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COMPARATIVE STUDY OF PERMEABILITY IN SANDSTONES AND SHALES-USING MERCURY INJECTION CAPILLARY PRESSURE (MICP) TECHNIQUE

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ABSTRACT

The various types of pores distributions and pore throats in shale and sandstone samples could be easily investigated and evaluated by Mercury Injection Capillary Pressure (MICP) techniques. The focus of this research was to figure out the pore size distribution, particularly in correlation with permeability, from MICP measurements. In this project, the rock samples were taken from Sarawak Basin in Malaysia, in the form of chips. In general, MICP profiling is very dependent on simple size due to both conformance and accessibility of pores. Due to conformance, a correction approach was applied in which we considered the samples' pore volume compression before it was intruded into samples. Mercury injection is used to help in determining the numerous properties of tight shale storage, because there are instances where cuttings from a drilled formation may be limited, and the source rock needed may not be available to be evaluated unless a whole core is taken. Core profiles and cuttings may seem like more plausible choices in calibrating well logs. The distribution of pores and their identification in a single sample may provide the required information on the permeability, porosity and bulk volume of the formation. It could be seen in the results that MICP is highly applicable to capture and figure out the pores size distribution and density in a shale and sandstone sample. From that point of view, the transportation of the gas from the source rock, through the fractures flow paths, into production wells could be understood. Therefore, MICP is a recommended method in analyzing the formation's permeability via its pore size distribution. However, the objective of this study could not be fully envisaged as it has been observed that there are several limitations when it comes to studying shale gas, and these limitations are not only from the unavailability of equipment but also the problem about the number of samples available.

Keywords: sandstone; shale; pore size distribution; permeability.

INTRODUCTION

Initially, resources which were recovered from reservoirs with difficult nature have been labeled as 'unconventional resources' (Haskett and Brown, 2005). Unconventional plays not only include fractured reservoirs, but also coal-bed methane (CBM), tight gas, oil sands and gas / oil shale. There are instances in which the depositional model or reservoir character has caused assessment difficult since the commercial gas and oil reservoirs exploration and production. When standard probabilistic methodology was applied, it could be seen that the production and volumetric assessment of these unconventional reservoirs were causing huge problems, and the evaluations often dependent on deterministic shortcuts of rules of thumbs.

The most abundant known sedimentary rock on Earth is shale. It is widely acknowledged that shale would accommodate large amounts of hydrocarbons, which hinder profitable extraction due to its insufficient geological resource delineation, lack of flow rates; due to the shale formation's low permeability when practiced with conventional gas production technology, and high finding with high development costs. US natural gas supplies has been enhanced by shale gas production for years even if it is at very low rates, ranging from 1% to 2%, of its total supply up until the Millennium turn. Through the huge-scale shale gas extraction from Texas' Barnett play during the early 21st century, shale gas production has gone through vital change when the up and raising US wellhead gas prices mentioned was stimulated (Rogner and Weijermars, 2014).

Going further into shale gas as a formation, in contrast to several types of sedimentary rocks containing deposits of natural gas, examples such as sandstone or limestone, shale rocks usually have very low permeability which makes gas production a far more costly, challenging, and complex process. 'Tight gas' that comes with sandstones, or 'low permeability' with 'coal bed methane' limestone, shale gas is considered as 'unconventional gas'. Both unconventional and conventional deposits do host natural gas, further production methods are applied for better understanding if the formation is of unconventional or conventional deposits.

Moreover, shale has come into attention of the oil and gas industry due to its increasing nanoscale pores ability which is able to contain abundant amount of hydrocarbon. It has now become globally focused in the field of exploration and development in unconventional gas and oil studies.

The study's motivation is based upon the understanding and investigation of hydraulic fracturing on the low permeability of potential tight and shale gas reservoirs, mainly on a focused scale is on its pore throat size. One of the example while looking at it on an international scale is that, back in 2014, hydraulic fracturing process has been used for over a decade in Europe as an industrial process to stimulate geothermal

wells, water wells, and conventional oil and gas wells, although it is still in its early phases of fracturing wells and horizontal drilling for shale gas extraction.

Releasing natural gas and oil from deep beneath the Earth surface is the process of hydraulic fracturing. Back in the 1940s, hydraulic fracturing has been used to stimulate gas and oil wells, increase production rates and geothermal energy recovery. It is mostly crucial in shale gas development. Since these hydrocarbons are trapped within rock formations from low to very low permeability, the releasing process of shale gas reservoirs could only be done by stimulating the shale formation through formation via hydraulic fracturing. Extraction is maximized when fracking process is combined with horizontal drilling by allowing numerous fractures along the shale bed which in turn allows the gas to be extracted commercially. This is when the information on the pore throat size of the formation is important. It would be able to decide if hydraulic fracturing is either necessary or not.

The study aims to help in making decision on whether hydraulic fracturing or other necessary steps should be practiced producing formation on potential tight and shale gas with low to ultra-low permeability. To be able to do so, studying the formation is utmost important, while applying the selected method which is the mercury intrusion capillary pressure method and making final decision on if hydraulic fracturing should be carried out for the conventional / unconventional formation. Therefore, the objectives of this study are to:

a) Investigate further on ultra-low permeability formation.

b) Study its roles in the reason of low permeability.

c) Investigate the issues concerning experiments for shale.

The scope of the study is on the mercury intrusion capillary pressure test and its relation to ultra-low permeability on shale gas. This study also focuses on the understanding of pore throat size and its relation to hydraulic fracturing. Throughout the experiment that will be applied, information could be extracted and would be of use to the production process.

BACKGROUND

Shale Gas Reservoirs

Shale has low pore connectivity with relatively ultra-tight rock, and exceedingly low permeability of 10-18 to 10-21 m^2. If hydraulic fracturing is applied to the formation, natural fractures would enable the formation to connect for it would break up the shale matrix, so that follow paths are constructed for the improvement on the reservoir's connectivity. Once the free gas is released along the fractures created hydraulically, the matrix-stored gas could also transport slowly to the connecting pores. In the end, absorption begins to occur for the absorbed gas in the pore surfaces because of the pressure gradient's reduction. Hence, both the matrix gas flow pattern and gas storage in the shale matrix are crucial for past production prediction.

The ranges of a shale matrix's pore size are from several hundred nanometers to several nanometers. To mention that shale matrix's pore size range is small, comparably, pore throat sizes are smaller, and much closer to the free path of the mean gas molecule. Henceforth, viscous flow does not govern the nano-scaled gas channels gas flow, in which the drag force along gas molecular and the rough collision of the pore surface with its pore wall become important. The main flow regimes in a basic shale reservoir are slip and transition flows. Applying Knudsen number (K_n) expressed as:

$$K_n = \lambda / r \quad (1)$$

Where;
λ = ration between gas molecular mean free path
r = gas flow characteristic size in porous media

Per the gas molecular dynamics theory, the Knudsen number considers on the frequency of both molecule-molecule and molecule-wall collisions. Therefore, gas flow model should have the combination of both slip flow and Knudsen diffusion. A true molecular simulation should also be done by using Lattice Boltzmann Method (LBM) or Molecular Dynamics (MD). In this case, the preferable approach would be LBM.

A certain study has mentioned that Chen *et al.* (2015), by using the Monte Carlo (MCMC) method, has simulated the three-dimensional (3D) nanoscale porous structures on shale; which is a reconstruction method that is based off the SEM images of shale samples. Subsequently, the nanoscale gas flow could be simulated by combining it with LBM. This approach is seen to be computationally intensive although being quite fundamental. With adequate accuracy but simpler methods, using typical shale reservoir data could be proposed in a research.

Pore scaled simulation process is a crucial way to interpret micro / nano scale of the fluid flow. Permeability tests would consume time since the shale matrix is of low connectivity, porous media. Thus, a practical way to attain shale reservoirs' basic hydraulic parameters may be provided by pore network models, to enable the prediction of reservoir gas production and understanding shale gas migration.

For this study, the proposed network model is the mathematical micro / meso / nano-pore, in which the coordination numbers that would satisfy the typical shale reservoir data of pore size and pore throat size. In this pore network, there exist clusters and isolated pores. Other than that, the statistics law needs to be abided by the pore-network models, then justified by applying a numerical simulation (Zhang *et al*, 2015).

Capillary Pressure Test

In the method of determining the capillary pressure, it is characterized by the known fact that the liquid which "wets" the solid which is one of the fluids present within the pore spaces in the solid. For the "wetting" liquids, the surface forces act on such that the fluid would fill the voids within the solid instantaneously. The forces mentioned would contradict the fluid withdrawal from the pores of the solid. Moreover, there is also another type of system which could as well be considered as a part of the capillary pressure studies. This would involve "non-wetting" fluid, which is mercury, and the porous solid which would form a contact angle of larger than 90° against the solid. The act of the surface forces involve goes against the entrance of liquid into the solid. Pressure should also be applied to the liquid to create penetration on the solid pores.

Fields such as petro-physics and reservoir engineering have interest on how porosity and permeability would relate to pore aperture size distribution and its size in general; the major point is that so permeability could be obtained. By using mercury injection data, the evaluations of the cap rock sealing capacity could also be derived. On the other hand, for water-saturated rock, hydrocarbon entrapment and migration are the results of both the opposing interplay of capillary pressure and buoyancy pressure. When a hydrocarbon filament has been established through the rock pores following the expulsion of a source rock, hydrocarbons would then migrate through the carrier beds. If the pressure needed to enact a connected filament through the biggest interconnected water-saturated pore throats, calculation on vertical hydrocarbon column needed for hydrocarbon migration is possible (Schowalter, 1979). This mentioned pressure is the displacement pressure is one of important information on hydrocarbon entrapment and migration.

The mercury injection test could determine the displacement pressure that corresponds to the pore aperture size. But due to lack of core, insufficient core material to permit sampling or more importantly cost considerations, one must know that these would be the issues if mercury injection test is unavailable for practice. Thus, an estimation of displacement pressure should be readily available, from other data such as permeability and porosity, would be useful.

Going further into this study, the Washburn equation assumes the opening of a pore is circular in cross-section and in cylindrical shape. The force that is applied along the line of mercury contact, solid, and mercury vapour is the net force that tends to resist the entry of mercury into pores. Washburn (1921) initially proposed that determining the pore aperture size distribution in porous rocks could be established by the usage of mercury injection as a laboratory method. The mentioned statement could be shown as below, which is also known as Washburn equation:

$$P_c = 2\gamma \cos\theta \, /r \qquad (2)$$

Where;

P_c = capillary pressure

γ = surface tension of mercury (Hg = 480 dynes/cm)

θ = contact angle of mercury in air (140°)

r = radius of pore aperture for cylindrical pores

One of the disadvantages in measuring rock porosity is that the pores are not always cylindrical in shape. The capillary pressure against mercury saturation is basically plotted on semilog or arithmetic plots. This could be the case, albeit the saturation scale at times inverted so that it escalates from right to left. The threshold, displacement and entry pressures are terms that are referred to the initial part of the mercury injection curve. The entry pressure of the mercury injection capillary pressure plot is the point which the curve of the mercury first enters the rock pores (Pittman, 2001).

Pore Size Distribution

Having complex origin with high heterogeneity are the main traits of shale as a tight gas reservoir. At nanoscale level, shale reservoirs have complex geometry that goes from low to ultra-low permeability. Investigations on the pore size and distributions of different shale formations from previous studies have also been done. With a range of 5 - 750 nm organic nanopore distribution, the North America Barnett shale has approximately median size of 100 nm. While the pore size in South China ranges between 2 - 900 nm with its marine shale, the North China has continental shale pore that ranges from 2 - 35 nm. Gas could accumulate since shale is able to provide good spaces from its abundance of nanoscale pores, which is an important role when it comes to gas storage and migration (Li *et al*, 2016).

There would be no certain definition of "pore size" or "pore diameter". Each method of determining the pore size would define the pore size with respect to the pore model that would be best suited to the measured quantity of an experiment. For simplicity measure, an explanation on the void space would be restricted to the "solid" surface that is enclosed by the pore spaces only. There are terms that differentiates the interconnected with relative larger pore spaces and pore spaces which are relatively narrower as shown in Figure-1.

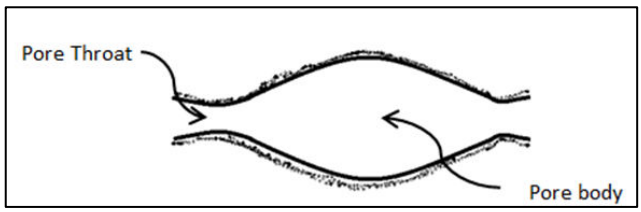

Figure-1. Schematic View of Pore Throat and Pore (Kantzas *et al*, 2016).

The pore throats (pore necks) would be the narrow constrictions, interconnecting relatively bigger spaces. While the pore bodies would be the pore spaces

with relatively bigger pore size. Pore size distribution would be the pore sizes of a large variety of pores. It is also known as the characteristic of the pore sizes on the distribution of the pore volume for its probability density function. In a sense that is pores were separated as objects, then a size would be set by some consistent definition.

In another study done by Comisky *et al* (2011) is by using the MICP test to find out the sample's pore size distribution. The result of this experiment was to acquire the percentage of intrusion of mercury on different distributions of the sample. The figures below (Figure-2, Figure-6) are plots of MICP measurement with various sample size distributions from a single sample.

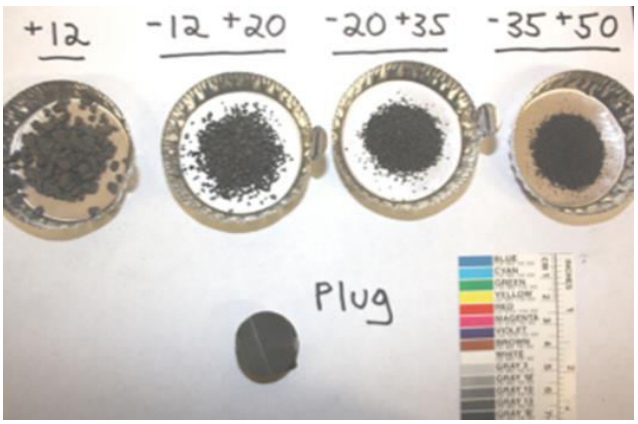

Figure-2. Particle Sizes of the Single Sample (Comisky *et al*, 2011).

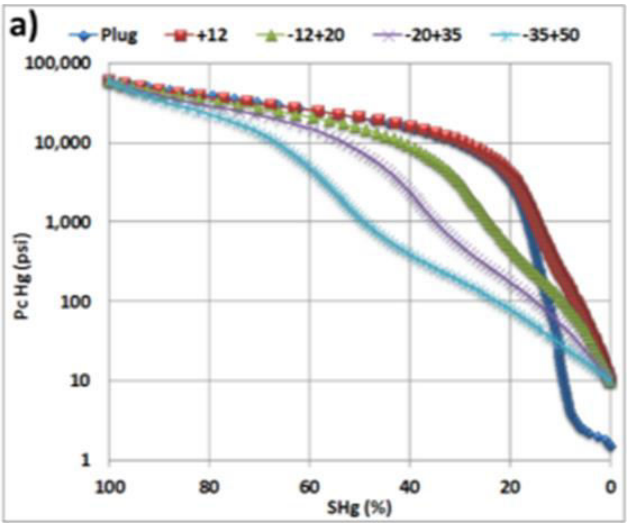

Figure-3. Mercury Intrusion Pressure against Mercury Saturation as Pore Volume Percentage of Plotted Raw MICP Data (Comisky *et al*, 2011).

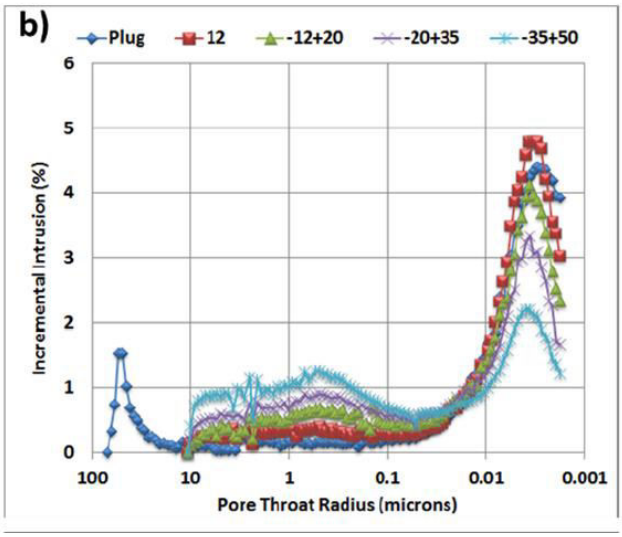

Figure-4. Pore Size Distribution against Incremental of Uncorrected MICP Measurement (Comisky *et al*, 2011).

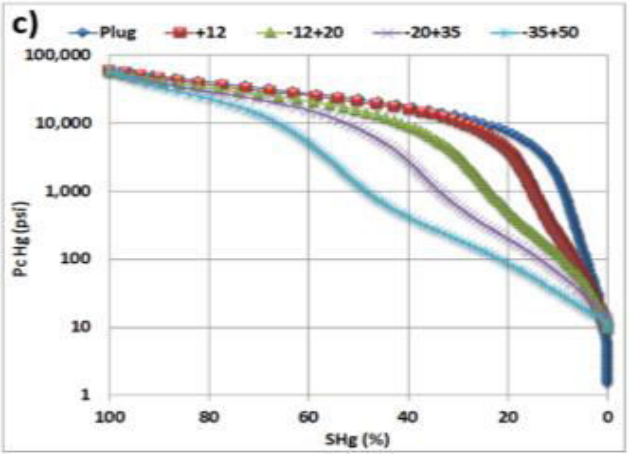

Figure-5. Mercury Intrusion Pressure against Conformance-Corrected Pore Volume Percentage (Comisky *et al*, 2011).

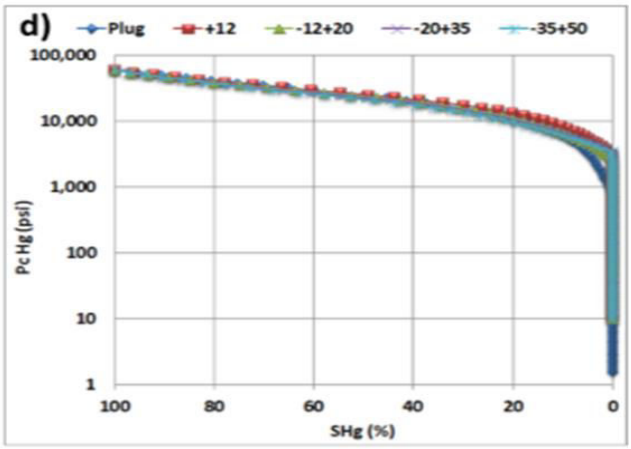

Figure-6. Mercury Intrusion Pressure against Mercury Saturation as Pore Volume Percentage of Plotted Raw MICP Data (Comisky *et al*, 2011).

Later, we would be able to see the major difference between the measured, raw-uncorrected MICP profiles as a function of particle size for any given sample. 0.01 to 0.001 microns are the range for finer pore throat sizes with high pressure portion of the MICP curve while all size classes portray about 0.003 microns for its peak.

One of the basis of Comisky *et al.,* study was measuring and comparing MICP with a few other methods such as FTIR and LPP helium porosity, together with grain density and bulk density profiles upon various sample sizes from the similar relative stratigraphical interval. Two-third butt of a core is taken as a puck, then is cut into a 1-inch core plug. The remaining puck were both crushed and homogenized by using mortar and pestle. Once the crushing process is done, the remaining crushed material would be measured by using series of US standard mesh sizes (12, 20, 35, and 50) were used to break out a few sample size classes of material.

This US standard mesh sizes can be converted and measured in terms of microns (μm). The size distribution for this comparison study ranges from plug to +12, -12+20, -20+35, and -35+50. For all the cases, it would be observed that on any given sample, the MICP porosity would be the largest for the finest particle size range (-30+50) and smallest for the core plug. This is expected of the core plug since it is still in uncrushed form and has been subjected to incomplete intrusion due to its pore size limitation.

The beginning intrusion would be 10 psi. when injection is started from the said value, it is observed that there is not much of a difference between the raw data in Figure-4 and conformance-corrected in Figure-5 curves. This is applied to all the samples except for plug. It is intruded with the approximately given pressure to fill the void between the grains. This is called conformance, for which it is the initial pressure that would finally fill up the sample. Subsequent peaks seen in this graph reflects the filling up process of the mercury in pores of individual grains. As the pressure increases, smaller pores are filled with time. The widening gap of the intrusion and extrusion curves would provide the value of mercury entrapment.

From Figure-4, we can see that at the beginning of the experiment, 10 psi was the beginning pressure to first fracture the sample. As it has been fractured, proper intrusion would then take place and the subsequent peaks would also be the continuous conformance to fill the void in the sample. As the pore throat radius gets smaller, more and more pressure would be needed to fill in the sample, and it is seen that the highest incremental the intrusion percentage could get to be almost 5%. After conformance-corrected value is applied, and the intrusion value has been amended, Figure-6 would be the result. Mercury could only intrude the pore space once it reaches a pressure of about 5 000 psi. The pore throat size is very small, to a point that only a pressure of more than 5 000 psi could finally intrude the sample, as in to prove that permeability in shale is extremely low.

It is in a sense that when the pore throat size decreases, the permeability would also be low. Even in small pore sizes, there are possibilities of having trapped pores with adsorbed gas. Once the MICP test is applied, the fractures would enable the gas to be transported from a high concentration area to a lower concentration area of gas. By figuring out the pore size distribution, the decision on how gas transportation occurs could also be determined as shown in Table-1 below.

Table-1. Pore Size Distribution.

Size Distribution	Transportation
≤ 2nm	Diffusion
> 2nm ≤ 50nm	
≥ 50nm	Darcy

METHODOLOGY

An experimental study is carried out for Liquid Permeability. A permeability study apparatus will be used in this experiment. Before the equipment below is used, the core must be cleaned and flushed, while the BPS equipment must be cleaned to remove any residual liquid / impurities. The core holder is cleaned by using distilled water for about 6 hours. While the core holder is in going through cleaning process, the preparation on brine is started. To prepare the injection liquid of 30 000 ppm brine, 30 g of sodium chloride (NaCl) is stirred with 1 L of distilled water.

Before Operation

a) The machines are made sure to be in a good condition.
b) The equipment is warmed up for one (1) hour before they are used.
c) Safety is prioritized.

Operating the Bench Top Permeability Equipment

a) The equipment and the PC are switched on.
b) All connections are made sure to be properly connected.
c) The core sample is then installed.
d) The overburden pressure on the coreholder is set.
e) The back-pressure regulators are also set.
f) The test is then run.
g) Any leak off measurement is taken.
h) The testing is stopped.
i) The equipment and PC are turned off.

After Operation

a) The machine and the valves (if any) are turned off.

b) Housekeeping is done.

Important Information

- The valves must be kept closed always until the switch is turned on.

- Maximum ΔP is 250, while the coring must be sized at 3-inch length with 1.5-inch diameter and the highest pressure would be 9950 psi.
- The core must be saturated for 6 hours before it goes through the liquid permeability experiment.

RESULTS AND DISCUSSIONS

The main objective of this study is to investigate the ultra-low permeability in shale sample. Due to time limitation, the equipment for shale study in UTP and the availability of the needed sample, it is difficult to obtain the specific results. However, it is decided that permeability measurement shall be done on sandstone samples to prove that it is not an easy task to find permeability in shale.

There are three (3) cores used for this project. These cores are ordered from Barrier and they are composite cores. Out of the three samples used, there are two tight core samples for which they portray low permeability as shown in Table-2 below.

Table-2. Dataset on Three (3) Different Cores.

Core ID	Core K	Core GB2	Core P
Length (cm)	7.45	7.50	7.42
Diameter (cm)	3.75	3.77	3.76
Viscosity (cp)	1.00	1.00	1.00
Flowrate (cc/min)	0.50	0.50 – 1.00	0.50
Permeability (mD)	0.89	111.82	8.85

Results from Core K

For the first core, it is a tight sample and this is the second result for the sample. Core K had a length of 7.45 cm, diameter of 3.75 cm, brine viscosity of 1.00 cp, flowrate set to 0.50 cc/min and the last permeability result before the logging was switched off is at 0.89 mD.

From both Figure-7 and Figure-8, it could be concluded that beginning from 16th minute until 55th minute into the run, the ΔP has remained stable at a 98.11 psi - 96.5 psi range.

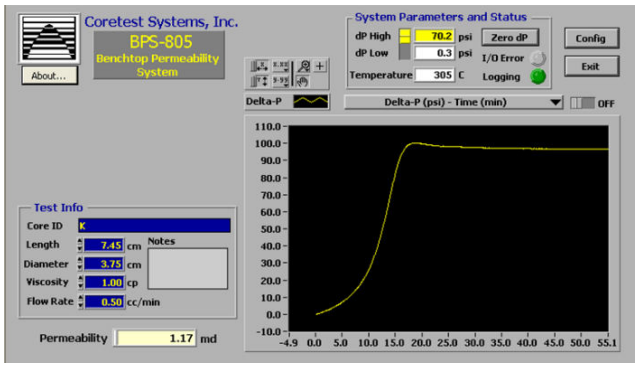

Figure-7. Delta-P (psi) against Time (min) for Core K.

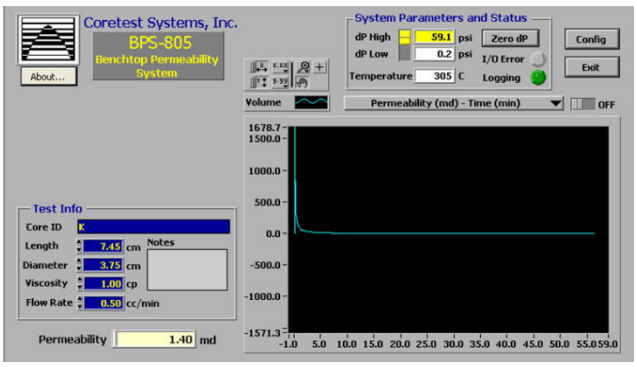

Figure-8. Permeability (mD) against Time (min) for Core K.

When the ΔP has remained stable (straight line), then the permeability reading is taken. In this case, it is 0.89 mD. Since it is a tight sample, the experiment was carried out twice just to confirm its properties and it is proven from the permeability reading.

Results on Core GB2

For the second core, it is not a tight core sample and this is the first result for the sample. In this case, the flowrate for this run is changed once. Core GB2 had a length of 7.50 cm, diameter of 3.77 cm, brine viscosity of 1.00 cp, flowrate set to 0.50 cc/min - 1.00 cc/min and the last permeability result before the logging was switched off is at 111.82 mD.

From both Figure-9 and Figure-10, it could be concluded that beginning from as early as 5th to 165th minute, when the flowrate is at 0.50 cc/min, the ΔP is at the range of 0.90 psi - 1.10 psi.

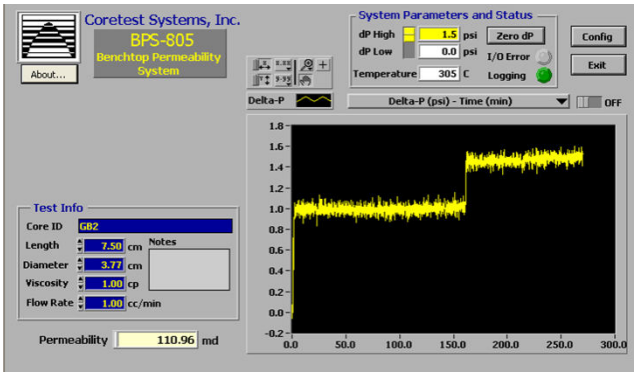

Figure-9. Delta-P (psi) against Time (min) for Core GB2.

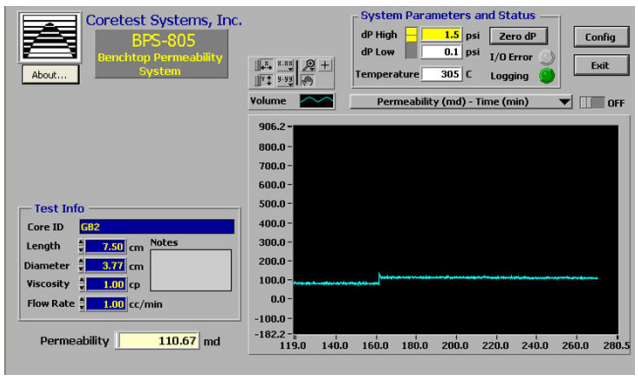

Figure-10. Permeability (mD) against Time (min) for Core GB2.

While the last recorded permeability for this flowrate would be 85.64 mD. When the flowrate is at 1.00 cc/min, the last ΔP recorded would be at a range of 1.40 psi - 1.60 psi with a permeability of 111.82 mD.

Results on Core P

For the third core, it is a tight core sample and this is the final test run result for the samples. In this case, the flowrate is not changed and the test was only done once instead of twice. Core P had a length of 7.42 cm, diameter of 3.76 cm, brine viscosity of 1.00 cp, flowrate set to 0.50 cc/min and the last permeability result before the logging was switched off is at 8.85 mD.

From both Figure-11 and Figure-12 from Core P graph results, the ΔP appears to remain stable once it reaches the 20th minute up until the 120th minute, with the range values of 9.01 - 9.27 psi. As for the last recorded permeability, the value would be 8.85 mD.

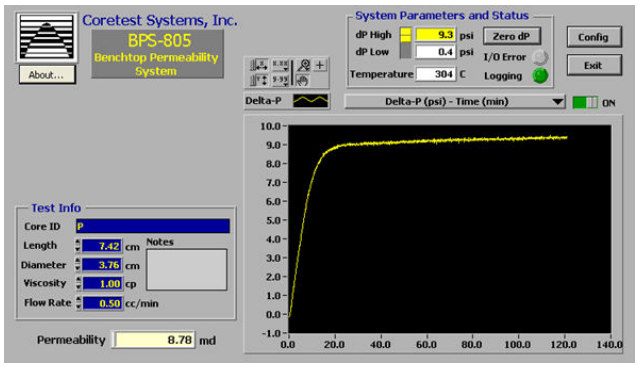

Figure-11. Delta-P (psi) against Time (min) for Core P.

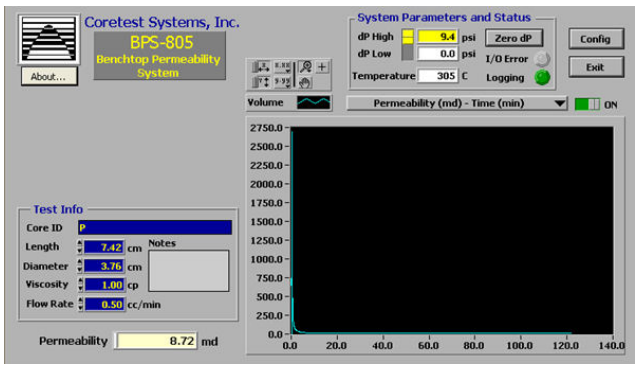

Figure-12. Permeability (mD) against Time (min) for Core P.

CONCLUSIONS

Pore dimension is important in determining factors such as capillary pressure, porosity and permeability. By using the method of mercury injection capillary pressure for porous media such as shale gas reservoirs, mercury is forced to go under pressure into the pores of the chips of the solids tested, it is found that similar results could be seen from the mercury injection method to the porous diagram technique which is practiced figuring out capillary pressure.

In this sense, after figuring out the relation on the pore throat size with its permeability can give a clearer decision making to the necessity of hydraulic fracturing or if there are any other means / systems possible to replace hydraulic fracturing. This method (MICP) is far more favourable compared to the previous methods due to several advantages, which are:

a) By using MICP, the capillary pressure ranges are observed to be from 5 up to 10 times scale compared to other conventional methods.

b) Irregular, small shaped pieces, such as drill cuttings or chips of drill cuttings (about 3^3cm of sample), could be handled in similar manner to regular, larger shaped samples such as permeability plugs or cores.

c) It would only take a few hours for an entire capillary pressure curve of 20 to 30 points compared to weeks of being able to obtain the results.

It is noted that completing the overall objectives could not be done for there are limitations to the equipment used. The pressure that should be applied would be about 60 000 psi from a certain case study, but the pressure on the equipment available is less than that.

There is also complication when it comes to the sample that should be studied. The outcrop sample available (shale) is insufficient in size to be made into cores. Even the equipment to cut the core is not able to carry out coring on the size of outcrop sample available.

Hence, proving the best possible solution is to use the Mercury Injection Capillary Pressure equipment.

Although, it is recommended that equipment of higher capability (pressure) be available and used for further studies in the future. It is also recommended that there is more equipment available and could cater to shale studies.

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