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## Mass flow rate and permeability measurements in microporous media

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## 11 1. Introduction

12 The determination of the permeability of porous media like the micro and  
13 nanoporous membranes or ultra-tight shale-gas reservoirs is still a challenge up  
14 to now. The low porous membranes find a broad application in medicine [1] and  
15 biotechnology for separation and filtration [2]. The recent development of low  
16 porous ceramic media with high thermal, chemical and structural stability and  
17 the ability to have catalytic properties has opened up new horizons for this kind  
18 of membrane applications, for example, in high-temperature gas separation and  
19 catalytic reactions [3]. Another type of porous media, the ultra-tight shale-gas  
20 reservoirs of tiny pores (in nanoscale) play a significant role in securing hydro-  
21 carbon energy because of their potential to offset declines in conventional gas  
22 production [4]. In all of these type of applications, the porous media permeabil-  
23 ity has to be known.

24 Permeability is a measure of how readily a fluid can flow through a porous  
25 material [5]. Gas permeability is an important parameter to understand the  
26 transport characteristics of a fluid through a porous medium, which can be  
27 obtained from the mass or volume flow rate. For the determination of low per-  
28 meability, either the steady-state or the transient methods can be used. The  
29 steady-state method needs a precise flow meter to measure very slow flow, so  
30 when the permeability is very low the conventional gas flowmeters may be in-  
31 appropriate [6]. Therefore, the transient "pulse-decay" or "draw-down" tech-  
32 niques, [7], [8], [9], [1], [6], are also used to determine the low permeabilities.  
33 By using these techniques, the permeability can be calculated directly from  
34 the pressure variations in time, without going first through the mass flow rate  
35 measurements [8], [1].

36 The primary objective of the present work is to develop the transient method  
37 to measure the pressure evolution in time in high and low-pressure tanks gen-  
38 erated by the gas flow through a homogeneous porous medium. This experi-  
39 mental methodology, based on the constant volume technique, was initially de-  
40 veloped for the isothermal and non-isothermal measurements of the mass flow  
41 rate through the microchannels [1], [1], [1]. We provide here the physical justi-  
42 fications of the exponential fitting of the pressure variations with time as well  
43 as the physical conditions of its implementation. From the measured pressure  
44 variations in time, the mass flow rate through the porous medium is deduced.  
45 It is shown that the gas permeability can be easily obtained directly from the  
46 pressure variations with time without going first through the mass flow rate cal-  
47 culations. The main advantages of the proposed approach are: its simplicity, the  
48 possibility of further extraction of the Klinkenberg coefficient and average pore  
49 size, and, finally, its further generalization for the case of temperature gradient  
50 driven flows.

51 The paper is organized as follows. After a brief introduction, the experimen-  
52 tal apparatus and methodology are presented in Section 2. Then, in Section 3,  
53 the relation between the pressure variations in the tanks and a porous medium  
54 permeability is established by introducing the pressure relaxation time, which  
55 properties are analyzed in detail in Section 4. The behaviors of the measured

56 mass flow rate and permeability are discussed in next two Sections. The paper  
57 is closed with final comments and conclusions.

## 58 **2. Experimental methodology**

### 59 *2.1. Experimental apparatus*

60 The experimental setup is a high vacuum system capable of measuring up to  
61 5 decades of pressure. In the presented experiment the mean pressure is varied  
62 from 75 Pa up to 131 kPa. This large pressure measurement range is achieved  
63 by using three pairings of four capacitance diaphragm manometers (CDM) with  
64 full-scales: 133 kPa - 133 kPa, 133 kPa -13.3 kPa and 13.3 kPa - 1.33 kPa. Four  
65 high purity gas bottles with test gases, Helium, Neon, Nitrogen, Argon (Air Liq-  
66 uide, France) are used. The pumping is performed by a two-stage diaphragm  
67 vacuum pump (DVP) and a turbomolecular pump (TMP), see the schematic  
68 of the experimental setup in Fig. 1. Each side of the porous medium is con-  
69 nected to two reservoirs of volumes  $V_1$  and  $V_2$  for the high and low-pressure,  
70 respectively. Both tanks' volumes, including the volumes of the valves, con-  
71 necting tubes and pressure sensors, are measured accurately, and these volumes  
72 are equal to  $V_1 = 255.8 \pm 5.5 \text{ cm}^3$  and  $V_2 = 238.8 \pm 5.1 \text{ cm}^3$ , for the high and  
73 low-pressure tanks, respectively. Several leakage tests were performed for this  
74 setup showing the absence of the detectable increase in pressure, measured with  
75 the lowest F.S. pressure sensor of 1.33 kPa over a period of 30 minutes. In  
76 addition, for all realized measurements we did not detected any linear increase  
77 of the mean pressure.

78 Two microporous samples, used in the experiments and mentioned in the  
79 following as the first and second discs, have a cylindrical shape (disc) with the  
80 same diameter and thickness (in main flow direction) equal to  $D = 9.5 \pm 0.01 \text{ mm}$   
81 and  $L = 2.3 \pm 0.01 \text{ mm}$ , respectively. The structure of these microporous discs  
82 is the same as that used to support the active layers of micro-to-ultra filtration  
83 ceramic membranes.

84 For such ceramic microporous medium, depending on manufacturer, the  
85 porosity is in the range 15 – 30% with pore diameter ranging from 1 to 10  
86  $\mu\text{m}$ . The total volume of each porous disc is  $0.14 \text{ cm}^3$ , so by taking 30% of  
87 porosity a gas volume inside the medium is approximately  $0.047 \text{ cm}^3$ , which is  
88 much smaller than the volume of each tank.

89 The experiments are performed within a narrow temperature range, exclud-  
90 ing any heat source in an environment. The temperature is measured using the  
91 thermocouple with the accuracy of 0.6 K.

### 92 *2.2. Mass flow rate measurements*

93 The constant volume technique, used previously for the measurements of  
94 the mass flow rate through the microchannels [1], [1], [1], [1], was implemented  
95 here to measure the mass flow rate through different samples of a microporous  
96 ceramic medium. This technique allows deducing the mass flow rate from the  
97 pressure variation in time. The mass flow rate through a microporous medium is

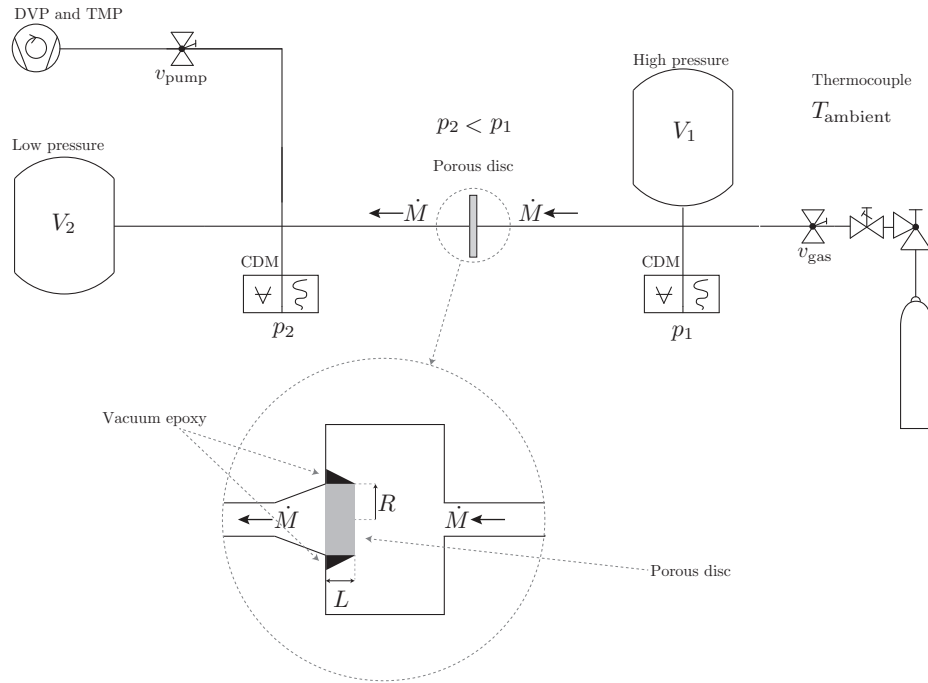


Figure 1: Schematic of the experimental setup. A diaphragm vacuum pump (DVP) and a turbomolecular pump (TMP) is connected to the experimental setup separated by a valve  $v_{\text{pump}}$ . The high pressure and low-pressure side are separated by a porous ceramic media which is fixated with vacuum epoxy glue. Each side of the porous media has a reservoir with the high-pressure tank volume of  $V_1 = 255.8 \pm 5.5 \text{ cm}^3$ , low-pressure tank volume  $V_2 = 238.8 \pm 5.1 \text{ cm}^3$  and capacitance diaphragm manometers (CDM) measuring pressure  $p_1$  and  $p_2$ . The room temperature is measured with a thermocouple. The vacuum system is connected to a gas delivery system with four gas bottles, Helium, Neon, Nitrogen, and Argon.

98 generated by setting an initial pressure drop between the reservoirs, see Fig. 1.  
 99 This method requires very large tank volumes relative to the volume occupied  
 100 by a gas inside a microporous medium: in our experimental setup this ratio is  
 101 larger than  $10^3$ . The applied method is similar to the Brace method [1] (pulse  
 102 decay method), usually used to analyze the permeability of the porous samples.  
 103 In addition, in this work we take into consideration the effects of rarefaction.  
 104 Even if the permeability can be deduced directly from the pressure (or pressure  
 105 difference) variation in time, we prefer to start by providing the expressions of  
 106 the mass flow rate through a microporous medium and the conditions of its  
 107 derivation. The mass flow rate could be a useful quantity to characterize a  
 108 porous sample; it can be used to derive the characteristic pore dimensions and  
 109 the gas-surface interaction characteristics.

110 Under the quasi-steady conditions, *i.e.* when the flow through a porous  
 111 medium is established, we assume that the gas temperatures in each tank, de-  
 112 noted  $T_1$  and  $T_2$ , are in thermal equilibrium with the walls of the tanks and  
 113 that both tanks are in thermal equilibrium with the environment. Therefore,  
 114 we assume that the ambient temperature, denoted  $T_{\text{ambient}}$ , determines the gas  
 115 temperatures  $T_1$  and  $T_2$  in each tank such that:

$$T_1 \approx T_2 \approx T_{\text{ambient}} = T. \quad (1)$$

116 A possible variation in temperature of the gas leaving and entering the tanks  
 117 could directly perturb the significance of the measurement. To make this clearer,  
 118 let us write the ideal gas law in each tank in the following form:

$$p_1 V_1 = M_1 \mathcal{R} T, \quad p_2 V_2 = M_2 \mathcal{R} T, \quad (2)$$

119 where  $\mathcal{R}$ ,  $p_i$ , and  $M_i$ ,  $i = 1, 2$ , are, respectively, the specific gas constant, the  
 120 pressure and the mass of the gas in tank  $i$ . In the present study the maximum  
 121 considered pressure is of the order of atmospheric pressure; therefore we do  
 122 not consider here the real gas effects. However, the proposed approach can be  
 123 generalized to take into account the real gas effects by using, for example, the  
 124 van der Waals equation instead of the ideal gas law.

125 Under our experimental conditions, the volume of each reservoir is constant  
 126 during an experiment, so it is possible to differentiate each expression in Eqs.  
 127 (2), as it has been done in Ref [1]:

$$dM_i = \frac{V_i}{\mathcal{R} T} dp_i \left( 1 - \frac{dT/T}{dp_i/p_i} \right), \quad i = 1, 2. \quad (3)$$

128 If the relative temperature variation in a tank is negligible in relation to the  
 129 relative pressure variation in time, then the mass flow can be considered to be  
 130 isothermal. Therefore, by defining a specific small time interval,  $dt$ , it is then  
 131 possible to obtain from Eqs. (3) the isothermal mass flow rates  $dM_1/dt$  and  
 132  $dM_2/dt$  as:

$$\frac{dM_i}{dt} = \frac{V_i}{\mathcal{R} T} \frac{dp_i}{dt}, \quad \text{if} \quad \epsilon_i = \frac{dT/T}{dp_i/p_i} \ll 1, \quad i = 1, 2. \quad (4)$$

133 Following the differentiation technique, we consider here the variation of any  
 134 thermodynamic parameter,  $dM$  and  $dp$ , sufficiently small to approximate  $dM/dt$   
 135 and  $dp/dt$  as the time derivative of the mass (*i.e.* mass flow rate  $\dot{M}$ ) and the time  
 136 derivative of the pressure, respectively. If the relative temperature variation is  
 137 small compared to the pressure variation, the values of  $\epsilon_i$  are small in Eqs. (4),  
 138 so by adjusting the sign,  $dM_i/dt$  can be considered as the mass flow rate  $\dot{M}_i$   
 139 through the microporous media

$$\dot{M}_1 = -\frac{dM_1}{dt} = -\frac{V_1}{\mathcal{R}T} \frac{dp_1}{dt}, \quad \dot{M}_2 = \frac{dM_2}{dt} = \frac{V_2}{\mathcal{R}T} \frac{dp_2}{dt}. \quad (5)$$

140 In the frame of the quasi-stationary flow assumption, *i.e.* when the flow through  
 141 a porous medium is established, it is clear that the mass flow rate leaving the  
 142 first tank is necessarily equal to the mass flow rate entering the second tank and  
 143 also to the mass flow rate at any point inside the porous medium

$$\dot{M}_1 = \dot{M}_2 = \dot{M}. \quad (6)$$

144 Along with this study, we will continue to admit relation (6) at any time. How-  
 145 ever, the assumption of the mass conservation, Eq. (6), neglects possible stor-  
 146 age of gas in the porous medium. For the small ceramic porous sample used  
 147 in present experiment, this hypothesis of the absence of the gas accumulation  
 148 inside a porous medium is justified. In the same time, for the porous media such  
 149 as the coal and shales, the compressible fluid storage needs to be accounted [5].  
 150 The experimental verification of mass conservation for the porous samples used  
 151 in present experiments is discussed in Section 5.2.

152 Sometimes it is convenient to express the mass flow rate in function of the  
 153 pressure difference between two tanks. From the mass conservation law, the  
 154 mass of a gas leaving the first tank is necessarily equal to the mass of a gas  
 155 entering the second tank, so Eq. (6) is valid. From Eqs. (5) and (6) we can  
 156 obviously deduce:

$$\dot{M}(t) = -\frac{V_0}{\mathcal{R}T} \frac{d(\Delta p(t))}{dt}, \quad V_0 = \frac{V_1 V_2}{V_1 + V_2}. \quad (7)$$

157 From the previous reasoning, it is clear that the mass flow rate can be  
 158 calculated using expressions (5) and (7), when the pressure variation in each  
 159 tank or the pressure difference between them in time is known. During the  
 160 experiments, the pressure variations in time in each tank are measured and  
 161 then fitted by using the exponential fitting function.

### 162 2.3. Exponential pressure fitting

163 The experimental procedure starts by setting the initial pressure difference  
 164  $\Delta p_0$  between the tanks at time  $t_0$  as:

$$\Delta p_0 = p_1(t_0) - p_2(t_0) = p_{01} - p_{02}, \quad (8)$$

165 where  $p_{01}$  and  $p_{02}$  are the initial pressures in the high and low-pressure tanks,  
 166 respectively. This first step is done by opening the valve  $v_{\text{pump}}$ , see Fig. 1,

167 for a short time, and then closing it. Further, we have a closed system with a  
 168 pressure difference. Afterward, the gas begins to flow through a microporous  
 169 medium from the high-pressure tank to the low-pressure tank up to the same  
 170 final equilibrium pressure,  $p_f$ , is reached in both tanks, see Fig. 2. The time-  
 171 dependent pressure difference between the tanks is noted as:

$$\Delta p(t) = p_1(t) - p_2(t). \quad (9)$$

172 One example of the pressure evolution in each tank as well as of the pressure  
 173 difference between two tanks is shown in Fig. 3.

174 Usually, when using the "pressure pulse technique", [1], the small pressure  
 175 "pulses" (pressure differences between the tanks) are applied to the system, *i.e.*  
 176  $\Delta p_0 \ll p_m$ , where  $p_m = 0.5(p_1 + p_2)$  is the mean pressure between two tanks. We  
 177 will discuss below that finally we do not really need to respect this restrictive  
 178 condition and the [an](#) arbitrary pressure difference between the tanks can be  
 179 used in the system if both tanks volumes are equal. However, the restriction for  
 180  $p_1/p_2$  ratio exists when the tanks volumes are different, see Section 3.1.

181 The authors of Ref. [1] proposed to use a linear fit of the natural logarithm  
 182 of an exponential function for the pressure variation in time, while we suggest  
 183 to use directly the exponential pressure fit to describe the pressure difference  
 184 decay in time in the following form:

$$\Delta p(t) = \Delta p_0 \exp(-(t - t_0)/\tau), \quad (10)$$

185 where  $\tau$  is the pressure relaxation time, which is constant during an experiment.

186 The same exponential representation of the pressure evolution in time in the  
 187 first  $p_1(t)$  and second  $p_2(t)$  tanks is written in the form [1]:

$$\begin{aligned} p_1(t) &= p_f + (p_{01} - p_f) \exp(-(t - t_0)/\tau_1), \\ p_2(t) &= p_f + (p_{02} - p_f) \exp(-(t - t_0)/\tau_2), \end{aligned} \quad (11)$$

188 here  $\tau_1, \tau_2$  are the gas pressure relaxation times in the reservoir 1 and 2, respec-  
 189 tively,  $p_f$  is the final pressure. The pressure variations with time  $p_i(t)$  in each  
 190 tank 1 and 2 can be thus associated with an exponential decay. In practice the  
 191 pressure relaxation times  $\tau_i$  are obtained from the fit of the measured pressure  
 192 evolution in each tank, see Fig. 3. The properties of the pressure relaxation  
 193 time are discussed in Section 4.

194 By using Eq. (7) we can now express the mass flow rate using the exponential  
 195 representation of the pressure difference in time, Eq. (10), and then its analytical  
 196 derivative, so the mass flow rate expression becomes

$$\dot{M}(t) = -\frac{V_0}{\mathcal{R}T} \frac{d(\Delta p(t))}{dt} = \frac{V_0}{\mathcal{R}T} \frac{\Delta p_0}{\tau} \exp\left(-\frac{t - t_0}{\tau}\right). \quad (12)$$

197 From Eqs. (5) we can also express the mass flow rate using the exponential  
 198 representations of the pressure variation in time in each tank, Eqs. (11), and



199 then their analytical derivatives, so the mass flow rate expressions become:

$$\begin{aligned} \dot{M}(t) = \dot{M}_1(t) &= -\frac{V_1}{\mathcal{R}T} \frac{d(p_1(t))}{dt} = \frac{V_1}{\mathcal{R}T} \frac{p_{01} - p_f}{\tau_1} \exp\left(-\frac{(t - t_0)}{\tau_1}\right), \\ \dot{M}(t) = \dot{M}_2(t) &= \frac{V_2}{\mathcal{R}T} \frac{d(p_2(t))}{dt} = \frac{V_2}{\mathcal{R}T} \frac{p_f - p_{02}}{\tau_2} \exp\left(-\frac{(t - t_0)}{\tau_2}\right). \end{aligned} \quad (13)$$

200 Equations (12) and (13) provide the time-dependent expressions for the mass  
 201 flow rate. Therefore, with one experiment we can calculate the mass flow rate  
 202 for various different pressure ratios between the reservoirs, see Section 4 for  
 203 more details. The conditions related to the implementation of this technique,  
 204 as the choice of the tank volume size, possible pressure ratio and the impact of  
 205 the thermal effects are discussed in Section 3.1.

### 206 3. Relation with Darcy law and the permeability

207 The Darcy law [1] allows us to relate the instantaneous discharge (or vol-  
 208 umetric) flow rate through a porous medium,  $Q$ , to the pressure drop over a  
 209 given distance  $L$ , which is the thickness of a porous sample:

$$Q = \frac{KS}{\mu} \frac{(p_1 - p_2)}{L}, \quad (14)$$

210 where  $S$  is the surface of the porous sample,  $\mu$  is the fluid viscosity,  $K$  is the  
 211 permeability. Initially, the Darcy law was derived for an incompressible fluid  
 212 with constant viscosity and the permeability  $K$  refers to the hydrodynamic  
 213 (intrinsic permeability). In this article we use the Darcy law for the gases, *i.e.*,  
 214 a compressible fluid and we do not make any preliminary assumption of the  
 215 rarefaction level of a gas.

216 The viscosity coefficient for a gas depends on gas temperature and gas nature  
 217 and it is calculated as [1]:

$$\mu = \mu_{\text{ref}} \left( \frac{T}{T_{\text{ref}}} \right)^\omega, \quad (15)$$

218 where  $\omega$  is the gas viscosity index,  $\mu_{\text{ref}}$  is the gas viscosity at temperature  
 219  $T_{\text{ref}} = 273.15$  K [1]. The reference values of the viscosity,  $\mu_{\text{ref}}$ , for each gas used  
 here as well as the viscosity index  $\omega$ , are given in Table 1.

Table 1: Parameters of the gases used in present experiments

Gas	$\mu_{\text{ref}} \times 10^{-5}$ [Pa.s]	$\omega$	$\mathcal{R}$ [J/kgK]	Molar mass $\mathcal{M}$ [g.mol <sup>-1</sup> ]
He	1.865	0.66	2077.1	4.003
Ne	2.976	0.66	412.02	20.18
N <sub>2</sub>	1.656	0.74	296.80	28.00
Ar	2.117	0.81	208.13	39.95

220 For the liquid flows the volumetric flow rate is constant along a porous  
 221 sample. For the gases only the mass flow rate is conserved along the porous  
 222

223 sample. To express the permeability for a gas we can rewrite Eq. (14) in  
 224 following form by replacing the pressure difference through a sample by the  
 225 pressure gradient [5]:

$$Q = -\frac{KS}{\mu} \frac{dp}{dx}. \quad (16)$$

226 By using the relation between the volumetric and mass flow rate

$$Q = \frac{\dot{M}}{\rho} = \dot{M} \frac{\mathcal{R}T}{p}, \quad (17)$$

227 then by integrating along the porous sample and by using the mass conservation  
 228 property, we obtain the expression, analogous to Eq. (14), which relates the  
 229 mass flow rate and the permeability

$$\dot{M} = \frac{KS}{\mu} \frac{\Delta p}{L} \frac{p_m}{\mathcal{R}T}. \quad (18)$$

230 Then, using Eqs. (5) and admitting the mass conservation again, Eq. (6), and  
 231 using Darcy law in form (18), we can relate the pressure variation in each tank  
 232 to the gas permeability:

$$V_1 \frac{dp_1}{dt} = -p_m \frac{K}{\mu} \frac{S}{L} (p_1 - p_2), \quad V_2 \frac{dp_2}{dt} = p_m \frac{K}{\mu} \frac{S}{L} (p_1 - p_2). \quad (19)$$

233 To obtain previous relations we replaced the local pressure in Eq. (17) by the  
 234 mean pressure, which does not vary during an experiment. This mean pressure  
 235 is constant during an experimental run, when the tanks volumes are equal,  
 236  $V_1 = V_2$ , and it varies only slightly when these volumes are slightly different, see  
 237 Section 3.1, where the conditions of the mean pressure constancy are provided.

238 From Eqs. (19) we obtain the differential equation for the pressure difference  
 239  $\Delta p(t)$  between the tanks:

$$\frac{d(\Delta p(t))}{\Delta p(t)} = -\frac{dt}{\tau}, \quad (20)$$

240 where

$$\tau = \frac{1}{p_m} \frac{\mu}{K} \frac{LV_0}{S}. \quad (21)$$

241 This differential equation, subjected by the initial condition,  $\Delta p(t = t_0) = \Delta p_0$ ,  
 242 is easily solved and the variation in time of the pressure difference between the  
 243 tanks is obtained in the form of Eq. (10).

244 From previous discussion it is clear that we do not use here any assumption  
 245 about the smallness of the pressure "pulses" compared to the mean pressure in  
 246 the tanks. Therefore, this technique can be implemented for any pressure dif-  
 247 ference between the tanks under the condition of equality of the tanks volumes.  
 248 When the tanks volumes are different some restrictions have to be respected in  
 249 order to keep the mean pressure close to a constant value during an experiment  
 250 duration, see Section 3.1. In addition, to integrate the differential equation  
 251 (20) the relaxation time  $\tau$  has to be time independent. By analyzing Eq. (21)

252 one can see that only mean pressure can be time-dependent. The experimental  
 253 conditions of the mean pressure constancy in time are discussed in Section 3.1.  
 254 The permeability depends on time only through the mean pressure, so the mean  
 255 pressure constancy in time ensures that  $\tau$  is constant in time and so justifies  
 256 the integration of Eq. (20) and consequently the use of its solution, Eq. (10),  
 257 for the experimental data (pressure) treatment. Therefore, the permeability  $K$   
 258 of a porous sample can be derived directly from the pressure measurements.

259 The experimental curve of pressure variation in time can be fitted by expo-  
 260 nential expression, Eq. (10), with  $\tau$  as a fitting parameter. Then, the perme-  
 261 ability can be found from the analytical expression for the pressure relaxation  
 262 time  $\tau$ , Eq. (21), as:

$$K = \frac{1}{p_m} \frac{\mu LV_0}{\tau S}. \quad (22)$$

263 Other expression for the permeability can be obtained from (18):

$$K = \frac{\dot{M}}{p_1^2 - p_2^2} \frac{2\mu RTL}{S} = \frac{\dot{M}}{\Delta p p_m} \frac{\mu RTL}{S}. \quad (23)$$

264 Both expressions, Eqs. (22) and (23), can be used to derive the permeability  
 265 from the measurements.

### 266 3.1. Conditions of constancy of mean pressure

267 In this section we establish the conditions of the constancy of mean pressure  
 268 during the measurement procedure, which leads to the pressure relaxation time  
 269 constancy and justifies the use of expression (22) for the permeability calcula-  
 270 tions. In addition, as it has been pointed out in Ref. [8], in general case, the  
 271 constancy of the mean pressure is an important point, especially for high pres-  
 272 sure experiments, where the viscosity and compressibility factor may change as  
 273 a function of pressure. In presented here experiments the implemented pres-  
 274 sure and temperature conditions allow to us to stay under the ideal gas flow  
 275 assumptions and the viscosity does not change with pressure. However, it is  
 276 still important to have a constant mean pressure as for low permeable porous  
 277 media the permeability can be a function of mean pressure due to rarefaction  
 278 effects.

279 A pressure difference between the tanks is fixed initially, at  $t_0$ , equal to  
 280  $\Delta p_0$ , Eq. (8), then the gas flows through the porous medium up to the final  
 281 stage, when a pressure equality in both tanks is reached, see Fig. 2. A relation  
 282 between the pressure variation in each tank, *i.e.* from the initial pressure in  
 283 each tank,  $p_i(t_0)$ ,  $i = 1, 2$ , to the final equilibrium pressure  $p_f$ , reached in the  
 284 system, can be calculated *a priori* as it is closely related to the tanks volumes  
 285 ratio. From the ideal gas law and admitting again the mass conservation along  
 286 the microporous medium at any time, we can write the following relation for  
 287 two tanks if they are maintained at the same temperature

$$dp_1 V_1 = -dp_2 V_2. \quad (24)$$

288 The previous relation is then integrated in time from an initial (at time  $t_0$ ) state  
 289 of a gas in each tank,  $p_i(t_0)$ , to its final state,  $p_f$ . It is worth to underline that in  
 290 Eq. (24) the expressions are exact (perfect) differential, and so their integration  
 291 does not depend on the form (linear or exponential) of the pressure variation in  
 292 time. Therefore, we obtain

$$\frac{p_f - p_2(t_0)}{p_1(t_0) - p_f} = \frac{V_1}{V_2}. \quad (25)$$

293 Without the loss of generality we can assume here that in the beginning of each  
 294 experiment we have  $p_1 > p_2$ . It is clear from Eq. (25) that by adjusting the  
 295 tanks volumes ratio we could control the pressure variation between initial and  
 296 final stages. From Eq. (25) we obtain the estimation of the maximal variation  
 297 of the mean pressure  $p_m$  with time, from its initial value  $p_m(t_0)$  to its final value  
 298  $p_m(t_f) = p_f$ :

$$\frac{p_f}{p_m(t_0)} = \frac{p_1(t_0)V_1 + p_2(t_0)V_2}{(V_1 + V_2)p_m(t_0)}. \quad (26)$$

299 It is clear from Eq. (26) that when the tanks volumes are equal, the mean  
 300 pressure  $p_m(t)$  does not vary in time, i.e., between its initial value,  $p_m(t_0)$ , and  
 301 its final value,  $p_f$ , so  $p_m(t_0) = p_f$ . When the volumes are different we can  
 302 estimate the maximal amplitude of mean pressure variation between its initial  
 303 state  $p_m(t_0)$  and its final state  $p_f$ , using Eq. (26) and the ratio of tanks volumes.  
 304 To do this expression (26) can be rewritten as:

$$\frac{p_f}{p_m(t_0)} = \frac{2(1 + k_V k_p)}{(1 + k_V)(1 + k_p)}. \quad (27)$$

305 We introduced here the tanks volumes ratio  $k_V = V_1/V_2$  and the initial pressure  
 306 ratio  $k_p = p_1(t_0)/p_2(t_0)$ . By using Eq. (27), we can calculate the variation of  
 307 the mean pressure during an experiment, *i.e.* the ratio between its final and  
 308 initial values  $p_f/p_m(t_0)$ .

309 In our experimental conditions two tanks volumes are related as  $k_V =$   
 310  $V_1/V_2 = 1.071$ , so from Eq. (27) we can find that for the initial pressure ratio  
 311 between the tanks,  $k_p$ , equal to 1.5, 2 and 3, the ratio  $p_f/p_m(t_0)$  is equal 1.0069,  
 312 1.0114 and 1.0171, respectively. Therefore, the initial pressure ratio equal to  
 313 2 leads to approximately 1% of deviation of the mean pressure from its initial  
 314 value. Under our experimental conditions the experimentally evaluated value of  
 315  $p_f/p_m(t_0)$  was found lower than 1%.

#### 316 4. Pressure relaxation time

317 By fitting the measured time variation of the pressure difference between  
 318 two tanks using exponential law, Eq. (10), we obtain a function  $\Delta p(t)$  that  
 319 describes the relaxation process with the help of a single fitting parameter  $\tau$ ,  
 320 that is the characteristic time of the experiment or pressure relaxation time.  
 321 Similar expressions for the pressure variation in each tank, Eqs. (11), involve

322 the relaxation parameters  $\tau_1$  and  $\tau_2$ , which can also be obtained by the fitting  
 323 of the pressure variation in one (high or low-pressure) tank. From different  
 324 expressions of the mass flow rates, Eqs. (12) and (13), and using the mass  
 325 conservation property in form (24), we can find that the ratios between the  
 326 characteristic times are finally independent from the tanks volumes and are  
 327 equal to one:

$$\frac{\tau_1}{\tau_2} = \frac{\tau_1}{\tau} = \frac{\tau_2}{\tau} = 1. \quad (28)$$

328 This analytical finding, Eq. (28), was confirmed experimentally. As it can be  
 329 seen from Table 2 for the most cases the difference between three relaxation  
 330 times,  $\tau_1$ ,  $\tau_2$  and  $\tau$ , is small, of the order of 1%.

331 Finally, to obtain the mass flow rate through the microporous medium and  
 332 its permeability, see Section 6, we can use either the exponential fit of the  
 333 pressure difference between the tanks (in the case of the use of the differential  
 pressure sensors), or just pressure evolution in a tank.

Table 2: Relaxation times  $\tau_1$ ,  $\tau_2$  and  $\tau$ , in seconds (s) for the second disc, measured in the high and low-pressure tanks and by using the pressure difference between two tanks, respectively. For each of four gases the relaxation time is provided for two pressure differences between the tanks. The mean pressure of each experiment is given in fifth column. The last column provides the conduction time  $\tau_c$ , Eq.(29).

	$\tau_1$ [s]	$\tau_2$ [s]	$\tau$ [s]	$p_m$ [ $10^5$ Pa]	$\tau_c$ [s]
HELIUM	54.11	53.94	54.03	1.13	0.96
	66.94	66.87	66.90	0.79	0.68
	153.83	155.60	154.72	0.04	0.03
NEON	92.38	92.48	92.43	1.16	3.11
	119.98	118.59	119.28	0.79	2.13
	333.25	331.46	332.34	0.03	0.08
NITROGEN	62.30	61.75	62.03	1.12	7.45
	92.98	92.04	92.50	0.70	4.64
	371.56	369.32	370.44	0.03	0.23
ARGON	79.27	78.61	78.94	1.11	8.11
	112.34	111.33	111.83	0.74	5.41
	444.07	444.22	444.15	0.04	0.26

334

#### 335 4.1. Gas conduction time

336 Now we can compare the gas conduction time  $\tau_c$  to the gas relaxation time  $\tau$   
 337 to have an additional estimation of the importance of the thermal effects. If the  
 338 tank represents an infinite heat sink at constant temperature to the gas, then,  
 339 the time it takes for the gas to reach equilibrium with the tank can be modeled.  
 340 In Ref. [1] the transient heat conduction equation was solved analytically and  
 341 the solution was presented as the infinite series of the Bessel functions. When  
 342 keeping only the first leading term of the series the characteristic conduction  
 343 time can be estimated as:

$$\tau_c = \frac{\rho R_{res}^2 Pr}{2.4\mu}, \quad (29)$$

344 where  $\rho$  is the gas density,  $R_{res}$  is the characteristic reservoir dimension,  $Pr$  is  
 345 the Prandtl number. The reservoir characteristic dimension (its radius) is equal  
 346 to  $19.6mm$ , the Prandtl number is equal to  $2/3$  and  $0.71$ , for the monoatomic  
 347 and polyatomic gases, respectively. The gas conduction time, Eq. (29), is pro-  
 348 portional to the gas density and so to the gas pressure under our experimental  
 349 conditions. It depends also on the gas nature through the gas viscosity. The  
 350 value of the gas conduction time for some experimental conditions are provided  
 351 in Table 2, last column. For all considered cases the pressure relaxation time  $\tau$   
 352 is much longer than the gas conduction time  $\tau_c$ . Therefore, we have a new ex-  
 353 perimental confirmation that the gas temperature remains close to the constant  
 354 temperature during the measurements.

#### 355 4.2. Properties of the pressure relaxation time

356 As the mass flow rate through a porous medium, Eqs. (12) and (13), and its  
 357 permeability, Eq. (22), depend on the pressure relaxation time it is interesting  
 358 to study its properties.

359 Figure 4 shows the pressure relaxation time, expressed in seconds, as a func-  
 360 tion of the inverse molecular mean free path,  $\ell^{-1}$ . The equivalent molecular  
 361 mean free path is defined as following [2]:

$$\ell = \frac{\mu v_0}{p_m}, \quad (30)$$

362 where  $v_0$  is the most probable molecular speed

$$v_0 = \sqrt{2\mathcal{R}T}. \quad (31)$$

363 It is clear from Eq. (30) that the inverse equivalent mean free path is a function  
 364 of the mean pressure. By analyzing Fig. 4 and Table 2 we can conclude that the  
 365 pressure relaxation time is proportional to the molar mass. That is, the short-  
 366 est relaxation time is obtained for Helium, which has smaller molar mass, the  
 367 longest relaxation time is found for Argon, which has greatest molar mass, see  
 368 Table 1. All gases have similar behaviors as a function of the inverse molecular  
 369 mean free path, which is a function of mean pressure. For the low mean pressure  
 370 (large mean free path) the relaxation time is quasi-constant, then it decreases  
 371 linearly with pressure increasing (the mean free path decreasing). This behavior  
 372 is related to the number of collisions (molecule-molecule and molecule-wall col-  
 373 lisions): when the intermolecular collisions are numerous (small mean free path)  
 374 the relaxation time is short. With increasing of the molecular mean free path  
 375 (decreasing of pressure) the number of intermolecular collision decreases which  
 376 leads to the increase of the relaxation time, because the gas reaches its equilib-  
 377 rium state through the intermolecular collisions. When the molecular mean free  
 378 path becomes large enough the number of the intermolecular collisions becomes  
 379 negligible in comparison to the number of collisions with the wall (Knudsen  
 380 diffusion regime) and the relaxation time becomes constant, see Fig. 4(b). In  
 381 this case, the pressure relaxation time is determined only by the morphological

382 parameters of a porous medium, *i.e.*, mean pore size, porosity, tortuosity and  
 383 particularities of gas-surface interaction.

384 It is worth to underline that the pressure relaxation time measured for two  
 385 discs is different even if the same gas is considered. This fact lets us conclude  
 386 that the internal structure of the microporous discs could be different. We  
 387 comment on this observation in Section 6.3.

388 The pressure relaxation time can be normalized by the characteristic time  
 389 of the flow, which is defined as follows:

$$t_c = \frac{L}{v_0}. \quad (32)$$

390 This characteristic time depends on the gas nature through the most probable  
 391 gas velocity  $v_0$ , Eq. (31). The relaxation time, normalized by the characteristic  
 392 time, is shown in Fig. 5, as previously in function of the inverse molecular mean  
 393 free path. It is interesting to note that now all gases follow the same curve,  
 394 so all gases have the same pressure relaxation time for the same value of the  
 395 inverse molecular mean free path. Similar behavior of the relaxation time of the  
 396 thermal creep flow was observed in Refs. [1], [1], [2], where the gas flow driven  
 397 by only a temperature gradient through the microchannels of the circular and  
 398 rectangular cross-sections was studied.

399 By taking into account the definition of the characteristic time, Eq. (32),  
 400 we can rewrite expression of the permeability, Eq. (22), in the following form  
 401 by using the equivalent molecular mean free path, Eq. (30),

$$K = \frac{V_0 t_c}{S} \ell. \quad (33)$$

402 When the pressure relaxation time is used, the previous expression of the per-  
 403 meability allows calculating the permeability of a porous sample for different  
 404 gases. This is because the normalized relaxation time is the same for all the  
 405 considered here gases for a given value of molecular mean free path.

406 Another formula for the permeability can be derived by introducing gas  
 407 relaxation time [2],  $t_f$ , which is inversely proportional to the collision frequency  
 408 of the gas molecules, and it can be calculated as

$$t_f = \frac{\mu}{p_m} = \frac{\ell}{v_0}, \quad (34)$$

409 so the expression of permeability becomes

$$K = \frac{V_0 L t_f}{S} \frac{t_f}{\tau}. \quad (35)$$

410 Previous expression shows that the gas permeability depends on sample di-  
 411 mensions,  $S$  and  $L$ , and on the tanks volume,  $V_0$ , used in experiments. The  
 412 increasing (or decreasing) in this volume leads to the corresponding change in  
 413 the pressure relaxation time, so that the ratio  $V_0/\tau$  remains the same. There-  
 414 fore, besides the geometrical characteristics, the permeability depends on the  
 415 ratio between two characteristic times: gas and pressure relaxation times.

416 **5. Mass flow rate**

417 The mass flow rate was calculated from the pressure measurements in each  
 418 tank by using the techniques, explained in Section 2.3. Three noble gases and  
 419 one diatomic gas were used: Helium, Argon, Neon and Nitrogen.

420 *5.1. Measurement uncertainty of the mass flow rate*

421 The classical uncertainty calculation technique is used to estimate the mea-  
 422 surement uncertainty of the mass flow rate, when it is calculated from the pres-  
 423 sure or pressure difference exponential evolution, Eqs. (12) or (13). To ensure  
 424 the upper limit of uncertainty, we use the maximum uncertainty on the mass  
 425 flow rate measurements in the case of the pressure difference fit, Eq. (12), which  
 426 reads:

$$\frac{\delta \dot{M}}{\dot{M}} = \frac{\delta V_0}{V_0} + \frac{\delta(\dot{\Delta p})}{\dot{\Delta p}} + \frac{\delta T}{T} + \varepsilon_0, \quad \text{where} \quad \frac{\delta(\dot{\Delta p})}{\dot{\Delta p}} = \frac{\delta \tau}{\tau} + \frac{\delta p_1}{p_1} + \frac{\delta p_2}{p_2}. \quad (36)$$

427 In previous expression the uncertainty on the pressure difference in time involves  
 428 the uncertainty on the fitting parameter  $\tau$  and the uncertainty on the pressure  
 429 sensors, provided by the manufacturer. The uncertainty on  $\tau$  was obtained from  
 430 the difference in magnitude of a 95% confidence interval for  $\tau$  to represent the  
 431 experimental data. The parameter  $\varepsilon_0$  in Eq. (36) represents the uncertainty  
 432 coming from the non-isothermal effects [1] and it is equal to the maximum value  
 433 in time of the ratio  $(dT/T)/(dp/p)$ . To evaluate the value of  $\varepsilon_0$  the standard  
 434 deviation and the mean temperature is used for  $dT/T$  term calculations, while  
 435 the pressure difference and mean pressure are used to evaluate the pressure  
 436 differential and pressure, respectively. The maximal relative uncertainties of  
 437 each term in Eq. (36), obtained for two microporous discs, are summarized in  
 438 Table 3. The uncertainty of the mass flow rate lies in the range 3.6 – 5.1%.

	$\frac{\delta V_0}{V_0}$	$\frac{\delta \dot{\Delta p}}{\dot{\Delta p}}$	$\frac{\delta T}{T}$	$\varepsilon_0$	$\frac{\delta \dot{M}}{\dot{M}}$
Uncertainty	3.0%	< 1.9 %	< 0.2 %	< 1.2 %	3.6% -5.1 %

Table 3: Measurement uncertainties of the mass flow rate, when the pressure difference between the tanks is used for the calculation. The maximal values obtained for two discs are given.

439

440 *5.2. Results on the mass flow rate*

441 Typical pressure variations over time in both reservoirs are shown in Fig. 3.  
 442 The exponential shape decay of the pressure in each tank and the pressure differ-  
 443 ence between two tanks are clearly visible on this figure. The exponential fit of  
 444 the pressure (and pressure difference) variation curves during the total measure-  
 445 ments duration with the pressure relaxation time as a single fitting parameter  
 446 allow for very smooth reproduction of the experimental pressure recording.



447 In the current experimental setup the measured mass flow rate through the  
 448 microporous media lies in the range  $5 \cdot 10^{-7} - 5 \cdot 10^{-12}$  [kg s<sup>-1</sup>]. This range can  
 449 be extended by modifying the experimental setup configuration.

450 The typical mass flow rate variations in time, calculated from the pressure  
 451 variation in inlet tank,  $-\dot{M}_1$ , in outlet tank,  $\dot{M}_2$ , are shown in Fig. 6. It is clear  
 452 that all three curves practically coincide, which confirms the mass conservation  
 453 property within instrumental uncertainty.

## 454 6. Permeability data

### 455 6.1. Measurement uncertainty on the permeability

456 The uncertainty of the permeability measurements, when using Eq. (22), is  
 457 calculated by the classical way similar to the calculation of the uncertainty on  
 458 the mass flow rate, where the maximum uncertainty is used:

$$\frac{\delta K}{K} = \frac{\delta V_0}{V_0} + \frac{\delta L}{L} + \frac{\delta S}{S} + \frac{\delta \mu}{\mu} + \frac{\delta p_m}{p_m} + \frac{\delta \tau}{\tau}. \quad (37)$$

459 The relative measurement uncertainty on the permeability is presented in Table  
 460 4, where only the maximum values (of two porous discs) for each term of Eq.  
 461 (37) are given, so the permeability uncertainty lies in the range 5.0 – 6.4%.

	$\frac{\delta V_0}{V_0}$	$\frac{\delta L}{L}$	$\frac{\delta S}{S}$	$\frac{\delta \mu}{\mu}$	$\frac{\delta p_m}{p_m}$	$\frac{\delta \tau}{\tau}$	$\frac{\delta K}{K}$
Uncertainty	3.0%	0.5%	0.1%	1.0%	< 0.6 %	< 1.4 %	5.0% -6.4 %

Table 4: Measurement uncertainties of the microporous media permeability. The maximum of uncertainty of two discs is provided.

462

### 463 6.2. Permeability results

464 The microporous media permeability, calculated using Eq. (22) from the  
 465 measured mean pressure and pressure relaxation time, is shown in Fig. 7 as  
 466 a function of inverse gas mean pressure. The results, obtained for two micro-  
 467 porous discs and different gases, are presented and plotted in the log-log axis.  
 468 Theoretically, for the large values of mean pressure, the permeability has to be  
 469 constant, and the same for all gases tested for each disc. This part of the per-  
 470 meability curve corresponds to the classical Darcy law (intrinsic permeability),  
 471  $K_\infty$ , where a porous medium permeability does not depend on the nature of a  
 472 fluid flowing inside. However, as it is clear from Fig. 7, only for the first disc  
 473 with Helium this regime is reached in the present measurements, see "plateau"  
 474 in Fig. 7(a). Larger mean pressure values, above atmospheric pressure, need to  
 475 be applied to reach this regime for other gases and the second disc.

476 When the mean pressure decreases the permeability increases, and it be-  
 477 comes larger than the intrinsic permeability, the phenomenon of the apparent

478 permeability appears, so-called Klinkenberg effect [2]. In this case, the perme-  
 479 ability is usually expressed as

$$K = K_{\infty} \left( 1 + \frac{b}{p_m} \right), \quad (38)$$

480 where  $b$  is a correction factor. As it is clear from Fig. 7 the apparent perme-  
 481 ability becomes gas dependent: for a fixed mean pressure, the permeability is  
 482 higher for lighter gases. However, if the same curves are plotted as a function of  
 483 the mean free path,  $\ell$ , see Fig. 8, the data for different gases are located on the  
 484 same curve. A similar property was also observed for the pressure relaxation  
 485 time  $\tau$ , see Section 4.2 and Fig. 5 This finding leads to an interesting property:  
 486 when the characteristic time is determined for a microporous medium for one  
 487 gas, then the permeability can be calculated using Eq. (33) for other gases.

488 If we compare expression derived in the present paper to calculate the per-  
 489 meability, Eq. (22), with the classical expression of apparent permeability, Eq.  
 490 (38), we find that Eq. (22) has the same asymptotic properties as Eq. (38),  
 491 which can be confirmed by analyzing the measured data. When the mean pres-  
 492 sure tends to infinity, then the product  $\tau p_m$ , tends to a constant value, see Fig.  
 493 9 and we find the constant intrinsic permeability. For the small values of the  
 494 mean pressure  $p_m$  the pressure relaxation time keeps its constant value, see Fig.  
 495 4, therefore the apparent permeability tends to infinity.

496 Recently several papers were published, where the dependence of the cor-  
 497 rection factor  $b$  in Eq. (38) from the pressure is discussed [2], [2]. Usually the  
 498 permeability is plotted as a function of inverse pressure in the linear-linear coor-  
 499 dinate system. If we plot the experimental permeability curves, shown on Fig. 7,  
 500 but using the linear-linear instead of logarithmic-logarithmic scale for both axis  
 501 we can observe the typical behavior of the permeability: it seems to be increase  
 502 linearly as the mean pressure decreases demonstrating the well-known Klinken-  
 503 berg effect [2], [1]. From this evident linear dependence of the permeability on  
 504 the mean pressure ( $K = A + B/p_m$ ) we could conclude that the correction factor  
 505  $B$  is pressure independent and can be used for large pressure variation range.  
 506 However, the linear-linear representation can mask some behavior, because it is  
 507 difficult to present correctly the different order of magnitude of the parameter  
 508 variation in linear scale. If we try to fit the whole permeability experimental  
 509 curve with the same expression,  $K = A + B/p_m$ , we cannot find the same pair of  
 510  $A$  and  $B$  coefficients for the whole curve, see Fig. 11b). The relative deviation  
 511 between the measured and fitted experimental points,  $(K^{exp} - K^{fit})/K^{fit}$ , for  
 512 helium curve is shown of Fig. 12. It is clear that this curve fit very well the  
 513 low pressure range and the large deviation exists in the high pressure (Darcy  
 514 regime). This result demonstrates that the permeability points cannot be fitted  
 515 with unique  $b$  coefficient in the large pressure range when using the Klinkenberg  
 516 expression. Of course, the more detailed study of this property is needed to find  
 517 the numerical values of the correction factor  $b$ .

### 518 6.3. Difference between two discs

519 Two porous discs considered in the present study were fabricated to have the  
520 same expected properties, *i.e.* the smallest average pore size of the order of  $3 \mu$   
521 m. However, the measured permeability has very different values for two discs,  
522 especially for the low pressure. Therefore we assume that two discs have different  
523 internal structure. The tomographic analysis confirms this experimental finding:  
524 the averaged pore size was much larger for the first disc compared to the second  
525 one. Therefore, this technique can be used for the non-destructive analysis of  
526 the permeability of the microporous media. In addition this technique can be  
527 implemented to derive the characteristic pore size of a microporous sample.

## 528 7. Conclusion

529 The experimental procedure for the measurements of mass flow rate and  
530 permeability through the microporous media is proposed and analyzed. In the  
531 frame of this procedure the pressure evolution in each tank (or the evolution of  
532 the pressure difference between two tanks) is successfully fitted with an expo-  
533 nential function using one fitting parameter: the pressure relaxation time. The  
534 simple expressions for the mass flow rate and the permeability, derived from  
535 the exponential fitting of the pressure relaxation in each tank, are proposed. It  
536 was found that besides of the sample dimensions the gas permeability can be  
537 characterized by the ratio between gas relaxation time (inverse of gas collision  
538 frequency) and the pressure relaxation time. With the present experimental  
539 setup we measured the mass flow rate in the range  $5 \cdot 10^{-7} - 5 \cdot 10^{-12}$  [kg/s]  
540 and the permeability in the range  $10^{-14} - 10^{-11}$  [m<sup>2</sup>]. However, we are not  
541 restricted to these ranges of parameters with the present experimental setup  
542 (volume configuration and sample size). We estimate that we could measure  
543 at least 50 times lower mass flow rate and permeability. To go further in low  
544 permeability measurements the experimental setup has to be modified. The  
545 proposed approach is the first very promising stage to evolve towards measure-  
546 ments of even lower permeabilities and also the characteristic dimension (pore  
547 size) of membranes used for microfiltration ( $> 100$  nm) and ultrafiltration ( $> 10$   
548 nm).

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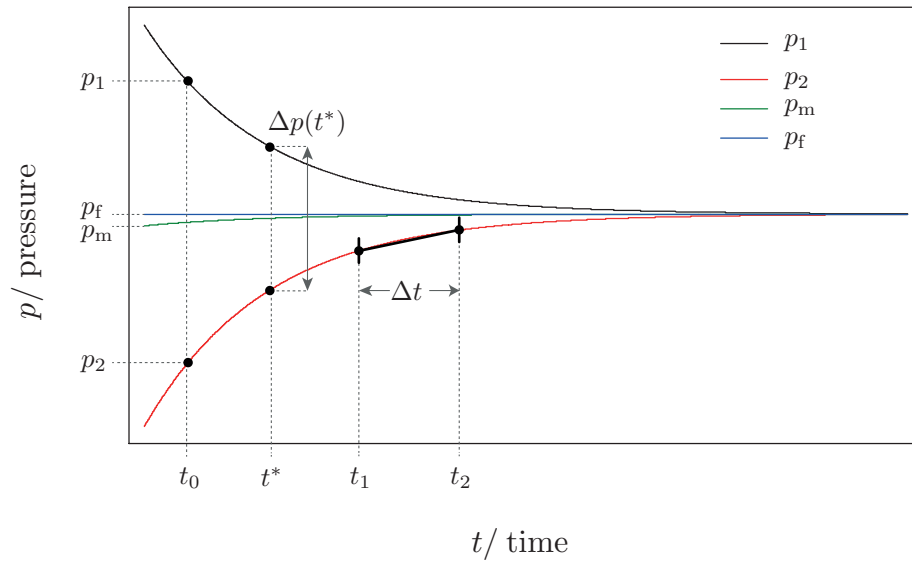


Figure 2: Schematic representation of upstream,  $p_1$ , and downstream,  $p_2$ , pressure response as a function of time. Where  $p_m$  denotes mean pressure and  $p_f$  the final mean pressure.

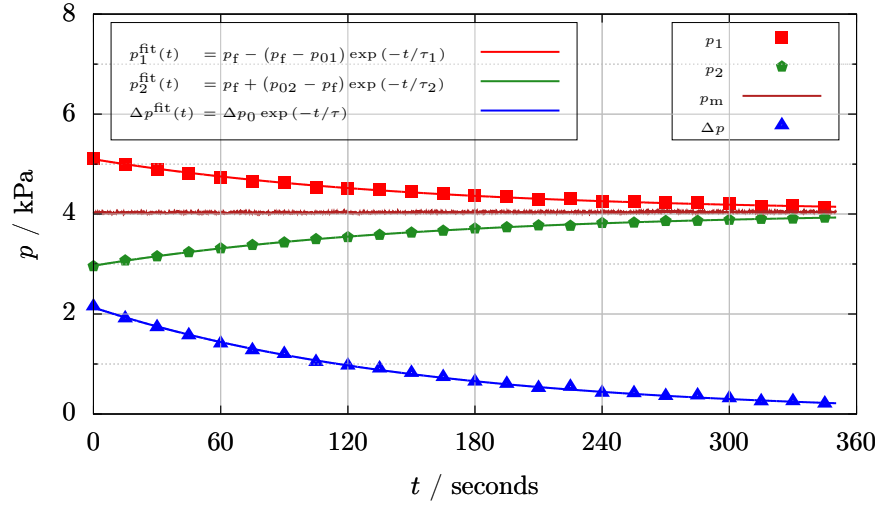
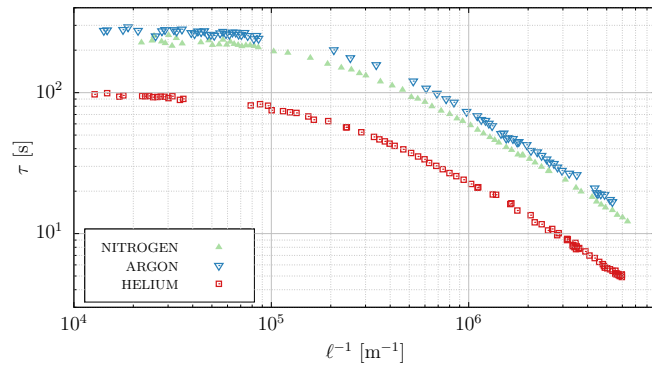
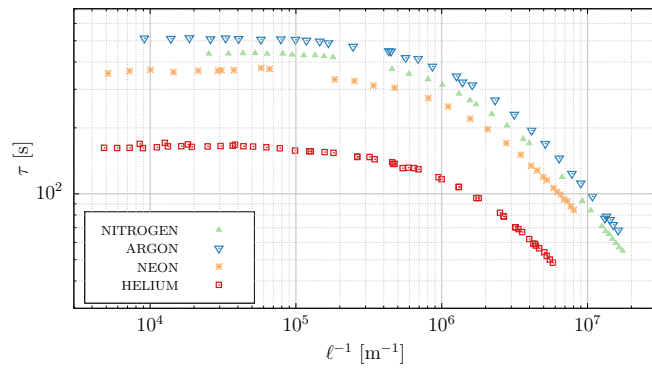


Figure 3: Color on-line: pressure evolution as a function of time. Red squares  $\square$  is upstream tank pressure,  $p_1$ , green pentagons  $\diamond$  is the downstream tank pressure,  $p_2$ , magenta line — is the mean pressure, blue triangles  $\triangle$  is the pressure difference between tanks. Red, green and blue lines represent the exponential pressure fits, which correspond to Eqs. (11) and (10).



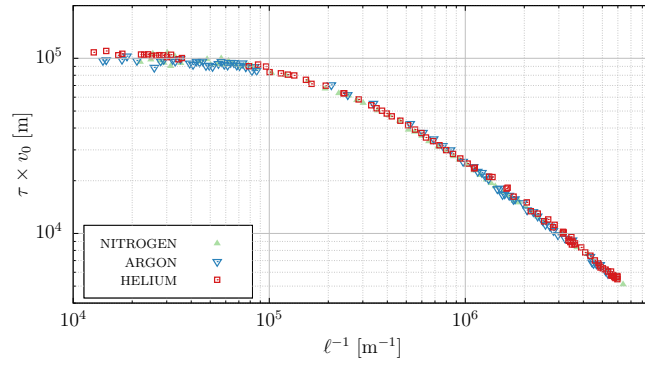
(a)



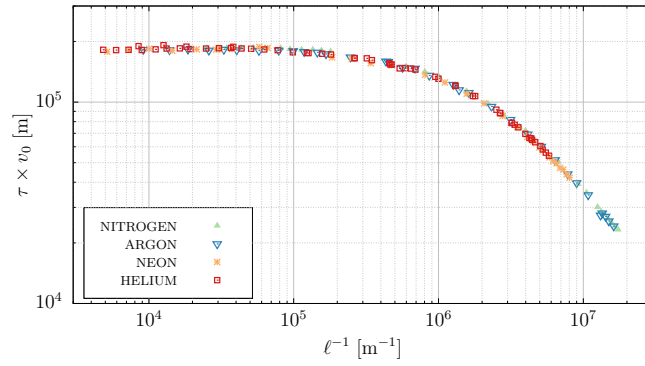
(b)

Figure 4: Color on-line: variation of the relaxation time as a function of inverse molecular mean free path,  $\ell^{-1}$ , for different gases and two porous media: (a) first disc, (b) second disc.





(a)



(b)

Figure 5: Color on-line: variation of the relaxation time, normalized by the characteristic time, Eq. (32), as a function of inverse molecular mean free path,  $\ell^{-1}$ , for different gases and two porous media: (a) first disc, (b) second disc.

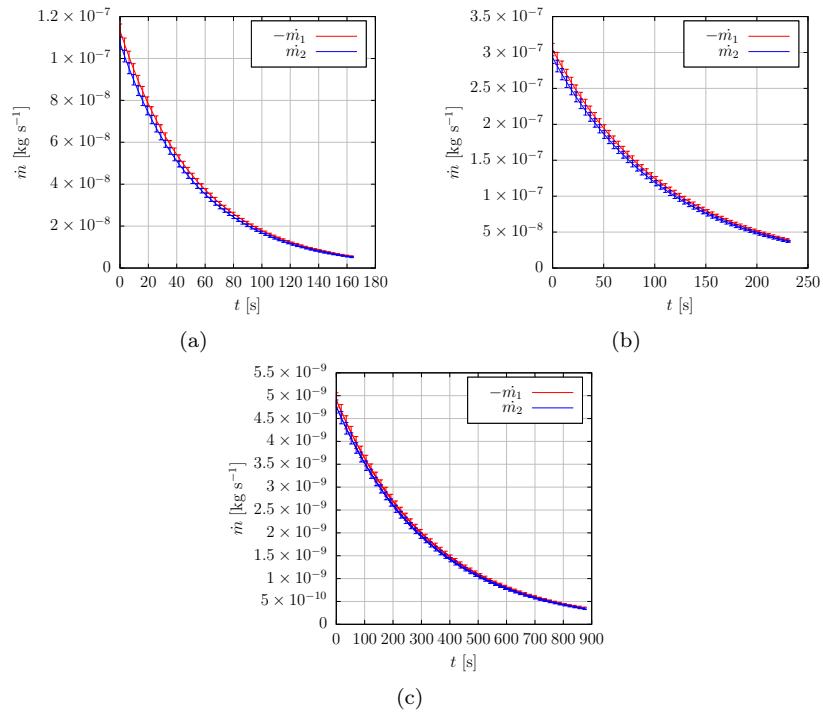
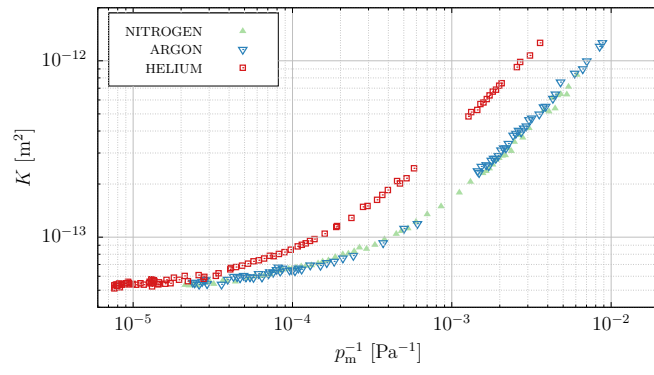
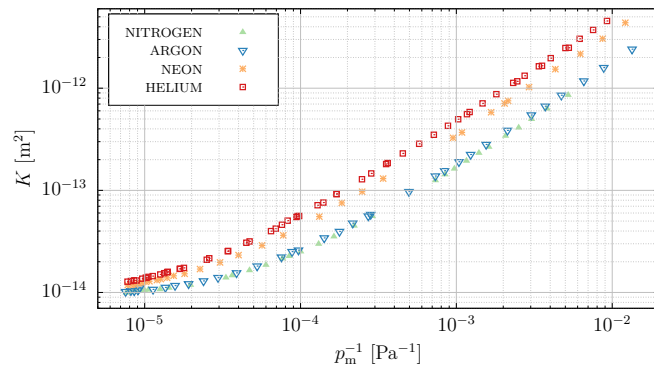


Figure 6: Color on-line: three examples of the mass flow rate variation, calculated from the pressure variation in the first tank,  $M_1$ , in the second tank,  $M_2$ . a) Helium at  $p_m = 1.13 \times 10^5$  Pa, first row in Table 2, b) Argon at  $p_m = 0.74 \times 10^5$  Pa, row 11 in Table 2 c) Neon at  $p_m = 0.03 \times 10^5$  Pa, row 6 in Table 2.

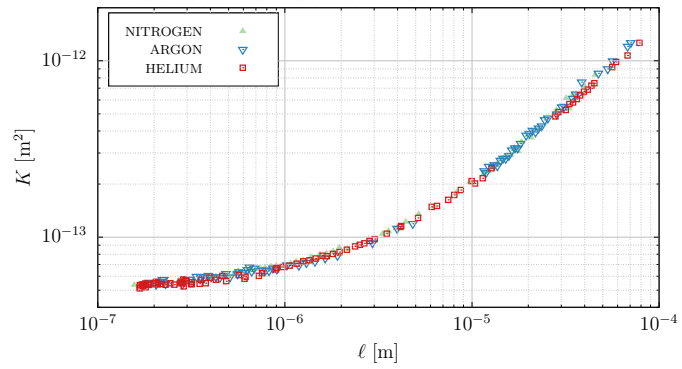


(a)

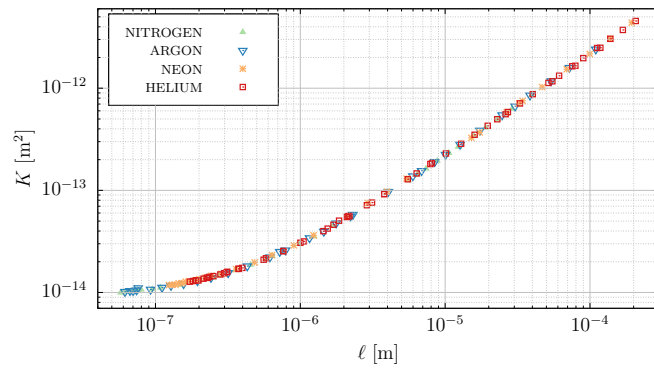


(b)

Figure 7: Color on-line: variation of the permeability of micro porous media, as a function of inverse mean pressure for four gases and two porous media: (a) first disc, (b) second disc.

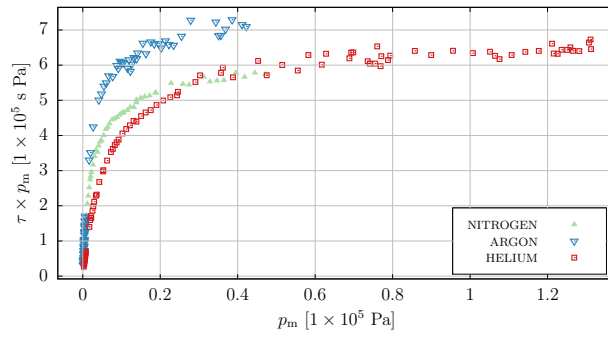


(a)

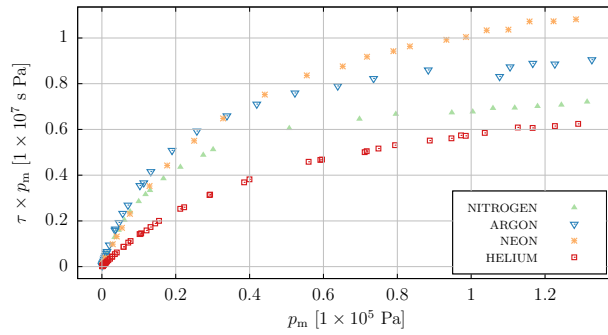


(b)

Figure 8: Color on-line: variation of the permeability of micro porous media, as a function of mean free path for four gases and two porous media: (a) first disc, (b) second disc.

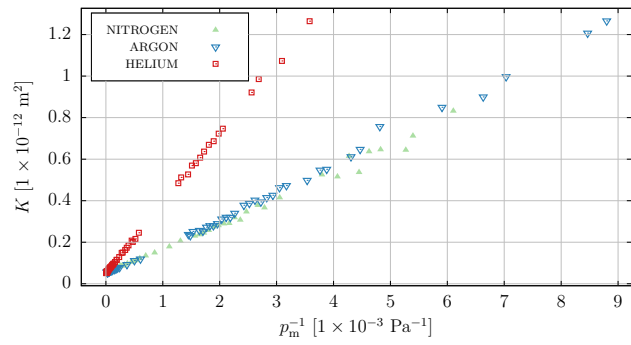


(a)

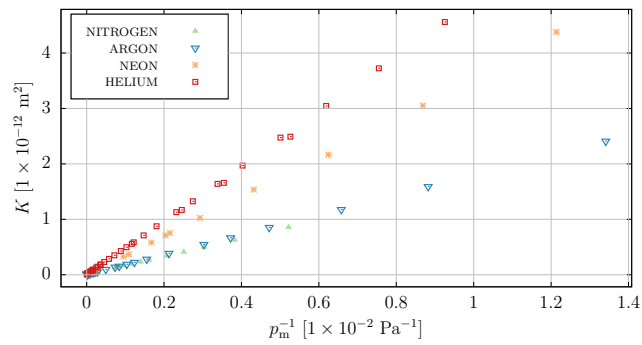


(b)

Figure 9: Color on-line: variation of the quantity  $\tau p_m$  as a function of mean pressure for two porous media: (a) first disc, (b) second disc.



(a)



(b)

Figure 10: Color on-line: Permeability of micro porous media, Eq. (22), as a function of inverse mean pressure for four gases and two porous media: (a) first disc, (b) second disc.

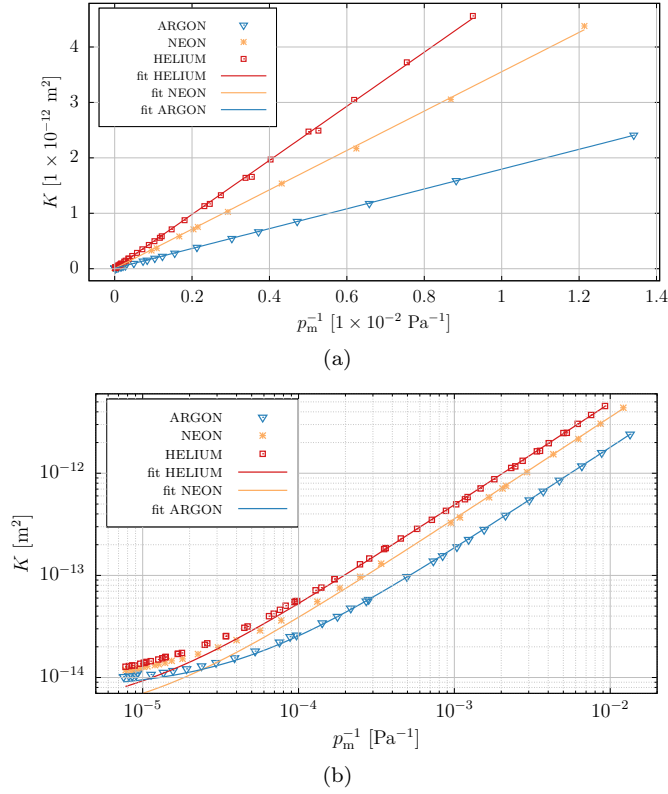


Figure 11: Color on-line: (a) measured permeability for three gases (symbols) for the first disc with the linear fit of the experimental data (solid lines); (b) the same data as on (a) but plotted in log-log coordinates.

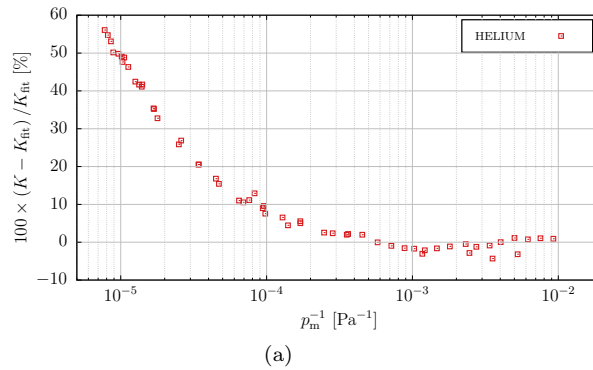


Figure 12: Color on-line: the relative error for the permeability:  $(K_{exp} - K_{fit}) / K_{fit} \times 100\%$  for the measurements of Helium for the first disc,  $K_{exp}$  is the measured value,  $K_{fit}$  is the fitted value. The same experimental points and linear fit are also presented on Fig. 11.