DETERMINATION OF PARTIAL INTENSITIES OF COMPONENTS OF A MOLECULAR FLUX

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The method described here was devised for determination of the partial intensities of components of a molecular flux, i.e., the products of the concentrations of the component molecules and their mean velocities.

If the mean velocities are known, however, this method can be used to determine the composition of the flux, i.e., the partial concentrations of the components.

To consider the main problems involved in determination of the composition of a flux of low-density gas we turn to some earlier research.

The device used in [1–3] to analyze the composition of a flux is schematically illustrated in Fig. 1.

Part of the investigated flux passes through diaphragm 1 of the device into chamber 2, which is continuously pumped out by diffusion pump 3. The analyzer 4, which is a mass spectrometer, is connected to the chamber.

The composition of the mixture in the analyzers of this type of apparatus differs from the composition of the mixture in the investigated flux. This difference is due primarily to the different conductance of any channel for different components and to the dependence of the pumping speed of the diffusion pump on the kind of gas being pumped.

The effect of the first factor can be allowed for in the analysis of the experimental data, but the selective action of the pump connected to the analyzer chamber cannot be allowed for in any way.

In fact, it is usually assumed that the pumping speed of a diffusion pump is inversely proportional to the square root of the molecular weight of the gas being pumped. In fact, as was shown in [4,5], it is a complex function of such parameters as the heating power, temperature of the cooling water, and pressure drop on the pump.

The following elementary arguments illustrate what has been said. We use the following symbols: w is the mean translational velocity at entrance to diaphragm 1 (Fig. 1); F is the cross-sectional area of diaphragm; $n^{(1)}$ and $n^{(2)}$ are the concentration of components 1 and 2 in binary mixture entering diaphragm; $n^{(1)'}$ and $n^{(2)'}$ are the steady-state concentrations of same components in analyzer; $Q^{(1)}$ and $Q^{(2)}$ are the pumping speeds of pump 3 for different components. (As in [1–3], we assume here that the mean velocities of the components are the same.)

Then, neglecting the resistance of the channel between the diaphragm and the pump, the conditions for conservation of components 1 and 2 of the mixture can be written in the following form:

$$n^{(1)} Fw = n^{(1)'} Q^{(1)}, \qquad n^{(2)} Fw = n^{(2)'} Q^{(2)}.$$

Hence

$$\frac{n^{(1)}}{n^{(2)}} = \frac{n^{(1)'} Q^{(1)}}{n^{(2)'} Q^{(2)}},$$

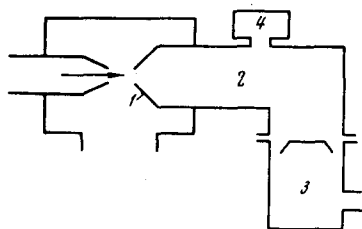where $Q^{(1)}$ and $Q^{(2)}$, generally speaking, are unknown quantities.

In the device used in [6–9] the analyzer was connected on the fore-vacuum side of pump 3. The experimental data obtained by means of this device will be valid only in the case in which an additional diffusion pump is not used to evacuate the analyzer. If such a pump is used, neglect of the selective nature of its action can lead, as in the case of the devices described above, to an appreciable error.

A similar comment can be made in the case of the device described in [10]. This device had another fault: the geometry of the diaphragm was such that the selective action of the main pump affected the results.

In addition to the selective action of the diffusion pump the following factors must be taken into account in determination of the composition of a flux: 1) the passage of molecules of residual gas (background) along with the molecules of the investigated flux into the working chamber;

2) gas release from the walls of the device.

It is obvious that the background will have little effect in investigations of the composition of relatively dense fluxes, but will have a significant effect in determinations of concentrations in rarefied fluxes. Gas release from the walls is not of particular importance in work with inert gases, but may be of great importance in investigations of the composition of a flux containing gases such as nitrogen or oxygen.

The device described below was used to investigate a molecular flux in an apparatus with a gasdynamic source, the idea for which originated with Kantrowitz and Grey [11]. Apparatus of this type is now widely used and has been described in many papers. A review of these papers and an extensive bibliography is contained in [12].

A schematic illustration of the apparatus and the proposed measuring device for the determination of partial intensities is shown in Fig. 2.

The source of the molecular flux in the apparatus is supersonic nozzle 1, in which the gas mixture is accelerated to velocities exceeding the thermal velocities in antechamber 2. Part of the flux at the exit from the nozzle passes through the first diaphragm 3 into the collimator chamber. The second diaphragm 4, situated at the entrance to the working chamber, forms the molecular beam. The nozzle and collimator chambers and the working chamber are evacuated by vacuum pumps.

The measuring device is evacuated by an additional diffusion pump. In this respect the proposed device is similar to the devices used earlier. There is a significant difference, however, in that it is not the steady concentrations in a cross section of the channel connecting the diaphragm and pump which are measured, but the molecu-
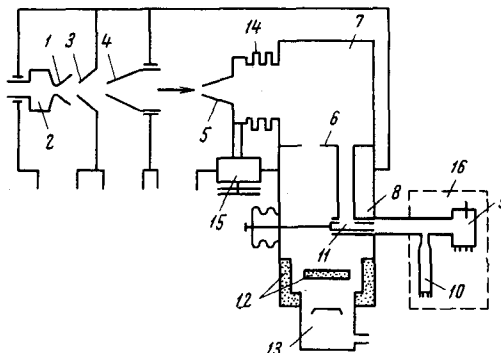

Fig. 1

Fig. 2

lar fluxes of the components through this channel. These fluxes are measured with the sampling nozzle in two positions: in the investigated flux and outside it. This means that with the proposed device the selective action of the diffusion pump can be ignored and the presence of residual gas in the working chamber, gas release, and leakage into the device can be taken into account.

The device for determination of the partial intensities consists of the following main parts: nozzle 5, thin partition 6 with an orifice, the first analyzing chamber 7, the second analyzing chamber 8, an IPDO-1 partial-pressure meter with a RMO-4S omegatron 9 [13], a VIT-1 vacuum guage with an LM-2 ionization gauge, slide valve 11, carbon trap 12, an N-1S oil-vapor pump 13, sylphon 14, a position-fixing device 15 with three translational degrees of freedom, and removable oven 16.

Nozzle 5 of the device provides for entry of part of the investigated flux. It is made in such a way that practically all the flux entering it passes into the first analyzing chamber.

The purpose of partition 6 with the orifice is to produce drops in the partial pressures of the components between the first and second analyzing chambers. If the conductance of the orifice is known the molecular fluxes of the components through the channel of the device can be determined from these drops.

Slide valve 11 allows omegatron 9 to be connected to the first or second analyzing chambers and the partial pressures of the components in these chambers can thus be determined.

Carbon trap 12 prevents the entry of oil vapor into the analyzing chambers.

Diffusion pump 13 pumps out the gas entering the device. In conjunction with the carbon trap it can produce a pressure of not more than 10^{-6} and 10^{-7} mm Hg, respectively, in the first and second chambers when there is no admission of gas.

Sylphon 14 and the position-fixing device are used to move nozzle 5 out of the investigated beam.

The removable oven is used to outgas the RMO-4S omegatron and LM-2 gauge. The oven provides for heating of the RMO-4S and LM-2 and the adjacent tubing to 400° C.

Thus, the device described above allows measurement of the partial pressures of the components in the first and second analyzing chambers for two positions of the nozzle—in the investigated beam and outside it.

To relate the indicated pressures to the intensities of the respective components in the investigated flux we find it useful to introduce the following additional symbols: $p_c^{(\nu)}$ is the partial pressure of ν-th component in working chamber; $p_k^{(\nu)}$ is the partial pressure of ν-th component in k-th analyzing chamber when the nozzle is in the investigated flux; $p_k^{(\nu)*}$ is the partial pressure of the ν-th component in the k-th chamber with the nozzle out of the flux; $N^{(\nu)}$ is the number of molecules of the ν-th component entering the first analyzing cham-

ber in unit time due to gas release and leakage; $K_1/(\mu^{(\nu)})^{1/2}$ is the conductance of nozzle; $K_2/(\mu^{(\nu)})^{1/2}$ is the conductance of orifice connecting analyzing chambers for ν-th component; $w^{(\nu)}$ is the mean velocity of molecules of ν-th component.

Using the introduced symbols we can easily write the conditions for conservation of the number of molecules in the first analyzing chamber with the nozzle in and out of the flux,

$$n^{(\nu)} F w^{(\nu)} + p_c^{(\nu)} \frac{K_1}{\sqrt{\mu^{(\nu)}}} + N^{(\nu)} + p_2^{(\nu)} \frac{K_2}{\sqrt{\mu^{(\nu)}}} =$$

$$= p_1^{(\nu)} \frac{K_1}{\sqrt{\mu^{(\nu)}}} + p_1^{(\nu)} \frac{K_2}{\sqrt{\mu^{(\nu)}}} \,,$$

$$p_c^{(\nu)} \frac{K_1}{\sqrt{\mu^{(\nu)}}} + N^{(\nu)} + p_2^{(\nu)*} \frac{K_2}{\sqrt{\mu^{(\nu)}}} =$$

$$= p_1^{(\nu)*} \frac{K_1}{\sqrt{\mu^{(\nu)}}} + p_1^{(\nu)*} \frac{K_2}{\sqrt{\mu^{(\nu)}}} \,.$$

From these conditions we can obtain

$$n^{(\nu)} F w^{(\nu)} = \frac{K_2}{\sqrt{\mu^{(\nu)}}} \times$$

$$\times \left[\frac{K_1}{K_2} (p_1^{(\nu)} - p_1^{(\nu)*}) + (p_1^{(\nu)} - p_1^{(\nu)*}) - (p_2^{(\nu)} - p_2^{(\nu)*}) \right]. \quad (1)$$

In the device described here the ratio K_1/K_2 is 0.033, and in principle can be made even smaller; equality (1) can be replaced by the following approximate equality:

$$n^{(\nu)} F w^{(\nu)} = \frac{K_2}{\sqrt{\mu^{(\nu)}}} [(p_1^{(\nu)} - p_1^{(\nu)*}) - (p_2^{(\nu)} - p_2^{(\nu)*})]. \quad (2)$$

Thus, if $p_k^{(\nu)}$ and $p_k^{(\nu)*}$ are known, equality (2) can be used to determine the partial intensities of the mixture components. If the velocities $w^{(\nu)}$ are known, Eq. (2) can be used to determine the absolute concentrations of the component molecules in the investigated flux.

The described device was used to determine the partial intensities of the components in a molecular flux of nitrogen and helium.

During the experiments we altered the pressure in the antechamber of the apparatus. Such parameters as the weight composition of the mixture entering the antechamber (50% N_2 + 50% He), the Mach number at the entrance to the first diaphragm (5.2), the distance between the exit sections of the first and second diaphragm (47.5 mm), the diameters of these sections (2.9 and 2.7 mm), and the distance between the entrance section of the second diaphragm and the entrance section of the nozzle (273 mm) were kept constant.

For the components of the binary mixture used for this work we can write relationships similar to (2) by putting $\nu = 1$ for nitrogen and $\nu = 2$ for helium.

Then the ratio W of the intensities of nitrogen and helium in the flux will be determined by the formula

$$W = \frac{n^{(1)} w^{(1)}}{n^{(2)} w^{(2)}} =$$

$$= \left(\frac{\mu^{(2)}}{\mu^{(1)}} \right)^{1/2} \frac{(p_1^{(1)} - p_1^{(1)*}) - (p_2^{(1)} - p_2^{(1)*})}{(p_1^{(2)} - p_1^{(2)*}) - (p_2^{(2)} - p_2^{(2)*})} \,. \quad (3)$$

The relationship, corresponding to formula (3), between the ratio of the nitrogen and helium intensities in the flux and the pressure p_0 mm Hg in the forechamber is shown in Fig. 3 (curve 1).

Curve 2 in Fig. 3 is plotted without consideration of gas release from the walls of the device and the penetration into it of molecules of residual gas from the working chamber. To plot this curve we used the formula

$$W = \left(\frac{\mu^{(2)}}{\mu^{(1)}} \right)^{1/2} \frac{p_1^{(1)} - p_2^{(1)}}{p_1^{(2)} - p_2^{(2)}}$$
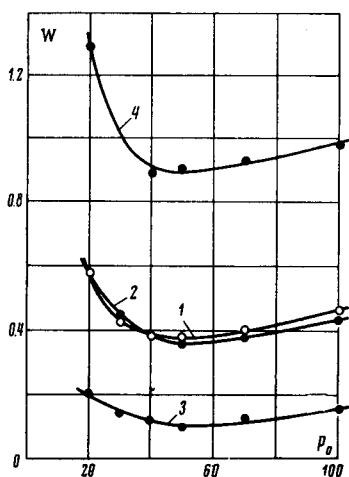
obtained from formula (3) with $p_k^{(\nu)*} = 0$.

Fig. 3

577

To construct curve 3 we used the formula

$$W = \left(\frac{\mu^{(2)}}{\mu^{(1)}} \right)^{1/2} \frac{p_2^{(1)}}{p_2^{(2)}},$$

i.e., we assumed, as before, that the gas release and background could be neglected and also that the pumping speed of the diffusion pump for the ν-th component is inversely proportional to $\sqrt{\mu^{(\nu)}}$.

Finally, curve 4 was plotted on the assumption that the composition established in the first analyzing chamber was identical to the composition of the molecular flux, i.e., that

$$W = \frac{p_1^{(1)}}{p_1^{(2)}}.$$

A comparison of the curves illustrated in Fig. 3 shows that for a quantitative determination of the intensities of components of a molecular flux the gas release, the leakage, and, in particular, the selective action of the diffusion pump must, in fact, be taken into account. It is wrong to assume that the composition of the molecular flux is the same as the composition established beyond the nozzle.

In conclusion it should be noted that with the same accuracy of determination of partial pressures which can be obtained with the IPDO-1 instrument the relative error in determination of the ratio of intensities of the mixture components by means of the proposed system of analysis does not exceed 10%.

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