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AN EXPERIMENTAL STUDY OF THE FILTERING CAPACITY OF A THERMAL
DIFFUSION CHAMBER CONTAINING A CONTROL ELEMENT

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Experimental results are presented on the deposition of a finely divided aerosol in a planar thermal-diffusion flow chamber containing a control element [1].

Diffusiophoresis has been used [2-7] to remove aerosols from gases (directional motion of the aerosol particles in a concentration gradient). The particle velocity is directly proportional to the concentration gradient in the diffusing component [8, 9], so thermal-diffusion chambers (TD chambers) are used to produce high stationary gradients [1, 3, 6, 7]. A major disadvantage of such a filtering device is that there is a positive supersaturation in the working space [10], which leads to vapor flow to the aerosol particles (by heterogeneous condensation), which tends to reduce the concentration gradient and thus reduces the filtration performance. This is very important if the particle concentration is more than 10^{10} - 10^{11} m^{-3} , but it can be eliminated in a TD chamber with a control element, whose operation has been described in detail in [11]. The control element takes the form of a heated grid permeable to the vapor molecules placed between continuously wetted surfaces differing in temperature, one of which produces the vapor and the other absorbs it. If the temperature of the grid is equal to the temperature of the heater (evaporator), the state of the vapor-gas mixture will be that of negative supersaturation in the region of the control element, which is called the working zone, but with a vapor concentration gradient determined as in an ordinary TD chamber by the temperatures of the cooler and heater and by the distance between them.

A quantitative measure of the filtration performance is provided by the breakthrough coefficient K_b , which is equal to the ratio of the aerosol concentrations at the outlet and inlet. The performance is commonly measured by a method based on nephelometry of a mono-disperse aerosol before and after the filter [12]. If the aerosol is finely divided, some enlargement of the particles is produced before nephelometry in the supersaturated vapor of a high-boiling substance in a device of KUST type [13]. This method cannot be used to measure the breakthrough coefficient for a TD chamber because of the enlargement of the particles by condensation at the outlet from the TD chamber due to cooling of the air flow with its high vapor content in the pipelines of the nephelometer and KUST.

During preliminary studies, it was shown that the aerosol may be deposited completely in the TD chamber.

Zero value of K_b (the absence of particles at the exit from the TD chamber is readily detected) is attained at a certain value of the volume flow rate Q^* through the TD working zone, which is subsequently called the critical value. The value of Q^* will be dependent on the working and geometrical parameters of the TD chamber, and also on the physical characteristics of the aerosol and vapor-gas medium.

We have measured Q^* ($K_b = 0$) for various conditions in the TD chamber and various parameters for a finely divided sodium chloride aerosol (Knudsen number $Kn \gg 1$) with the

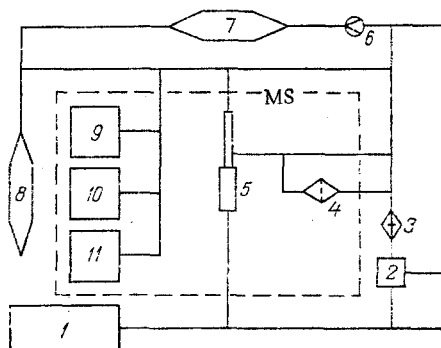


Fig. 1. Block diagram of the apparatus: 1) flow generating unit; 2) highly dispersed aerosol generator; 3) unselective diluent block; 4) fan filter; 5) KUST particle enlarger; 6) rheometer; 7) thermal diffusion chamber containing control element; 8) indicating thermal diffusion chamber; 9) FÉN-58 nephelometer; 10) VDK-4 ultramicroscopic counter; 11) AZ-2M modified aerosol particle counter.

apparatus shown in Fig. 1. The main units were as follows: flow thermal-diffusion chamber with control element (TDC), indicator flow thermal diffusion chamber (ITDC), highly dispersed aerosol generator (HDAG), measuring system (MS), and unit providing the flow of purified air (FGU).

The air flow generating unit consists of compressors type UK-40, damping vessels, a coarse filter (wadding), a fine filter (FPP-15), an electrical filter, and vessels containing silica gel and activated charcoal. The degree of purification in the air is monitored either directly by the nephelometer after passing through the KUST or by means of the indicating thermal-diffusion chamber.

The highly dispersed aerosol generator is of mixer type (a detailed description is given in [14]), and this provides a sodium chloride aerosol with a fairly narrow spectrum (standard geometrical deviation β_g not more than 1.5) in the range of particle sizes $r_g \in (20-100) \cdot 10^{-10}$ m with a numerical concentration in the range $10^9-10^{14} \text{ m}^{-3}$, which was dependent on the particle size.

The measurement system was used in determining and checking the aerosol parameters, the thermal conditions in the TD chamber, and the critical flow rate.

The particle sizes were determined by a diffusion method based on measuring the breakthrough coefficient in a model system. We used a fan filter for this purpose [15-17]. The methods of measuring the breakthrough and the apparatus were similar to those described in [12, 13, 18] (FÉN-58 nephelometer, KUST enlarger, and unselective diluters).

The numerical particle concentration was measured after enlargement (with allowance for the dilution factor at high concentrations) with a VDK-4 ultramicroscope, while at low concentrations ($<10^9 \text{ m}^{-3}$) it was measured with a modified AZ-2M counter connected to a PP-15 scaler (not shown).

The thermal conditions in the TD chamber was monitored by means of copper-constantan differential thermocouples and an F-30 digital voltmeter (we measured the temperatures of the heaters, coolers, and control element at several points). The measurement error was 0.1° .

The volume flow rate of the air reaching the TD chamber was determined with a capillary rheometer with an error of 3%.

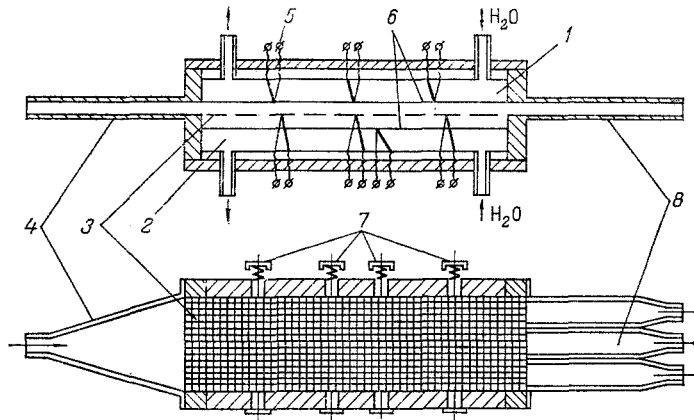


Fig. 2. Scheme for thermal-diffusion chamber containing control element.

Figure 2 shows the essential scheme of the TD chamber containing the control element. The planar heaters 1 and cooler 2 were enclosed in cloth (cambric) 6 and formed two parallel planes of size 0.5×0.1 m separated by a distance of $15 \cdot 10^{-3}$ m; their temperatures were maintained at a given level by water thermostats of UT-15 type. The heater and cooler were continuously wetted with distilled water until water films appeared on the working surfaces using a special supply system not shown. Between these planes at a distance of $6 \cdot 10^{-3}$ m from the heater was the control element 3, which was made from a nickel grid of wire diameter $50 \cdot 10^{-6}$ m and cell dimensions $(75 \times 75) \cdot 10^{-12}$ m². The grid was heated by a precision controlled voltage source. To prevent the grid from sagging as a result of expansion on heating, there was the automatic tensioning system 7. The temperatures of the working surfaces of the plates and control element were monitored with copper-constantan differential thermocouples 5.

In the space between the heater 1 and the control element 3 (working zone of the TD chamber), the flow containing the aerosol was introduced by the heated nozzle 4 (the flow temperature was equal to the temperatures of the heater and control element). The aerosol passed through the working zone in the chamber and reached the heated three-section exit nozzle 8 (heated to prevent vapor concentration), whose central part was joined to the aerosol indicator. This design of exit nozzle enabled us to neglect the influence of edge effects on the results.

The aerosol at the exit from the filter chamber was recorded by the particle-enlargement method in an ordinary planar flow TD chamber placed vertically. The aerosol was introduced into the indicator through a thin-walled nozzle placed along the central axis. To prevent turbulence and consequent jet spread, the nozzle was surrounded by a sheath flow of purified air.

The enlargement of the particles to visible size was observed with dark-field illumination through the lateral transparent walls of the indicator TD chamber using a KM-8 cathetometer.

The methods involved measuring the volume flow rate for an aerosol with known parameters supplied to the TD filter chamber (the chamber had previously been brought to a steady thermal state) while checking that no particles appeared at the exit.

Table 1 gives the basic parameters characterizing the working conditions and the results for Q^* as referred to the working zone. The molar water-vapor concentrations at the working surfaces of the TD chamber $C_{10}(T')$, $C_{1d}(T'')$ were calculated from the data of [19], and these with film wetting were equal to the saturated vapor concentrations at the corresponding temperatures.

Table 1 shows that as the temperature difference increases (and correspondingly the difference in the concentrations), the flow rate of completely purified air increases.

It is of interest to relate Q^* to the modulus of the vapor concentration gradient $|\vec{\nabla}C_1|$.

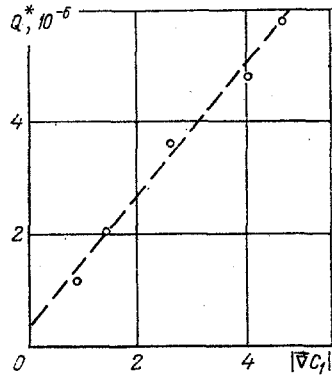


Fig. 3

Fig. 3. Dependence of the critical flow rate Q^* (m^3/sec) on the concentration gradient for water vapor $|\vec{\nabla}C_1|$ (m^{-1}).

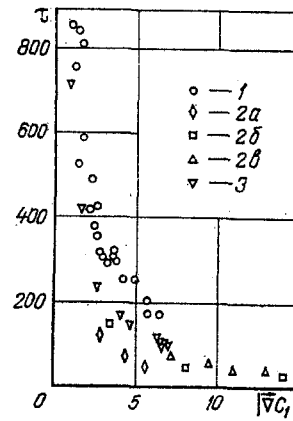


Fig. 4

Fig. 4. Dependence of the aerosol deposition time τ (sec) on the concentration gradient for water vapor $|\vec{\nabla}C_1|$ (m^{-1}) for a channel of height $d = 0.02$ m: 1, 2) experimental data of [21] and [3] correspondingly ($1 - \langle C_1 \rangle \in (1-2)\%$; $2a - \langle C_1 \rangle \in (55-58)\%$; $2b - \langle C_1 \rangle \in (44-55)\%$; $2c - \langle C_1 \rangle \in (36-48)\%$; 3) results from our experiments ($\langle C_1 \rangle \in (10-16)\%$).

Figure 3 shows the dependence of Q^* on $|\vec{\nabla}C_1|$ as calculated for the linear distribution of the vapor concentration in the working zone [11]. The experimental data are described satisfactorily by the linear function

$$Q^* = Q_0 + a|\vec{\nabla}C_1| \quad (1)$$

where $Q_0 = (0.36 \pm 0.13) \cdot 10^{-6} \text{ m}^3/\text{sec}$; $a = (1.17 \pm 0.04) \cdot 10^{-6} \text{ m}^4/\text{sec}$.

The value of Q_0 differs from zero because of deposition by diffusion. We estimated Q_0 from de Marcus's formula [20] for the working conditions, and this agreed satisfactorily with our measured Q_0 .

Measurements have been made [3] on a filter based on a planar TD chamber. Tobacco smoke was used (particle radius about $4 \cdot 10^{-7}$ m), and the mass flow rate was measured for purified air at which the aerosol was deposited completely. The results were presented as the total deposition time as a function of heater temperature for various cooler temperatures when the separation was $2 \cdot 10^{-2}$ m.

Values have been published [21] for the rates of diffusio-phoretic deposition of a highly dispersed aerosol: ($\langle r \rangle \approx 3 \cdot 10^{-8}$ m). The data were obtained under isothermal conditions in a planar slot channel, where the water-vapor concentration difference was produced by the use of hygroscopic substances (solutions of sulfuric acid and phosphorus pentoxide). These data can be used to calculate the total deposition times.

Our conditions also allowed us to calculate the deposition times τ , which are given along with the data of [3, 21] in Fig. 4 in the form of $\tau = \tau(|\vec{\nabla}C_1|)$ for a channel having $d = 0.02$ m.

In all the experiments, τ decreased as $|\vec{\nabla}C_1|$ increased, and τ was the less the higher the mean vapor content $\langle C_1 \rangle$ in the filter. In fact, in the experiments of [21] $\langle C_1 \rangle \in (1-2)\%$, while in ours $\langle C_1 \rangle \in (10-16)\%$, and in [3] $\langle C_1 \rangle \in (36-58)\%$. This result occurs because the deposition rate for a finely divided aerosol is determined in the main by the speed of the Stefan flux [22], which is inversely proportional to $(1 - \langle C_1 \rangle)$.

A major characteristic of the filter, which enables one to estimate the energy consumption, is the amount of vapor deposited in the filter in order to purify unit mass of air I_1/I_2 .

The specific vapor flow rate for each experiment was determined from

TABLE 1. Measured Specific Vapor Flow Rates I_1/I_2 and Ones Calculated from Formulas in the Theory of Aerosol Particle Deposition [23]

$T'=T''', K$	T'', K	$C_{10}(T')$	$C_{1d}(T'')$	$Q^*, 10^{-6}$ m^3/sec	$r_g, 10^{-10}$ m	I_1/I_2	
						theor.	exp.
320,6	318,2	0,108	0,096	1,21	32	0,856	0,670
321,3	317,2	0,112	0,091	2,05		0,858	0,655
323,0	315,0	0,122	0,081	3,64		0,863	0,684
324,6	312,6	0,132	0,071	4,84		0,868	0,810
325,3	311,3	0,136	0,066	5,82		0,870	0,778
324,2	298,9	0,129	0,033	7,92	26	0,860	0,763
325,9	303,0	0,140	0,042	8,10		0,869	0,780
327,5	307,0	0,152	0,053	8,07		0,878	0,806
329,8	312,0	0,169	0,069	8,30		0,893	0,823
332,2	317,0	0,189	0,090	8,33		0,912	0,847

$$\left(\frac{I_1}{I_2}\right)_{\text{exp}} = \frac{D_{12}n(T') m_1 F}{n_2(T'') m_2 Q^* d} \ln \frac{1 - C_{1d}}{1 - C_{10}} \quad (2)$$

The results from deposition theory in [23] were used in calculating analogous quantities for the same conditions (see Table 1):

$$\left(\frac{I_1}{I_2}\right)_{\text{theor}} = \frac{m_1}{m_2} \frac{\Phi_1(s)}{1 + A} \quad (3)$$

where

$$\Phi_1(s) = \left(\frac{1}{2} - \frac{1}{1 - \exp \omega} - \frac{1}{\omega} \right) \frac{1}{(1 - C_{10}) \Psi} + \frac{m_2 - m_1}{m_1} - \beta_0 \left[1 + \frac{(m_2 - m_1) [1 - C_1(d_1)]}{2m_1 s \Psi \exp s} \left(\frac{\omega [1 - \exp(\omega + 2s)]}{(\omega + 2s)(1 - \exp \omega)} + \frac{1 - \exp 2s}{2s} \right) \right]; \quad (4)$$

$$\Psi = \frac{1}{s} \left(1 - \frac{1}{s} \right) \exp s - \frac{\exp s - 1}{s(1 - \exp \omega)} + \frac{1}{s^2} + \frac{\exp(\omega + s) - 1}{(\omega + s)(1 - \exp \omega)}; \quad (5)$$

$$s = \ln \frac{1 - C_1(d_1)}{1 - C_{10}}; \quad (6)$$

$$\beta_0 = \frac{(m_2 - m_1) A}{m_1 (1 + A)}; \quad (7)$$

$$\omega = \frac{1}{\mu} n D_{12} m_1 s. \quad (8)$$

For an aerosol with $Kn \gg 1$ [22],

$$A = \frac{m_2^{1/2} - m_1^{1/2}}{\langle C_1 \rangle m_1^{1/2} + \langle C_2 \rangle m_2^{1/2}} \frac{(m_1 m_2)^{1/2}}{\langle C_1 \rangle m_1 + \langle C_2 \rangle m_2} \langle C_2 \rangle. \quad (9)$$

The calculated and measured values for the specific mass vapor flow rate differ slightly (discrepancy about 10-20%), but they are in qualitative agreement. Also, I_1/I_2 increases with the concentration gradient. The reason for the discrepancy appears to be that the calculation of I_1/I_2 in [23] neglected Brownian motion. Estimates show that the Brownian diffusion increases I_2 (with I_1 constant) by about 15-25%. Therefore, one assumes that the theory of [23] for particle deposition in TD chambers can be used in engineering calculations on filters based on TD chambers with control elements.

Similar experiments were performed on the atmospheric aerosol, which has a wide particle size spectrum. The above regularities are completely applicable to a filter working with a polydisperse aerosol.

The main advantage of a filter of this design is that it can give ultrafine gas purification from aerosols over wide ranges in particle size and concentration.

NOTATION

T' , T'' , T , temperatures of heater, control element, and cooler, respectively; $C_{10}(T')$, $C_{1d}(T'')$, $C_1(d_1)$, molar concentrations of saturated water vapor near heater and cooler surfaces and vapor concentration near control element; $\langle r \rangle$, r_g , mean and geometric mean radii of aerosol particles; Kn , Knudsen number; λ , mean free path of gas molecules; d_1 , d , heater control element and heater-cooler distances; F , F_1 , surface area of heater and of the control element free for molecular vapor flow (total area of grid cells); $|\nabla C_1|$, absolute water vapor concentration gradient; Q^* , Q_0 , critical flow rates through thermal diffusion chamber for $|\nabla C_1| \neq 0$ and $|\nabla C_1| = 0$, respectively; τ , deposition times in the thermal diffusion chamber channel; $\langle C_1 \rangle$, $\langle C_2 \rangle$, mean relative concentrations of vapor molecules (vapor content) and air; I_1 , I_2 , mass fluxes of water vapor and clean air; m_1 , m_2 , molecular masses of water and air; μ , dynamic viscosity; n_1 , n_2 , concentrations of water vapor and air; $n = n_1 + n_2$; D_{12} , mutual diffusion coefficient of air-vapor mixture.

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DIFFUSION SEPARATION OF HYDROGEN FROM GAS MIXTURES

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Experiments are described and a model is evaluated for the process of production of superpure hydrogen from mixtures with ammonia and nitrogen by selective diffusion through thin metallic membranes.

Current predictions indicate that the worldwide demand for hydrogen for use in energy production and industry will increase by a factor of at least 4-5 times by the end of the present century [1, 2]. In a number of industries (foremost ferrous metallurgy and chemical technology) there is an increasing need for hydrogen of very high purity (more than 99.999%).

In principle hydrogen of such high purity can be obtained either by the conventional electrolytic method, or by using selective diffusion through membranes of palladium and its alloys [3-5]. In contrast to the majority of other metals, glasses, polymers, etc., those materials are highly permeable to hydrogen, while remaining practically impermeable to other gases [6]. Among the advantages of the diffusion method are simplicity of construction, easy mobility and long lifespan of the equipment, low cost, high (up to 99.9999%) purity of the product, simplicity, and safety in use [7, 8]. Calculations performed with data from experimental industrial tests of diffusion equipment indicate that the unit cost of hydrogen produced by this method in equipment operating at the 500-1000 m³/h level is 40-70 rubles/1000 m³, i.e., 2-3 times less expensive than hydrogen produced electrolytically, while having a significantly higher purity.

At the present time the diffusion method is used for hydrogen production in a wide range of small scale (up to 100 m³/h) devices. Attempts to design and utilize larger-scale separators based on tube type diffusion cells (undertaken mainly in the USA) have not met with success, due to technological unreliability of diffusion cell construction and losses related to cell combination. The latter is to a great degree the result of difficulties in mathematical modeling of the mass-exchange processes in real diffusion separators with consideration of inhomogeneity in mixture composition and pressure in the cells, hydraulic resistance, etc. Examples of theoretical studies can be found in [9-11].

New possibilities for development of large-scale diffusion technology have appeared as a result of multifaceted studies of metal physics and welding technology which have made possible construction of high-output diffusion cells with highly reliable sealing properties. The equipment in which these cells are used consists of a group of alternating supply and output channels, separated by thin walls which are permeable only to hydrogen. In the present study we will consider hydrogen diffusion separation in cells with planar channels

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