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Abstract

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## PHYSICAL CHEMISTRY

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### MEASUREMENT OF THE VELOCITY OF MOTION OF AEROSOL PARTICLES IN THE FIELD OF DIFFUSION OF WATER VAPOR

(Presented by Academician M. M. Dubinin, 20 IX 1963)

Diffusiophoresis of aerosol particles is important for their absorption by droplets of aqueous fog both in the atmosphere and in installations for dust collection using atomized salt water, as well as in inducing precipitation with granulated "dry ice." Under laboratory conditions the phenomenon of diffusiophoresis was observed by Facy<sup>(1)</sup>, Waldmann and Schmitt<sup>(2,3)</sup>. The aim of the present work is to measure the velocity of motion of aerosol particles in the field of diffusion of water vapor on the basis of the jet method developed by us.

**Theory.** Two limiting cases of diffusiophoresis should be distinguished: small and large particles, corresponding to the conditions  $r \ll \lambda$  and  $r \gg \lambda$ , where  $r$  is the particle radius and  $\lambda$  is the mean free path of gas molecules.

The first case was considered by B. P. Deryagin and S. P. Bakanov<sup>(4)</sup> (in this case the presence of aerosol particles does not substantially alter the distribution of molecular velocities in the diffusing gas mixture, found by Chapman and Enskog, which makes it possible to calculate the resultant force acting on an aerosol particle), who obtained for the velocity of steady-state diffusiophoresis in the system of the center of mass of the mixture of diffusing molecules the expression \*

$$U = -D_{12} \text{grad } n_1 \frac{n(m_1 m_2)^{1/2}}{\rho n_2} \frac{(m_{12}^{1/2} - 1)}{(n_{12} m_{12}^{1/2} + 1)}, \quad (1)$$

where  $n_1$  and  $n_2$  are the numbers of molecules of the components in 1 cm<sup>3</sup>;  $n = n_1 + n_2$ ;  $m_1$  and  $m_2$  are the molecular masses;  $\rho$  is the density of the mixture;  $D_{12}$  is the coefficient of mutual diffusion of the mixture;  $m_{12} = m_1/m_2$ ;  $n_{12} = n_1/n_2$ .

On passing from this coordinate system to the laboratory system with walls that neither absorb nor evaporate water vapor, equation (1) is transformed as follows:

$$U = -\frac{m_{12}^{1/2} - 1}{m_{12}m_{12}^{1/2} + 1} \frac{D_{12}}{n_2} \text{grad } n_1. \quad (2)$$

If the first component of the mixture is absorbed by a drying substance on one wall and evaporates from the opposite moist wall,  $U$  is given by the expression:

$$U = -\frac{1}{n_1 + n_2m_{21}^{1/2}} \frac{D_{12}n}{n_2} \text{grad } n_1. \quad (3)$$

Expressing  $n_1, n_2$  through the mole fractions  $N_1 = n_1/n$  and  $N_2 = n_2/n$ , instead of (3) we obtain:

$$U = -\frac{1}{N_1 + N_2m_{21}^{1/2}} \frac{D_{12}}{N_2} \text{grad } N_1 = \frac{1}{N_1 + N_2m_{21}^{1/2}} \frac{d \ln N_2}{dy}, \quad (4)$$

where  $N_1 + N_2 = 1$ .

Choosing the direction of the  $y$ -axis along the diffusion flux of vapor, we take as boundary conditions, at  $y = d$ ,  $N_2 = N_2(d)$ , and at  $y = 0$ ,  $N_2 = N_2(0) = 1$ , where  $d$  is the distance over which the relative humidity drops from 100% to 0. Taking into account the adopted boundary conditions and assuming, in the first approximation, the linearity of  $\ln N_2$ , equation (4) may be written—

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\* Later an identical relation was obtained by Waldmann.

written in the form:

$$U = \frac{1}{N_1 + N_2m_{21}^{1/2}} \frac{D_{12}}{d} \ln \frac{N_2(d)}{N_2(0)}. \quad (5)$$

Since  $\ln(N_2(d)/N_2(0)) \approx \ln[1 - N_1(d)] \approx -N_1(d)$ , instead of (5) one may write

$$U \approx -\frac{1}{N_1 + N_2m_{21}^{1/2}} \frac{D_{12}}{d} N_1(d), \quad (6)$$

where  $N_1(d)$  is the molar fraction of water vapor at the evaporating surface;  $m_{21} = m_2/m_1$ ;  $N_1$  and  $N_2$  are values averaged over the interval  $0, d$ .

**Measurement procedure.** Between parallel horizontal planes  $a$  and  $b$  (Fig. 1), exactly midway between them, a flat aerosol jet is passed, emerging from a rectangular nozzle and having, in cross-section, a height of 0.01 cm and a width of 0.05 cm. The aerosol jet is located inside an enveloping filtered flow having the same linear velocity, which prevents widening of the jet. With a vertical

Fig. 1. Schematic of the apparatus for measuring the velocity of motion of aerosol particles in the field of diffusion of water vapor

Figure 1: Fig. 1. Schematic of the apparatus for measuring the velocity of motion of aerosol particles in the field of diffusion of water vapor

concentration gradient of water vapor, the aerosol jet was displaced from its initial

Fig. 1. Schematic of the apparatus for measuring the velocity of motion of aerosol particles in the field of diffusion of water vapor

mean position. This displacement of the jet was measured with microscope  $M$  from the end part of the cuvette under side illumination.

Into the cuvette, made of organic glass, two cassettes  $a$  and  $b$  were inserted, facing one another with stretched brass meshes. Purified and degreased cotton wool was placed in the lower cassette, and phosphorus anhydride in the upper one. In the lower cassette, on the outside, a hollow tube was soldered in, intended simultaneously for inserting a thermocouple and water when wetting the cotton plug.

The experimental procedure was as follows. Cassettes  $a, b$  were inserted into the cuvette. With microvalves  $H_1, H_2, H_3$  closed, air was pumped out to a specified pressure, measured by manometer  $Q$ . The system was then disconnected from the pump by means of stopcock  $H_4$ . Using microvalve  $H_1$ , a flow of filtered air with a definite linear velocity was established. After this, a flow with aerosol was passed through microvalve  $H_2$ . The volumetric rate of the total air flow that had passed through the cuvette was measured with rheometer  $R_2$ . At pressures exceeding 200 mm Hg, the velocity of diffusiophoresis was also measured at a constant pumping rate using a water-jet pump. In the absence of a water-vapor diffusion field (with dry cotton wool), the jet occupied the mean position between the planes, which was taken as zero. Opening microvalve  $H_3$ , the cotton wool was wetted with a measured amount of distilled water, and then, already with microvalve  $H_3$  closed, the magnitude of the jet displacement, the pressure in the system, and the volumetric rate of the total flow were measured simultaneously. In the presence of a gradient of water-vapor concentration, the displacement of the jet was measured. Owing to the vessel ( $W \sim 50$  l) connected in series into the system, it was possible to carry out measurements without a noticeable change

pressure changes in the system at the low velocities and measurement times used.

Aerosols of vaseline oil and paraffin were obtained by heating these substances at constant temperature and with a vapor-blowing rate measured by rheometer  $R_1$ ; the aerosol of magnesium oxide was obtained by burning magnesium wire. The vaseline-oil particles were relatively monodisperse, with radius  $r_{av} \sim 0.25 \cdot 10^{-4}$

cm. The polydisperse aerosols of paraffin and magnesium oxide had approximately  $r_{av} \sim 0.5 \cdot 10^{-4}$  cm.

The temperature of the evaporation surface was measured by thermocouple  $T$ . Since  $D_{12}$  is inversely proportional to the pressure  $p$ , then as the latter decreased the temperature of the evaporation surface could decrease. To avoid this, the temperature of the evaporation surface was kept constant ( $15.5^\circ$ ) by regulating the temperature of the room air. The temperature of the condensation surface rose by at most  $1.5-2^\circ$  (at pressures below 280 mm). An estimate of the resulting thermophoretic force, directed in this case opposite to the acting diffusion force, was made using the formula of B. V. Deryagin and S. P. Bakanov<sup>(5)</sup> for the case  $r \ll \lambda$ . The thermophoresis velocity for this temperature gradient is at most  $1.0 \cdot 10^{-3}$  cm/sec, i.e., 1% of the diffusiophoresis velocity of the particles measured at 150 mm.

The diffusiophoresis velocity of aerosol particles  $U$  was determined in the pressure interval 150-660 mm. Knowing the horizontal component of the velocity  $\bar{V}$ , the displacement  $y$  of the jet, and the distance  $l$  from the jet inlet to the observation plane, for the vertical component of the velocity of the aerosol particles, i.e., for the diffusiophoresis velocity, we obtain:

$$U = \frac{y}{l} \bar{V} \text{ cm/sec}, \quad (7)$$

where  $V$  is the average velocity of the particles along their path to the observation point. It can be determined from the equation:

$$\bar{V} = \frac{1}{y} \int_0^y V(y) dy = V_{\max} \left[ 1 - \frac{1}{3} \frac{y^2}{(d/2)^2} \right], \quad (8)$$

where  $\bar{V}_{\max}$  corresponds to the particle velocity in the middle position of the jet, and  $d/2$  is half the distance between the planes.

The experiments were carried out at an average linear flow velocity of 0.35 cm/sec. The maximum experimental error was 10%. A linear gradient of the concentration of water vapor as a function of  $y$  was established at some distance from the nozzle. The length of the initial section with an unestablished vertical concentration gradient amounted to 10% of the distance from the nozzle to the observation plane.\*

**Results of the measurements and their discussion.** In Fig. 2, the measured diffusiophoresis velocities of aerosol particles of vaseline oil were plotted as a function of  $1/p^2$ . The theoretical curve was calculated by formula (6). In doing so it was assumed that  $N_1 = 0^{**}$  and  $D_{12} = 0.22$  cm/sec<sup>\*\*\*</sup>. The mole fraction  $N(d)$  of water vapor was determined as the ratio of the pressure of saturated water vapor at the given temperature of evaporation of the surface to

Fig. 2 and Fig. 3: plots of aerosol-particle velocity versus  $1/p^2$

Figure 2: Fig. 2 and Fig. 3: plots of aerosol-particle velocity versus  $1/p^2$

the total pressure in the system  $P$ . In calculating the theoretical curve, the inverse dependence of the diffusion coefficient on pressure was taken into account.  $d = 0.7$  cm, allowing for the thicknesses of the grids.

We see that in the pressure interval 150–400 mm, which corresponds to  $r/\lambda \sim 0.7 \div 2$ , the experimental data are satisfactorily described

\* A theoretical estimate of the length of the initial section  $a$ , characterized by an unestablished vertical concentration gradient, was made by S. S. Dukhin:  $a \sim \frac{1}{2}d \text{Re}$ , where  $d$  is the distance between the planes and  $\text{Re}$  is the Reynolds number.

\*\* Since, under the experimental conditions,  $N_1 \ll 1$ , the adopted assumption  $N_1 = 0$  introduces no substantial error into the calculation of the theoretical curve.

\*\*\*  $D_{12}$ , the diffusion coefficient of water vapor in air, was calculated as  $D_{12} = D_0(T/T_0)^2$ , where  $D_0 = 0.203$  cm/sec,  $T_0 = 273^\circ\text{K}$ ,  $T = 273 + 15.5^\circ\text{K}$ .

the theoretical formula of Derjaguin and Bakanov (6), which is therefore applicable not only in the region  $r/\lambda \ll 1$ . In the pressure region above 400 mm, a noticeable deviation of the experimental data from the theoretical curve is observed; the former are approximately twice as large as those calculated from formula (6).

The velocity of motion of aerosol particles of paraffin and magnesium oxide was also investigated at pressures exceeding 400 mm (see Fig. 3). Agreement, within the experimental error, of the data obtained for three substances

Fig. 2. Velocity of motion of aerosol particles of petroleum jelly oil as a function of  $1/p^2$

Fig. 3. Velocity of motion of aerosol particles as a function of  $1/p^2$ .  $a$ —petroleum jelly oil,  $b$ —magnesium oxide,  $v$ —paraffin

(petroleum jelly oil, paraffin, magnesium oxide) with different particle radii shows that the dependence of the velocity of diffusiophoresis on particle size is absent for  $r/\lambda \gg 1$ . This also proves the absence of spreading of a polydisperse aerosol jet in the field of diffusion of water vapor.

In addition, a comparison was made of the behavior of particles soluble and insoluble in water. Measurements of the diffusiophoresis velocities of particles of ammonium chloride and petroleum jelly oil at  $p \sim 760$  mm showed that these velocities coincide within the experimental error.

**Conclusions.** The experimental data for the velocity of diffusiophoresis of aerosol particles in the field of diffusion of water vapor, obtained in the present

work in the pressure interval 150–400 mm, which corresponds approximately to  $r/\lambda \sim 0.7 \div 2$ , are in satisfactory agreement with the formula of Derjaguin and Bakanov, which applies to the case of small particles.

It was found that the velocity of motion of aerosol particles satisfying the condition  $r \gg \lambda$  is approximately twice as large as the velocity calculated from the above-mentioned formula.

On the basis of the experimental data, it may be assumed that the dependence of the velocity on particle size is also absent for  $r/\lambda > 2$ .

No difference was experimentally detected in the behavior of particles soluble and insoluble in water.

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