CONDENSATION ONSET AND GROWTH DYNAMICS OF CLUSTERS

IN FREELY EXPANDING CO_2 FROM A SONIC NOZZLE

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A gas in supersonic adiabatic expansion reaches supersaturation, with the possibility of cluster formation. The process of condensation is controlled by the stagnation parameters, nozzle geometry, and type of gas. One of the methods employed in flow diagnostics is the Rayleigh-scattering method. It is characterized by the fact that (a) measurements can be made in the region within which nucleation and cluster growth occur, and (b) the scattered signal strength varies strongly with the cluster size distribution function.

This study makes use of Rayleigh scattering [1] to follow the dynamics of cluster formation and growth in the homogeneous condensation of CO_2 discharged from a sonic nozzle into a vacuum. The experimental findings extend and supplement previously reported data [2-6].

Rayleigh-Scattering Method. When a sounding laser beam of intensity I_r propagates in a medium with a monomer concentration N_1, dimer concentration $N_2, \ldots$, and i-mer concentration N_i, the scattered polarized-component fraction is given by

$$I/I_r = K\alpha^2 \sum_{i=1} N_i i^2, \tag{1}$$

where α is the molecule polarizability, and K is a scaling constant. Structural asymmetry of the molecules gives rise to a depolarized scattered component, but two to four orders of magnitude weaker [7], which has been omitted from consideration here. Equation (1) applies provided two conditions hold, viz., the geometric cluster size must be much smaller than the light wavelength [8], and molecular bonding in the cluster must have little effect on the electronic levels of the molecules. In order to analyze the condensation process in supersonic gas expansion, it is convenient to separate the monomer contribution in Eq. (1). To do this we introduce the total molecular concentration in the flow, $N = \Sigma(N_i i)$; the mass fraction of condensate, $q = (1 - N_1/N)$; the molecular concentration in the nozzle forechamber, N_0; and the scattering intensity at that concentration, $I_0/I_r = K\alpha_2 N_0$. Setting in (1) the condensate mass fraction and normalizing to $I_0 I_r$, we get for the relative scattered intensity

$$I/I_0 = (1 - q)\, N/N_0 + \sum_{i=2} N_i i^2/N_0. \tag{2}$$

If the clusters are of a single size i, Eq. (2) reduces to

$$I/I_0 = [(1 - q) + q\bar{i}]N/N_0. \tag{3}$$

Given a distribution function of finite but sufficiently small width N_i, we can use Eq. (3) for rough estimates, taking $\bar{i}$ to be the mean cluster size.

Experimental Setup. The experiments were performed in a 0.1-m^3 vacuum chamber, with the gas source fixed to a micrometric coordinate mechanism at its center (the coordinates could be adjusted to within 20 μm). The chamber was fitted with an NVZ-500 pump. The light beam was let in and out through optical windows. The gas was fed into the nozzle forechamber through a heat exchanger placed after the reducer, enabling the stagnation temperature to be maintained to within not more than 1 K. The stagnation pressure was measured with standard pressure and vacuum gauges to within not more than 0.5%. The sonic nozzles used for the experiments had a critical cross section diameter $d_* = 1.5\text{-}5.9$ mm. The generatrix of the inner surface made an angle of 15° with the outer surface, which was plane. The stagnation

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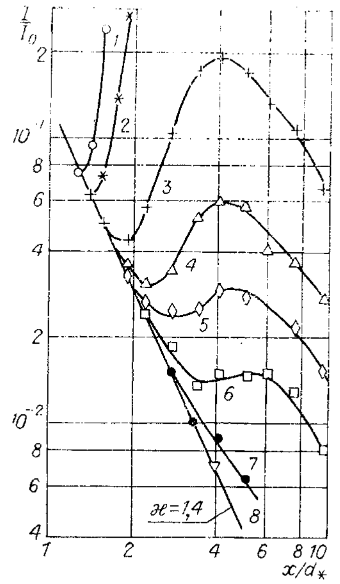

Fig. 1

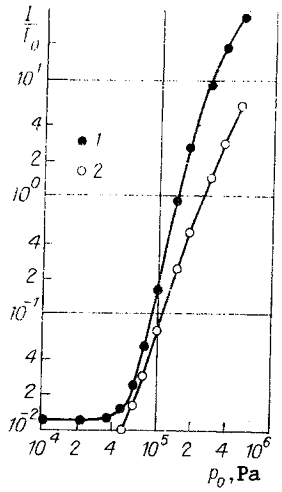

Fig. 3

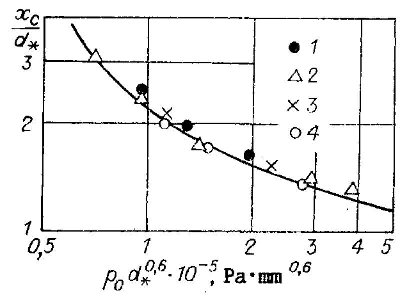

Fig. 2

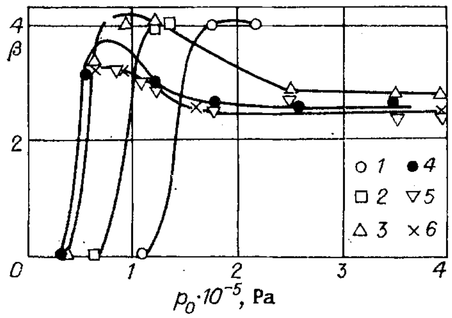

Fig. 4

temperature was kept constant (T_0 = 295.5 K) in all the experiments. The stagnation pressure range was $p_0 = 10^4$-$5\cdot10^5$ Pa. The light source was the second harmonic of a neodymium laser (0.53 μm wavelength).

Experimental Results. Figure 1 shows axial profiles of the relative scattered intensity for a nozzle with d_* = 3.05 mm at $p_0 = 1.98\cdot10^5$, $1.49\cdot10^5$, $0.98\cdot10^5$, $0.73\cdot10^5$, $0.60\cdot10^5$, $0.49\cdot10^5$, $0.37\cdot10^5$, and $0.098\cdot10^5$ Pa (curves 1-8). The gas expansion is initially nearly isentropic, with a specific-heat ratio $\kappa = 1.4$. The appearance of clusters in the flow causes the scattered signal to increase. We define the coordinate of condensation onset x_c/d_* as the last experimental point on the isentrope. Given that the measurement error at each point is < 10%, this definition corresponds (by Eq. (3)) to the condition $q(\bar{i} - 1) < 0.1$. When the stagnation pressure and nozzle edge diameter were varied, the curve describing the condensation-onset coordinate as a function of $p_0 d_*^{0.6}$ was found to be generalized (Fig. 2); the exponent of d_* is adopted from [9, 10], and the points 1-4 are for d_* = 1.55, 3.05, 3.99, 5.89 mm.

In Fig. 3 the relative scattered intensity at two distances from the nozzle edge (x/d_* = 0.3 and 9.0, points 1 and 2 (d_* = 3.05 mm)) is plotted against the stagnation pressure. The curve shape depends on the contribution of each term in Eq. (1). Specifically, before condensation begins ($q = 0$) the relative scattered intensity is proportional to the relative density, which depends on x/d_* in the case of isentropic expansion. After the onset of condensation, the contribution of the condensed phase in the scattered signal increases with the stagnation pressure, and thus beyond a certain pressure p_0 the first term in Eq. (2) can be neglected. On the basis of (3), the expression for the relative scattered intensity in advanced condensation hence becomes

$$I/I_0 = q\bar{i}N/N_0. \tag{4}$$

Assuming that the heat of condensation liberated in the flow does not appreciably affect N/N_0 and that the condensate mass fraction is a slowly varying function of p_0 [10], we can use the experimental data to follow the growth dynamics of the mean cluster size as it varies with the stagnation pressure. Following [6], we represent the mean cluster size as a power function of the stagnation pressure, viz., $I/I_0 \sim i \sim p_0^{\beta}$. The exponent β is a function of p_0 and x/d_*. The variation of β with p_0 for $x/d_* = 1.5$, 2.2, 3.4, 4.4, 6.1, 9.0 (points 1-6) is plotted in Fig. 4. It shows that as p_0 increases, β tends to 2.5. The mean cluster size consequently grows proportionally to $p_0^{2.5}$ in advanced condensation. This approximates the data given in [11] and differs from the results derived in [6].

The experimental results, together with Eq. (4), thus provide the means of estimating the mean cluster size (e.g., for $p_0 = 10^5$ Pa and $x/d_* = 10$, and taking q = 20%, we get $\bar{i} = 300$). A more rigorous analysis, however, should take into account the cluster size distribution function, the variation of condensate mass fraction with the discharge parameters, and the heat of condensation liberated in the flow. If a distribution function of finite width is taken into account, the value of $\bar{i}$ calculated from Eq. (4) cannot be smaller than the actual mean cluster size (the first moment of the distribution function).

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