RECONSIDERING KLINKENBERG'S PERMEABILITY DATA

Douglas Ruth and Rasoul Arabjamaloei, University of Manitoba

This paper was prepared for presentation at the International Symposium of the Society of Core Analysts in Trondheim, Norway, 27-30 August, 2018.

ABSTRACT

The foundational paper by Klinkenberg contains a very rich data set for gas flow in porous samples over a range of mean pressures from 1 *kPa* to 2000 *kPa*. Based on his data, Klinkenberg proposed a correlation between pressure drop and flow rate that depends on both the Darcy permeability (the permeability at infinite mean pressure) and the ratio of a coefficient, now generally termed the Klinkenberg coefficient, and the mean pressure. Klinkenberg's approach to analyze his data was to determine the Darcy permeability at a high mean pressure, then calculate Klinkenberg coefficients at lower values of mean pressures. He found that values of the calculated Klinkenberg coefficient remained constant for a certain range of mean pressures, but changed significantly at low mean pressures. Klinkenberg clearly stated that his results did not show a strictly linear function of effective permeability with the inverse of mean pressure – it appears that this observation has never been studied in detail. Based on an approach published by Arabjamaloei and Ruth, Klinkenberg's data have been reanalyzed using three methods: by optimizing the Darcy permeability and the Klinkenberg coefficient simultaneously; by holding the Darcy permeability constant but optimizing the value of the Klinkenberg coefficient to obtain a single value for all mean pressures; by optimizing Darcy permeability, the Klinkenberg coefficient, and a second Klinkenberg coefficient divided by mean-pressure-squared. It is shown that the last approach is successful in correlating all of Klinkenberg's data to within $\pm$ 5%. However, the improvements due to the modified Klinkenberg equation are marginal and do not explain all the disagreement. For this reason, a second data set, published by Ash and Grove, was explored. This data set, which has been largely ignored in the literature, provides convincing evidence for Klinkenberg's ideas, once the data are reanalyzed to account for shortcomings in the ranges of experimental pressures. Based on ideas documented by Carman for mixed viscous/ diffusive flows, the results are used to derive estimates of an effective pore diameter and the tortuosity.

INTRODUCTION

Two foundational papers on low-pressure flow in porous media, one by Klinkenberg [1] and the other by Ash and Grove [2], have had very different impacts in the literature. Based on data from Google Scholar at the time of writing the present paper, the paper by

Klinkenberg has been cited 1853 times while the paper by Ash and Grove has been cited only 15 times. The present paper will show that by combining the results from these papers with the model documented by Carman [3] (a similar treatment is also presented in Klinkenberg's paper for a single straight capillary), a very simple method can be derived to predict an effective pore diameter and tortuosity of a porous sample. The great advantage of the two papers is that they both contain sufficiently detailed experiment data to allow reanalysis of the results, a rare occurrence in the literature.

For the purpose of the present paper, three characteristic flow regions will be defined. When pressure is very low or pore sizes are very small, this will be termed the "purely diffusive flow" region. This region is often referred to as the "free molecular flow" region. When the pressure is very high or the pore sizes are large, this will be termed the "purely viscous flow" region. This region is often referred to as the "Poiseuille flow" or "Darcy flow" region. Between the two regions is as "intermediate flow" region. This region is also sometimes referred to as the "Knudsen flow", "slippage flow" or "Klinkenberg flow" region, although the Knudsen flow region often is defined to include also the purely diffusive flow region.

MATHEMATICAL BASIS

The analysis in this paper is based on a generalized Klinkenberg equation as proposed by Arabjamaloei and Ruth[4].

$$-\left(1+\frac{b}{P}+\frac{b_2}{P^2}\right)\frac{dP}{dx}=\mu\frac{v}{k_o} \quad (1)$$

Here b is the Klinkenberg coefficient, b_2 is a second Klinkenberg coefficient, v is the Darcy (bulk) velocity, and k_o is the Darcy permeability, the permeability at infinite mean pressure. The mass flow rate, $\dot{m}$, is related to the Darcy velocity by the equation

$$v=\frac{\dot{m}}{\rho\, A_b} \quad (2)$$

Here A_b is the bulk cross-sectional area and ρ is the density. For steady, compressible gas flow, the mass flow is constant along the sample but the Darcy velocity will vary with the density, hence pressure. Substituting Equation 2 into Equation 1 results in the equation

$$-\left(1+\frac{b}{P}+\frac{b_2}{P^2}\right)\frac{dP}{dx}=\mu\frac{\dot{m}}{\rho\, k_o\, A_b} \quad (3)$$

For an ideal gas and isothermal flow

$$\rho=\rho_m\frac{P}{P_m} \quad (4)$$

Here the subscript denotes the conditions at the arithmetic mean pressure. Substituting into Equation 3 and multiplying through by P

$$-\left(P+b+\frac{b_2}{P}\right)\frac{dP}{dx}=\mu\frac{\dot{m}}{k_o\, A_b}\frac{P_m}{\rho_m} \quad (5)$$

The right-hand side of Equation 5 is a constant for steady flow. Therefore the equation can be integrated over the bulk length, L_b, and from the pressure at its highest value, P_h, to its lowest value, P_l. The result is

$$\frac{{P_h}^2 - {P_l}^2}{2} + b\,(P_h - P_l) + b_2 \ln\frac{P_h}{P_l} = \mu \frac{\dot{m}}{k_o\,A_b}\frac{P_m}{\rho_m}\;L_b \tag{6}$$

Because the mean pressure, P_m, is equal to $(P_h + P_l)/2$, this equation can be rearranged as

$$P_m + b\; + \frac{b_2}{(P_h - P_l)}\ln\frac{P_h}{P_l} = \mu \frac{\dot{m}}{k_o\,A_b}\frac{P_m}{\rho_m}\,\frac{L_b}{(P_h - P_l)} \tag{7}$$

An effective permeability, k, is defined by the expression

$$k = \mu \frac{\dot{m}}{A_b\,\rho_m}\,\frac{L_b}{(P_h - P_l)} \tag{8}$$

Substituting this expression into Equation 7 and rearranging

$$k = k_o\,\left(1 + \frac{b}{P_m} + \frac{b_2}{P_m\,(P_h - P_l)}\ln\frac{P_h}{P_l}\right) \tag{9}$$

Three dimensionless groups will now be defined:

$$\textbf{Darcy Number} \quad Da = \frac{k}{k_o} \tag{10}$$

$$\textbf{Klinkenberg Number} \quad Kl = \frac{b}{P_m} \tag{11}$$

and

$$\textbf{Second Klinkenberg Number} \quad Kl_2 = \frac{b_2}{P_m\,(P_h - P_l)}\ln\frac{P_h}{P_l} \tag{12}$$

to yield

$$1 + Kl + Kl_2 = Da \tag{13}$$

At first sight, the second Klinkenberg number appears to be ill-behaved because as $P_h \to P_l$ this term goes to infinity. However, as $P_h \to P_l$ then $\ln(P_h/P_l) \to 0$ which compensates.

THE KLINKENBERG RESULTS

Klinkenberg included the following statement in his paper:

"*Fig.1, 2 and 3 show that the apparent permeability is approximately a linear function of the reciprocal mean pressure. The linear function, however, is an approximation… wherein the value of the constant b increases with increasing pressure.*"

To explore reasons for this behavior, the Klinkenberg data were reanalyzed using three different approaches. First, the data were fitted with Equation 5 but assuming b_2 is zero.

This resulted in values for k_o and b. Second, the data were fitted with Equation 5 holding the value of k_o equal to the value at high pressure and assuming b_2 is zero. Third, the data were fitted with Equation 5 allowing k_o, b and b_2 to vary. The results for the three samples for which Klinkenberg provided detailed data are shown in Table 1.

Table 1 The Fitting Parameters for the Three Models Considered

Sample/Property	#1	#2	#3
	Core Sample "A"	Jenna Glass Filter	Core Sample "F"
Varying k_o and b			
k_o *(mD)*	24.94	2.51	182.14
b *(kPa)*	11.33	55.56	6.759
Varying b only			
k_o *(mD)*	23.66	2.45	176.57
b *(kPa)*	12.02	57.07	7.038
Varying k_o, b and b_2			
k_o *(mD)*	24.39	2.46	177.81
b *(kPa)*	12.28	58.40	7.781
b_2 $(kPa)^2$	-0.661	-4.183	-1.165

Figures 1 through 3 show the errors between the fitted equations and the measured values of effective permeability. For Sample #1, using both k_o and b results in errors exceeding 5% at high pressures and in the vicinity of 10 *kPa*. When only b is used in the fit, the effective permeability is well predicted at high pressures (this should occur because this is the region used to predict k_o) but the error near 10 *kPa* is the greatest observed. Using all three parameters results in the best prediction. However, the improvements are marginal and the errors at high pressures and near 10 *kPa* are still relatively large.

The behavior of Sample #2 (Figure 2) is similar to that for Sample #1. Again, using both k_o and b results in the largest errors at high pressures. When only b is used in the fit, the effective permeability is better predicted at high pressures, although not as well as for Sample #1, but the error near 100 *kPa* has increased. Using all three parameters results in the best predictions. However, the improvements are again marginal and the error near 100 *kPa* is still relatively large.

The behavior of Sample #3 (Figure 3) is even more similar to that for Sample #1. Again, using both k_o and b results in the largest errors at high pressures and in the vicinity of 10 *kPa*. When only b is used in the fit, the effective permeability is better predicted at high pressures, although not as well as for Sample #1, but the error near 10 *kPa* is still large. Using all three parameters results in the best prediction. However, the improvements are again marginal and the errors near 10 *kPa* are still relatively large.

In summary, despite using a higher order correlation, there remains a systematic deviation in the Klinkenberg data. In order to obtain further insights into this problem, a second data set, published by Ash and Grove [2], was studied.

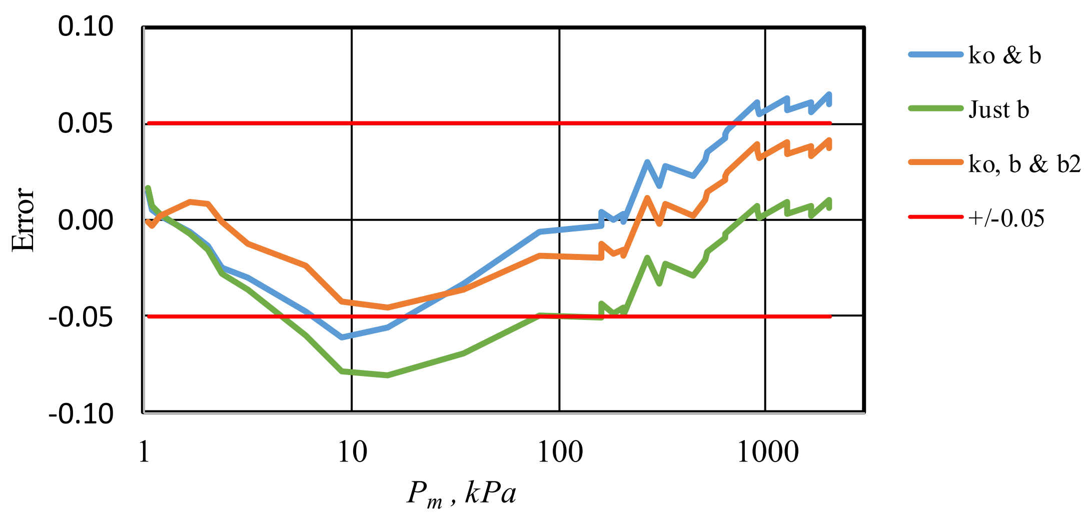

Figure 1 The Errors for Sample #1 (Core Sample "A"). These errors are the values predicted by the correlation equation minus the measured values, divided by the measured values.

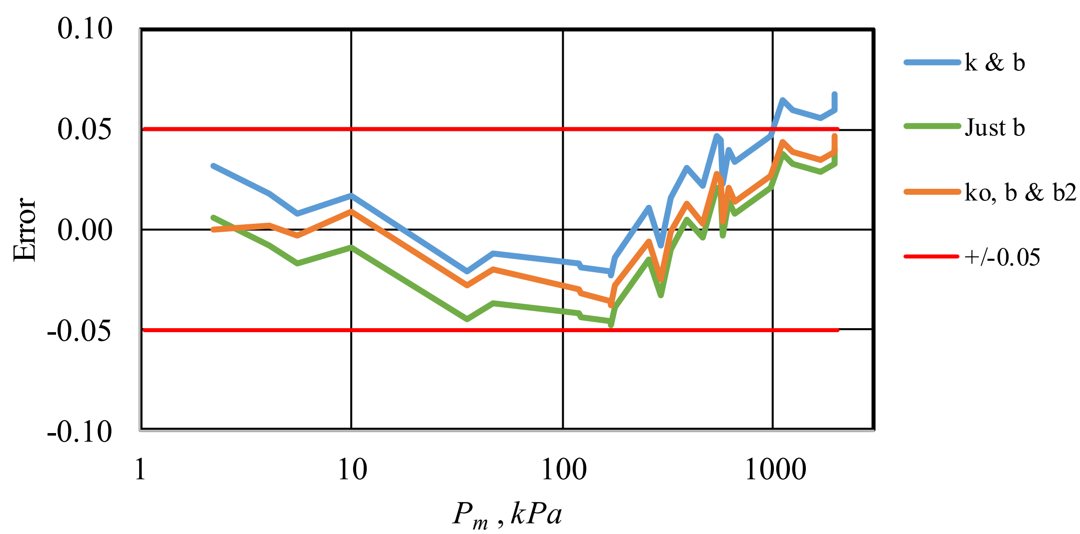

Figure 2 The Errors for Sample #2 (Jenna Glass Filter). These errors are the values

predicted by the correlation equation minus the measured values, divided by the measured values.

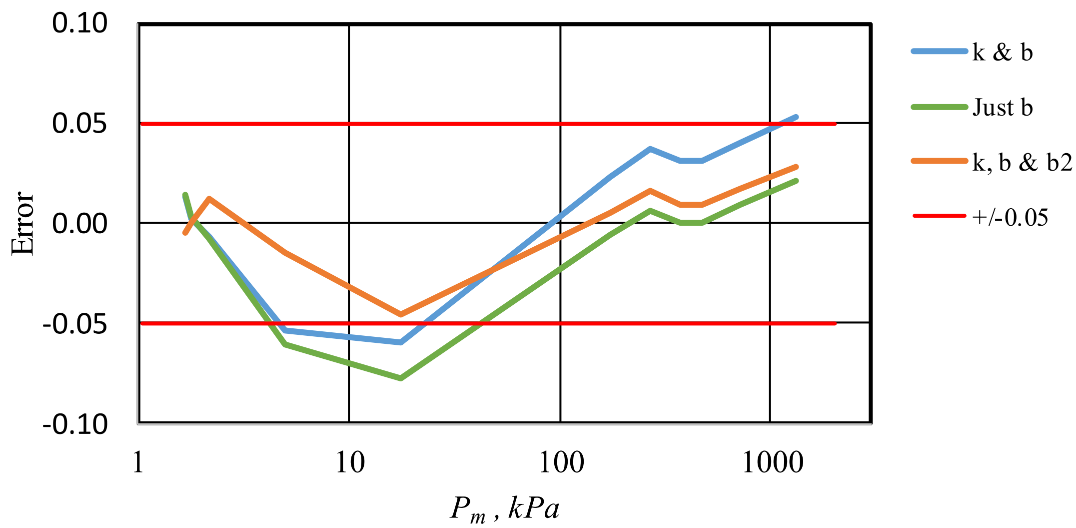

Figure 3 The Errors for Sample #3 (Core Sample "F"). These errors are the values predicted by the correlation equation minus the measured values, divided by the measured values.

THE ASH AND GROVE RESULTS

Ash and Grove [2] reported effective permeability, and upstream and downstream pressure data, for 10 gases on a single sample of ceramic porous media. However, the data is not in the same form as that used in the petroleum literature. The flow rate, G, was calculated using the declining pressure in a known upstream volume using the equation

$$G = \frac{dP_h}{dt} V_h \frac{T}{T_R} \tag{14}$$

Here t is time, V_h is the upstream chamber volume, T is temperature, and T_R is a reference temperature, in this case ambient. (The units of the flow rate in the original paper were *ergs/s*, the pressures were in *cm of Hg* and time was in *min*. In Equation 14 the units of G are *J/s*.) In the paper, flow rates are not actually reported; however, permeabilities, K, in units of cm^2/s are reported and G is related to K by the equation

$$K = \frac{G\, L_b}{A_b\, (P_h - P_l)} \tag{15}$$

All the variables in this equation are reported except G which can be calculated from Equation 15. Once G is calculated, the mass flow rate can be calculated using the equation

$$\dot{m} = \frac{G\ M}{\mathcal{R}\ T} \tag{16}$$

where M is the molecular weight of the gas and $\mathcal{R}$ is the universal gas constant. Once the mass flow rate is known, Equation 8 can be used to calculate the effective permeability.

The resulting effective permeability/ mean pressure data were fitted to determine the Darcy permeability and the Klinkenberg coefficient. When fitting an equation to experimental data, the form of the error equation used can lead to different values for the fitting parameters because different equations will "weight" the data points differently. In the present analysis, two different error equations were used

$$\sum_{i=1}^{i=N} \left(1 + \frac{b}{(P_m)_i} - \frac{k_i}{k_o}\right) = Error \tag{17}$$

and

$$\sum_{i=1}^{i=N} \left(k_o + \frac{k_o\ b}{(P_m)_i} - k_i\right) = Error \tag{18}$$

The results for fitting these two equations to the experimental data for the ten gases are shown in Table 2. These results are not at all what is expected. All the experiments were done on a single experimental sample and the expectation is that the Darcy permeability should be the same for all the gases. What is observed is that the Darcy permeability varies by a factor approaching 3 and the two error equations generally predict very different values for the same gas. In the original paper, the same observation was made. There was some attempt to explain the variations based on arguments involving adsorption and surface flow but the authors admitted the arguments were not convincing. We could speculate that this inconsistent behaviour may be why this work has been largely ignored in the literature.

Table 2 Calculated Darcy Permeabilities and Klinkenberg Coefficients

	Equation 17		Equation 18				Equation 17		Equation 18	
Gas	k_o (mD)	b (kPa)	k_o (mD)	b (kPa)		Gas	k_o (mD)	b (kPa)	k_o (mD)	b (kPa)
He	13.84	97.77	19.11	70.37		H	5.78	149.03	14.00	61.45
Ne	10.59	94.95	14.78	67.89		N	15.84	29.65	18.58	25.09
Ar	12.10	42.52	15.11	33.77		O	10.73	49.36	13.99	37.56
SO_2	8.59	22.37	25.14	7.45		C_2H_6	16.50	13.09	16.79	12.84
Kr	14.32	27.23	14.50	26.89		CO_2	9.33	34.39	29.74	10.19

When fitting data, it is important that the data covers the full range that the equation represents. In this case, if an accurate value of k_o is desired, at least some of the data points should have a sufficiently high mean pressure such that the Da is close to 1. Table 3 shows the minimum Darcy numbers calculated using the Darcy permeabilities based on fitting the data. Clearly, none of these Darcy numbers are even close to 1. In order to determine if this observation was the source of the scatter in the values of k_o, the data were reanalyzed by using only data points that had a Darcy number less than 10 (this was not possible for hydrogen) or the last three data points. Although this was not expected to yield accurate values for the Darcy permeability, this was the only way that at least three points would be used for each gas. The results for the recalculated Darcy permeabilities are shown in Table 4.

Table 3 The Minimum Experimental Darcy Number Calculated using Darcy Permeabilities based on Equation 13.

Gas	Minimum Da		Gas	Minimum Da
He	6.352		H	14.540
Ne	5.229		N	2.801
Ar	3.577		O	4.556
SO_2	2.782		C_2H_6	2.326
Kr	2.345		CO_2	4.397

Although the values for Darcy permeability still show variations, they are in much better agreement. Furthermore, the two error equations now predict very similar values. In order to proceed, the values for the 10 gases and the two error equations were averaged. This gave a value for k_o of $16.0\ mD$.

Table 4 Reanalysed Darcy permeabilities based on data points for which the Darcy number is near 1.

Gas	k_o (mD) (Eq.13)	k_o (mD) (Eq.14)		Gas	k_o (mD) (Eq.13)	k_o (mD) (Eq.14)
He	18.32	18.40		H	14.47	14.48
Ne	15.44	14.78		N	14.97	14.98
Ar	16.63	16.74		O	15.25	15.26
SO_2	15.51	15.52		C_2H_6	17.41	17.85
Kr	16.11	16.15		CO_2	15.74	16.11

Using the single value of the Darcy permeability, the data were reanalyzed to obtain new values for the Klinkenberg coefficients. The work of Carman suggests that the

Klinkenberg coefficient divided by the mean pressure should vary with the mean free path of the gas. The mean free path can be calculated from the equation

$$\bar{\lambda} = \frac{\lambda_c}{P_m} \tag{19}$$

where λ_c is the mean free path coefficient

$$\lambda_c = \mu \sqrt{\frac{\pi \mathcal{R} T}{2 M}} \tag{20}$$

Table 5 shows the results and Figure 4 shows a plot of b_1 versus λ_C

Table 5 Calculated values for the Klinkenberg coefficient and the mean free path coefficient.

Substance	$b\ (kPa)$	$\lambda_C \times 10^3$		Substance	$b\ (kPa)$	$\lambda_C \times 10^3$
He	84.34	5.72		H	53.73	3.57
Ne	62.64	3.99		N	29.33	1.90
Ar	31.82	2.05		O	32.70	2.06
SO_2	11.87	0.904		C_2H_6	13.54	0.962
Kr	24.28	1.57		CO_2	19.69	1.29

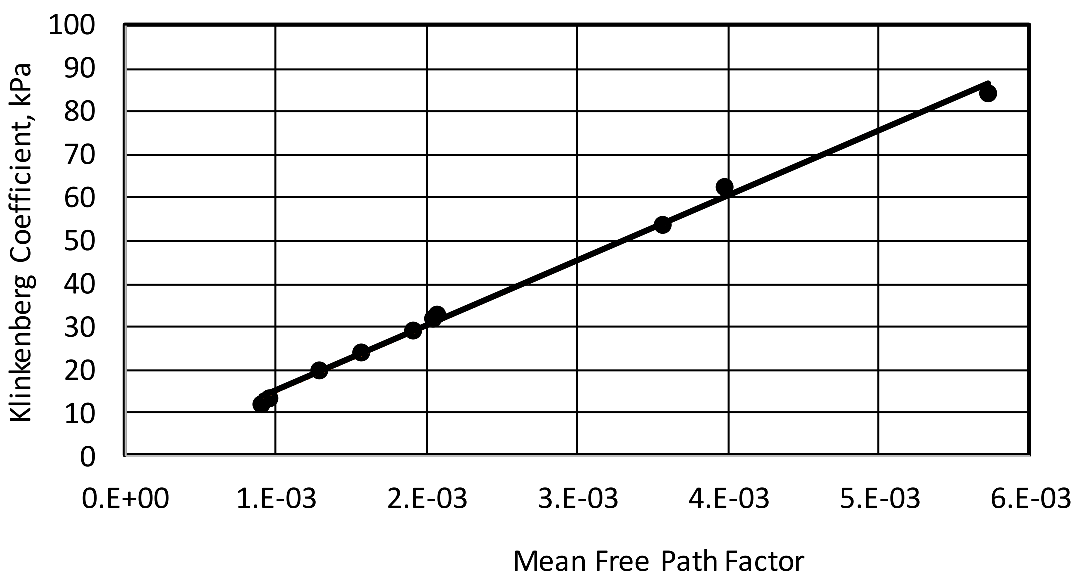

Figure 4 The Klinkenberg Coefficient as a Function of the Mean Free Path Coefficient

As observed in Figure 4, the correlation between the two variables is remarkably good. In fact, the regression coefficient is 0.9962. It can be concluded that when the Ash and Grove data is analysed by taking care to separately analyse the data that contains information on the viscous flow region, the results conform well with the expectation that all the different gases will have behaviours that can be accounted for by changes in the mean free path coefficient.

Figure 5 summarizes the errors between the experimental values and the calculated values of permeabilities using the data from Table 5. In general the errors are small, much below ±0,05. However, the sulphur dioxide results show anomalously large errors; there is no apparent reason for this behaviour. It is observed that the errors are generally positive. This may be due to the value of Darcy permeability used in the analysis. The Darcy permeability can easily be in error because it was calculated from data that did not include values for Darcy numbers near 1.

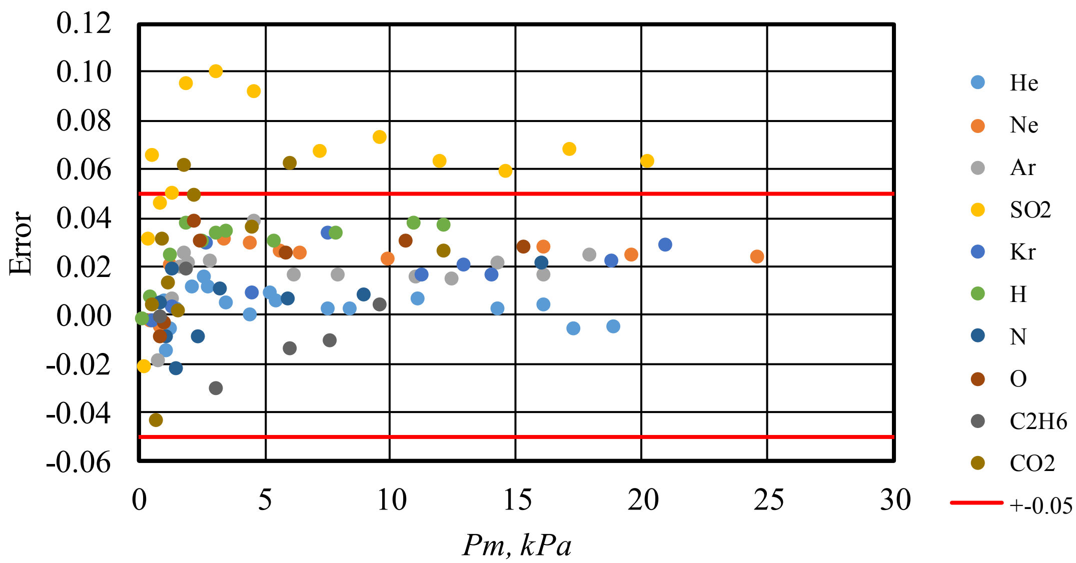

Figure 5 The Errors for the Ash and Grove data. These errors are the values predicted by the correlation equation minus the measured values, divided by the measured values.

USING KLINKENBERG DATA TO PREDICT SAMPLE STRUCTURE

The Ash and Grove work was motivated by a desire to determine pore properties based on flow in capillary tubes collected into a parallel tube model. The equation for the effective permeability (as developed in Carman [3] but based on the earlier work of others, most notably Adzumi [5]) is

$$k = \frac{\phi\, \delta^2}{32\, \tau^2}\left(1 + 8\,\frac{\bar{\lambda}}{\delta}\right) \qquad 21$$

Here ϕ is the porosity, δ is an "effective" tube diameter, τ is the tortuosity, and $\bar{\lambda}$ is the mean free path of the gas given by Equation 19. Comparing this equation with Equation 9, and ignoring the second Klinkenberg term, the Darcy permeability may be identified as

$$k_o = \frac{\phi\,\delta^2}{32\,\tau^2} \tag{22}$$

and the Klinkenberg coefficient by

$$b = \frac{2\,\mu}{\delta}\sqrt{\frac{\pi\,\mathcal{R}\,T}{M}} \tag{23}$$

Given values for ϕ, k_o and b, these equations should allow the prediction of the effective diameter and the tortuosity. Based on the Ash and Grove data, the values for these variables are $\delta = 1.91\times10^{-6} \mp 0.14\,m$ and $\tau = 1.46 \mp 0.11$. The value for the diameter compares favorably with values that were calculated by Ash and Grove based on a number of different approaches (0.41×10^{-6} to $2.84\times10^{-6}\,m$). The medium used for this study was a ceramic which would be expected to have a very uniform and systematically packed structure. As such, a reasonable expectation for the tortuosity would be $\sqrt{2}$ or 1.414 which is remarkably close to the value based on the Ash and Grove data.

At very low pressures, or for samples with very small flow passages, the flow becomes purely diffusive and viscosity effects become minimal. The onset of this behavior is characterized by the Knudsen number which is defined as

$$Kn = \frac{\bar{\lambda}}{\delta} \tag{24}$$

and Equation 21 may be written as

$$k = \frac{\phi\,\delta^2}{32\,\tau^2}(\,1 + 8\,Kn\,) \tag{25}$$

As the Knudsen number becomes large, this equation goes to

$$k = \frac{\phi\,\delta^2}{4\,\tau^2}Kn \tag{26}$$

If experiments are conducted in the diffusive region, this equation may be used to model the results. The measurement of diffusive properties of porous media has great utility. As pointed out by Klinkenberg in a separate paper [6], diffusion is an analogy for electrical conductivity in porous media. For a parallel tube model, the formation factor F is given by (Ruth, Lindsay and Allen [7])

$$F = \frac{\tau^2}{\phi} \tag{27}$$

Therefore, once effective pore diameters and tortuosities are determined, formation factors can be predicted without the need to saturate the samples with an electrically conducting liquid. The present work clearly demonstrates that when experiments are conducted to capture and analyze flow in both the diffusive and viscous regions, gas flow experiments give the results predicted from simple theories of flow in tubes. Therefore, they should allow calculation of meaningful values for effective pore diameters and tortuosities. It is the opinion of the authors that diffusive experiments represent a very important but underutilized opportunity to gain a much better understanding of rock samples.

A word of caution is required. It should always be possible, by using very low pressures, to conduct experiments in moderate to high permeability samples that range over the diffusive, intermediate, and viscous flow regions. However, for low permeability samples, it may be difficult to perform experiments in the viscous flow region without using very high pressures. Therefore, the potential to use this technique to determine pore structure on tight samples needs further investigation.

CONCLUSIONS

The work reported in this paper supports the following conclusions:

1. Using a model equation with a second order dependence on mean pressure leads to a better correlation between mean pressure and permeability. However, the improvements are marginal.
2. Even with a second order model, the Klinkenberg data show a systematic deviation from the predicted values in the intermediate flow region between purely viscous and purely diffusive flow.
3. When reanalyzed to reduce the impact of lack of data near a Darcy number of 1, the Ash and Grove data provide very consistent results for the permeability of the sample to various gases.
4. Based on the Ash and Grove data, the Klinkenberg coefficient varies in a linear fashion with the mean free path coefficient with a very high regression coefficient.
5. Using the derived Darcy permeabilities and Klinkenberg coefficients, very reasonable values for the effective pore diameter and tortuosity are predicted for the sample used by Ash and Grove.
6. In order to best implement a method to calculate effective pore diameter and tortuosity of a sample, accurate data must be collected in both the purely viscous and the purely diffusive flow regions.
7. Diffusion experiments could represent a very important technique for studying samples of porous media.

REFERENCES

1. Klinkenberg, L.J., "The Permeability of Porous Media to Liquids and Gases", *API Drilling and Production Practice*, (1941), 200-213.
2. Ash, R. and D.M.Grove, "Low-Pressure Gas Flow in Consolidated Porous Media. I. Flow through a Porous Ceramic", *Transactions of the Faraday Society*, (1960), **56**, 9, 1357-1371.
3. Carman, P.C., "Flow of Gases through Porous Media", Butterworths, London, (1956).
4. Arabjamaloei, R. and D.W.Ruth, "Lattice Boltzmann Based Simulation of Gas Flow Regimes in Low Porous Media: Klinkenberg's Region and Beyond, *Journal of Natural Gas Science and Engineering*, (2016), **31**, 405-416.
5. Adzumi, H., "Flow of Gaseous Mixtures through Capillaries I, II,and III", *Bulletin of the Chemical Society of Japan*, (1937), **12**, 199-226, 285-303.
6. Klinkenberg, L.J., Analogy between Diffusion and Electrical Conductivity in Porous Rocks", *Bulletin of the Geological Society of America*, (1951), **62**, 559-564.

7. Ruth, D.W., Lindsay, C., and Allen, M., "Combining Electrical Measurements and Mercury Porosimetry to Predict Permeability", *Petrophysics*, (2013), **54**, 6, pp.531-537.