EXPERIMENTAL STUDY OF GAS FLOW THROUGH FINE-POROUS MEDIA IN THE TRANSITION PRESSURE RANGE

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Abstract

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EXPERIMENTAL STUDY OF GAS FLOW THROUGH FINE-POROUS MEDIA IN THE TRANSITION PRESSURE RANGE

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The question of the nature of the dependence of the specific gas flow (the permeability g* of a porous medium or capillary) on the mean pressure $\bar{p}$ is closely connected with physical ideas about the transport mechanism at different values of the Knudsen number ($\mathrm{Kn} = \lambda/d$, where λ is the mean free path of gas molecules and d is the diameter of the capillary or the characteristic geometrical dimension for a porous medium).

One of the most difficult questions arising in the theoretical treatment of gas flow in the transition pressure region ($\mathrm{Kn} \simeq 1$) is the correct allowance for viscous flow. It is often assumed ([1,2]) that the total flow through a porous medium can be represented as the sum of a "diffusive" flow G_D, constant over the entire pressure range, and a viscous flow G_V, increasing with pressure from $\bar{p} = 0$. The dependence of the specific flow on pressure should then be linear. The appearance of a minimum on the curve $g = f(\bar{p})$ for long capillaries is usually explained ([3]) by a decrease, with increasing pressure, in the fraction of long-free-path molecules in the flow. The absence of this minimum for many porous media is explained from this point of view by the fact that a porous medium should not have long through capillaries and may rather be represented as a set of short curved tubes. There are also other explanations, different from that mentioned above, for the linear dependence $g = f(\bar{p})$ for porous media ([4]). On the other hand, transport theories developed in ([5,6]) predict the appearance of a minimum in the specific flow during the flow of gases through porous media.**

It should be noted, however, that the validity of the assumption of the appearance of a viscous flow (the hypothesis of additivity of G_V and G_D) at the very lowest pressures, when the concept of normal viscosity loses its physical meaning, raises serious doubts. Recently several experimental works ([7–11]) have been published in which various nonlinear dependences $g = f(\bar{p})$ are described, including curves with a minimu—

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* By permeability we shall mean a dimensionless parameter characterizing the probability of transfer through a porous medium and determined by the struc-

tural properties of the latter. The permeability can be expressed in terms of the flow by the formula

$$g = \frac{G \text{ mol/sec}}{\Delta p S_r \cdot v_t / 4\, RT},$$

where Δp is the pressure drop across the specimen; S_r is the cross-sectional area of the porous medium; v_t is the thermal velocity of the gas molecules; T is the temperature of the experiment; R is the universal gas constant.

** In work (^{5}), in the graphical representation of the dependence G/G_{Kn} on Kn^{-1}, an error in scale was made. When the curve is plotted on the correct scale, the "minimum" of the curve is much less pronounced. Allowance for the viscous flow in accordance with the corrections proposed in (^{6}) leads to a strong "stretching" of the minimum (see Fig. 3).

mometers. However, such results were obtained mainly either on so-called consolidated porous bodies (low-permeability graphites, ceramics with low porosity), or on very highly porous specimens (cotton wool, cardboard).

We investigated the dependence of the specific flow of various gases on the mean pressure and temperature in compacts made from highly dispersed powders over a wide range of porosity δ.

The experimental procedure, based on measuring the time required for gas to flow through a porous specimen into a calibrated volume (see, for example, (12)), made it possible to determine the permeability with an accuracy of ±1% over a wide range of mean pressures (from 25 mm Hg to 5-7 ata). Measurements of g were made at two temperatures of the porous medium: -78.5° (a mixture of solid CO_2 with acetone) and $\sim 20^\circ$, with the temperature maintained at the prescribed level to within $\pm 0.5^\circ$. Pure gases He, Ar, CF_2Cl_2 (Freon 12) were used in the experiments.

Special attention was paid to cleaning the internal surface of the porous specimens of sorbed vapors. For this purpose, a method of "washing" the specimen with the pure working gas was used (evacuation for 1-2 hours at a pressure of $\sim 10\,\mu$ Hg, followed by pumping through pure gas at $\Delta p \approx 1$ ata for 1-2 hours). Such an operation cleans the internal surface of the porous medium well and, according to some authors (13), may even replace the usual method of surface cleaning in preparing a specimen for sorption measurements.

As is known, the process of establishing flow in porous bodies that have low permeability and sufficiently large length h can be rather slow. A calculated and experimental estimate of the magnitude of the "time lag" (time-lag; see, for example, (12)) for our specimens showed that the flow regime in each of our measurements could be considered steady 10–20 sec after the onset of the flow process. The gas-flow regime in the specimens we investigated, even at the highest pressures $\bar{p}$ (maximum Δp), remained laminar in all cases. As is

Figure 1

Figure 1: Figure 1

known, the exact value of the critical Reynolds number for porous media has not been established. The value $\mathrm{Re}_{\mathrm{cr}}$, calculated for our specimens, proved to be 2 orders of magnitude smaller than the lowest value of this criterion determined for porous media (~ 0.1, according to (14)).

Table 1

No.	Specimen	h, cm	δ	d, μ*	S_{BET}, m^2/g**
1	Iron I	6.88	0.42	1.84	0.47
2	» III	4.00	0.28	0.77	0.47
3	» II	0.52	0.48	0.10	0.85
4	» III	1.4	0.87	0.31	16.3
5	» III	0.78	0.77	0.12	16.3
6	» III	0.51	0.65	0.072	16.3
7	» III	0.45	0.60	0.055	16.3
8	» III	0.35	0.49	0.030	16.3

* The value of the characteristic geometrical size (diameter) d, determined by generally accepted means from the ratio gV/g_0 (see, for example, (15, 16)). The constants in this calculation were chosen according to (1).

** S_{BET} is the total specific surface area, determined by low-temperature adsorption of nitrogen.

The investigation was carried out on many porous media of different nature (powders of metals, oxides, salts) and degree of dispersion. The measurement procedure we selected made it possible, in particular, to carry out an experiment on one and the same compact, varying its porosity stepwise by compression at ever higher pressures. Such a procedure makes it possible

cover a wide range of effective sizes d while maintaining a practically unchanged value of the internal surface, shape, and particle sizes. Table 1 gives some parameters characterizing the properties of several specimens for which typical results were obtained, shown in the coordinates g—$\bar{p}$ and g/g_0—Kn^{-1}* in Figs. 1 and 2.

Fig. 1. Dependence of the permeability of the specimen on the mean pressure at different temperatures.
Specimen No. 7. Argon. a—$t_1 = 20°$, b—$t_1 = -78.5°$

Analysis of the experimental results makes it possible to note the following principal experimental facts:

Figure 2

Figure 2: Figure 2

Fig. 3. Comparison of experimental and theoretical dependences of g/g_{Kn} on Kn^{-1}. 1—$G = G_V + G_M$; 2–experiment; 3–according to (5^6) at $\delta = 0.8$; 4–according to (6) at $\delta = 0.8$; 5–according to (6) at $\delta = 0.5$

Figure 3: Fig. 3. Comparison of experimental and theoretical dependences of g/g_{Kn} on Kn^{-1}. 1—$G = G_V + G_M$; 2–experiment; 3–according to (5^6) at $\delta = 0.8$; 4–according to (6) at $\delta = 0.8$; 5–according to (6) at $\delta = 0.5$

A. In the pressure region where $\lambda \geq d$, noticeable deviations of the function $g = f(\bar{p})$ from linearity are observed (“flattening” of the curve); moreover, the absolute level of the mean pressures at which the nonlinearity begins to appear depends on the nature of the gas and is observed approximately at one and the same d/λ.

Fig. 2. Dependence of g/g_0 on Kn^{-1} for various specimens and gases at different temperatures. Points without strokes: $t_1 = 20^\circ$; points with strokes: $t_1 = 78.5^\circ$

B. Changing the temperature of the experiment in measurements in the region $\lambda \geq d$ does not lead to a substantial ($\sim \eta$) change in the slope of the curve $g = f(\bar{p})$.

C. In the pressure region where $\lambda \leq d$, for all the gases used by us there is a strict linearity of the function $g = f(\bar{p})$; moreover, changing the temperature of the experiment leads to a substantial and regular

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[*] g_0 is the permeability value obtained by extrapolating the linear part of the dependence for $g = f(\bar{p})$ from the region of the highest pressures to $p = 0$. In other words, g_0 is determined by the specific “slip flow” corresponding to the value of the diffusion flux at high pressures [2]. g_{Kn} is the permeability value corresponding to $\mathrm{Kn}^{-1} \to 0$.

(to the viscosity η) change in the slope of the straight line $g = f(\bar{p})$ (since G_V for different gases and temperatures are recalculated, the dependence of g/g_0 on Kn^{-1} in this range of Knudsen numbers is universal).

On the basis of the first two observations it may be assumed that the change noted above in the character of the curve $g = f(\bar{p})$ in the region $\lambda \simeq d$ is apparently connected with the disappearance of viscous flow. The value g_{Kn}, determined from the experimental points in Fig. 2, proves to be greater–

Fig. 3. Comparison of experimental and theoretical dependences of g/g_{Kn} on Kn^{-1}. 1—$G = G_V + G_M$; 2–experiment; 3–according to (5^6) at $\delta = 0.8$; 4–

according to (6) at $\delta = 0.8$; 5—according to (6) at $\delta = 0.5$

than the corresponding specific "slip flow" g_0 by an average of 7%. Therefore it is evident that, in order to determine a reliable value of g_0, it is necessary to use extrapolation from the pressure range where $\mathrm{Kn} < 1$.

Thus, the experimental material at our disposal indicates that the widely accepted assumption of additivity of the flow ($G = G_V + G_D$, where $G_D = \mathrm{const}$, while G_V exists over the entire pressure range) is not valid for the types of porous media studied. On the other hand, the method of treatment proposed in works ([5],[6]) leads to an even greater discrepancy with the experimental data (Fig. 3).

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