Permeability Prediction of Sandstones Based on Mercury Intrusion Method

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Abstract. The use of the mercury intrusion method has been one of the most relevant trends in determining the permeability of porous media in the past decades. In this paper, general knowledge of sandstone reservoir evaluation is delineated including the pore distribution of sandstones and air permeability measurement. Based upon the paradigmatic study conducted by Purcell, a schematic diagram illustrating apparatus used in mercury intrusion is shown and introduced, and the relevant procedure is also outlined. Four significant permeability prediction models are described respectively and compared based on researches focusing on tight rocks. By doing so, this article reveals that the performance of the models is different despite the painstaking analysis and the significance of these studies. The contribution of this present study is providing a general reference of permeability prediction by mercury intrusion method as well as its previous momentous studies, giving a comparison among the given models.

Keywords: Sandstone, Permeability, Mercury intrusion method.

1. Introduction

With the rapid development of the oil industry, researches on oil reservoirs have become one of the major scientific fields. Over the past decades, various well-established methods evaluating permeability and hydraulic conductivity of porous rocks have been lucidly discussed earlier [1-3]. Meanwhile, the mercury intrusion method was applied in various aspects of rock observation because of its efficiency and relatively cheaper expenses [4-7].

To increase the accuracy as well as the efficiency of fluid permeability measurement in sandstone, which is one of the major reservoirs containing oil resources, more and more studies evaluating the permeability of the rock based on the method of mercury intrusion began to emerge. Mercury injection technique, also known as mercury porosimetry experiment, is well-known for its simplicity and power as a characterization tool [8]. It is also one of the essential methods that are widely used to accurately calculate pore size distribution, which is a critical factor in reservoir evaluation. Purcell first combined air permeability of drilling cuttings with mercury capillary data for a mathematics model, a brief introduction of the apparatus that is used for the determination of mercury capillary pressures is also concluded in his research [9]. It was then followed by a great number of works in which improved the given equation by considering different factors such as types of sandstone reservoirs, sedimentary environments, and size of the samples, etc. [10-14].

This paper aims to first delineate fundamental knowledge of sandstone reservoir as well as its pore size distribution, then give the background of some suggested methodologies in the measurement and
evaluation of porosity and permeability, after which an apparatus is described whereby the data for capillary pressure curves are measured. Based on studies conducted before, several mathematics equations between pore size distribution and permeability will be listed and described. This article focuses on the significance and highlight of these scientific researches, by listing and comparing the academic achievements. This paper is purposed to provide a general reference of some noticeable achievements in this field, providing a brief introduction of mercury intrusion as well as permeability calculation advancements.

2. Methodology

2.1. Sandstone pore distribution
There are four types of sandstone pore space: Inter-granular pores, dissolution pores, micro-pores, and fractures, where fractures can co-occur with any other pore space. The first three types, however, are related to the structure of the rock. After sedimentation, sandstones initially have inter-granular pores with large sizes and generally have good permeability. However, the permeability of the rocks does not become better with the increase in the number of dissolution pores, but depending on the degree of connectivity of the pore throats, when the pores are connected, the permeability of the rock is good. Sandstone containing more clay minerals will then have many micro-pores, high specific surface area, small pore size, low permeability, and high water content saturation. Fractures make up a small proportion of the rock, but they can increase the permeability of rocks. The first three pore types are related to the structure of the rock and the pores can change with diagenesis processes [15].

2.2. Air permeability measurement
The gas permeability method was one of the early methods of direct laboratory measurement of permeability, where core samples were held stationary by an instrument, and under certain temperature and pressure conditions, air or nitrogen was introduced to measure the difference in air pressure and gas flow at the inlet and outlet ends of the rock sample to calculate the permeability. Zhao claims that early methods of direct measurement of permeability were based on Darcy's Law [16, 17], until Kundt and Warburg first proposed the gas slip effect [18], which is also known as the Klinkenberg slip theory, i.e. that the gas transport at the rock wall is not negligible and that this slip phenomenon leads to a large actual flow rate at the outlet, and that when using liquids for permeability measurements, there is often a physical and chemical reaction between the clay minerals and the fluids [19]. The experimental conditions are harsh, with problems such as fluids moves at an uneven rate in the pores of the core sample (the schematic diagram is shown in Figure 1).

As the inability of the liquid to be injected into the core sample at low pressure, the early scholars mostly used gas to obtain the permeability of the samples more accurately.
Laboratory methods of measuring permeability are divided into steady-state and non-steady-state methods. The steady-state method is based on Darcy’s law and correlated by a Klinlenberg-correction equation [16]. This method has a long measurement period and the test conditions are temperature dependent, which is prone to errors, so the test conditions are relatively harsh. Brace proposed the pulse decay method based on the non-steady-state percolation theory, which dispenses with the need to record the pressure difference between the inlet and outlet end faces compared to the steady-state method, and instead uses a relatively mature technique to record the parameters associated with the applied pressure pulse, improving the efficiency of the experiment [20].

In addition, to address the additional microfractures generated during coring, Luffel proposed the core grinding method, which eliminates the effect of microfractures by grinding the core [21]. However, Cui argued that Luffel’s method destroyed the sample and he proposed the ISSP method, which is characterized by simulating the conditions of the formation [22].

Although many of these non-steady-state methods have been widely discussed and studied, there are still efforts to improve the steady-state method, and Li found that the slip effect could be attenuated by applying a tail pressure, but the slip-free permeability obtained by this method deviated from that obtained by applying the Kerr equation correction, and many studies have since improved the method based on the steady-state method [23-25].

However, Zhao argues that the limitation of the methods above cannot be brushed aside [16]. Initially, despite the inaccuracy claim made by scholars, the steady-state methods are still based on the slip theory [26, 27]. Secondly, the non-steady-state method usually requires a change in the grain size or structure of rock samples. Moreover, the apparatus used still needs advancement in accuracy as well as technology.

2.3. Theory
Principle: Mercury with a contact angle greater than 90° cannot enter the pores of the sample at normal atmospheric pressure, while under external pressure it can overcome the resistance caused by the surface tension of the mercury. The higher the external pressure, the smaller the pore size in the sample occupied by the mercury [28].

However, the mercury intrusion method is not suitable for all kinds of samples, Li Yang et al. claimed that the mercury injection method enjoys its benefits when characterizing the macro-porous structures (>50 nm), while it is better to apply different methods such as carbon dioxide adsorption and low-temperature nitrogen adsorption for characterization of micro-pores (<2 nm) and mesopores (2-50 nm), respectively [29].

Besides, Deng Yu et al. used the mercury injection method and logging curves to make a comprehensive quantitative evaluation of the pore structure of sandstones and conglomerates, and the experiments demonstrated the applicability of the combined mercury-pressure and logging evaluation system in sandstones and conglomerates [30].

2.4. The apparatus of mercury injection method
An apparatus that is suitable for using the mercury intrusion method to determine the capillary pressure of porous media is shown in Figure. 2.
Being carried out by Purcell, figure 1. illustrates the main equipment that is used in Purcell’s capillary pressure determining process [9]. The apparatus contains a mercury displacement pump A, a sample holder B, a manifold system C, which is shown schematically, and equipment D, which measures the volume of the liquid from the pump. As it is shown in detail, several essential components in equipment B are as follows: At the top and the bottom of the sample holder are two lucite windows, the sample is held in the chamber E in the middle part of equipment B, between the two windows. The reference marks, F, are marked at approximately the midway point of the two lucite windows, on the conduit G connecting the pump, the sample holder system B, and the manifold system C.

Due to the complexity of the manifold system, it is demonstrated schematically as it is connected to a vacuum system which is isolated from the manifold system, a pressure gouge at a scale of 0-200 psi, and another gauge that is capable to measure the pressure ranging from 0 to 2000 psi, so the system can measure various of gas pressure ranging from high vacuum to around 2000 psi. To this manifold are also connected a high-pressure nitrogen bottle, a U-Tube manometer, and a tube to the atmosphere.

As for the measurement, the decline of the mercury-gas interface from the upper reference line F indicates the entrance parameter of the mercury, while the degree of penetration determines to control the mercury meniscus back to its original reference mark. And the whole recession-return procedure contributes to the determination of the amount of mercury that is injected into the porous media under a sequence of different pressures.

According to Fu Xin, the steps are as follows: 1. Open nitrogen valve, mercury piezometer; 2. Take a sample, select an appropriate sample tube to encapsulate the sample, apply sealing grease, and weigh; 3 [31]. Place sample in a low-pressure chamber, load measuring cylinder; 4. Carry out low-pressure analysis, remove sample tube at end and weigh again and record; 5. Start high-pressure
analysis, sample tube fed upwards into the sample chamber and drop together, transfer sample tube to the high-pressure chamber after reaching halfway; drop sample chamber; 6. When finished, release the exhaust valve, spin out the sample chamber and later remove the sample tube and clean, wipe and dry it to end the experiment.

As reported by Liu et al., during the procedure of mercury intrusion, samples are taken, the different samples obtained are dried, the temperature is controlled at 80-90 °C, a suitable expansion gauge is selected and weighed for mass, the gauge is closed and a series of other steps are carried out, the sample is evacuated in a low-pressure area to below 6.65 Pa for sample testing; thereafter the sample is removed from the low-pressure station and weighed to the high-pressure station for testing [28].

The mercury intrusion method is only valid for pore studies within a certain size range of pores, generally large and some medium pores. Liu et al. used the fractal dimension as a parameter measuring the compressibility during the given experiment, and defined D as different fractal dimensions (D) at different pressure intervals [28]. The different stages of the pressurization process divided the testing stages of the mercury compression method into three stages (forward mercury stage, incoming mercury stage, and backward mercury stage), and correspondingly three different D values (D1, D2, D3) could be calculated where D1 characterized the stage at which the mercury has not yet occupied the inner pores of the sample grains in the advancing mercury phase, the D2 value reflects the width of the pressure interval (the harder the sample is to compress, the larger the D2 value, i.e. the larger the pressure interval for mercury compression). So it can be seen that the D2 value stands for the most effective and accurate stage in the mercury compression experiment; D3, as quantitative data, yet qualitatively expresses the compressibility of the sample: the harder the sample is, the less easily the coal is compressed, and thus the smaller the D3 value is.

The following should be noted: 1. Distinguish the difference between inter-granular pores and intra-granular pores. It is generally accepted that as the pressure increases, mercury occupies the inter-granular firstly and intra-granular pores secondly, and that the pressure threshold contained in this rule is related to the size of the sample particles and the way in which they are stacked. 2. The compressibility of the sample. It can also be caused by the compression of the coal itself [27]. These two points again demonstrate the limitations of the mercury compression method in studying the pore size of the sample.

3. Models

3.1. Purcell Model

As the nature of capillary pressure that has been lucidly carried out by Leverett, Hassker, etc., considerable attention has been directed towards the estimation of the percentage of irreducible water none of the other application had been studied [5-7,32,33]. Purcell first correlated gas permeability and capillary pressure data based on the researches on the role that capillary pressure plays in reservoirs [8]. The project lays out and justifies the equation showing the relationship between the permeability of porous medium and capillary pressure curve, the equation is shown as equation (1):

$$K = 0.66Ff \int_0^{\rho_{100}} \frac{d\rho}{(P_c)^2}$$

In which K is the permeability of porous media in millidarcies, f is percent porosity, ρ is the proportion of liquid-occupied pore space, and Pc is the capillary pressure.

An experimental procedure that enables this experiment to be done accurately is also described.

The main highlight of the project is that it fills the gap in the prediction method for small and irregular samples (e.g. drilling cuttings), is faster (in hours, compared to days in most cases), and requires a reduced number of experiments.
3.2. Thomeer Model and Swanson Model
Using the 'log-log plot', which is the logarithmic curve of capillary pressure found that the fitted curve pattern was close to a hyperbola and established the relevant mathematical expression [11,12]. The shape of the hyperbola is related to the pore geometry. Empirically, he formed a mathematical correlation between permeability and the hyperbolic function, and this method made a significant improvement over the earlier methods e.g. Thomeer mainly studied the mathematical expression relating to capillary pressure curves, but thanks to his achievement, Swanson enhanced the capability of estimating permeability in small rock samples such as sidewall core and drill cuttings by developing a nomograph that permits direct estimation of permeability based on mercury intrusion data [11]. An equation between \((\frac{S_b}{pc})_{max}\) and permeability is also developed, in which \(S_b\) is the saturation of mercury in the rock sample and \(pc\) is the threshold capillary pressure of the hyperbola [1, 34]. Efforts have been taken to revise the mathematical expression given by Swanson by Li Jianming, who used the parameter of \((\frac{S_b}{pc^2})_{max}\) that is more sensitive to the change/variation of permeability to replace \((\frac{S_b}{pc})_{max}\) in Swanson’s study [34].

3.3. Pittman Model
Considering the expensive expenditure of mercury intrusion, Pittman empirically carried out a series of equations to calculate pore aperture radii corresponding to the percentiles ranging from 10th to 75th of mercury saturation, establishing a relating diagram using the given data of porosity and permeability [13]. In the study, the pore throat radius corresponding to this maximum value is called \(r_{apex}\), i.e. the inflection point, and Pittman’s model is proposed. Since the saturation of the non-wetting phase and the capillary pressure reach a maximum at the inflection point, this point is chosen to establish the relationship parameter between capillary pressure and permeability, and the parameter is regarded as a sign that the fluid in the non-wetting phase flows from large pores with large porosity and good connectivity to small pores with small porosity and poor connectivity.

The highest correlation coefficient was measured when the Hg saturation was 25%, and the empirical formula (2) was obtained by analysis:

\[
\log r_{apex} = -0.117 + 0.475 \log k - 0.099 \log \Phi \tag{2}
\]

where \(k\) is the permeability in \(mD\), \(r_{apex}\) is the maximum value of the radius corresponding to the apex in \(\mu m\), and \(\Phi\) is the porosity in percent, the equation (3) is another form in terms of \(k\):

\[
k = 4.6042 r_{apex}^{2.1503} \Phi^{0.2084} \tag{3}
\]

Berg, who recognized the scarce of mercury intrusion test data as a limitation, developed a former method to approximately calculate dominant pore size by estimating grain size initially, whereas that method was also challenged by Pittman’s approach that avoids the estimation of particle size [35].

3.4. Rezaee Model
Rezaee gave a model for the relationship between permeability and pore throat radius based on the data from mercury intrusion and NMR [10, 34]. Rezaee added a mathematical model of porosity and pore throat radius in dense sandstone reservoirs, which are characterized by weak permeability. The authors argue that the quantification of permeability is an important factor in the economic assessment of resources in a reservoir, allowing for a complex interrelationship between porosity, connectivity between pores, grain size, diagenesis, and other factors.

The structural characteristics of ordinary rocks provide a good reference for some of the petrophysical factors and can be used to directly pre-evaluate the reservoir by using the sedimentary environment and the control of permeability by the depositional process; however, the case of dense sandstones is extreme and the evaluation methods and equipment used in ordinary reservoirs do not apply to dense sandstones. Using the Rezaee method of mercury pressure, the radius \((r_{10})\) of the dense sandstone reservoir at a mercury saturation of 10% best correlates with permeability: in dense sandstone reservoirs, micropores, and intergranular mesopores have a strong controlling effect on permeability.
By Winland and Pittman's study, $r_{35}$ and $r_{25}$ are proposed as the most suitable radii for permeability prediction respectively [13, 14].

The difference between the above three radii is mainly since the pore structure is complex, e.g. in this experiment, 10% saturation corresponds to a grain size of 0.1-1 µm, and at the same 10% saturation, the radius of ordinary sandstone must be >0.1-1 µm, which is not a good representation of the grain size controlling permeability. In contrast, the pore size of a dense sandstone reservoir at 35% saturation is nanometre-scale and provides minimal control of permeability.

4. Model comparison

Indeed, the studies above all enjoy significant achievements, whereas many scholars started to analyze the limitation of these models. One initial drawback that is commonly claimed is that the models are only suitable to conventional clastic rocks with high porosity as well as permeability, none of the models have been established aiming to accurately measure samples whose permeability are in nanodarcy or micro-darcy scale e.g. tight rocks [8, 36, 37].

To compensate, Nooruddin used 225 carbonate rock samples from the Jurassic period to compare several permeability-prediction models based on high-pressure mercury intrusion data [8]. A parameter is defined whose figure plays a role as the accuracy index, based on that result, the Swanson model and the Winland model performed comparatively well, followed by that of Thomeer and Pittman, whose average accuracy index is half as much as the best two models [12, 14, 38]. Nooruddin also mentions that the use of uncorrected air permeability makes the influence of the slip phenomenon apparent in low-permeability samples, which is considered to be another demerit that is shared by models of Swanson, Winland and Pittman.

Tang also addresses that the models, derived from single pore radius e.g. Swanson model, perform badly when measuring permeability in carbonate samples as the texture and structure of carbonates are more complex than clastic rocks, especially sandstones [12, 36, 37]. In comparison, the Purcell model compensates for the effect of mercury intrusion parameter, porosity, and lithology [9]. Likewise, the Thomeer model is also well-applied in the well-logging evaluation of carbonate formation in the mid-east according to Tang [11, 37].

Using the measured porosity and permeability of limestone samples from the Kurdistan region of Iraq, Rashid compared 16 different permeability prediction methods [36]. The study claims that the Swanson model performed very badly in predicting the permeability of tight carbonate rock, while it also lists that poiseuille-based models are expected to provide more accurate permeability due to the more fitting parameters and the specificity of these methods. However, most of the methods in the latter ones only perform well in certain types of rocks, as they are prone to give much less accurate permeability data when evaluating the rocks that they were not calibrated, though Winland method and Pittman model performed creditably when asked to predict the permeability of tight carbonate rocks. Rashid concluded that most of the methods he studied are designed and calibrated mostly for high porosity and permeability types of rocks, some of them even not been Klinkenberg-corrected. Apart from that, carbonate rocks like limestone are different in pore microstructure and pore connectedness as they are likely to be affected by post-depositional diagenesis and become more complex compared to conventional clastic rocks. Hence, as Rashid addresses, these factors cause bad performance in the evaluation of permeability in carbonate rocks.

5. Conclusion

The mercury intrusion method is widely used as a major strategy of determining the pore distribution as well as predicting permeability in porous rocks due to its simplicity and reasonable expenses, and thus is considered to be one of the most efficacious ways of reservoir evaluation as well as irreducible water determination.

All of the models mentioned above are based on painstaking research and careful calculation, while their performance differs from one model to another. Purcell model, the earliest prototype in the research of mercury intrusion, is calibrated and applied in the following studies. Although being
accepted by Nooruddin, the Swanson model is claimed to perform negatively in Rashid’s study. Moreover, Thomeer’s model, Winland method, and Pittman’s model are well-accepted by the three scholars above.

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