GENERATION OF AN INTENSE MOLECULAR BEAM OF CO_2 BY A GASDYNAMIC METHOD

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The idea of using the core of a jet of expanding gas to form a molecular beam has led to the construction of complex gasdynamic facilities and has prescribed a number of specific requirements for the creation of high-intensity molecular beams. Two basic requirements are high pumping speed and a skimmer, the first element in the beam-generating system, which does not react appreciably on the jet. The present article gives the results of an experimental investigation of conditions for creating a molecular beam from a jet of carbon dioxide downstream of a sonic nozzle. The position of maximum intensity with room temperature gas in the source is given by the group of variables $(p_0 d_*)^{0.4}$. Kn_∞. By measuring the intensity and by mass-spectrometric analysis of a molecular beam for specific conditions we have established the CO_2 pressure in the stagnation chamber at which condensation begins. The investigations were carried out in a molecular-beam generator with cryogenic pumping.

The interaction of a jet with a skimmer has been the subject of lengthy discussion [1-5], which has sought to elucidate the discrepancy between experimental and theoretical values of the absolute beam intensity. A detailed study of the effect of skimmer geometry on the molecuar beam intensity has been given in [6, 7]. However, the flow conditions in these references differed widely, while the jet structure was diffuse, as a rule. Penetration of the background gas into the jet core appreciably changes the molecular-beam parameters [8-11]. This makes it difficult to correlate the results of beam-intensity measurements for the different skimmers.

Additional problems arise in expansions with condensation. The effect of the skimmer on the characteristics of a condensed molecular beam, in particular, have not been studied.

The present paper gives the results of an investigation of the intensity of a CO_2 molecular beam formed from a jet downstream of a sonic nozzle, over a wide range of stagnation pressure (from expansion of a homogeneous gas to flow with condensation), with nozzle–skimmer distance varying from $x = l_{n-sk}/d_* = 1$ to 134 calibers. Special attention was paid to the study of conditions for obtaining a molecular beam with maximum intensity, and with weak interaction between the gas flow and the skimmer.

As well as measuring the total beam intensity, we conducted a mass-spectrometric analysis of its composition. This allowed us to determine the conditions for the start of condensation in the expanding flow and to determine the effect of this on the intensity of the molecular beam.

The experiments were carried out on a molecular-beam generator with cryogenic pumping [12]. A general view of the generator and the arrangement of the instrumentation is shown in Fig. 1, where 1-3 are sections of the skimmer, the collimator, and the working area, 4-6 are nitrogen–helium cryogenic pumps, 7 is a lamp, 8, 9 are windows, 10 is the CO_2 source, 11 is the skimmer, 12 is the collimator, 13 is a screen, 14 is the effuser source, 15 is a modulator, 16 is a total intensity sensor, and 17 is the sensor of a type MX-7301 mass spectrometer. The experimental instrumentation 10-17 was mounted on traverse devices which allowed it to be moved under vacuum conditions for adjustment and for calibration, and during the experi-

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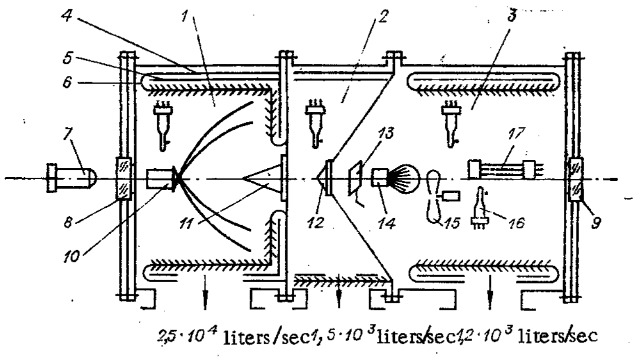

Fig. 1

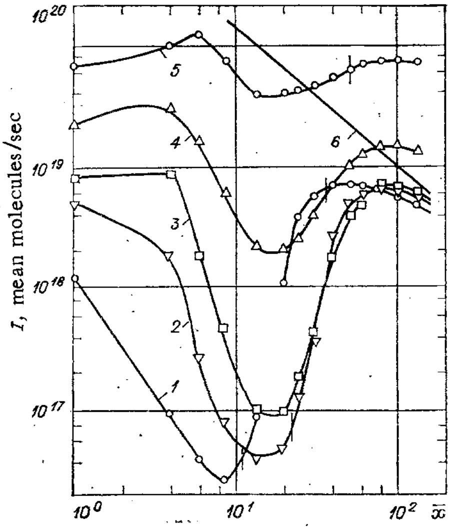

Fig. 2

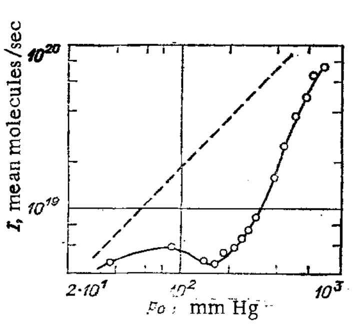

Fig. 3

ment. The adjustments were carried out using the lamp for step-by-step adjustment of the skimmer, collimator, source, sensor, and effuser source (during calibration).

The following items were used in the experiments: a sonic nozzle of diameter 1.91 mm; an aluminum skimmer in the shape of a truncated hollow cone of internal opening angle 40°, and external angle 45° with entrance aperture diameter of 4.11 mm, and length of conicl part 116 mm; a collimator with entrance aperture diameter 3.24 mm, a total intensity sensor, a type LM-2 ionization tube with sensitive aperture diameter 3 mm; a type MX-7301 analyzer with collimating aperture diameter 2.5 mm. The skimmer–detector distance was 887 mm. The method of synchronous detection [12] was used in recording the molecular beam.

The experiments were conducted with technically pure carbon dioxide. The gas temperature in the source was maintained at room value, and the pressure was varied from 33 to 742 mm Hg. The rarefaction at sections of the molecular-beam generator was varied (in mm Hg) from $8 \cdot 10^{-6}$ to $6 \cdot 10^{-5}$ in the skimmer, from $1 \cdot 10^{-6}$ to $4 \cdot 10^{-6}$ in the collimator, and from $9 \cdot 10^{-7}$ to $2 \cdot 10^{-6}$ in the working section. The vacuum characteristics of the molecular-beam generator have been published in [13].

Figure 2 shows several typical relationships between the absolute intensity I as a function of the nozzle–skimmer distance x, normalized by the nozzle diameter. The pressure in the source was 33.2, 134.2, 210.1, 361.5, and 742 mm Hg (curves 1-5, respectively). The numeral 6 denotes the theoretical dependence of I_T on x, obtained for $\gamma = 1.4$ and $p_0 = 33.2$ mm Hg in the case of free-molecular flow over the skimmer [14]. Thevalue of the adiabatic exponent is not known. However, for qualitative comparison of the calculated data with the experiments one can take $\gamma = 1.4$. This is close to the actual values, since the vibrational–translational relaxation was frozen [15] in all of the investigations.

It has been established experimentally that for $p_0 < 190$ mm Hg the expansion of CO_2 takes place without condensation. For these conditions we examine the variation of molecular beam intensity as a function of the relative nozzle–skimmer distance.

TABLE 1

p_0, mm Hg	33.2	83,8	134,2	152,5	184,8
I/I_T	0,35	0,25	0,17	0,15	0,14
x	49,5	84,5	111	119	133
M_∞	17,0	21,4	23,8	24,5	25,8

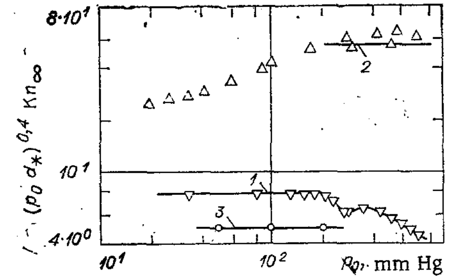

Fig. 4

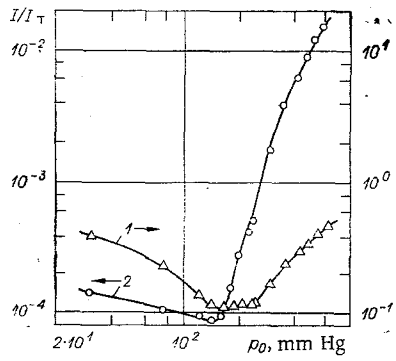

Fig. 5

We compare the skimmer with the mass flux sensor. The basic condition for normal functioning of the ρu-sensor is that the nose density shock should be swallowed. This occurs for $\mathrm{Re}_{\infty,2}=\rho_\infty u_\infty d/\mu_2 > 10$ [16] (here ρ_∞, u_∞ are the gas density and velocity at the skimmer entrance; d is the skimmer diameter; and μ_2 is the gas viscosity behind the shock). The values $\mathrm{Re}_{\infty 2}=10$ in Fig. 2 are indicated by a vertical line. In all cases these values lie to the right of the beam intensity minimum, for which $\mathrm{Re}_{\infty,2}$ varies in the range 19-23, i.e., the nose shock is swallowed in the region of the minimum. Therefore, the nature of the relationship $I = f(x)$ in the section before the minimum intensity is determined exclusively by processes within the skimmer, which vary in accordance with the variations in the Mach and Reynolds numbers at the skimmer entrance.

Since no flow investigations were made inside the skimmer, our representations of the nature of the dependence of the intensity must be constructed from the general qualitative observations.

The flow in the skimmer can be considered as flow in a nozzle with a given velocity distribution at the entrance, for $M_\infty > 1$. For flow emerging from this nozzle the collimator performs the role of a skimmer.

We note that factors such as the appearance of shock waves inside the nozzle, boundary-layer growth, and withdrawing the skimmer from the nozzle should lead to a reduction in the absolute beam intensity

The presence of a minimum following the sharp decrease in intensity in the region where the nose shock is swallowed is evidence of attenuation of the effects of conversion of kinetic flow energy into thermal energy in the shock waves. This may be associated with a corresponding adjustment in the shock structure (for example, transition from a Mach disk to an x-shaped configuration and to a general disappearance of shocks on the flow axis). The effect of these factors continues to be seen even at greater x distances.

As is known [16], measurments made by ρu-sensors give low values of mass flux for $\mathrm{Re}_{\infty,2} \leqslant 10$. The reason is that for increased flow rarefaction the displacement thickness increases and the effective diameter of the sensor inlet aperture decreases.

In our case in the region $\mathrm{Re}_{\infty,2} \leqslant 10$, in spite of the decrease in density of the incident stream with increase of x, there is a sharp increase in the recorded intensity following the minimum. This is evidence of improved penetration of the viscous region inside the skimmer, and of a gradual transition to molecular conditions in the flow over the skimmer and the internal flow.

Figure 3 shows the molecular beam intensity as a function of pressure in the source for x=134, i.e., maximum nozzle–skimmer distance in the present experiments. Here the broken line for the particular case $\gamma=1.4$ shows the theoretical dependence of the intensity on p_0.

Figure 3 shows that in the region $p_0 < 190$ mm Hg, where the CO_2 expands without condensation, the beam intensity varies very little with increase in pressure. Factors inhibiting the increase in intensity may

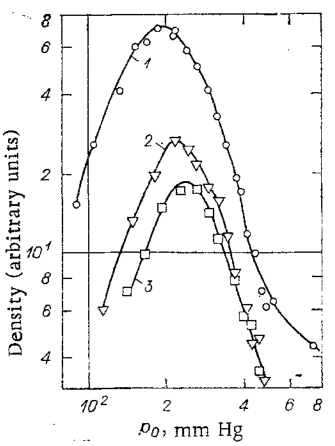

Fig. 6

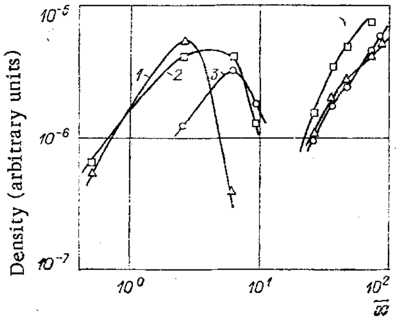

Fig. 7

include: 1) increase of interaction with the skimmer; 2) attenuation in the nonequilibrium expansion and associated decrease in the adiabatic exponent; and 3) the development of a free-condensation process, an increase in the number of dimers, trimers, and so on.

We now consider the effect of pressure p_0 on beam intensity at constant Knudsen number (Kn_∞) at the skimmer entrance. The parameter Kn_∞ is determined in terms of the incident stream parameters (we use a viscosity–temperature relationship with exponent 0.87 [17]) and the skimmer diameter. The parameter Kn_∞, along with the Mach number (M_∞), which can be found, for example, from the relations given in [18], determine the conditions for interaction of the gas with the nose of a skimmer of given geometry and constant temperature.

With an increase of p_0 and with $Kn_\infty = \text{const}$ the nozzle–skimmer distance increases, and therefore, if the adiabatic index is held constant, the Mach number at the skimmer entrance increases. Under these conditions an increase in M_∞ must lead to less scattering of molecules during interaction with the skimmer, and there must therefore be an increase in the relative beam intensity, normalized to the theoretical value at some value of $\gamma = \text{const}$. The data in Table 1 are evidence of a contrary tendency. (The table shows values of normalized beam intensity I/I_T in the region $p_0 = 33$-185 mm Hg, and also the values of nozzle–skimmer distance and Mach number obtained for $Kn_\infty = 1.5$ and $\gamma = 1.4$.)

From what has been said it follows that the presence of flow interaction with the skimmer cannot explain the relative decrease in the beam intensity to the right of the maximum on the curves (Fig. 2) for the conditions examined.

We turn now to examination of the relaxation effects associated with energy exchange in the internal degrees of freedom.

According to estimates using the technique of [19], the translational relaxation may be frozen in the range of p_0 and x investigated; and the process of vibrational–translational CO_2 relaxation is frozen [15]. The quantity $p_0 d_*$ varied approximately in the range 60 to 400 mm Hg · mm, and there was not rotational equilibrium in the jet [20, 21]. Therefore, the CO_2 expansion can proceed with adiabatic exponent γ even greater than 1.4, and an increase in p_0 leads to a decrease in γ. For a jet with free boundaries the decrease in the exponent with increase of $p_0 d_*$ is accompanied by a deviation from the equilibrium density distribution below the isentropic value and a decrease in Mach number on the jet axis. For example, for a variation in γ from 1.5 to 1.4 the calculated beam intensity decreases by approximately a factor of 3.3.

A third factor is the precondensation process, which also leads to a decrease in the mean-mass adiabatic index, as a result of the release of condensation heat. However, this effect can hardly be decisive prior to the onset of condensation, because of the small fraction of particles formed in the total flow.

Therefore, the nature of the dependence of I on p_0 shown in Fig. 3 for expansion without condensation ($p_0 < 190$ mm Hg) is determined mostly by the nonequilibrium nature of the expansion and by the associated changes in γ.

A matter of very great interest, from the viewpoint of finding the optimum nozzle–skimmer distance, is the position of the maximum on the intensity curves. In our work we tried to correlate the available experimental information, in order to determine conditions for which a maximum intensity is found for a given molecular-beam system. Reduction of the experimental results of the present work and of [6, 9] has shown a correlation between the group p_0d_* which describes the conditions of gas expansion in the jet at a given stagnation temperature, and Kn_∞, which describes the conditions for flow over the skimmer. It was shown that with room temperature CO_2 and N_2 as sources the position of the maximum was described by a constant value in the group $(p_0d_*)^{0.4} \cdot Kn_\infty = \text{const}$. Figure 4 shows the parameter $(p_0d_*)^{0.4} \times Kn_\infty$ as a function of pressure in the source. Curve 1 refers to the present results (skimmer of 40/45°), and curve 2 shows the results of [6] with N_2 (skimmer 25/32°, with diameters of 0.76 and 1.12 mm). Curve 3 shows the data of [9] with CO_2 (30/40 skimmer). In the present work the group $(p_0d_*)^{0.4}\,Kn_\infty = 7.7$ (mm Hg · mm)$^{0.4}$ up to $p_0 \approx 190$ mm Hg. In [6] (see Fig. 4, curve 2) the deviation of the quantity $(p_0d_*)^{0.4} \cdot Kn_\infty$ from a constant value is evidently connected with the effect of the molecular background at the expansion section. It was shown in [9-11] that the increase in background pressure at the skimmer section and constant p_0 leads to a displacement in the position of the maximum intensity upstream with a simultaneous decrease in absolute intensity level. It should be noted that the penetration of gas from the surrounding volume is due not only to the level of the background pressure, but also to the flow conditions, i.e., to the penetration of the jet. It was shown in [22] that the parameter $Re_L = Re_*/(p_0/p_1)^{1/2}$ can be used as a criterion for penetration of the jet by molecules of the surrounding space, where Re_* is the Reynolds number based on parameters at the sonic section and the nozzle exit diameter, and p_1 is the background pressure. In [6] (see Fig. 4, curve 2) the effect of background became noticeable for $p_0 \leq 150$ mm Hg; $p_1 \leq 4 \cdot 10^{-3}$ mm Hg and $Re_L \leq 50$.

We now consider the special features apparent in the present work as regards the dependence of the intensity of a CO_2 molecular beam on pressure in the region $p_0 \geq 190$ mm Hg. It can be seen in Fig. 3 that, starting at $p_0 \approx 190$ mm Hg, the subsequent increase in pressure in the source leads to an increase in beam intensity. Even more evident is a change in the nature of the dependence of intensity in p_0 in Fig. 5, which shows the dependence of the ratio I/I_T normalized to the theoretical intensity, on p_0 at the points of maximum (curve 1) and minimum (curve 2). The decrease in I/I_T changes at $p_0 \approx 190$ mm Hg to a sharp increase. The effect observed cannot be explained on the basis of the usual ideas of expansion of a homogeneous gas stream. To explain it we must therefore look to ideas regarding condensation in an expanding stream.

A derivation that an increase in intensity is associated with the condensation process was first made in [23] and was confirmed in [24, 25] in the study of condensed particles. The condensation process leads to a reduction in the mean-mass adiabatic index, and therefore, to a reduction in the number density of particles on the jet axis, compared with the isentropic distribution. This is confirmed by the results shown in Fig. 4. In Fig. 4 it can be seen that in the region $p_0 = 190$–742 mm Hg the parameter $(p_0d_*)^{0.4} \cdot Kn_\infty$ does not remain constant with increase in p_0, but diminishes continuously. It is interesting that the nozzle-skimmer distance at which the maximum and minimum intensities are obtained remains practically unchanged (see Fig. 2) in spite of the increase in p_0.

The increase in molecular-beam intensity seen in Figs. 3 and 5 in the region $p_0 > 190$ mm Hg is apparently a result of nonequilibrium enrichment of the jet axis by heavy clusters of particles. On collision with the walls in the intensity sensor the clusters decompose right down to monomers; there is an increase in the number of particles within the sensor volume and in the recorded signal.

The increase in the number of particles in conversion to monomers on the jet axis may be associated with the fact that the thermal velocity of the clusters decreases in the condensation process, i.e., the dispersion of the cluster particles diminishes. In this case an increase in intensity with increase of p_0 is due, firstly, to the increase in the total amount of condensate, and, secondly, to an upstream shift in the condensation front [26]. In the present experiments the effect of condensation and separation for $p_0 > 190$ mm Hg is apparent even at distance $x \leq 4$, and the beam intensity in the initial expansion section not only does not decrease, but even increases somewhat with increase in x (see Fig. 2).

From Figs. 2 and 5 (curve 2), we can observe that the presence of condensate particles in the flow has the strongest effect on the increase of intensity in the region where there is maximum influence of viscous effects, i.e., in the regions of minimum absolute molecular-beam intensity. Evidently scattering of clusters is appreciably less than for monomers.

The use of mass-spectrometric diagnostics has enabled us to obtain the density distribution of dimers, trimers, and quadrimers in the molecular beam of CO_2 as a function of p_0 and x. Figure 6 shows the dependence of density of dimers, trimers, and quadrimers (curves 1-3) on p_0 for $x = 77$. An increase in the nozzle–skimmer distance does not change the nature of the dependence. The maximum for the dimers is seen at $p_0 = 190$ mm Hg.

The reduction in concentration of dimers, trimers, and quadrimers for $p_0 > 190$ mm Hg is evidence that there is a transition in the process of nuclei formation to a process of actual condensation, i.e., a vigorous growth of droplets after nuclei of critical size are reached. The origin of this process may be connected with the moment of formation of maximum dimer concentration, and assumptions were made in [25, 27, 28] to describe thus, by the quantity $p_0 d_*$ at fixed gas temperature in the source. In our case $p_0 d_* \approx 365$ mm Hg · mm.

Figure 7 shows the dimer density as a function of x for source pressures: 120, 200, and 300 mm Hg (curves 1-3, respectively). A decrease in the number of dimers is observed only in the region where there is greatest interaction of the flow with the skimmer.

The results presented show how complex is the dependence of beam characteristics on the physical process in the expansion of gases and on the conditions of interaction between the flow and skimmer. In many cases [3, 6, 8, 10, 11] a decisive factor is the flow regime and the residual gas pressure in the expansion section. The data obtained have improved the conditions for choice of optimum parameters of a molecular-beam system. This question requires further development.

The molecular beam generator described here, as the first measurements show, can be used to study the process of nonequilibrium gas condensation.

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