given concentration was prepared by filling the cylinder with the components under pressure using the valves 6 and 9. Before the experiment began the cylinder containing the mixture was stored for 24 h to allow the components to mix.

In setting up the experiment it was observed that the air flow had an effect on the intensity I(Ar) to be measured. This effect was taken into account by taking all the measurements of the concentrations at fixed times and correcting I(Ar) with a correction computed from a preliminary measurement of the incident air flow.

In the experiments a mixture with an initial concentration of 20% Ar + 80% He was used. The gas flowed into the vacuum through an orifice pierced by a needle in a copper plate of thickness 0.3 mm. The

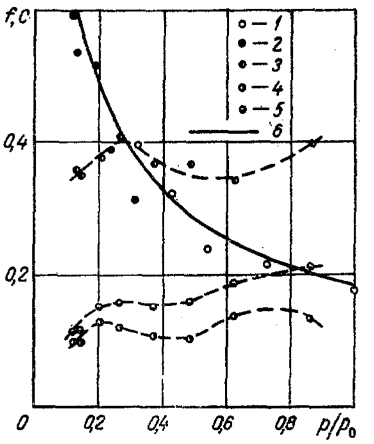

Fig. 4. Flow of the mixture CO_2 + He (p_0 = 4.74 mm Hg): 1, 2) CO_2 concentration in the cylinder; 6) calculation for molecular conditions; 3, 4, 5) flow-rate coefficient for the orifice for He, CO_2, and total.

shape of the channel thus obtained is shown in Fig. 2. The smallest diameter d = 0.203 ± 0.001 mm was measured using a microscope.

The experiments were made at different ranges of Knudsen numbers. In both experiments the pressure in the vacuum chamber was maintained at not more than 10^{-3} mm Hg.

Experiment 1. The mixture with initial cylinder pressure p_0 = 2190 mm Hg was released into the vacuum chamber until the pressure built up to 44.1 mm Hg in 15,880 sec. The results for the concentration, shown in Fig. 3a as a function of the pressure, correspond to a range of Knudsen numbers Kn = $2.0 \cdot 10^{-4}$–$1.2 \cdot 10^{-2}$. Each point is the average of three measurements. No increase in the concentration of argon in the cylinder was observed.

Experiment 2. The mixture with initial pressure p_0 = 42.3 mm Hg flowed from the cylinder until the pressure reached 2.94 mm Hg in 16,000 sec. The range of Knudsen numbers was 0.012–0.150. As in the first experiment, the pressure and concentration in the cylinder were measured. Each point is the average of three measurements. From Fig. 3a we see that the argon concentration in the cylinder increases from 20 to 21.8%.

To illustrate the experimental results qualitatively, Fig. 3a shows the computed curve for the molecular flow of a mixture from the cylinder through a diaphragm of diameter 0.2032 mm obtained from (16) and (18).

It should be noted that there is a marked difference in the composition of the mixture in the cylinder from the initial value beginning at Kn = $1.2 \cdot 10^{-2}$, which corresponds to the beginning of transitional conditions.

Figure 3b gives the flow-rate coefficient of the nozzle, computed from (9) and from the experimental results. To the left on the vertical axis is shown the limit of the flow-rate coefficient for a thin diaphragm for a mixture with f = 0.2 in molecular conditions. The qualitative disagreement is explained by the fact that the shape of the nozzle is different from that of a thin diaphragm.

The limit of the experimental curve for C_Σ(Re) in the region of Re = 20 is evidently explained by the merging of the boundary layers at the critical section. This is confirmed by calculation of the relative displacement thickness of the boundary layer at the critical section. The Reynolds number Re = 20 corresponds to $\delta^*/d = 0.152$. For a velocity distribution in the shape of a second degree parabola, $\delta^* = \delta/3$. Hence in our case we can assume approximately that $\delta = 0.456d$.

Since, when Kn = 0.151, the sample filling the chamber in the determination of the concentration is of the same order of size as that of the whole gas in the cylinder, at low pressures the method described above can no longer be used. Hence in experiments at large Kn, we developed and used a method of analyzing the mixture He + CO_2 based on freezing the CO_2 with liquid nitrogen, for which we used the trap 12 (Fig. 2) which can be cut off from the cylinder 4 by the valve 13. The trap can be cooled to a low temperature by placing it in a Dewar flask 11 filled with liquid nitrogen. The thermocouple lamp 14 controls the pressure in the trap. To measure the pressure in the trap we used a U-shaped oil-filled manometer with a scale accuracy of ±0.01 mm.

The concentration was determined from pressure measurements in the closed frozen trap. The first measurement was taken when the trap was filled with the mixture to be analyzed. Then the trap was set in the Dewar flask filled with liquid nitrogen and the CO_2 in the cavity of the trap was frozen at its walls. The remaining helium was pumped through the valves 9, 6, and 10 until the pressure did not exceed $5 \cdot 10^{-4}$ mm Hg. Then the valves were closed and the whole system was heated to the temperature which it had at the first pressure measurement. The second measurement was now of the pure CO_2 remaining in the cavity of the trap. The concentration of the CO_2 in the original mixture was

$$f = \frac{p_{\mathrm{II}}}{p_{\mathrm{I}}}.$$

The accuracy of the concentration measurement in the method described above is determined by the accuracy of the pressure measurements, the accuracy of observation of isothermal conditions, the absence of an incident air flow on the system, and the degree of degassing of the internal surfaces. The error in the determination of the concentration did not exceed ±5%.

A mixture with the molar composition 18.5% CO_2 + 81.5% He was released from the cylinder through the nozzle with initial pressure 4.74 mm Hg until the pressure was 0.5 mm Hg, which corresponds to a change in Kn from 0.15 to 1.33.

From the results in Fig. 4, we see that the CO_2 concentration in the cylinder increases from 20 to 60%. For comparison the curve obtained from (16), (18) for a thin diaphragm is given. The open and black circles denote the experimental results obtained using various freezing traps with an interval of 14 h. The discrepancy of 18% between the results before and after the interval provides a basis for assuming that as the gas flows out there is a difference between the composition of the mixture in the cylinder 4 (Fig. 2) and in the pipes leading the mixture to the nozzle 2. With the elements of the gas mains used (the cross section of the valve at the cylinder was 3 mm, the diameter of the pipes was 10 mm, the pipe length was 1 m) concentration diffusion could not equalize the compositions of the gases.

Figure 4 shows also the flow-rate coefficient for the orifice: the total for the He + CO_2 mixture and for the components, computed from the experimental results using (9), (10), (11). The significant difference in the flow-rate coefficients for the components is evidence that the local velocities of the components were different. The velocity of the atoms of helium is significantly greater than that of CO_2 molecules, which results in an increase in the proportion of the heavy component in the cylinder.

The above effect has to be taken into account in mass-spectrographic measurements and in all those cases when we have have to deal with the flows of gaseous mixtures in transitional or free-molecular conditions.

NOTATION

C	is the flow-rate coefficient;
G	is the actual mass flow rate;
N	is the flow of particles;
m	is the molecular weight;
N_A	is Avagadro's number;
γ	is the ratio of specific heats;
p	is the pressure;
R	is the universal gas constant;
f	is the molar concentration of the heavy component;
t	is the time;
n	is the particle density;
I	is the intensity of component radiation;
δ	is the boundary layer thickness;
η	is the viscosity;
$Re = 4G/\pi d\eta_0$	is the Reynolds number;
$Kn = \bar{l}/d$	is the Knudsen number;
$\bar{l}$	is the particle mean free path;
c_V	is the specific heat at constant volume;
δ^*	is the displacement thickness of the boundary layer.

Subscripts

1	denotes the heavy component parameter;
2	denotes the light component parameter;
0	denotes the stagnation parameter;
T	denotes the theoretical parameter;
Σ	denotes the mixture parameter.

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