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Impact of nanoscale pore sizes on the flow of gas mixtures in marcellus shale reservoir

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**Impact of nanoscale pore sizes on the flow of gas mixtures in marcellus
shale reservoir**

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**Thesis submitted to Statler College of Engineering and Mineral Resources
at West Virginia University
in partial fulfillment of the requirements
for the degree of**

**Master of Science
in
Petroleum and Natural Gas Engineering**

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**Keywords: Viscosity, Nanopores, Pore Proximity, Confinement, z-Factor.
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Abstract

THE IMPACT OF PORE CONFINEMENT ON SHALE GAS PROPERTIES

Noor Mazher Alfashkhi

Pore proximity effect in shale reservoirs, unlike the conventional reservoirs, is very significant because the shale matrix contains pores in the range of nano meter that impact the phase behavior and fluid properties of the gas mixtures. The changes to the critical properties of the natural gas impacts the storage and flow in shale matrix. For optimization of gas extraction, understanding the gas properties is of great research interest.

In this research the impact of confinement on the gas viscosity and compressibility factor were evaluated using two typical composition of Marcellus shale gas. The results are to be used in formulating a new calculation method for gas storage and flow in shale reservoirs.

The results of the study indicate that the compressibility factor and viscosity of natural gas is changed by the pore proximity effect. The extent of gas properties change due to confinement depends on the composition of the gas mixture and the size of the nano-pores. Using two gas samples from Marcellus Shale, it was found that the viscosity and z-factor of confined gas deviated by an average of 20% and 6% respectively when compared to unconfined gases.

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CHAPTER 1 : INTRODUCTION

The Marcellus Shale is an abundant source of natural gas, and according to the U.S. Energy Information Administration, most of the natural gas consumed in the states comes from domestic production. The advances in technology such as hydraulic fracturing and horizontal drilling that uncovered a substantial amount of hydrocarbons accumulated in shale formations have increased the domestic production. Even though advances in technology have proven successful, the characteristics of the rock and the presence of nanoscale pores have created new challenges for the development of the reserves. Understanding the effects that nano pores have on the behavior of hydrocarbons within the shale formation can improve estimation of the gas in place and increase the production efficiency.

Reservoir analysis and prediction has been an integral necessity of the Oil & Gas industry for decades. Understanding reservoir components have allowed for better production efficiency and investment decision making. Conventional methods of modelling hydrocarbon reservoirs have been utilized by reservoir engineers, geologists, and industry professionals for years. With the development of new technology, unconventional resources have brought a wave of growth within the United States. Shale has taken its place as being the most explored and produced unconventional hydrocarbon reservoir in the modern era of oil & gas. With the advent of an industry overcome with unconventional shale production, its analysis and development of its production techniques, became a significant topic for lots of researchers.

When pore sizes decrease, the flow mechanism of fluids through the pores is changed and no longer follow Darcy's law of fluid flow (Curtis et al., 2010). Therefore, properties of the gas such as compressibility, and viscosity will be analyzed for different pore sizes. The results of the analysis will then be used to determine the gas in place and measure the gas flow rate in shale formations.

Chapter 2 : Literature review

2.1 viscosity of gas mixtures

Viscosity is the internal resistance of fluid to flow. For a mixture of gases, the viscosity can be evaluated if the composition as well as the viscosity of each component is available (McCain, 1990).

$$\mu_{ga} = \frac{\sum_{i=1}^N y_i \mu_i \sqrt{M_{gi}}}{\sum_{i=1}^N y_i \sqrt{M_{gi}}} \quad (2.1)$$

Where, μ_{ga} is the viscosity of the gas mixture at atmospheric pressure, y_i is the mole fraction of each component of the gas mixture, μ_i is the viscosity of each individual component at atmospheric pressure, and M_{gi} as well as N are the molecular weight and number of components in the gas mixture respectively.

The viscosity of each component of natural gas mixture at a given pressure and temperature can be determined by using a correlation chart (Appendix) that rely on the pseudo reduced values of temperature and pressure (Carr et al., 1954).

2.2 Gas Compressibility factor

Assumptions in the current kinetic theory of gases (McCain, 1990) states that gas molecules which are completely separated from each other have the same average velocity, have the same mass, the interactions among gas particles is negligible, and the molecules are perfectly round and are elastic in nature. Gases following these assumptions, are known as ideal gases.

In reality however, gas particles do interact with each other and only at high temperature and pressure they stay far apart and so their interaction as well as their relative volume are insignificant. Real gases under such conditions are said to behave ideally.

To account for the deviation of gases from their ideal state, a correction factor is used. This correction factor is called gas compressibility factor or *Z-factor*. The *z-factor* can be estimated using Standing and Katz chart (appendix) that utilizes the pseudo-reduced temperature and pressure obtained from the pseudo-critical pressure and temperature. The pseudo-critical pressure and temperature may simply be expressed as the summation of the molar critical pressure and temperature of the gas mixture components (McCain, 1990). Hence:

$$p_{pc} = \sum y_i p_{ci} \quad (2.2)$$

$$T_{pc} = \sum y_i T_{ci} \quad (2.3)$$

Where p_{pc} and T_{pc} are the pseudo-critical pressure and temperature of the gas mixture, p_{ci} and T_{ci} are the critical pressure and temperature of component i in the gas mixture while y_i is the mole fraction of component i in the gas mixture.

$$T_{pc} = \sum y_i T_{ci} \quad (2.4)$$

$$P_{pc} = \sum y_i p_{ci} \quad (2.5)$$

$$T_{pr} = T_r / T_{pc} \quad (2.6)$$

$$P_{pr} = P_r / P_{pc} \quad (2.7)$$

Where p_{pc} and T_{pc} are the pseudo-critical pressure and temperature of the gas mixture, p_{ci} and T_{ci} are the critical pressure and temperature of component i in the gas mixture while y_i is the mole fraction of component i in the gas mixture.

2.3 Impact of Pore Confinement on the Gas Critical Temperature and Pressure

When defining gases, properties such as pressure and temperature are usually used. Any change in pressure and temperature inside the reservoir will cause a change to the behavior and maybe the phase of the gas. Figure 2-1 shows when each phase starts and end, and the critical point (point C in Fig 2-1) in which the gas turns to liquid.

Figure 2-1 phase diagram (Clark 2004)

Over the years many studies have been conducted to analyze the effect pore confinement has on the critical properties of gas from their bulk values. Singh et al. (2009) studied the shift in critical properties and proposed equations describing the shift in methane, n-butane, and n-octane. Later, Devegowda et al. (2012) extended the work of Singh and proposed correlations (table 2-1) for measuring the deviation of critical pressure and temperature of the heavier gases confined in the nano-pore sizes of 2, 4, and 5 nm. Figures 2-2 and 2-3 show the deviation in critical temperature and pressure with respect to the molecular weight of the gas.

Table 2-1 Proposed Correlation for determining the shift of critical pressure and temperature

ΔP_c	2 nm	$0.085\ln(\text{MW}) - 0.0693$
	4 nm	$-0.085\ln(\text{MW}) + 0.1193$
	5 nm	$-0.077\ln(\text{MW}) - 0.041$
ΔT_c	2 nm	$0.0636 (\text{MW})^{0.2129}$
	4 nm	$0.0229 (\text{MW})^{0.2319}$
	5 nm	$0.0153 (\text{MW})^{0.241}$

$$\Delta T_c = \frac{T_{cb} - T_{cp}}{T_{cb}} \quad (3.1)$$

$$\Delta P_c = \frac{P_{cb} - P_{cp}}{P_{cb}} \quad (3.2)$$

Where T_{cb} and P_{cb} are the critical temperature and pressure of the bulk fluid, and T_{cp} as well as P_{cp} are their equivalent values in confined pores.

Figure 2-2 Impact of Nanopore Sizes on gas critical pressure (Devegowda 2012).

Figure 2-3 Impact of Nanopore Sizes on gas critical temperature (Devegowda 2012).

Chapter 3 : Methodology

The objective of this research was to study and understand the effect pore proximity has on the natural gas found in Marcellus shale formation. Mainly its effect on compressibility and viscosity of the gas.

To meet the objective of this study, two different gas compositions from two different areas within the Marcellus (Table 3-1), were utilized to measure and compare the viscosity as well as the deviation factor of the gas confined within the pores.

Table 3-1 Marcellus Shale Gas Compositions

Mole fraction		
component	sample 1	sample 2
CO ₂	0.00156	0.00089
N ₂	0.00465	0.00355
C ₁	0.77299	0.74308
C ₂	0.14775	0.15185
C ₃	0.04885	0.05874
i-C ₄	0.00573	0.00714
n-C ₄	0.01156	0.01717
i-C ₅	0.00239	0.0037
n-C ₅	0.0023	0.00537
C ₆	0.00129	0.005
C ₇₊	0.00093	0.0035

The relative shift in critical properties of each component were estimated by utilizing Singh et al. equations and Devegowda's et al. correlations for the confined pores listed in the literature review. A set of reservoir temperature and pressure (Table 3-2) were used in the evaluation process of gas properties.

Table 3-2 Reservoir Temperature and Pressure

P (psia)	T(°R)
3000	574.69
3100	576.69
3200	578.69
3300	580.69
3400	582.69
3500	584.69
3600	586.69
3700	588.69
3800	589.69
3900	592.69
4000	594.69

3.1 Impact of confinement on compressibility (Z) factor.

The pseudo-critical and pseudo-reduced values of pressure and temperature were calculated using the equations provide in the literature review, and the results were used for estimating the real gas deviation factor from Standing and Katz Z-factor chart (Appendix).

3.2 Impact of confinement on viscosity of gas mixtures.

The viscosity of the gas confined in 2, 4, and 5 nanometer pores were calculated using the compositional analysis as well as pseudo-reduced properties of the gas found in the previous section. The viscosity at one atmospheric pressure and reservoir temperature is determined first using the gas mixture composition equation (2.1), and then, multiplying the result by the viscosity ratio of each component obtained from Carr's diagram (Appendix) of the effect temperature and pressure has on the viscosity of natural gas.

Chapter 4 Results

4.1 Impact of Pore Confinement on the gas compressibility factor

The z-factor for the two samples of shale used in this study were determined using the compressibility chart mentioned in the literature review. The estimated values of the z-factor for the confined gas in the shale matrix are compared to the unconfined (base) mixture. The plots below show the deviation of z-factor for various reservoir pressures and pore sizes of 2, 4, and 5 nanometers in diameter.

Figure 4-1 Sample 1 z-factor at different reservoir pressures

Figure 4-2 Sample 2 z-factor at different reservoir pressures

4.2 Confinement effect on the viscosity.

The viscosity was measured using the method proposed in the methodology, and the results for the confined gas in 2, 4, and 5 nanometer sized pores were compared to the viscosity of unconfined (base) gas. The following tables (4-1 and 4-2) and Figures 6 and 7 show the percent change due to confinement as well as the deviation of viscosity compared the change in reservoir pressure:

Table 4-1 Percent change in viscosity from the unconfined gas (sample 1)

% Change in Viscosity (Sample 1)			
<i>P (psia)</i>	2 nm pore size	4 nm pore size	5 nm pore size
3000	-15%	-20%	-20%
3100	-15%	-19%	-20%
3200	-16%	-21%	-21%
3300	-15%	-19%	-20%
3400	-14%	-19%	-20%
3500	-14%	-17%	-21%
3600	-15%	-19%	-24%
3700	-14%	-19%	-24%
3800	-15%	-19%	-23%
3900	-13%	-19%	-23%
4000	-13%	-20%	-23%

Table 4-2 Percent change in viscosity from the unconfined gas (sample 2)

% Change in Viscosity (Sample 2)			
<i>P (psia)</i>	2 nm pore size	4 nm pore size	5 nm pore size
3000	-20%	-21%	-23%
3100	-19%	-21%	-23%
3200	-18%	-19%	-23%
3300	-19%	-21%	-22%
3400	-19%	-20%	-23%
3500	-19%	-19%	-23%
3600	-19%	-20%	-24%
3700	-18%	-19%	-22%
3800	-18%	-20%	-22%
3900	-18%	-19%	-22%
4000	-19%	-19%	-24%

Figure 4-3 change in viscosity for the change in reservoir pressure (sample 1)

Figure 4-4 change in viscosity for the change in reservoir pressure (sample 2)

Chapter 5 Conclusions

In this research three cases of confined pores with pore sizes of 2, 4, and 5 nanometers were used to study the impact they have on the viscosity and compressibility of the gas by comparing the results to the case of unconfined pores.

The results have shown that nanopores impact the viscosity and the compressibility factor of the gas. The most observed effect occurs when pore sizes are 2 nanometers in diameter, as shown in tables 5-1 and 5-2.

5.1 confinement effects on viscosity

Table 5-1 Percent Change of viscosity

pore size	sample 1	sample 2
2 nm	-15%	-19%
4 nm	-19%	-20%
5 nm	-22%	-23%

A decrease in pore size decreases the viscosity. Change in viscosity impacts flow rate, the higher the viscosity means that hydrocarbons will flow more slowly, therefore, when pores are 2 nanometers in diameter where the viscosity is at its lowest the hydrocarbons will flow much faster.

5.2 Confinement effects on z-factor

Table 5-2 Percent Change of compressibility factor

pore size	sample 1	sample 2
2 nm	17%	18%
4 nm	2%	1%
5 nm	-1%	-2%

While smaller pores have a positive impact on the viscosity of the gas, it impacts the estimation of original gas in place negatively. A smaller pore size leads to an increase in z-factor and therefore an over estimation of gas reserves.

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APPENDIX

Table A- 1 Critical temperature and pressure for unconfined gas

<i>component</i>	<i>Critical Temperature (R)</i>	<i>Critical Pressure (psia)</i>
CO₂	548	1072
N₂	227	492
C₁	344	673
C₂	550	709
C₃	666	618
i-C₄	733	530
n-C₄	766	551
i-C₅	830	482
n-C₅	847	485
C₆	915	434
C₇₊	973	397

Table A- 2 Critical Temperature and Pressure for Confined pores

<i>Component</i>	<i>2 nm pore</i>		<i>4 nm pore</i>		<i>5 nm pore</i>	
	<i>P_c (psia)</i>	<i>T_c R</i>	<i>P_c (psia)</i>	<i>T_c R</i>	<i>P_c (psia)</i>	<i>T_c R</i>
CO₂	801.45	469.99	1288.95	517.82	1428.33	527.13
N₂	386.73	197.65	572.67	215.74	638.42	219.25
C₁	560.89	304.50	751.46	329.01	844.40	333.73
C₂	553.02	477.80	829.53	522.27	923.88	530.89
C₃	461.92	571.15	743.18	629.30	823.52	640.62
i-C₄	383.71	622.29	649.79	689.94	717.52	703.15
n-C₄	398.92	650.31	675.53	721.00	745.95	734.80
i-C₅	340.12	698.75	599.78	778.74	660.55	794.39
n-C₅	342.22	713.04	603.53	794.68	664.67	810.66
C₆	299.68	764.71	546.62	856.11	600.72	874.02
C₇₊	269.04	807.97	505.11	908.14	554.12	927.81

Table A- 3 Shift in Critical Properties

Component	shift(Pc)			shift(Tc)		
	2 nm pore	4 nm pore	5 nm pore	2 nm pore	4 nm pore	5 nm pore
CO2	0.2524	-0.2024	-0.3324	0.1424	0.0551	0.0381
N2	0.2140	-0.1640	-0.2976	0.1293	0.0496	0.0342
C1	0.1666	-0.1166	-0.2547	0.1148	0.0436	0.0299
C2	0.2200	-0.1700	-0.3031	0.1313	0.0504	0.0347
C3	0.2525	-0.2025	-0.3326	0.1424	0.0551	0.0381
i-C4	0.2760	-0.2260	-0.3538	0.1510	0.0587	0.0407
n-C4	0.2760	-0.2260	-0.3538	0.1510	0.0587	0.0407
i-C5	0.2944	-0.2444	-0.3704	0.1581	0.0618	0.0429
n-C5	0.2944	-0.2444	-0.3705	0.1582	0.0618	0.0429
C6	0.3095	-0.2595	-0.3841	0.1642	0.0644	0.0448
C7+	0.3223	-0.2723	-0.3958	0.1696	0.0667	0.0464

Table A- 4 Estimated Z-Factors for Sample one

Sample 1			
Z (base)	Z (2nm pore)	Z (4nm pore)	Z (5nm pore)
0.74	0.87	0.775	0.76
0.75	0.88	0.78	0.768
0.76	0.89	0.785	0.77
0.77	0.905	0.795	0.775
0.78	0.915	0.8	0.779
0.785	0.925	0.804	0.784
0.795	0.935	0.81	0.79
0.805	0.945	0.82	0.794
0.815	0.958	0.83	0.789
0.83	0.967	0.834	0.802
0.84	0.975	0.844	0.809

Table A- 5 Estimated z-factors for sample two

Sample 2			
<i>Z (base)</i>	<i>Z (2nm)</i>	<i>Z (4nm)</i>	<i>Z (5nm)</i>
0.72	0.86	0.75	0.73
0.73	0.87	0.755	0.73
0.74	0.885	0.765	0.74
0.751	0.892	0.776	0.75
0.76	0.9	0.78	0.755
0.78	0.91	0.785	0.76
0.789	0.93	0.79	0.768
0.8	0.935	0.8	0.77
0.811	0.944	0.81	0.775
0.82	0.952	0.819	0.785
0.831	0.965	0.828	0.79

Table A- 6 Pseudo reduced values of temperature and pressure sample one

Component	2 nm pore		4 nm pore		5 nm pore	
	<i>Ppr (psia)</i>	<i>Tpr R</i>	<i>Ppr (psia)</i>	<i>Tpr R</i>	<i>Ppr (psia)</i>	<i>Tpr R</i>
CO2	5.46	1.63	3.95	1.50	3.52	1.48
N2	5.64	1.64	4.08	1.51	3.64	1.49
C1	5.82	1.65	4.21	1.52	3.76	1.49
C2	6.00	1.65	4.34	1.52	3.87	1.50
C3	6.18	1.66	4.47	1.53	3.99	1.50
<i>i</i> -C4	6.36	1.66	4.61	1.53	4.11	1.51
<i>n</i> -C4	6.55	1.67	4.74	1.54	4.23	1.51
<i>i</i> -C5	6.73	1.67	4.87	1.54	4.34	1.52
<i>n</i> -C5	6.91	1.68	5.00	1.54	4.46	1.52
C6	7.09	1.69	5.13	1.55	4.58	1.53
C7+	7.27	1.69	5.26	1.56	4.70	1.53

Table A- 7 Pseudo reduced values of temperature and pressure sample two

Component	2 nm pore		4 nm pore		5 nm pore	
	Ppr (psia)	Tpr R	Ppr (psia)	Tpr R	Ppr (psia)	Tpr R
CO2	5.50	1.59	3.96	1.46	3.54	1.44
N2	5.69	1.59	4.09	1.46	3.65	1.44
C1	5.87	1.60	4.23	1.47	3.77	1.45
C2	6.05	1.60	4.36	1.47	3.89	1.45
C3	6.24	1.61	4.49	1.48	4.01	1.46
i-C4	6.42	1.61	4.62	1.48	4.12	1.46
n-C4	6.60	1.62	4.75	1.49	4.24	1.47
i-C5	6.79	1.63	4.89	1.49	4.36	1.47
n-C5	6.97	1.63	5.02	1.50	4.48	1.47
C6	7.15	1.64	5.15	1.50	4.60	1.48
C7+	7.34	1.64	5.28	1.51	4.71	1.49

Table A- 8 Composition and critical viscosity of gas sample one

component	Mole fraction yi	Mi	viscosity	viscosity * yi
CO2	0.00156	44.01	0.016	0.00002496
N2	0.00465	28.01	0.019	0.00008835
C1	0.77299	16.04	0.012	0.00927588
C2	0.14775	30.07	0.0102	0.00150705
C3	0.04885	44.1	0.009	0.00043965
i-C4	0.00573	58.12	0.0083	0.000047559
n-C4	0.01156	58.12	0.008	0.00009248
i-C5	0.00239	72.12	0.0072	0.000017208
n-C5	0.0023	72.15	0.007	0.0000161
C6	0.00129	86.18	0.0066	0.000008514
C7+	0.00093	100.21	0.0062	0.000005766
total	1			0.011523517

Table A- 9 Composition and critical viscosity of gas sample two

component	Mole fraction y_i	M_i	viscosity	viscosity * y_i
CO₂	0.00089	44.01	0.016	1.42E-05
N₂	0.00355	28.01	0.019	6.75E-05
C₁	0.74308	16.04	0.012	0.008917
C₂	0.15185	30.07	0.0102	0.001549
C₃	0.05874	44.1	0.009	0.000529
i-C₄	0.00714	58.12	0.0083	5.93E-05
n-C₄	0.01717	58.12	0.008	0.000137
i-C₅	0.0037	72.12	0.0072	2.66E-05
n-C₅	0.00537	72.15	0.007	3.76E-05
C₆	0.005	86.18	0.0066	0.000033
C₇₊	0.0035	100.21	0.0062	2.17E-05
total	0.99999			0.011392

Figure A- 1 Standing and Katz z -factor chart (McCain, 1990)

Figure A- 2 Effect of temperature and pressure on viscosity of natural gas (Carr et al., 1954)