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MEASUREMENT OF UNSTEADY GAS FLOW UNDER ANISOTHERMIC CONDITIONS

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We describe a dynamic method for measuring unsteady gas flow under anisothermic conditions. We show that the value of the flow sensitivity determined under isothermal conditions can be used for molecular flow.

In measuring gas flow, the volume in which gas is generated or absorbed (reactor volume) and the volume directly connected to the recording equipment ("instrument" volume) may be at different temperatures.

Static and dynamic methods of measurement can be used. The static method can be used when the interconnected reactor and instrument volumes are hermetically sealed. The gas flow is determined from the rate of change of pressure in the instrument volume. The presence of a temperature gradient along the gas route from the reactor volume to the instrument volume requires corrections because of the thermomolecular pressure difference [1]. The dynamic method involves measuring the pressure drop of the gas across the port connecting the evacuation system (in the case of gas generation) or the pressure system (in the case of gas absorption) with the instrument and reactor volumes which, just as in the static method, are interconnected. In the molecular flow regime the gas flow through the port is proportional to the measurable pressure drop.

The fact that no information on the effect of anisothermicity on the results of dynamic measurements of gas flows is presently available in the literature stimulated the writing of the present paper.

In discussing the dynamic method in the presence of a temperature gradient (Fig. 1), we make three assumptions: 1) gas is generated in the reactor volume; 2) the gas flow regime is molecular; 3) the gas in the exhaust system is much less dense than that in the instrument volume ($n_0 \ll n_2$). In this case the equations for the balance of gas molecules are [2]

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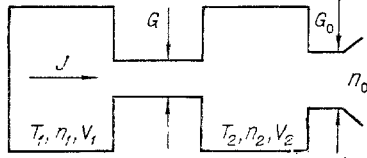


Fig. 1

Fig. 1. Anisothermic gas flow. T_1, T_2 , °K; n_0, n_1, n_2 , moles/m³; G, G_0 , m²; V_1, V_2 , m³; J , moles/sec.

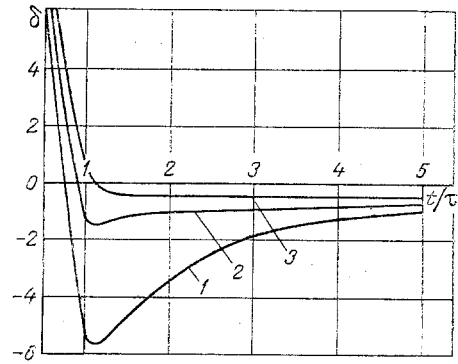


Fig. 2

Fig. 2. Kinetics of the variation of flow sensitivity. δ , %; t, τ , sec.

$$V_1 \frac{dn_1}{dt} = J - G(\langle v_1 \rangle n_1 - \langle v_2 \rangle n_2),$$

$$V_2 \frac{dn_2}{dt} = G(\langle v_1 \rangle n_1 - \langle v_2 \rangle n_2) - G_0 \langle v_2 \rangle n_2. \quad (1)$$

For many gas-generating processes (diffusion, desorption, radiolysis, etc.), the time dependence of the gas flow rate has the form

$$J(t) = J_0 \exp(\lambda t). \quad (2)$$

From the solution of system (1) for zero initial conditions ($n_1(0) = n_2(0) = 0$) it follows by taking account of (2) that

$$n_2(t) = A \{ \exp(\lambda t) + [(r_2 - \lambda)/(r_1 - r_2)] \exp(r_1 t) + [(r_1 - \lambda)/(r_2 - r_1)] \exp(r_2 t) \}, \quad (3)$$

where

$$A = G \langle v_1 \rangle J_0 / [V_1 V_2 (\lambda^2 + p\lambda + q)], \quad (3a)$$

$$r_{1,2} = -(p/2) \pm \sqrt{(p^2/4) - q}, \quad (3b)$$

$$p = (G \langle v_1 \rangle / V_1) + (G + G_0) \langle v_2 \rangle / V_2, \quad (3c)$$

$$q = GG_0 \langle v_1 \rangle \langle v_2 \rangle / (V_1 V_2). \quad (3d)$$

Since r_1 and r_2 are always negative, the second and third terms of Eq. (3) decrease with time. If $\lambda < 0$ and $|\lambda| < |r_1|$, after a time τ determined by the relation

$$\tau = \{ \ln [(\lambda/r_1)(r_2 - r_1)/(r_2 - \lambda)] / (r_1 - \lambda) \}, \quad (4)$$

the following linear relation is established between the gas flow and its molecular concentration in the instrument volume:

$$n_2(t) = \{ G \langle v_1 \rangle / [V_1 V_2 (\lambda^2 + p\lambda + q)] \} J(t). \quad (5)$$

Since $|r_1|$ is generally less than $|r_2|$, it can be shown by using Eqs. (3a)-(3d) and (5) that for $t > \tau$, if

$$|\lambda| < 0.01 |r_1| \quad (6)$$

the ratio $J(t)/n_2(t)$ can be given, with an error not exceeding 1%, in the form

$$J(t)/n_2(t) = G_0 \langle v_2 \rangle. \quad (7)$$

Thus, for fixed temperatures T_1 and T_2 , when inequality (6) is satisfied, the concentration and pressure of the gas in the instrument volume are proportional to the flow of gas evolved in the reactor volume, with the proportionality factor determining the sensitivity of the dynamic flow method depending only on T_2 (since $\langle v_2 \rangle \sim \sqrt{T_2}$) and not on T_1 . The solu-

tion of system (1) for $T_1 = T_2$ agrees exactly with (7). This permits the use of the value of the sensitivity determined under isothermal conditions ($T_1 = T_2$) for measurements of gas flow under anisothermic conditions ($T_1 \neq T_2$) without any correction for anisothermicity. The procedure for determining the flow sensitivity when a mass spectrometer is used as the recording instrument is described in [3].

Let us now assume that in the instrument volume the temperature is constant, and in the reactor volume it varies linearly with time

$$T_1(t) = T_{10}(1 + \alpha t). \quad (8)$$

In this case the mean thermal velocity of the gas molecules in the reactor volume is time dependent ($\langle v_1 \rangle \sim \sqrt{1 + \alpha t}$), and the system of equations (1) becomes nonlinear.

The numerical solution of this system by the Runge-Kutta method, taking account of the exponential time dependence of the flow rate (2), is shown by curves 1 and 2 of Fig. 2. The quantity δ characterizes the kinetics of the variation of flow sensitivity, and is given by the relation

$$\delta = \frac{J(t)/n_2(t) - G_0 \langle v_2 \rangle}{G_0 \langle v_2 \rangle} \cdot 100, \%. \quad (9)$$

For comparison, curve 3 of Fig. 2 shows the solution of the linear system (1). In all three cases it was assumed that helium was generated, and the parameter λ of Eq. (2) was taken equal to -0.020 sec^{-1} . Curve 3 was obtained for $T_1 = 300^\circ\text{K}$, $T_2 = 700^\circ\text{K}$, and the following values of the constants of system (1): $r_1 = -3.7 \text{ sec}^{-1}$, $r_2 = -2.7 \times 10^3 \text{ sec}^{-1}$, $\tau = 1.4 \text{ sec}$. Curves 1 and 2 were obtained with these same values of the constants in system (1). The relative heating rate $\alpha = 1.0 \text{ sec}^{-1}$ for the first curve, and $\alpha = 0.010 \text{ sec}^{-1}$ for the second. In both cases the initial temperature in the reactor volume was taken equal to $T_{10} = 300^\circ\text{K}$.

It can be seen from Fig. 2 that the deviation of the ratio $J(t)/n_2(t)$ from the constant value $G_0 \langle v_2 \rangle$ for curve 3 is no more than 0.5% for $t > \tau$. This deviation does not exceed 1% for $\alpha = 0.010 \text{ sec}^{-1}$ (curve 2) for $t > 3\tau$, and for $\alpha = 1.0 \text{ sec}^{-1}$ (curve 1) for $t > 5\tau$.

Thus, in measuring gas flows by the dynamic method it is not necessary to correct for anisothermicity. This is true for a constant temperature difference between the instrument and reactor volumes. If this temperature difference varies, then for a given rate of change of temperature a time can be indicated after which the flow sensitivity becomes constant and equal to the sensitivity determined under isothermal conditions. This time depends on the relative rate of gas generation λ , the parameters of the equipment used in the measurements (V_1 , V_2 , G , and G_0), and the required accuracy of the gas flow measurement.

NOTATION

A , constant, moles/ m^3 ; α , relative heating rate, sec^{-1} ; δ , quantity characterizing the kinetics of the variation of flow sensitivity, %; G and G_0 , geometrical characteristics of gas routes between the reactor volume and instrument volume, and between the instrument volume and the exhaust system, respectively, m^2 ; J and J_0 , gas flow rate and initial flow rate, respectively, moles/sec; λ , relative rate of change of flow, sec^{-1} ; n_0 , n_1 , and n_2 , molecular concentrations of gas in exhaust system, reactor volume, and instrument volume, respectively, moles/ m^3 ; p and q , constants having dimensions sec^{-1} and sec^{-2} , respectively; $r_{1,2}$, roots of characteristic equation for linear system (1), sec^{-1} ; T_1 and T_2 , temperatures in reactor and instrument volumes, respectively, $^\circ\text{K}$; T_{10} , initial temperature in reactor volume, $^\circ\text{K}$; t , time, sec; τ , time for flow sensitivity to become constant (for fixed T_1 and T_2), sec; V_1 and V_2 , reactor and instrument volumes, respectively, m^3 ; $\langle v_1 \rangle$, and $\langle v_2 \rangle$, mean thermal velocities of gas molecules at temperatures T_1 and T_2 , respectively, m/sec.

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