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Estimating permeability using median pore-throat radius obtained from mercury intrusion porosimetry

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Abstract
Mercury intrusion porosimetry (MIP) has been widely used to characterize the pore structure for various types of porous media. Several relationships between permeability and pore structure information (e.g., porosity and pore-size distribution) have been developed in the literature. This work is to introduce a new, and simpler, empirical equation to predict permeability by solely using the median pore-throat radius \( r_{50} \), which is the pore-throat radius corresponding to 50% mercury saturation. The total of 18 samples used in this work have a wide range of permeability, from \( 10^{-6} \) to \( 10^{3} \) mD, which makes the new equation more applicable. The predicted permeabilities by using the new equation are comparable with permeability values obtained from other measurement methods, as shown from ten samples with permeability data measured with nitrogen.

Keywords: MIP, median pore-throat radius, permeability, pore structure

1. Introduction
Permeability is an important parameter to characterize the ease with which a porous medium transmits fluids, and an accurate measurement or estimation of permeability has been a challenging task, especially for media with low permeability. Due to the limitations (e.g., required experimental apparatus and long measurement duration, especially for tight samples) associated with the permeability measurement, several empirical relationships have been published to predict permeability based on other parameter(s) that are relatively easier to obtain; but most of these relationships involve more than one parameter variable. As one of the first empirical relationships, Hazen (1911) derived a simple equation to calculate hydraulic conductivity of unconsolidated porous materials by only using effective grain size. The recent work of Rezaee et al (2006) concluded that the median pore-throat radius yields the best correlation coefficient for permeability, porosity and pore-throat size of carbonate rocks.

Over the past 30 years, mercury intrusion porosimetry (MIP) has become a well-established technique for characterizing porous media, since Washburn (1921) proposed the relationship between capillary pressure and the pore radius as the basic theory for MIP (Giesche 2006). Compared with other pore size characterization approaches (e.g. gas sorption), MIP is based on a simple principle and could cover a wide range of pore sizes (from about 3 nm to 300 \( \mu \)m for current models of MIP instruments), which makes it a powerful characterization tool. In addition, MIP measurement is less time-consuming, one MIP test is usually completed within 1 h for not-so-tight samples like sandstones and carbonates, while for tight samples (e.g., shales) it usually needs 2 h because of long evacuation time due to its nano-sized pores during low pressure analysis. Because of the extremely low permeability, which is usually in nano-darcy, it is expensive to measure the permeability of shale samples directly. As a result, estimating permeability of shale samples by MIP could be an alternative method.
Derivation of permeability from MIP data has been pursued by several researchers (Swanson 1981, Katz and Thompson 1986, 1987), among them the Katz and Thompson (1986, 1987) method (called KT method in this paper) is the one we use to calculate permeability and our new equation is derived based on these calculated values. The validation of the KT method will be provided in section 5.2 of this paper. We will also compare the permeability values measured by N2 with permeability calculated by using our new equation and the equation proposed by Rezaee et al (2006).

2. MIP background

As a non-wetting fluid for most porous media, mercury will not invade pores unless an external pressure is applied. The diameter of the pores invaded by mercury is inversely proportional to the applied pressure; the higher the pressure applied, the smaller the pores invaded by mercury. Washburn (1921) developed the following equation based on the assumption that all the pores are cylindrical in shape:

\[ \Delta P = \frac{2 \gamma \cos \theta}{R}, \]

where \( \Delta P \) is the pressure difference across the curved mercury interface; \( \gamma \) is the surface tension of mercury; \( \theta \) is the contact angle between mercury and the porous medium; and \( R \) is the corresponding pore-throat radius. Using \( \gamma = 485 \text{ dynes/cm} \) and \( \theta = 130^\circ \), equation (1) becomes

\[ \Delta P = \frac{90.43}{R}, \]

where \( \Delta P \) is in psia and \( R \) is in micrometres (\( \mu \)m).

During the sample analysis, MIP collects the data of applied pressure and cumulative intrusion volume at that specific pressure. Katz and Thompson (1986, 1987) introduced the following equation to calculate permeability based on the MIP data:

\[ k = \frac{1}{\phi \mu} (L_{\text{max}})^2 \frac{L_{\text{max}}}{L_c} \phi S(L_{\text{max}}), \]

where \( k \) (darcy) is the air permeability; \( L_{\text{max}} \) (\( \mu \)m) is the pore-throat diameter at which hydraulic conductance is maximum; \( L_c \) (\( \mu \)m) is the characteristic length which is the pore-throat diameter corresponding to the threshold pressure \( \text{P}_t \) (psia); \( \phi \) is porosity; \( S \) is the surface area per unit pore space that is composed of pore width of size \( L_{\text{max}} \) and larger.

The threshold pressure \( P_t \) is determined at the inflection point of the cumulative intrusion curve and the selection of \( L_{\text{max}} \) is dependent on \( P_t \). Webb (2001) described in detail the KT method (equation (3)) for predicting permeability, and the step-by-step data processing procedures to determine each parameter in equation (3) will be shown in section 4.

3. Materials and experimental procedure

In order to obtain a representative empirical relationship to predict permeability, a total of 18 samples with a wide range of permeability were tested by the MIP method; the samples are listed in table 1. Each sample (mostly cube-sized, with the largest linear dimension of about 1.5 cm) was oven-dried at 60°C for at least 48 h to remove moisture in pore spaces and then cooled to room temperature (\( \sim \)23°C) in a desiccator before the MIP test.

During an MIP test, each sample underwent two analyses: low-pressure and high-pressure analyses. The highest pressure produced by Micromeritics AutoPore IV 9510 (Norcross, GA) is 60 000 psia (413 MPa), and the pore-throat diameter corresponding to this pressure according to equation (2) is about 3 nm. The largest pore-throat diameter recorded by MIP is about 300 \( \mu \)m under low-pressure analysis. The samples were evacuated to 50 \( \mu \)m Hg (i.e. 0.05 Torr or 6.7 Pa), Equilibration time (the minimum time duration to achieve a stable mercury level before moving on to the next pressure value) was chosen to be 50 s.

Among these samples, ten of them were measured for permeability to nitrogen gas (\( k_{\text{measure}} \) in table 2), using rock cylindrical core samples (2.54 cm i.d., 4 cm height), by Core Laboratories Inc. (Aurora, CO or Houston, TX) following standard methods in API RP 40 (API 1998).

4. Data analysis of MIP tests

Besides permeability, several other pieces of useful information could be derived from MIP data, like bulk density, porosity and tortuosity (Hager 1998). In order to concentrate on the purpose of this study, we will only introduce the data-processing procedures related to permeability calculation. The KT method is the basic theory we use here to calculate the permeability and to derive our new equation. Although Webb (2001) has described the KT method in detail, it is still necessary to present this process clearly by giving an actual example calculation.

Here we discuss the permeability calculation process of sample 8 (white chalk) in table 1, which is a type of carbonate. After obtaining the raw data of the MIP test (as shown in figure 1), the first and important step is to define the threshold pressure \( P_t \), which is determined at the inflection point of the cumulative intrusion curve. This inflection point (414.6 psia) is defined as the highest point in the log differential intrusion curve, which is shown in figure 2. The characteristic length \( L_c \) corresponding to \( P_t \) can be calculated as 0.436 \( \mu \)m.

### Table 1. 18 samples tested by MIP.

<table>
<thead>
<tr>
<th>No.</th>
<th>Sample</th>
<th>No.</th>
<th>Sample</th>
<th>No.</th>
<th>Sample</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Asphalt</td>
<td>6</td>
<td>Dolomite</td>
<td>11</td>
<td>Barnett Shale no. 1*</td>
</tr>
<tr>
<td>2</td>
<td>Red brick</td>
<td>7</td>
<td>Gray chalk</td>
<td>12</td>
<td>Barnett Shale no. 2</td>
</tr>
<tr>
<td>3</td>
<td>Concrete</td>
<td>8</td>
<td>White chalk</td>
<td>13</td>
<td>Barnett Shale no. 3</td>
</tr>
<tr>
<td>4</td>
<td>Granite</td>
<td>9</td>
<td>Berea sandstone</td>
<td>14</td>
<td>Barnett Shale no. 4</td>
</tr>
<tr>
<td>5</td>
<td>Limestone</td>
<td>10</td>
<td>Indiana sandstone</td>
<td>15</td>
<td>Japan mudstone</td>
</tr>
<tr>
<td>6</td>
<td>Red brick</td>
<td>11</td>
<td>Dolomite</td>
<td>16</td>
<td>Yucca Mt. Tuff</td>
</tr>
<tr>
<td>7</td>
<td>Red brick</td>
<td>12</td>
<td>Gray chalk</td>
<td>17</td>
<td>Hanford basalt</td>
</tr>
<tr>
<td>8</td>
<td>Red brick</td>
<td>13</td>
<td>White chalk</td>
<td>18</td>
<td>Costa Rica basalt</td>
</tr>
</tbody>
</table>

* Barnett Shale samples 1–4 come from different depths in the same well.
Figure 1. Cumulative intrusion volume versus intrusion pressure for white chalk.

Figure 2. Log differential intrusion versus intrusion pressure for white chalk.

Table 2. Comparison between \( k_{\text{cal}} \) and \( k_{\text{measure}} \).

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Sample name</th>
<th>Porosity</th>
<th>( r_{50} ) (( \mu m ))</th>
<th>( k_{\text{cal}} ) (mD)(^a)</th>
<th>( k_{\text{measure}} ) (mD)(^b)</th>
<th>( k_{\text{measure}}/k_{\text{cal}} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>Red brick</td>
<td>0.212</td>
<td>0.662</td>
<td>1.42E + 00</td>
<td>30.7</td>
<td>21.54</td>
</tr>
<tr>
<td>3</td>
<td>Concrete</td>
<td>0.208</td>
<td>0.070</td>
<td>1.45E – 02</td>
<td>0.151</td>
<td>10.44</td>
</tr>
<tr>
<td>4</td>
<td>Granite</td>
<td>0.011</td>
<td>0.485</td>
<td>1.24E – 02</td>
<td>0.003</td>
<td>0.24</td>
</tr>
<tr>
<td>5</td>
<td>Limestone</td>
<td>0.145</td>
<td>1.930</td>
<td>3.78E + 01</td>
<td>18.3</td>
<td>0.48</td>
</tr>
<tr>
<td>6</td>
<td>Dolomite</td>
<td>0.091</td>
<td>0.436</td>
<td>4.09E – 01</td>
<td>9.8E – 02</td>
<td>0.24</td>
</tr>
<tr>
<td>7</td>
<td>Gray chalk</td>
<td>0.320</td>
<td>0.147</td>
<td>1.22E – 01</td>
<td>3.2E – 01</td>
<td>2.63</td>
</tr>
<tr>
<td>8</td>
<td>White chalk</td>
<td>0.346</td>
<td>0.197</td>
<td>2.40E – 01</td>
<td>4.3E – 01</td>
<td>1.80</td>
</tr>
<tr>
<td>9</td>
<td>Berea sandstone</td>
<td>0.248</td>
<td>11.491</td>
<td>6.13E + 02</td>
<td>9.1E + 02</td>
<td>1.48</td>
</tr>
<tr>
<td>10</td>
<td>Indiana sandstone</td>
<td>0.167</td>
<td>8.945</td>
<td>1.92E + 02</td>
<td>1.8E + 02</td>
<td>0.94</td>
</tr>
<tr>
<td>16</td>
<td>Yucca Mt. Tuff</td>
<td>0.096</td>
<td>0.021</td>
<td>7.33E – 04</td>
<td>2.0E – 04</td>
<td>0.27</td>
</tr>
</tbody>
</table>

\(^a\) \( k_{\text{cal}} \): permeability calculated by using KT method;  
\(^b\) \( k_{\text{measure}} \): permeability measured by \( N_2 \) (Klinkenberg effect has been taken into account).

according to equation (2), and \( V_t \) can be obtained directly from the cumulative intrusion curve, which is 0.0875 mL g\(^{-1}\). The next step is to obtain \( L_{\text{max}} \) and \( V_{L_{\text{max}}} \). \( V_t \) is subtracted from each cumulative intrusion volume \( V_i \) at each pressure in the dataset from \( P_t \) to the maximum pressure. Then the net volume \( (V_i – V_t) \) times the diameter-cubed for the corresponding pressure is plotted as a function of pore-throat diameter. As shown in figure 3, the pore-throat diameter of 0.349 \( \mu m \) corresponding to the maximum \( y \)-value is \( L_{\text{max}} \) and the cumulative intrusion volume corresponding to this diameter is denoted as \( V_{L_{\text{max}}} \), which is 0.149 mL g\(^{-1}\). The total intrusion volume \( (V_{\text{tot}}) \) for the sample of white chalk is 0.237 mL g\(^{-1}\). According to the definition of \( S(L_{\text{max}}) \), \( (V_{L_{\text{max}}})/(V_{\text{tot}}) \) can be calculated as 0.629.

The porosity (\( \phi \)) of white chalk is 0.346, which can be directly obtained from the MIP measurement. Until now all
Figure 3. Determination of $L_{\text{max}}$ in white chalk.

Figure 4. Log $k_{\text{cal}}$ versus log $r_{50}$ for all 18 samples in table 1.

the required parameters in equation (3) are known, and we can calculate the permeability of white chalk to be 0.239 mD. All the other samples were processed to obtain the permeability by the same procedures described above.

5. Results

5.1. Reproducibility of MIP tests

We perform triplicate MIP measurements on Berea sandstone and Barnett Shale no. 3 to evaluate the repeatability of MIP. Ideally, a repeatability test should be carried out on the same piece of sample. However, this is not possible in the case of MIP test because the sample is contaminated with mercury after MIP test. As a result, we choose three representative samples from the same rock block to check the repeatability of MIP. For Berea sandstone, we obtain the porosity as 22.86 ± 1.72% and $r_{50}$ as 11.89 ± 0.44 μm; for Barnett Shale no. 3, the porosity is 5.29 ± 0.59% and $r_{50}$ is 0.0031 ± 0.0002 μm. Considering the inherent heterogeneity of natural rock, this result is acceptable and the repeatability of MIP is verified.

5.2. Comparison between $k_{\text{cal}}$ and $k_{\text{measure}}$ (permeability measured by $N_2$)

Using regression analyses, Rezaee et al (2006) obtained a set of relationships between permeability, porosity and pore-throat size for 144 carbonate samples. They found that the following equation has the highest correlation coefficient between measured and calculated permeability:

$$\log k = -1.160 + 1.780 \log f + 0.930 \log r_{50}$$

where $k$ is the air permeability (mD); $\phi$ is porosity (%); $r_{50}$ is the median pore-throat radius corresponding to 50% mercury saturation (μm).

The new empirical equation (Gao–Hu equation in this paper), by plotting $\log k_{\text{cal}}$ (permeability calculated by using KT method) versus $\log r_{50}$ (as shown in figure 4), is obtained as follows:

$$\log k = 2.225 \log r_{50} + 0.214.$$
y = 2.055x + 0.499
R² = 0.723 (N=10)

Figure 5. Log \( k_{\text{measure}} \) versus log \( r_{50} \) for ten samples in table 2.

materials (concrete and red brick), there is only a slight difference between \( k_{\text{cal}} \) and \( k_{\text{measure}} \) for all other natural rocks. This is encouraging, considering the difficulty of permeability measurement, for difference within 1–2 orders of magnitude obtained by different approaches for the same sample is not uncommon. As a result, the KT method is reliable to calculate permeability and selection of \( k_{\text{cal}} \) as the sample permeability to derive the new equation is appropriate.

We also plot \( k_{\text{measure}} \) versus log \( r_{50} \), and the fitted equation and the \( R^2 \) value are shown in figure 5. Although the \( R^2 \) value is not high (0.723), it indeed indicates a relationship exists between permeability and \( r_{50} \).

5.3. Comparison between Gao–Hu equation and Rezaee equation

We derive a linear relationship between log \( k_{\text{cal}} \) and log \( r_{50} \), which is called the Gao–Hu equation in this paper. The comparison between the Gao–Hu and Rezaee equations is shown in figure 6, where \( k_{\text{cal}} \) is used as the sample permeability to compare the utility of these two equations. The permeability calculated using the Gao–Hu equation exhibits a better agreement than the Rezaee equation. All the permeability values calculated using the Gao–Hu equation are closely distributed along the \( y = x \) line in figure 6, while the permeabilities calculated using Rezaee equation deviate somewhat from the \( y = x \) line and this phenomenon becomes more obvious for samples with a low permeability. Rezaee et al (2006) pointed out that their equation has a good outcome when it is applied to carbonates and we can arrive at the same conclusion from figure 6 in which carbonates have been differentiated from other samples by using different symbols. As a result, both the Gao–Hu equation and the Rezaee equation are applicable to carbonates, while the Rezaee equation becomes less valid when dealing with tight samples like shale compared with the Gao–Hu equation.

In order to draw a more reliable conclusion, we compare the \( k_{\text{measure}} \) with permeability calculated by the Gao–Hu
equation and the Rezaee equation for eight natural rocks; the results are shown in figure 7. We add trend lines to each group of permeabilities, and the equations of the trend lines and the $R^2$ values together with the number of samples ($N$) are given in figure 7. Again, the permeability values calculated using the Gao–Hu equation are closely distributed along the $y = x$ line, which verifies the validity of the Gao–Hu equation.

6. Conclusions

This study presents a new equation which solely uses the median pore-throat radius to estimate the permeability of porous consolidated media. Compared with the existing relationships such as Rezaee equation, the advantages of our new equation include the simple form, high reliability and wide applicability. Nowadays, more and more unconventional reservoirs with relatively low permeability are investigated and explored, which makes the new equation have a more realistic meaning. Our results also show that the KT method is reliable to obtain permeability from MIP data, from the good agreement with the measured permeability using N2.

The reason that porosity could be ignored in our new equation may be that the effect of porosity on permeability is negligible, compared with the effect of median pore-throat radius. The sole use of median pore-throat radius to predict permeability indicates that $r_{50}$ may control the fluid flow in porous media.

For consolidated porous materials (rocks) this work is the first to only relate permeability to median pore-throat radius without considering other parameters. The samples we use here also have a wide range of permeability, from sandstone ($\sim 10^3$ mD) to shale ($\sim 10^{-6}$ mD), which makes the new method more applicable.

Although MIP has been developed for a long time, there are still some issues related to its application (El-Dieb and Hooton 1994, Diamond 2000). Hysteresis phenomenon (non-reversibility between mercury intrusion and extrusion curves) and different contact angles between mercury and different porous media have attracted a lot of researchers’ attention (Moro and Böhni 2002, Zhou et al 2010). In this paper, we used the median pore-throat radius from the mercury intrusion curve; the new equation may be improved as more data with rocks are collected in the future.

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