EFFECT OF TEMPERATURE ON DIFFUSIONAL INSTABILITY

Yu. I. Zhavrin and V. N. Kosov

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It is shown that in the case of unstable diffusion, the process tends to become more stable with an increase in temperature. A reduction in temperature in a stable system may result in the formation of convective flows, and the system will change over to the unstable state.

Experimental and theoretical study of differential instability in three-component gas mixtures under isothermal conditions in closed vessels has shown that the intensity of the process depends on the pressure, the diameter of the diffusion capillary, and the initial composition of the gas mixture [1-3]. The relations between the diffusion coefficients play an important role here as well [4, 5]. A simple physical explanation for the initiation of diffusional instability (instability) was given in [2, 6-8] and can be summarized as follows. Diffusion results in "stratification" of the phase boundary into two diffusion sublayers. The layer composed of a mixture of the light and heavy gases located above the boundary with a third gas whose density is greater than the density of the mixture (such as 0.5 He + 0.5 Ar—N_2) will be enriched by the heavy component due to the greater mobility of the molecules of the light gas − since the coefficient of diffusion of helium in nitrogen is higher than the coefficient of diffusion of argon in nitrogen. Diffusion below the boundary leads to enrichment of the layer with the light component. As a result, the density of the gas mixture becomes locally stratified. The mixture becomes hydrostatically unstable, which in a gravitational field may lead to the formation of convective flows.

Here, we will examine the effect of temperature on diffusional instability, since the diffusion coefficients of the components depend differently on temperature. Thus, a change in temperature will lead to a change in the relations between the diffusion flows.

We experimentally studied the temperature dependence of the process in the range 276-353 K. All of the measurements were made on a unit which employed the two-flask method. The procedure used to conduct the measurements was described in [2]. The length of the connecting capillaries was $(69.15 \pm 0.05)\cdot 10^{-3}$ and $(70.75 \pm 0.05)\cdot 10^{-3}$ m, while their diameters were $(4.00 \pm 0.05)\cdot 10^{-3}$ and $(4.25 \pm 0.05)\cdot 10^{-3}$ m. The volumes of the flasks were the same and were equal to $(76.9 \pm 0.6)\cdot 10^{-6}$ m^3. All of the experiments were conducted at different temperatures and pressures. The initial composition of the binary mixture was analyzed with an ITR-1 interferometer, while the composition of the mixture after diffusion was analyzed with a chromatograph.

We chose two three-component systems for our study: 1) He + Ar—N_2; 2) H_2 + N_2—CH_4. The composition of the original components in the binary mixtures was chosen so that, regardless of the test temperature, the density of the mixture was always lower than the density of the pure component. The mixtures were located in the top flask, while the pure gases were in the bottom flask. The essence of the experiments can be described as follows: We took a binary mixture with a composition such that diffusion would be unstable at the specified pressure and capillary diameter and an initial temperature T = 276 K. Then we changed only the temperature from test to test, up to the maximum value T = 353 K. The composition of the gas mixtures was analyzed at the end of each test on the basis of the contents of both flasks.

The results of measurement of the temperature dependence of the quantity of the mixture components that diffused into the pure component (nitrogen or methane) are shown in Table 1. Test duration was 1 h in every case.

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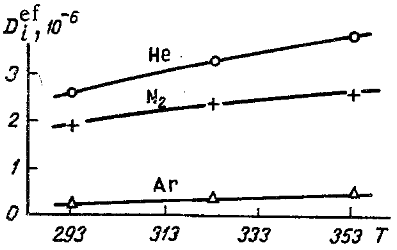

Fig. 1

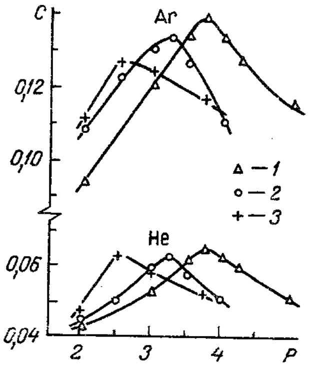

Fig. 2

Fig. 1. Dependence of the effective diffusion coefficients of helium, argon, and nitrogen on temperature: the system 0.6607 He + 0.3393 $Ar-N_2$, P = 2.54 MPa; curves show results calculated in accordance with [9]. D_i^{ef}, m^2/sec; T, K.

Fig. 2. Dependence of the quantities of helium and argon diffused into nitrogen at different temperatures and pressures: the system 0.4688 He + 0.5312 $Ar-N_2$; 1) T = 353 K; 2) 323 K; 3) 293 K. c, mole fractions; P, MPa.

TABLE 1. Temperature Dependence of the Quantities of Diffused Components (in mole fractions), P = 2.54 MPa

T, K	$0,6179H_2+0,3821N_2-CH_4$			$0,4688He+0,5312Ar-N_2$		
	H_2	N_2	CH_4	He	Ar	N_2
276	0,0589	0,0929	0,1518	—	—	—
293	0,0536	0,0855	0,1385	0,0793	0,1405	0,1983
313	0,0493	0,0770	0,1263	0,0709	0,1305	0,1913
333	0,0412	0,0690	0,1102	0,0661	0,1228	0,1856
353	0,0375	0,0660	0,1065	0,0549	0,1092	0,1531

It can be seen from the table that the quantity of diffused components decreases with an increase in temperature, which is not characteristic of diffusion, i.e., the intensity of the unstable process decreases.

For mixtures in which diffusion is stable, we measured the effective diffusion coefficients (EDC) of the components at different temperatures. Figure 1 shows the results of the experiment and calculations of the EDC performed in accordance with the Stefan–Maxwell scheme [9] for one such system (0.6607 He + 0.3393 $Ar-N_2$). It can be seen that the EDC increases monotonically with temperature and coincides with the calculated values within the range of the experimental error – 5-7%.

Thus, it follows from the data obtained that stable diffusion remains unchanged with an increase in temperature and is described by the usual diffusion laws, while unstable diffusion "diminishes" as it were and approaches the stable state. In fact, it is possible to select an initial composition of the gas system such that, in a certain temperature range, it will change from diffusionally unstable to diffusionally stable, i.e., no convective flows will develop in the system. This system can be described by Fick's law. Table 2 shows results of measurement of the EDC of the components of two such systems at different temperatures.

It is evident from the tabular data that at T = 353 K, the investigated systems act stably. This is evidenced by the coincidence of the experimental and theoretical EDC's. At the same time, the experimental EDC's increase sharply at a lower temperature (T = 293 K) and the system "breaks down" and exhibits distinctly unstable behavior.

It was noted in [1-3] that the maximum intensity of unstable diffusion in the system He + $Ar-N_2$ and H_2 + N_2-CH_4 at T = 298 K corresponds to a pressure on the order of 2.5 MPa. With a change in temperature, the position of the maximum will evidently correspond to different values of pressure. We conducted special studies of the pressure dependence of the quantity of diffused components at different temperatures (Fig. 2). It can be seen from

TABLE 2. Experimental and Calculated Values of the EDC D_i^{ef}, 10^{-4} m²/sec, of Three-Component Gas Mixtures

System $0.6192He + 0.3808 Ar-N_2$, P = 2.54 MPa						
T, K	meas.			calc.		
	He	Ar	N_2	He	Ar	N_2
293	0,027	0,026	0,030	0,027	0,003	0,017
323	0,035	0,012	0,026	0,034	0,005	0,023
353	0,038	0,008	0,026	0,037	0,008	0,025

System $0.6463H_2 + 0.3537N_2-CH_4$, P = 1.56 MPa						
T, K	meas.			calc.		
	H_2	N_2	CH_4	H_2	N_2	CH_4
293	0,055	0,076	0,062	0,049	0,004	0,033
323	0,056	0,036	0,049	0,055	0,007	0,037
353	0,062	0,009	0,043	0,061	0,009	0,042

the figure that up to pressures on the order of 2.5 MPa, the system again strives toward a stable state with an increase in temperature for a constant pressure. With an increase in both pressure and temperature the maximum of the curve is shifted toward higher pressures.

The experimental data obtained allows us to unambiguously conclude that an increase in temperature helps stabilize diffusion. Conversely, a reduction in temperature causes a "breakdown" in a diffusionally stable system, and the system becomes unstable. This fact can be explained qualitatively as follows.

With an increase in temperature, the nominally stratified region of the gas mixture "spreads out" along the capillary due to an increase in the diffusion coefficients of the components, and the system moves in the direction of stable diffusion. Proceeding on the basis of this, we can better understand the shift of the maxima of the instability curves shown in Fig. 2. In fact, at a temperature somewhat greater than the given temperature, the size of the diffusion layer increases without a change in the mean density. Thus, in order for the same value of mean density to be associated with a unit length of the diffusion layer (in the unidimensional case), this layer must be compressed, i.e., pressure must be increased.

The character of the distribution of mean density during diffusion in a three-component gas mixture (unidimensional problem) was examined in [10] as a function of pressure and concentration. We solved the same problem, but with allowance for the effect of temperature on the distribution of mean density. The results of numerical calculations are shown in Fig. 3, which illustrate that – as in [10] – regions of high and low density are formed at the boundary of the gases. However, the difference in densities at the boundary decreases with an increase in temperature, which leads to stabilization of the process.

It is possible to select an initial system composition such that the system will be stable at a certain temperature and will be described by the usual diffusion laws. Thus, according to calculations, the system 0.51 He + 0.49 $Ar-N_2$ will be stable throughout the temperature range 200-700 K at P = 1.0 MPa, while at 2 MPa or more, density will be locally stratified at the boundary in the low-temperature region and the system will become unstable. Figure 4 shows that, with an increase in temperature, the theoretical difference in mean density at the boundary decreases and ultimately reaches zero, while it increases sharply with a decrease in temperature and the system becomes diffusionally unstable.

The concentrational stability (instability) of a system is usually determined by the Rayleigh number. This number was transformed in the following manner in [2] for the mean values $\bar{\eta}$, $\bar{D}$, and $\bar{\rho}$:

$$R_d = \frac{gr^4\Delta\bar{\rho}}{\bar{\eta}L\bar{D}}. \quad (1)$$

It is evident from this definition of the Rayleigh number that its numerical value depends implicitly on temperature in terms of the density and viscosity of the system, as well as in terms of the diffusion coefficients of the components. Assuming the mixture to be ideal, $\rho \sim T^{-1}$, we obtain the following for the hard-sphere model $\eta \sim T^{1/2}$, $D \sim T^{3/2}$

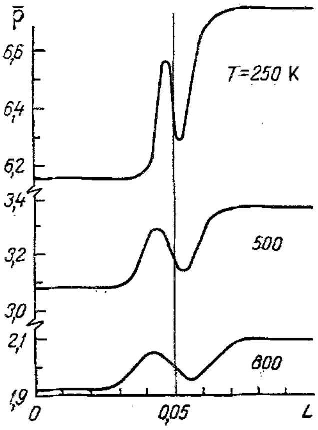

Fig. 3

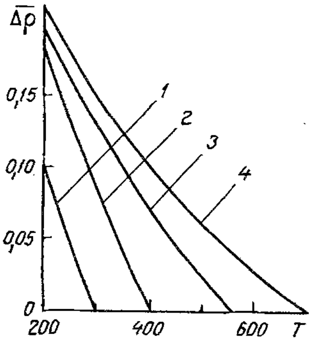

Fig. 4

Fig. 3. Distribution of mean density in the system 0.4 He + 0.6 Ar—N_2 at different temperatures, P = 0.50 MPa. $\bar{\rho}$, kg/m^3; L, m.

Fig. 4. Change in the difference in the mean density of the components in relation to temperature and pressure: the system 0.49 He + 0.51 Ar—N_2; 1) P = 2 MPa; 2) 3; 3) 4; 4) 5 MPa.

$$\frac{R_d^T}{R_d^{T_0}} = \left(\frac{T_0}{T}\right)^3. \tag{2}$$

It is evident from Eq. (2) that the diffusional Rayleigh number increases sharply with a decrease in temperature. This is evidence of the significant effect of temperature on unstable diffusion — which clearly follows from our experiments.

Thus, experimental and numerical study of the effect of temperature on the character of diffusional instability established that in regions corresponding to instability of the process, an increase in temperature helps stabilize diffusion. Meanwhile, a decrease in temperature may cause a diffusionally stable system to "break down" and become unstable. With an increase in temperature, the maximum of the intensity of the process moves toward the high-pressure region. Knowing the critical Rayleigh number for the given temperature, it is always possible to use Eq. (2) to find this number for another temperature and to determine the behavior of the system during the diffusion of the components of a three-component gas mixture. If the diffusion process is stable, it remains so with an increase in temperature, and the diffusion coefficients can be determined from the usual temperature relation. With an unstable regime, the convective flows of the components alter the expected concentration distribution so much that molecular mass transport can no longer be calculated just on the basis of Fick's law.

NOTATION

T, temperature; P, absolute pressure; D_i^{ef}, effective diffusion coefficient; D, interdiffusion coefficient; $\bar{D}$, mean diffusion coefficient of the system; R_d, diffusional Reynolds number; $R_d^{T_0}$, Rayleigh number at the temperature T_0; $\bar{\eta}$, mean value of the viscosity of the gas mixture; ρ, density; $\bar{\rho}$, mean value of the density of the gas mixture; $\Delta\bar{\rho}$, difference in mean density at the ends of the capillary; g, acceleration due to gravity; r, radius of capillary; L, length of capillary; c, mole fraction.

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STUDY OF THE SYNCHRONICITY OF PRESSURE PULSATIONS IN FLUIDIZED-BED REACTORS

A. P. Baskakov, N. F. Filippovskii, V. G. Tuponogov,
and A. V. Mudrechenko

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The interrelation between pressure pulsations and the movement of gas bubbles in a fluidized bed is studied experimentally. Cross-correlation functions are presented for different points of a full-scale reactor.

An orderly theory of the development and propagation of pressure pulsations in fluidized beds has yet to be presented, despite the large number of studies [1-3] devoted to this subject. The creation of such a theory may be aided considerably by the study of correlations between the pressures at different points of a fluidized bed and the motion of gas bubbles in the bed.

The change which occurs in pressure with the passage of a gas bubble through a measurement point has been studied fairly well [3], and the experimental results obtained coincide with calculations such as those performed by the Davidson model [4]. The maximum pressure is seen at the moment the frontal point of the bubble approaches the sensor. Then pressure decreases until the sensor touches the rear point of the bubble. This can be clearly seen from Fig. 1 (points A_1-B_1 and A_2-B_2). If the bubble passes next to the sensor without touching it (the middle pressure maximum in Fig. 1), the pattern remains basically the same. The disturbing effect of the bubble is manifest at a distance on the order of the bubble's diameter from the axis of its rise.

The results shown in Fig. 1 were obtained in a planar transparent unit with a cross section of 18 × 300 mm. The heights of the fluidized bed H = 400-550 mm. The bed was maintained at the fluidization limit by supplying air through a perforated grate, while excess flow was directed through a central nozzle 5 mm in diameter. One of the pressure-sampling tubes was located next to the nozzle, while a second tube was located 0.44 m above the nozzle. The motion of the bubbles was monitored with photodiodes located at different elevations above the nozzle and illuminated through the bed. The current from all diodes and from the extensometric pressure sensors was recorded on an N338-6P multichannel recorder.

The character of pressure oscillation near the nozzle (Fig. 1c) was determined by the superposition of small-scale, high-frequency pulsations connected with the separation of small gas bubbles from the cavity and large-scale pressure gradients caused by the escape

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