NONEQUILIBRIUM EXPANSION OF CARBON DIOXIDE GAS AT RETARDATION TEMPERATURES UP TO 1200°K

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INTRODUCTION

In the expansion of molecular gases, qualitatively differing relaxation processes may take place at the same time, e.g., nonequilibrium condensation, vibrational relaxation, and rotational relaxation. No theoretical methods have been developed for the consideration of such flows at the kinetic level, nor have there been any systematic experimental investigations specially aimed at establishing limitations to the influence of individual relaxation processes. In individual experiments regarding the jet expansion of carbon dioxide gas subjected to electron-beam diagnostics [1, 2], contradictions occur in the general conclusions relating to the expansion of the gas along the axis of the flow. According to [1] the adiabatic index $\gamma = 1.29$ for the expansion of CO_2; moreover, no influence of condensation on the gasdynamic structure of the flow is observed. According to [2], in which flow is considered from the initial stage of condensation, it is concluded that expansion takes place with a constant adiabatic index $\gamma = 1.4$.

In neither case was the whole course of the expansion considered, but only the change in parameters which occurred on passing downstream from a certain point on the axis of the jet to which the gas had expanded after subjection to a complex sequence of equilibrium and nonequilibrium processes. Hence the values of the adiabatic index given in [1, 2] do not constitute an exact characteristic of the whole expansion process.

The main aim of the present investigation is that of obtaining quantitative data as to the influence of vibrational relaxation and the initial stage of condensation under nonequilibrium conditions on the expansion of CO_2 in the jet behind a sonic nozzle. By the initial stage of condensation in the present case we mean the formation of clusters, with the subsequent freezing of their dimensions under conditions in which no condensation jump occurs.

The characteristics of the nonequilibrium expansion of CO_2 were studied on the basis of local density measurements with the aid of an electron beam.

§ 1. The experiments were carried out in a gasdynamic vacuum apparatus [3] provided with the means of measuring local density by reference to the radiation of the gas excited by the electron beam in the optical

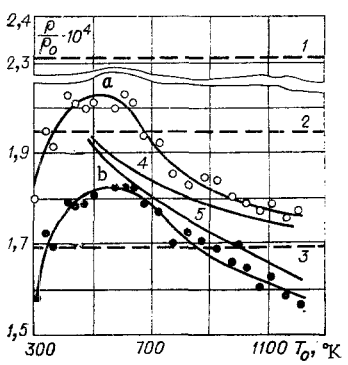

Fig. 1

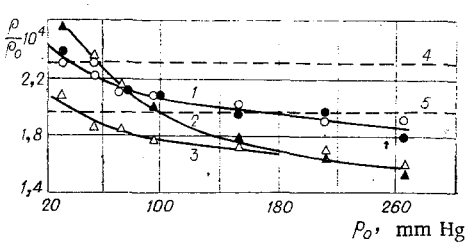

Fig. 2

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and x-ray parts of the spectrum. We used sonic nozzles with critical cross-sectional diameters of d = 2 and 2.85 mm. The retardation temperature T_0 was varied from room temperature to 1200°K by means of an ohmic heater [4]. The range of retardation pressures p_0 for the nozzles was 320-590 and 220-450 mm Hg, respectively.

A leading methodical characteristic of the experiments was the execution of measurements at fixed distances x from the tip of the nozzle for a constant gas flow and pressure in the vacuum chamber p_n, the retardation temperature being revised from room temperature to the maximum value at a fixed heating rate. This method avoided errors in determining the distance from the tip of the nozzle to the point of measurement and also almost entirely eliminated errors due to the scattering of the electron beam. The elimination of these errors was very important for jet expansion in vacuo, especially in regions with high density gradients. As a result of the measurements taken the error in determining the density was no greater than ± 5%.

Measurement of the density at a fixed point on the axis of the jet was carried out simultaneously by two methods: by reference to the x-ray brems radiation (using ordinary x-ray apparatus) and by reference to optical radiation in the spectral region of 2890 Å (transition $B^2\Sigma_u \rightarrow X^2\Pi_g$), using an SPM-2 monochromator. We found that the optical measurements gave systematic results approximately 10% lower than the x-ray values. Special methodical investigations confirmed the validity of the x-ray measurements. We also found that the systematic difference between the optical and x-ray measurements was due to differences in the spatial resolution of the two methods.

§ 2. Figure 1 shows the dependence of the relative density ρ/ρ_0 (ρ is the density at the point of measurement; ρ_0 is that in the retardation chamber) at a distance x/d = 20 from the tip of the nozzle on the retardation temperature. The light and dark symbols correspond to the results of the x-ray and optical measurements, respectively, for a nozzle with d = 2.85 mm. The relationships *a* and b are the results of averaging the measurements. The broken lines 1-3 indicate the relationships for expansion with $\gamma = 1.4$, 1.33, and 1.28, respectively. We see that the relative density does not remain constant on varying the retardation temperature and lies below the values corresponding to expansion with $\gamma = 1.4$ (for completely frozen vibrational relaxation and the absence of condensation). This means that, although (according to estimates) the vibrational degrees of freedom in CO_2 are frozen under our present experimental conditions in the supersonic region of expansion, the role of relaxation remains considerable in the neighborhood of the critical cross section. A characteristic feature is the presence of a maximum on the experimental curve (for $T_0 \approx 550$°K). The general form of the relationship for ρ/ρ_0 remains intact when x/d = 12 and 30; it may be explained as being due to the influence of condensation and vibrational relaxation processes.

An estimation of the intensity of condensation on the principle of [5] shows that under our experimental conditions the initial stage of condensation takes place close to room temperature. For pure CO_2 at $T_0 = 300$°K the characteristic cluster size is 5 molecules/cluster. There should be no condensation at $T_0 \geq 420$°K. Estimates based on [6] give the condition of no condensation at $T_0 \geq 490$°K.

A model parameter for vibrational relaxation at constant retardation temperature is the complex p_0d. For the values of $p_0d \leq 10^3$ mm Hg · mm existing in the temperature range covered here, the vibrational degrees of freedom are frozen in the supersonic range of expansion for $x/d \geq 1$. Nevertheless, a temperature rise should lead to a certain intensification of the role of vibrational relaxation, both as a consequence of the reduction in γ and as a consequence of the downstream displacement of the freezing zone attributable to the reduction in relaxation time. These considerations and the character of the ρ/ρ_0 relationship at high temperatures are confirmed by calculations based on the following: 1) the model of instantaneous freezing; curve 4 in Fig. 1 (obtained on the assumption of freezing at a distance x/d = 1 and subsequent expansion with $\gamma = 1.4$); 2) the model of the expansion of a nonviscous vibrationally relaxing gas with a finite relaxation time; curve 5 in Fig. 1 (obtained for $p_0d = 7.6 \cdot 10^3$ mm Hg · mm; vibrational relaxation time taken from [7]).

Thus when vibrational relaxation and condensation processes are operative, any attempt to characterize the expansion process by some "effective" adiabatic index may lead to considerable quantitative errors.

§ 3. Since the vibrational relaxation and condensation processes at a specified temperature level are determined by the model parameter p_0d^{α} (for condensation $\alpha < 1$, for vibrational relaxation $\alpha = 1$), it is interesting to pursue the influence of p_0 on relaxation effects in numerical form. Figure 2 illustrates the change in relative density at x/d = 20 as a function of the retardation pressure p_0 at $T_0 = 300$°K and d = 2.85 mm. Curve 1 is derived from x-ray measurements and curves 2 and 3, from optical; the light symbols refer to $p_n = 2 \cdot 10^{-2}$ mm Hg and the dark symbols, to $p_n = 7 \cdot 10^{-2}$ mm Hg. The horizontal lines 4 and 5 correspond to expansion with $\gamma = 1.4$ and 1.33, respectively. A reduction in pressure is accompanied by two groups of phenomena: weakening of the relaxation processes and intensified influence of viscous effects. Both lead to an increase in relative

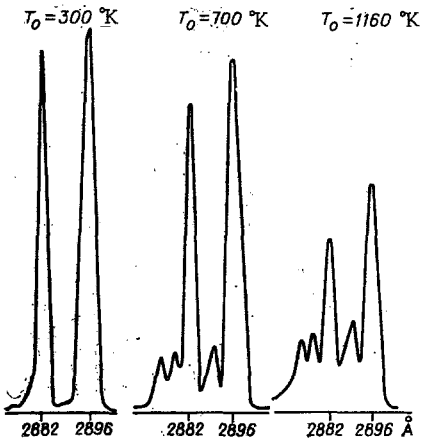

Fig. 3

density (the first by way of an increase in the effective adiabatic index and the second as a result of viscous dissipation in the expansion process, the merging of shock waves, and the penetration of gas into the core of the jet from the surrounding space). There is yet another effect of viscous origin: an increase in the effective diameter of the tip of the nozzle as a result of the influence of the boundary layer; however, this only becomes appreciable for $p_0 d < 10$ mm Hg · mm, i.e., outside the range of the conditions under consideration.

The initial stage of condensation occurs over the whole range of pressures embraced. According to [6] condensation ceases at $p_0 \lesssim 30$ mm Hg. At $p_0 \lesssim 30$ mm Hg vibrational relaxation only occurs in the subsonic part of the nozzle.

The results of the x-ray measurements at $p_n = (2\text{-}7) \cdot 10^{-2}$ mm Hg indicate that the penetration of the surrounding gas into the paraxial region for all values of p_0 is negligible, and the same applies to the influence of shock waves. Optical measurements at greater values of p_0 are sensitive to the rearrangement of the flow even for $p_0 \lesssim 180$ mm Hg owing to the poorer spatial resolution. For the minimal values of $p_n \approx 2 \cdot 10^{-2}$ mm Hg the curves obtained from the optical and x radiation run in an equidistant manner.

Since for low values of p_0 the effective adiabatic index approaches 1.4 (see Fig. 2), the influence of viscous effects in the process of free expansion may be estimated from calculations for the radial and jet expansion of a diatomic gas [8]. We find that for $x/d = 20$ the viscosity correction to the Mach number (in the sense of a reduction in the latter) is no greater than 10% and for $M \gtrsim 10$ does not lead to any marked change in density.

Thus the increment in relative density is due to the freezing of condensation and vibrational relaxation – two effects which up until now have not been separated. For $p_0 d < 10^2$ mm Hg · mm we may also speak of the effects of rotational relaxation, at least for large distances from the tip of the nozzle.

Basing our considerations on the x-ray measurements, on passing outside the limits of the complete freezing of vibrational relaxation and condensation at room retardation temperature we may take an approximate value of $p_0 \approx 30$ mm Hg at $d = 2.85$ mm. These quantities may be used to estimate the limits within which relaxation effects exert an influence in jets behind nozzles of other diameters, in accordance with the modeling of condensation by reference to the parameter $p_0 d^{0.6} = \text{const}$ [5] and vibrational relaxation by reference to the parameter $p_0 d$.

§ 4. The foregoing investigation required advanced accuracy and stability of operating conditions. Certain methodical features should be mentioned separately. For example, the absence of thermal insulation from the leading chamber resulted in a considerable scatter of the experimental data, evidently because of the uncontrollable inhomogeneity of the temperature field at the entrance into the nozzle.

The experiments showed that the use of the optical radiation of CO_2 (transition $B^2\Sigma_u \rightarrow X^2\Pi_g$) for measuring the density requires a certain amount of caution, since the shape and intensity of the bands depends very considerably on temperature, as may be seen from Fig. 3. For a constant value of the density the integrated intensity of the 2890-Å band does not depend on temperature in the range studied ($T_0 = 300\text{-}1200$°K). This may signify that the bands actually appearing belong to a single electron transition. The problem of measuring the density presents no difficulties on using all the bands of the system.

In view of the strong dependence of the shape of the bands on the temperature we may well pay some attention to the possibility of using the CO_2 radiation excited by an electron beam in the region of 2890 Å in order to measure the vibrational temperature.

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