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GAS FLOW TECHNIQUE FOR NON-DESTRUCTIVE POROUS MEDIA ANALYSIS

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Abstract

In this work the transport of rarefied gas in porous media caused by either pressure or temperature gradients is investigated. A gas in porous media becomes rarefied when either the scale is small, as for micro and nanoporous media, or when the pressure is low (vacuum conditions). The measurement methodologies for the respective gradients are developed, and the results are analyzed. For a pressure gradient driven gas flow. the permeability is an intrinsic property, a measure of how easily gas flow through the porous media. The gas flow behavior differs significantly depending on the degree of rarefaction. To characterize the rarefaction level of the gas flow inside a porous medium an additional intrinsic parameter sis proposed, the characteristic flow dimension. This parameter also has a physical interpretation, and its measure for a porous sample can be used to characterize the sample as a non-destructive analysis method. When the porous medium is subjected to a temperature gradient under rarefied conditions, the thermal transpiration effect, causes gas flows from the cold side toward the hot one. Both the transient and stationary properties of the thermal transpiration in porous media are analyzed. The developed methodologies are applied to analyze the microporous ceramic membranes.

KEYWORDS: Rarefied gas, porous media, microfluidics, Knudsen number, thermal transpiration, permeability.

The microporous membranes find a broad applications in medicine [1] and biotechnology for separation and filtration [2]. The recent development of low porous ceramic media with high thermal, chemical and structural stability and the ability to have catalytic properties has opened up new horizons for this kind of membrane applications, for example, in high-temperature gas separation and catalytic reactions [3]. Another type of porous media, the ultra-tight shale-gas reservoirs of tiny pores (in nanoscale) play a significant role in securing hydrocarbon energy because of their potential to offset declines in conventional gas production [4]. The determination of porous media permeability, a measure of how easily a fluid flows through a porous media, like the micro and nanoporous membranes or ultra-tight shale-gas reservoirs, is still a challenge up to now. The small scale of the microporous media causes the gas flow properties to change their characteristics even under atmospheric conditions and overpressure as such as in ultra-tight shale-gas reservoirs. The change of the gas flow properties is due to rarefaction effects. A gas becomes rarified either when the flow scale is small, as for micro and nanoporous media, or when the pressure is low (vacuum conditions). The rarefaction effect causes the permeability increase in several order of magnitude. Furthermore, under non-isothermal conditions, rarefaction effect causes gas to flow from the cold side to the hot side of the porous media. This is the working principle of a Knudsen pump, a gas pump without any moving parts.

This work investigates the changes of the gas flow properties due to rarefaction effects and contributes to the understanding of particularities of gas flows in microporous media. New permeability measurement method which takes into account the rarefaction effects is proposed. The analysis of experimental results allows us

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to extract a characteristic dimension of a porous sample which quantifies the effects of the rarefaction. This methodology opens up the prospect of a non-destructive technique of microporous media analysis. The study of the temperature gradient driven flow contributes to the understanding of the thermal effects at microscale. This study supports further microtechnology development such as the Knudsen pump. The developed general approach for the experimental analysis of isothermal and non-isothermal flow through the microporous media is applied to characterize the microporous ceramic membranes and sintered stainless steel porous media.

For the pressure gradient driven flow, a closed system is used consisting of two tanks with a known volume, V_1, V_2, only connected by the porous sample. The method applied here requires that the tank volume is relatively large compared to the volume occupied by a gas inside a microporous medium. In our experimental setup this ratio is larger than 10^3. The gas mass flow rate through a microporous medium is generated by setting an initial gas pressure drop between the reservoirs, see Fig. 1 in a closed system. For the temperature gradient driven flow experiments, we have again a closed system with two volumes, V_1 and V_2, maintained at temperatures T_1 and T_2, respectively, $T_1 > T_2$, and connected by a porous media, which is submitted to a temperature gradient, and by a bypass valve see Fig. 2. The absolute gas pressures, $p_1(t)$, and $p_2(t)$, are measured in each tank and the pressure difference $\Delta p(t)$ between the tanks. Initially, the tanks are connected with the bypass valve (kept

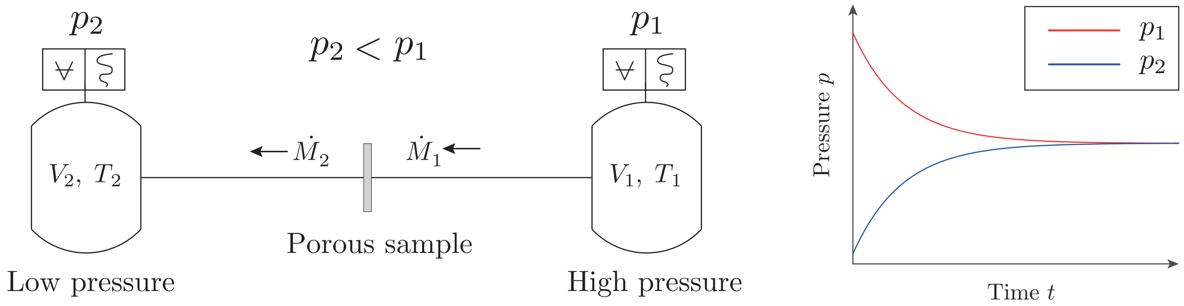

Figure 1: Principle of the pressure gradient driven flow experiment. A closed system with two tanks connected by a microporous medium, with an initial gas pressure difference causing gas to flow from high-pressure tank to low-pressure tank.

in open position) and the pressures on each side are equal, $p_1(t_0) = p_2(t_0)$, denoted in the following as p_{ini}. At this stage the temperature driven gas flow rate is established in the system, $\dot{M}_T$. By closing the bypass valve,

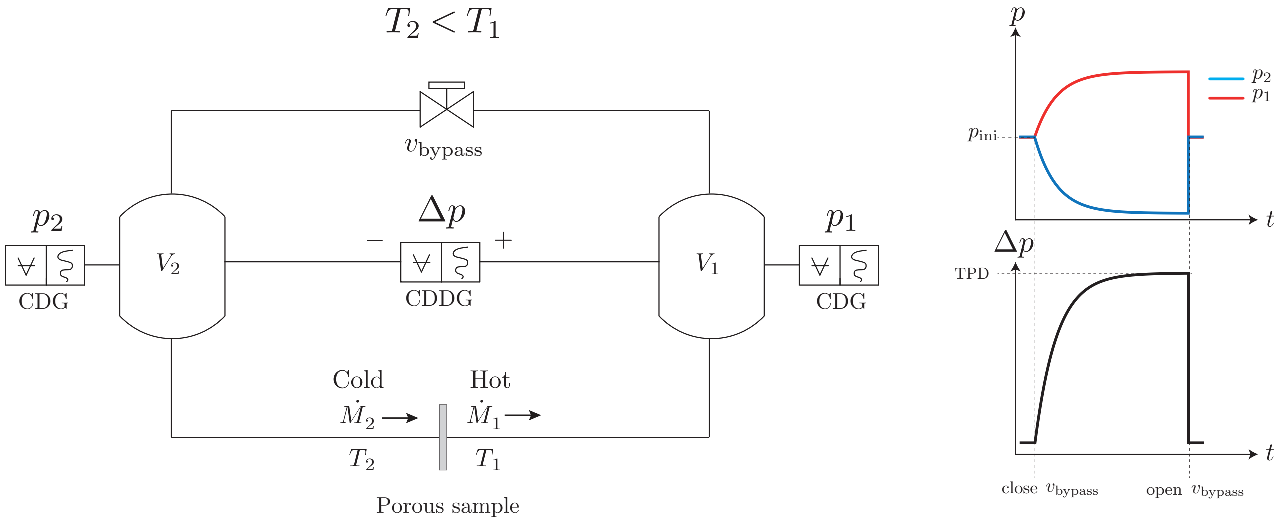

Figure 2: Sketch of the experimental setup.

the tanks are only connected by the porous media subjected to a temperature gradient. At this second stage, the pressure in the hot side increases and the pressure in the cold side decreases. The main driving force of the gas flow causing the build-up of the pressure difference at this stage is due to the temperature gradient driven flow $\dot{M}_T$. The gas flow toward the hot side generates the build-up of a pressure difference between the tanks. This generated pressure difference finally causes the return gas flow, $\dot{M}_p$, to the cold side. At the final stationary state, these two mass flow rates are in balance and constant in time. The established final pressure difference is called TPD.

Let us use the analytical expression for the mass flow rate through one tube and write it for a bundle of N capillaries, where the capillary length L_c can be different from the thickness L of the porous sample (disc). In this case, the mass flow rate through a bundle of N capillaries reads:

$$\dot{M} = \frac{N\pi a^4}{L_c}\frac{\Delta p p_m}{\mu v_0^2}\left(\frac{1}{4} + \frac{\sigma_p}{\delta}\right). \tag{1}$$

In the previous expression, four parameters are unknown: N, a, L_c and σ_p. To determine them from experimental data we can write the previous expression in the following form

$$S_0 = \dot{M}/M_{S0} = \frac{N\pi a^4}{L_c}\left(\frac{1}{4} + \frac{\sigma_p}{\delta}\right), \qquad M_{S0} = \frac{\Delta p p_m}{\mu v_0^2}. \tag{2}$$

Then we fit the previous expression according to the linear regression:

$$\mathcal{F}_S = \mathcal{A}_S X + \mathcal{B}_S, \qquad \mathcal{A}_S = \sigma_p \frac{\pi a^3 N}{L_c}, \qquad \mathcal{B}_S = \frac{\pi a^4 N}{4L_c}, \qquad X = \ell, \tag{3}$$

$\mathcal{A}_S$ and $\mathcal{B}_S$ are the fitting coefficients of the S-fit. The mass flow rate is fitted via the molecular mean free path, $X = \ell$. From the previous expressions, we find that that the slope of the fitting curve, $\mathcal{A}_S$ coefficient, depends on the gas nature only via the slip coefficient σ_p.

The S-type fit, Eq. (3), is realized in the hydrodynamic and slip flow regimes. From the fitting coefficients, $\mathcal{A}_S$ and $\mathcal{B}_S$, the effective flow dimension of the porous medium a, *i.e.* effective pore radius, can be found as

$$a = 4\sigma_p \frac{\mathcal{B}_S}{\mathcal{A}_S}. \tag{4}$$

To calculate the characteristic dimension of the porous medium, a, from the previous expression, we need only the information on the velocity slip coefficient, σ_p, which characterizes the gas-surface interaction. In addition, Eq. (4) is independent of the external geometrical parameters of a sample, so we are not restricted to only the cylindrical shape of the porous media. In the following, we assume that all the gases interact with the wall of the porous medium diffusively and the analytical value of this coefficient ($\sigma_p = 1.018$) is used for further calculations.

Applying the presented experimental methodology the effective pore dimensions are extracted from the mass flow rate measurements for two porous discs, see Table 1. By analyzing the data presented in Table 1, we can see that for the first porous sample the effective pore diameters, $2a$, calculated with different gases, are very close one to another. The uncertainty in the estimation of the characteristic pore dimension is of the order of 16% for Nitrogen and decreases up to 13.9% for Argon. The average pore dimension for the first disc, estimated with two gases, $2a = 3.6$ μm, is obtained with an uncertainty of 13.9%. For the second disc, which was taken from the same batch the characteristic dimension was found much larger, $2a = 22.$ μm. This results allows us to assume that the proposed technique could be used as a non-intrusive analysis of the porous simples.

1st disc

GAS	$2\,a$ [μm]	N [10^5]	$\mathcal{S}_\text{A}$ [10^5 m^2/m^3]	l_τ
N_2	3.7 ± 0.6	3.4 ± 1.0	1.5 ± 0.6	2.6 ± 0.4
Ar	3.6 ± 0.5	3.8 ± 1.0	1.5 ± 0.6	2.5 ± 0.4
AVR	3.6 ± 0.5	3.8 ± 0.9	1.5 ± 0.6	2.5 ± 0.4

2nd disc

GAS	$2\,a$ [μm]	N	$\mathcal{S}_\text{A}$ [10^4 m^2/m^3]	l_τ
He	25 ± 3	4300 ± 900	2.2 ± 0.7	4.7 ± 0.6
N_2	20 ± 2	8000 ± 1000	2.7 ± 0.6	3.8 ± 0.4
Ar	25 ± 4	4000 ± 1000	2.0 ± 1	4.7 ± 0.9
AVR	22 ± 3	6000 ± 1000	2.4 ± 0.9	4.3 ± 0.6

Table 1: Estimation of the porous media characteristic dimension, a, the number of capillaries N, and the surface to volume ratio, $\mathcal{S}_\text{A}$, by using S-type fit and the porosity obtained from the micro-computed tomography, $\varepsilon = 13.6\%$.

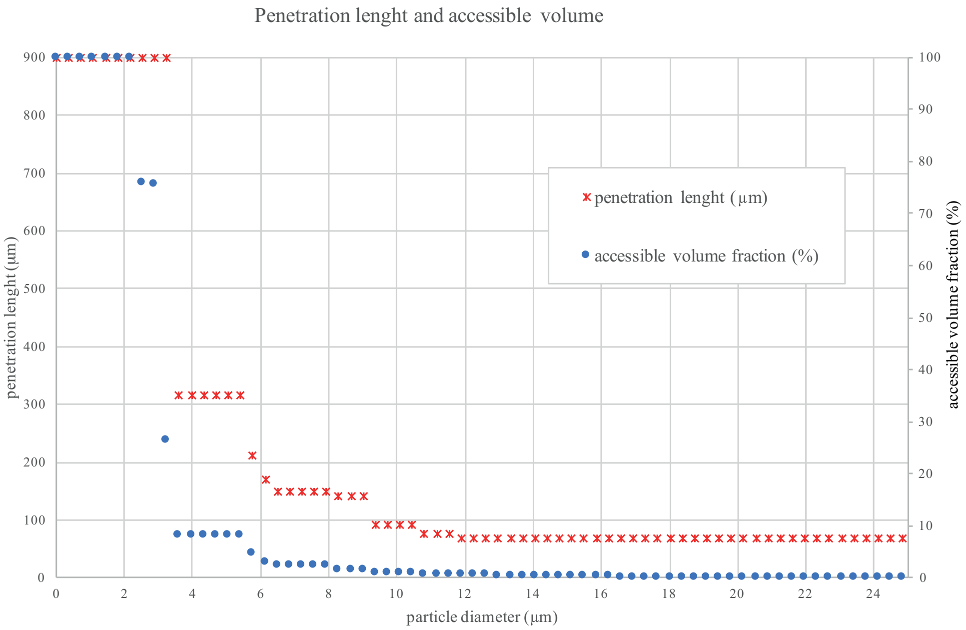

Figure 3: Microtomographic analysis: penetration length as a function of a particle diameter.

The uncertainty of the effective pore dimension is calculated using the square root of the summation of the fitting coefficients uncertainty, which is calculated from the limits of a 95% confidence interval of the fitting parameters.

The iMorph computer analysis of the tomographic data can represent the porous structure of a sample as a system of the pores connected by the constrictions (throats). From the result of the tomographic analysis of the first sample, it was also found that only the particles with diameter of around 3.5 μm are able to cross the sample, see Fig. 3. From this analysis, we can conclude that the proposed gas flow methodology allows us to estimate the effective pore size which determines the flow through a porous medium.

The classical model of the porous media presentation as a bundle of capillaries was revised. The original methodology was suggested to determine the characteristic flow dimension. The experimental procedure is developed to determine the effective pore size (characteristic flow dimension) and the number of capillaries, related to the model a bundle of capillaries. The experimentally obtained effective pore dimension is in very good agreement with the results of the mercury porosimetry and micro-computed tomography. The use of additional information on the sample porosity allows finding the tortuosity and the surface-to-volume ratio, which were close to that calculated from the tomography analysis. The characteristic flow dimension, a newly defined intrinsic property of the porous media, can be used to characterize the gas rarefaction effects.

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