Measurements of air permeability of sandstones using a pycnometer, novel experimental and theoretical approach

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Addition of auxiliary hardware to a gas pycnometer forms a permeameter- pycnometer instrument which allows for air permeability measurements as well as density and porosity. Characterization of sandstone samples using the linear (Darcy) model and capillary flow model (corrected Poisseuille equation) is presented.

Gas flow rates through sandstones are substantially higher then liquids and generally much lower pressures can be used for gas permeability measurements. It appears that the pressure range used in typical gas (Helium) pycnometers is sufficient for such measurements and large range of permeability coefficients can be obtained. Moreover, the typically used Hassler cells for high pressure applications can be replaced by simple hardware which can be easily attached to a pycnometer sample chamber for rock core holding. The pycnometer offers volume (density) measurements and if the geometrical volume can be determined, the porosity of such core can be obtained.

Three samples of sandstones cores of different permeability coefficients were acquired from Kocurek Industries, Inc., Caldwell, Texas. The received samples were of perfect cylindrical shape with 1" diameter and 2 " of length. Some of their listed properties are presented in the table 1 below.

Number	Name	Permeability [mD]	Porosity [%]	Permeability by	Homogeneity
1	Bentheimer	1500 - 3500	23 - 26	KCl/N2	Yes
2	San Saba	70 - 85	19 - 21	N2	Yes
3	Colton	0.1 - 2	7 - 15	N2	Yes

Table 1. Literature data of the received samples

Using the specimens as received, the density and porosity of the samples were measured using the VersaPyc XR pycnometer and air was used as the carrier gas. The results are presented in the table below and the extra significant figures in some columns were preserved for further calculations.

Number	Name	Mass [g]	Volume by pycnometer, Vp [mL]	Density by pycnometer [g/mL]	Geometrical volume, Vg [mL]	Porosity [%]
1	Bentheimer	50.6696	19.0286	1.968	25.741	26.077
2	San Saba	53.7726	20.0871	2.089	25.741	21.965
3	Colton	60.5655	22.4586	2.353	25.741	12.748

Table 2. Measured density and porosity of received samples

The porosity ε is a dimensionless quantity and is defined as the ratio of void volume Vv to geometrical volume Vg. If multiplied by 100, it is expressed as % of Vv/Vg. The void volume Vv is the difference between the geometrical volume Vg and the volume Vp obtained by pycnometer.

$$\varepsilon = \frac{Vv}{Vg} = \frac{Vg - Vp}{Vg} \qquad \text{(Eq. 1)}$$

Before carrying out the permeability measurements, the core samples, except the top and bottom, were uniformly covered with epoxy. High strength, aluminum filled, two-parts epoxy of high viscosity was used to prevent deep migration into the pores of samples. Holding the sample by the ends horizontally in a rotary machine, the thickness of epoxy layer and its uniformity along the side surface of the cylinder was satisfactory to be used for O-rings mounting.

A simple three-part holding system was developed to hold the sample and easily attach it to the pycnometer sample chamber. The bottom of the sample is exposed to either vacuum or pressure conditions inside the sample chamber while the top can be exposed to either atmospheric pressure or high external pressure for dealing with cores with very low permeability coefficients.
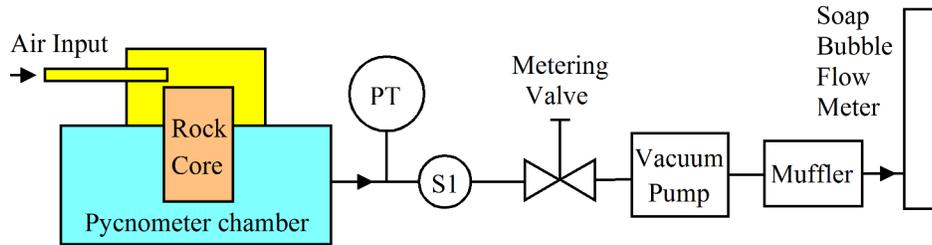
Fig. 1. Vacuum mode setup

The simplified flow schematic presented above shows the vacuum mode setup for measuring flow rates through rock cores. The top of the sample is exposed to atmospheric pressure and the bottom is kept under various vacuum levels, which are indicated by the absolute pressure transducer PT. When the solenoid S1 is opened and the vacuum pump activated, the metering valve adjustments allows for setting various vacuum levels in the pycnometer sample chamber. The metering valve was temporarily installed between the S1 and the vacuum pump for the vacuum mode. After a steady state is established, the flow rate is measured using the soap bubble flowmeter and digital stopwatch. In order to reduce vibrations from the vacuum pump output, the specially designed muffler is added between the vacuum pump output and the flowmeter.
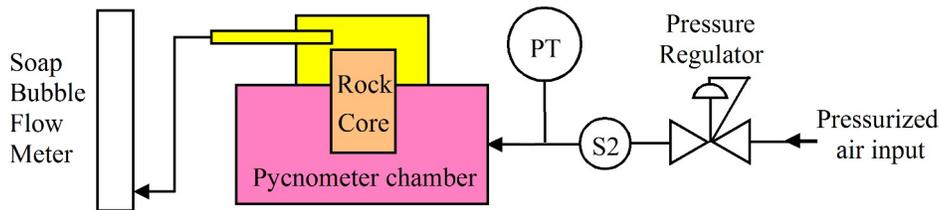
Fig. 2. Pressure mode setup

The pressure mode setup is envisioned as the typical hardware arrangement without any internal customization of the pycnometer. If the solenoid 2 is turned on, the precision pressure regulator embedded inside the instrument can set various pressure levels inside the sample chamber of the pycnometer. The pressure transducer, PT, measures the pressure values. The flow of air out of the sample is directed to the bubble flow meter. Technically one pressure setpoint is sufficient for determining the permeability coefficient, but for further discussion of the theory multiple pressures setpoints were used.
The flow rates measurements versus set pressures for the San Saba sample were carried out using the vacuum and pressure mode setups described above. This sample was selected because it was just

convenient to obtain the whole range of pressures using the 344 kPa pressure range transducer and flow rates values using only one bubble flowmeter of 50 mL volume range. The results are presented on the graph below.

San Saba core sample

$f(x) = 0.000127 \, x + 0.015205$
$R^2 = 0.999853$
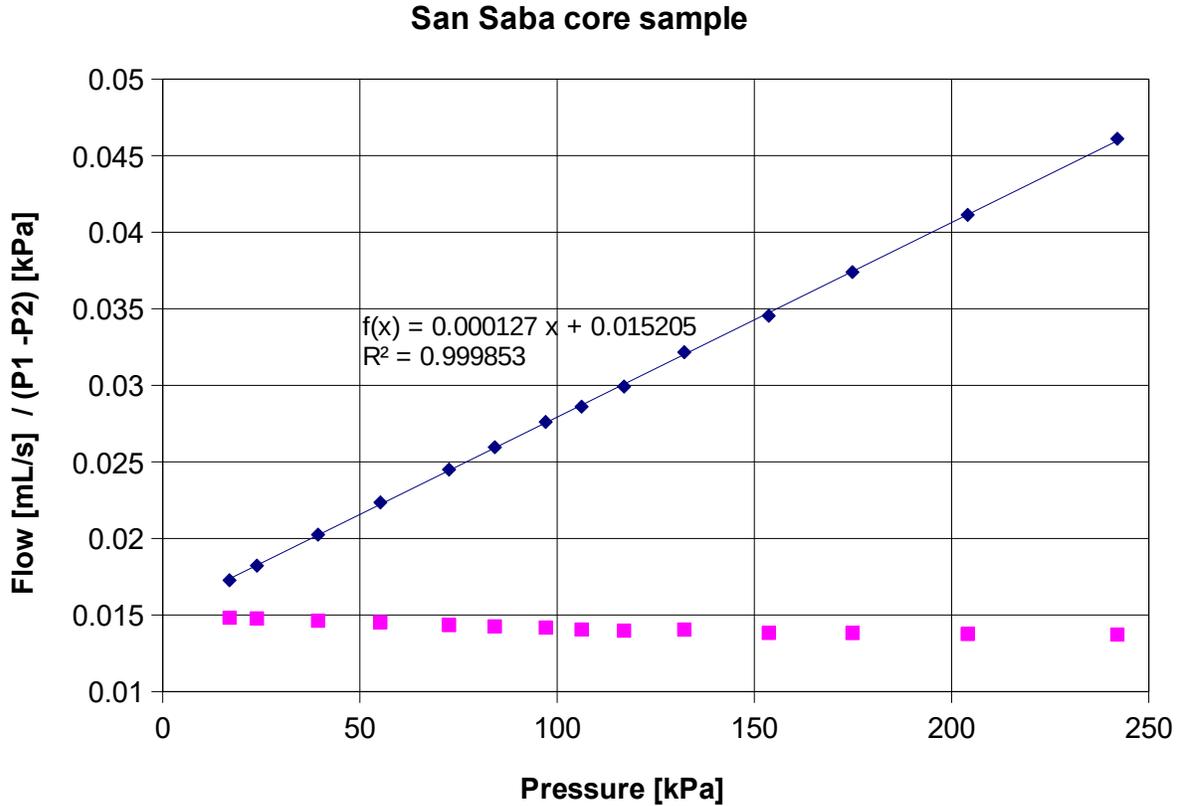
Fig. 3. Permeability results for Darcy (blue) and capillary (red) flow models

The pressure values on the abscissa are the final pressure values measured by the pressure transducer at the bottom of the sample while the top of the sample was kept at atmospheric pressure. The ordinate is the quotient of flow rate [mL/s] and absolute value of pressure differences [kPa] on opposite sides of the sample. The series of blue diamonds correspond to the so-called Darcy model calculations.

The series of pink squares corresponds to the capillary flow model where the ordinate values are calculated as the same flow rate multiplied by pressure P1 and divided by the absolute difference of the squares of pressures P1 and P2.

For non-compressible fluids, the simple relation of flow rate versus pressure differential is commonly used for permeability calculations (linear Darcy model):

$$Q = \frac{k \cdot A}{\mu \cdot h} \cdot (P_1 - P_2) \qquad \text{(Eq. 2)}$$

where:
Q - flow rate [mL/s],
k - permeability coefficient [mD]
A - cross section of sample area [cm^2]
μ - dynamic viscosity [cP] = [mPa*s]
h - height (length) of the sample [cm]
P_1 - P_2 - pressure differential across the sample [kPa]

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Somehow the same form of the equation 2 is often used for compressible fluids, namely gases. Using the dynamic viscosity of air at 23 C as 0.0183 mPa*s, the sample diameter as 2.54 cm and its length of 5.08 cm, the permeability coefficient k can be written as

$$k \, [mD] = 1859.01 \cdot \frac{Q}{P_1 - P_2} \qquad \text{(Eq. 3)}$$

As it can be seen from the above chart, the $Q/(P_1-P_2)$ ratio for the Darcy model is not a constant but it rather exhibits a linear relationship and the $Q/(P_1-P_2)$ ratio depends on the pressure used. The linear profile remains the same in the vacuum region and above atmospheric pressures. In the pressure range used for this sample, from 16.75 to 242 kPa, k [mD] varies from 32 to 86. So, if the experimental conditions are not specified and if the particular formula is not stated, comparison of literature k values (70-85) is difficult.

Using Poisseuille equation of flow through a capillary and corrected for compressible fluids (gases), and assuming N such capillaries of having the same (representative, equivalent) radius R and height h, the relationship of flow and pressure can be written as:

$$\text{Total Flow } Q = N \cdot Q_c = \frac{N \cdot \pi \cdot R^4}{16 \cdot \mu \cdot h} \cdot \frac{\left(P_1^2 - P_2^2\right)}{P_2} \qquad \text{(Eq. 4)}$$

where Qc is the flow through a single capillary, **μ** is the dynamic viscosity, **P_1** and **P_2** are the pressures at the sample input and output. An important consequence of this equation is that both pressure values, P_1 and P_2 must be known, not just a difference between the two as in incompressible fluids case. In most typical experiments the P_1 or P2 is just the atmospheric pressure (for convenience of experimental setup) and is called the reference pressure. In general case, the reference pressure can be of any value, from vacuum to much higher pressures then the atmospheric.

In general it is more appropriate to consider various numbers N_i of different radii R_i and carry out a summation over the index i, but for simplicity reasons the same size of capillaries is being assumed. Since the gas flows faster through the larger size capillaries and the exponent of four practically ensures that only the largest sizes of capillaries need to be considered.

Combining the constants from equation 4 into an overall constant k_c, the equation can be rearranged as

$$k_c = \frac{N \cdot \pi \cdot R^4}{16 \cdot \mu \cdot h} = Q \cdot \frac{P_2}{P_1^2 - P_2^2} \qquad \text{(Eq. 5)}$$

The series of red squares on the Fig. 3 have the ordinate values calculated using the right hand side of the equation 5. The k_c values remain fairly constant versus the pressure range used. It can be easily concluded that the Eq. 4 is more appropriate for description of gas (air) permeability in sandstones then Eq. 2. The slightly decreasing trend of k_c values vs pressure can be related to a more complex relationship.

The equation 4 can be re-written in a different form as

$$Q = k_c \cdot \left(P_1 - P_2\right) \cdot \frac{\left(P_1 + P_2\right)}{P_2} \qquad \text{(Eq. 6)}$$

From the mathematical point of view, the Eq. 6 can be considered as the Eq. 2 multiplied by the correction factor $(P_1+P_2)/P_2$. Perhaps the most popular is the so-called Klinkenberg correction to account for discrepancy of results when experimental flow-pressure results for gases are interpreted using Darcy formula (Eq. 2). This is basically not needed if the capillary model (Eq. 4) is used for interpretation of such experimental data. The further explanation of using the "gas slippage" concept does not seem to be valid at

all. As shown on the Fig. 3 (the series of blue diamonds), the linear dependence of the $Q/(P_1-P_2)$ vs pressure is the same in the vacuum region and in the pressurized region. The overall problem just seems to be arising from using the wrong equation for gas permeability calculations.

The interpretation of the straight line fitted through the series of data points for the Darcy model can yield interesting results. Extrapolation to the zero pressure yields the minimum k value, which roughly corresponds to the k_c value for the capillary model. Assuming $P_1 = 0$, and comparing Eq. 2 to Eq. 4 (or Eq. 6), the expression evaluates just to comparing the constants.

$$\frac{k_{min} \cdot A}{\mu \cdot h} = \frac{N \cdot \pi \cdot R^4}{16 \cdot \mu \cdot h} \qquad \text{(Eq. 7)}$$

The cross-sectional area of all capillaries can be written as $N \cdot \pi \cdot R^2$ and it can be interpreted as the area of voids Sv in the base of a cylindrical sample. From the definition of porosity and assuming uniformity of voids throughout the sample, it can be stated that the ratio of voids on the surface, Sv, to the geometrical surface area, A, is the same as volume of voids, Vv, to the geometrical volume Vg.

$$\frac{Sv}{A} = \frac{Vv}{Vg} = \varepsilon \qquad \text{(Eq. 8)}$$

So it can be written that

$$N \cdot \pi \cdot R^2 = A \cdot \varepsilon \qquad \text{(Eq. 9)}$$

Substituting the $N \cdot \pi \cdot R^2$ by A·ε in Eq. 7 and after simplification, the constant k can be expressed as

$$k_{min} = \frac{R^2}{16} \cdot \varepsilon \qquad \text{(Eq. 10)}$$

Since the k_{min} can be obtained from calculations of flow-pressure data and other parameters, and the porosity ε can be obtained using a pycnometer, the radius R of a representative (imaginary) capillary can be easily evaluated.

$$R = 4 \cdot \sqrt{\frac{k_{min}}{\epsilon}} \qquad \text{(Eq. 11)}$$

Similar relationship between R and k can be found in literature but since derivations were based on the Darcy (linear) model and presumably for non-compressible fluids, the numerical factor is different. After calculations of the $k_{min} = 0.2785 \cdot 10^{-9}$ [cm^2] and using the porosity value ε = 0.21965, the calculated value of R is equal to $1.424 \cdot 10^{-4}$ [cm] = 1.424 [μm].

Once the capillary radius R is calculated, the number N of such capillaries can be estimated using for example Eq. 9. The estimate of the N value yields 17463217 or $1.7463217 \cdot 10^7$. Now, having the number of the imaginary capillaries and their radius R, it is possible to calculate the total surface area of them and therefore, use this number as an estimate of the surface area of the sample S_a.

$$S_a = 2 \cdot \pi \cdot R \cdot h \cdot N \qquad \text{(Eq. 12)}$$

The Sa value amounts to 79391 cm^2 for the whole sample. Dividing this value by the mass of the sample, the specific surface area per gram is equal to 1476 [cm^2/g]. Such value should be considered as a lower end estimate as obviously a much more complex geometry of passages does exist in any such samples and smaller size passages can contribute to a greater extent to the overall specific surface area.

The outlined above procedure for some characterization of a rock core sample requires several different pressures and corresponding flow data to obtain the useful parameters. It might not be so simple to

accomplish that for various samples using a single flowmeter or specific pycnometer due several orders of magnitude of the permeability coefficients. For example, the sample named Bentheimer requires large flow rates to cover reasonable pressure range and the vacuum range is very limited due to low capacity of the vacuum pump used. Samples of rather low permeability like the Colton one, requires much higher pressures to be used then the typical range used in any gas (helium) pycnometer and for flow rates measurements, the bubble flowmeters of 1 and 1 mL range are needed. To overcome the technical difficulties, it is better to use the capillary flow model instead of the linear (Darcy) model. A single measurement of flow vs pressure is sufficient as the k_c constant is fairly the same and it is close to the k_{min} value.

In some previous applications notes, the Eq. 4 was proposed for evaluation of cement fineness (specific surface area) instead of other simplified approaches. The packed bed of cement (or any fine porous powder) can be considered similar to a core rock, except that the packed bed must have a container for preserving its shape while the rock core sample holds its shape without any container.

In the application note entitled "*Absolute method for determination of specific surface area using gas (air) permeability technique*", the following equation was proposed for evaluation of cement fineness:

$$S_a = \frac{A_b}{2 \cdot m_b} \cdot \sqrt{\frac{\epsilon^3 \cdot V_b \cdot (P^2 - P_o^2)}{\mu \cdot P_o \cdot Q}} \quad \text{(Eq. 13)}$$

Sa – specific surface area [cm^2/g]
A_b –geometrical surface area of the powder bed [cm^2]
m_b – mass of the powder used for bed formation [g]
ϵ – bed porosity [dimensionless]
V_b – geometrical volume of the powder bed [cm^3]
P – pressure at the top of the powder bed [kPa]
Po – Pressure at the bottom of the powder bed [kPa]
μ – dynamic viscosity [Pa·s]
Q – flow rate [mL/min]

Using the flow-pressure data and porosity obtained from pycnometric measurements, the equation 13 was tentatively applied to the sandstone samples. The following table present results of the calculations.

Number	Name	Avg k_c [mL/(s·kPa)]	S_a [cm^2/g]	R [cm]	N
1	Bentheimer	$7.154 \cdot 10^{-1}$	290	$9.136 \cdot 10^{-4}$	$5.039 \cdot 10^{5}$
2	San Saba	$1.423 \cdot 10^{-2}$	1545	$1.360 \cdot 10^{-4}$	$1.914 \cdot 10^{7}$
3	Colton	$1.188 \cdot 10^{-4}$	17767	$1.214 \cdot 10^{-5}$	$2.465 \cdot 10^{9}$

Table 3. Characterization parameters using the absolute method for specific surface area measurements

In summary, using a very simplistic theoretical approach and simple instrumentation, the parameters, like density, porosity, permeability coefficients, minimum specific surface area, representative radius of capillary, and a number of such capillaries can be obtained. Such parameters can be useful in characterization of similar classes of materials like sandstones.