

COMPENSATION EFFECT IN GASDYNAMIC SEPARATION OF MIXTURES

S. V. Dolgushev and V. M. Fomin

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Increasing the efficiency of operation of apparatus designed for separation of gaseous mixtures remains an important problem in chemical, petroleum, and gas industries, nuclear power engineering, and other high-technology industries. The gasdynamic method is a promising separation method with which we have already had industrial experience (see, for example, [1-5]). Since the separation efficiency in this method is mainly due to the difference between the masses of the particles, the efficiency can be considerably increased if we provide conditions for condensation of the heavy component. Clusters and droplets of condensate appearing in this case in the flow have much greater mass than single molecules; consequently, the separation factor may considerably increase [6-8].

Accordingly, it is of interest to study the pure condensation effect in the general scheme of the separatory process, which should include cooling of the mixture upon expansion in an ultrasonic nozzle until conditions are right for the beginning of condensation of one of the components, formation of droplets of condensate and their separation from the gas phase at the outlet of the nozzle. This schemes may involve additional elements such as: creation of conditions for intensive coalescence of droplets [7] at the outlet of the nozzle, stimulation of condensation of the gas to be separated by laser irradiation [8], etc.

In this paper we have numerically investigated condensation enrichment of a separable mixture by one of the components upon passage through an ultrasonic nozzle. The calculations were carried out for a helium – nitrogen mixture often encountered in many production processes. It is assumed that the helium atoms do not exert an effect on the processes of formation and growth of nitrogen droplets, which has experimental support (see, for example [9]).

As the parameter characterizing the effectiveness of enrichment of the gas phase by helium, here we use a quantity we call the condensation separation factor m , defined by the formula

$$m = (P_1/P_2)_+ / (P_1/P_2)_-, \quad (1)$$

where P is the pressure; the subscripts 1 and 2 mark quantities belonging to helium and nitrogen respectively the subscripts + and – indicate the conditions at outlet of the nozzle and at its inlet.

Although such a definition of the separation factor is generally accepted [4], we should bear in mind that there we take into account only the pure condensation effect. The real separation factor of course, will be smaller even if we can extract a large part of the droplets of condensate at the outlet of the nozzle. The separation of droplets from a gas condensate mixture having supersonic velocity may not be complete; some of the droplets stay in the flow.

The calculations were done in the quasi-one-dimensional approximation using the classical theory of homogeneous condensation [10]. Relative to the properties of the forming gas – condensate mixture, we made the following assumptions: 1) before onset of condensation, the flow is isentropic; 2) the flow is one-dimensional, nonviscous, and steady; 3) the gas phase is described by the equation of state for an ideal gas; 4) the change in entropy of mixing is neglected; 5) the pressure of the mixture is due only to chaotic motion of the gas molecules, the droplets of condensate do not make a contribution; 6) there is no heat exchange with the external medium; 7) the condensed phase is uniformly distributed in the gas phase and has the same velocity as the gas phase; 8) the volume of the condensed phase is small compared with the total volume of the gas-condensate mixture; 9) phase transition of the gas – solid phase or liquid – solid phase type are not considered; 10) the droplet size is so small that droplet growth occurs in the free-molecule regime; 11) the effect of the droplet size on the surface tension coefficient and other properties is not taken into account; 12) the nucleation rate for condensation nuclei is described by the classical formula, obtained successively in papers by Becker, Dering, Frenkel, and Zel'dovich [10, 11]:

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$$I = (\zeta / \rho_c) (P_2 / kT)^2 \sqrt{2\sigma\mu_2 / \pi N_A} \exp(-4\pi\sigma r_*^2 / 3kT).$$

Here I is the rate of appearance of condensation nuclei per unit volume; ρ_c is the density of the condensed nitrogen; k is the Boltzmann constant; T is the temperature of the gas; σ is the surface tension coefficient; μ_2 is the molecular weight of the nitrogen; N_A is Avogadro's number; ζ is the sticking coefficient for the nitrogen molecule onto the droplet, which here we take to be equal to unity; r_* is the radius of the droplet of critical size, determined from the Thomson–Helmholz formula [10]

$$r_* = 2\sigma\mu_2 / [\rho_c R T \ln(P_2 / P_{2s})],$$

where R is the universal gas constant; P_{2s} is the saturation pressure of nitrogen, calculated as a function of temperature from the Clausius–Clapyeron formula

$$\ln P_{2s}(T) = A - B/T$$

(A , B are parameters).

The growth rate of a droplet whose size is significantly smaller than the free path length of the gas molecules is found from the Hertz–Knudsen formula [11]

$$\dot{r} = \zeta P_2 [1 / \sqrt{T} - (P_{2s} / P_2) / \sqrt{T_c}] / (\rho_c \sqrt{2\pi R / \mu_2})$$

(T_c is the temperature of the droplet, which is assumed to be equal to the saturation temperature of nitrogen at the specified pressure P_2 [11]).

The mathematical model used here for flow with condensation includes equations for the three moments of the droplet size distribution function, equations for conservation of mass, momentum, and energy of the gas–condensate system, and the equation of state of the gas phase [11]:

$$ud\Omega_0/dx = I/\rho; \quad (2)$$

$$ud\Omega_1/dx = Ir_*/\rho + \Omega_0\dot{r}; \quad (3)$$

$$ud\Omega_2/dx = Ir_*^2/\rho + 2\Omega_1\dot{r}; \quad (4)$$

$$udq/dx = 4\pi\rho_c\Omega_2\dot{r} + (4/3)\pi\rho_c Ir_*^3/\rho; \quad (5)$$

$$\rho uF = \rho_* u_* F_* = C_*; \quad (6)$$

$$udu + dP/\rho = 0; \quad (7)$$

$$(3.5 - \alpha_{10})R_0T + 0.5u^2 - (q/\gamma_{20})L_e = H_0; \quad (8)$$

$$P = (1 - \mu_0 q / \mu_2) \rho R_0 T. \quad (9)$$

Here ρ , u are the density and velocity of the mixture $N_2 + He +$ nitrogen droplet; x is the coordinate measured along the axis of the nozzle, starting from its critical cross section; C_* is the gas flow rate in the critical cross section of the nozzle; R_0 is the gas constant of the mixture before onset of condensation; q is the mass fraction of condensate; μ_0 is the molecular weight of the mixture before onset of condensation; H_0 is the enthalpy of stagnation; α_{10} , α_{20} are the molar concentrations of helium and nitrogen respectively before onset of condensation; L_e is the effective heat of condensation, calculated per unit mass of the gas–condensate mixture ($L_e = \gamma_{20}L$); γ_{20} is the mass concentration of nitrogen in the mixture before condensation; L is the specific heat of condensation for pure nitrogen; the subscript * marks parameters in the critical cross section of the nozzle. The moments Ω_i of the distribution function $f(r)$ of the droplets of condensate with respect to their radii r are defined as:

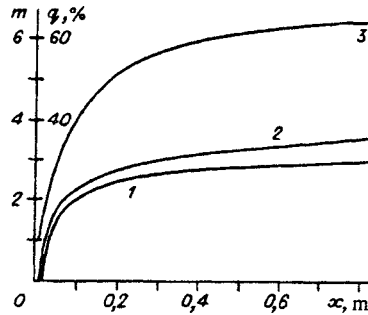


Fig. 1

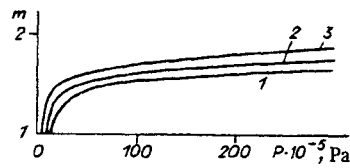


Fig. 2

$$\Omega_i = \int_{r_0}^{\infty} r^i f(r) dr.$$

The thermodynamic properties of the condensed and gaseous nitrogen and also the parameters of the phase equilibrium curve were taken from [12], where calculations were carried out for condensation of pure nitrogen for nozzle parameters under stagnation conditions close to those given in this paper, and good agreement was obtained with experimental data.

The system of equations (2)-(9) were converted to the form required for using the programs for integration of systems of ordinary differential equations. This system was also made dimensionless. Integration of the system of equations obtained was carried out from a point located in the supersonic part of the nozzle at which the saturation conditions were not yet achieved. The initial conditions are the equilibrium values of the gasdynamic parameters and the conditions that there are no droplets of condensate present: $q = 0$, $\Omega_i = 0$, $i = 0, 1, 2$. Over the course of integration of the system of equations from below along the flow, saturation conditions are achieved for which $P_2 > P_{2s}$, the right-hand side of the equations in (2)-(5) becomes different from zero, and here we begin to calculate the increase in the values of q and Ω_i .

The solution was accomplished by the Runge-Kutta method, fourth-order approximation with local accuracy $\varepsilon = 10^{-5}$. The ultrasonic conical nozzle for which we performed the calculations is characterized by the following parameters: radius of the critical cross section of the nozzle, 10^{-3} m; length of the supersonic part, 1 m; aperture semiangle, 10° . The stagnation temperature was specified as close to room temperature, allowing small variations toward a decrease and increase. The stagnation pressure ranged approximately from 10^5 to $3 \cdot 10^7$ Pa.

The graphs in Figs. 1-4 illustrate the results obtained. In Fig. 1, we present the dependences on the axial coordinate x of the mass concentration of condensate q (curve 1) obtained when solving the system of equations (2)-(9), and the equilibrium value of this quantity (curve 2) found using the formulas describing equilibrium condensation. These dependences correspond to stagnation parameters $P_0 = 3 \cdot 10^6$ Pa, $T_0 = 300$ K and initial composition of the mixture 5% He + 95% N_2 . From the graphs we see that the delay of onset of condensation under the given conditions is insignificant, although then the nonequilibrium value of the mass fraction of condensate markedly lags behind its equilibrium value.

The major increment in the friction of condensate occurs in a relatively short section of the nozzle beyond the saturation point of length about 0.2-0.3 m, after which a slow increase in this quantity occurs. For $x > 0.5$ m, the increase in q practically stops. This is explained by two factors: 1) the drop in the gas density and consequently the reduction in the growth rate of droplets of condensate; 2) the decrease in the supersaturation as a result of release of the heat of condensation into the gas phase. Negative feedback occurs between the temperature of the gas and the condensation process: a drop in temperature of the gas and the condensation process: a drop in temperature leads to onset of condensation; release of the heat of condensation tends to increase the gas temperature and to eliminate supersaturation.

Curve 3 in Fig. 1 is the dependence of the condensation separation factor, calculated from formula (1), on the axial coordinate x for parameters in the forechamber equal to $P_0 = 5 \cdot 10^6$ Pa, $T_0 = 300$ K and initial composition of the mixture

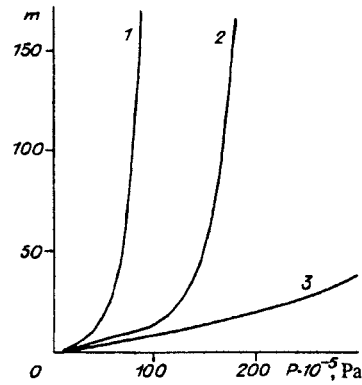


Fig. 3

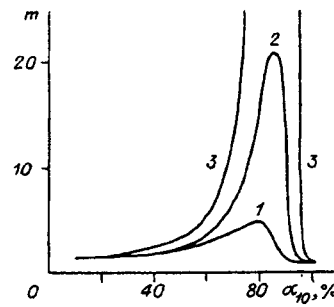


Fig. 4

70% He + 30% N₂. The behavior of the $m(x)$ dependence qualitatively duplicates the $q(x)$ dependence qualitatively duplicates the $q(x)$ dependence.

In Figs. 2 and 3, we show the dependence of m on the stagnation pressure for three values of the stagnation temperature: $T_0 = 250, 300,$ and 350 K (curves 1-3). The graphs in Fig. 2 correspond to a low initial concentration of helium (5%) with respect to molecular composition; the graphs in Fig. 3 correspond to a relatively high initial molar concentration of helium (70%). There is a substantial qualitative difference in the behavior of the curves of the $m(P_0, T_0)$ dependence for high and low initial He concentrations. For small values of α_{10} in the range of stagnation pressure $P_0 \sim (5-15) \cdot 10^5$ Pa, there is a section of steep growth in the condensation enrichment factor m . Then the curve for the $m(P_0)$ dependence rapidly becomes flat, going to the asymptote where $m \approx 1.5-1.9$. An increase in stagnation temperature does not lead to qualitative changes in the graphical curves; they are only slightly lowered on the (P_0, m) plane.

The graphs of the $m(P_0)$ dependence for $\alpha_{10} = 70\%$ (high initial helium concentrations) look different. In this case, a considerably higher value of the condensation enrichment factor is reached (several hundreds), the curves of the $m(P_0)$ dependence over the entire extent of the considered range of P_0 values show steep growth, similar to exponential. A decrease in the stagnation temperature leads to an increase in the steepness of the $m(P_0)$ curves. In the pressure region $P \leq 10^6$ Pa, these curves almost coincide ($m \approx 1$, no condensation enrichment). Such a difference in the behavior of the $m(P_0)$ curves for high and low values of the initial helium concentration may be explained by the different role of the heat of condensation and the different rate of cooling of the mixture in these two limiting cases. The indicated factors are noted in [13, 14] and are as follows. An increase in the helium fraction in the mixture increases its specific heat, which leads to a smaller increase in temperature than in the case of a large nitrogen fraction. Thus the thermal resistance to condensation becomes less.

More rapid flow of the mixture with a higher helium concentration (the smaller the molecular weight of the gas, the higher its velocity in the nozzle) also promotes more efficient condensation even though it leads to a delay in onset of condensation. In our opinion, here we should add two more factors which enhance condensation separation in an N₂-He mixture with an increase in the helium fraction. The first involves the fact that the specific heat of condensation appearing in Eq. (8) and defined above becomes less effective. The second factor is connected with the increase in the ratio of the specific heats (for constant pressure and constant volume respectively). This leads to a more rapid drop in the temperature of the mixture as it moves in the nozzle and consequently to a weaker manifestation of the heat condensation.

In order to illustrate the dependence of the condensation separation factor on the initial composition of the mixture, in Fig. 4 we present the calculated curves for the $m(\alpha_{10})$ dependence for three values of the stagnation pressure: $P_0 = 3 \cdot 10^6, 5 \cdot 10^6,$ and 10^7 Pa (curves 1-3 respectively). The stagnation temperature for all the curves is equal to 300 K. The peak values of m are the following: for $P_0 = 3 \cdot 10^6$ Pa and $\alpha_{10} = 78\%$, $m = 5.1$; for $P_0 = 5 \cdot 10^6$ Pa and $\alpha_{10} = 85\%$, $m \approx 21$; for $P_0 = 10^7$ Pa and $\alpha_{10} = 89\%$, $m \approx 537$.

The reason for the abrupt increase in m with an increase in the helium fraction in the mixture, as indicated above, is the decrease in the effect of the heat of condensation and the higher expansion rate of the mixture in the nozzle. The dependence has a "resonance" character close to $\alpha_{10} = 85\%$. An increase in the stagnation temperature leads to an abrupt increase in m , which may be explained by the practically complete "freezing out" of nitrogen, when the value of (P_1/P_2) in formula (1) goes to infinity.

The decrease in m with a reduction in stagnation pressure and the abrupt fall-off of the $m(\alpha_{10})$ dependence for $\alpha_{10} > 90\%$ are explained by the fact that for low partial pressures of nitrogen, the saturation conditions either are not achieved over the length of the nozzle, or if they are nevertheless achieved, this occurs for such a low density of N_2 molecules that the processes of nucleation and growth of droplets cannot lead to the appearance of an appreciable fraction of condensate.

The average sizes of the nitrogen droplets for all calculation variants were $\sim 10^{-9}$ - $5 \cdot 10^{-8}$ m.

Such rough calculations were also done for a helium–methane mixture, but the value of m obtained in this case were considerably smaller even though the results are qualitatively identical. This is explained by the significantly higher heat of condensation of methane and the smaller ratio of its specific heats, which leads to reduction of the cooling rate in the nozzle.

The major conclusions of this paper are the following.

1. The efficiency of condensation separation is increased with an increase in the stagnation pressure and a decrease in the stagnation temperature of the mixture.

2. There is a substantial quantitative and qualitative difference between the dependences of the condensation separation factor m on the stagnation parameters P_0 and T_0 for high and low initial helium concentrations in the $N_2 + He$ mixture. The nitrogen molecules are most efficiently bound into clusters and droplets for high helium concentrations. The graphs for the $m(P_0)$ dependences show that the fraction of nitrogen going into the condensed phase is maximum for $\alpha_{10} \approx 80$ - 90% . For $\alpha_{10} > 90\%$, the efficiency of condensation separation abruptly drops down to unity. Such a dependence of the condensation separation factor on the initial composition of the mixture is explained by the different degree of influence on the condensation from the heat release as molecules are bound into droplets and the cooling rate of the gas in the nozzle for high and low concentrations of the light gas (helium).

3. Although the value of the average droplet size is not large ($\sim 10^{-9}$ - $5 \cdot 10^{-8}$ m), they have a mass larger than the mass of the nitrogen molecules, which should increase the efficiency of gasdynamic separation of nitrogen and helium.

In conclusion, we should note that the quantitative results presented require experimental verification. However, the comparison of the experimental and calculation results done previously in many papers within the framework of the classical theory of homogeneous condensation (see, for example, [12, 14-18]) allows us to hope that our analysis gives results which are at least qualitatively correct.

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