Journal of Petroleum Science and Engineering 133 (2015) 226–237

Contents lists available at ScienceDirect

Journal of Petroleum Science and Engineering

journal homepage: www.elsevier.com/locate/petrol

# Experimental investigation on the characteristics of gas diffusion in shale gas reservoir using porosity and permeability of nanopore scale

Changjae Kim, Hochang Jang, Jeonghwan Lee*

*Department of Energy and Resources Engineering, College of Engineering, Chonnam National University, 77 Yongbong-ro, Buk-gu, Gwangju 500-757, Republic of Korea*

ARTICLE INFO

*Article history:*
Received 2 September 2014
Received in revised form
20 March 2015
Accepted 10 June 2015
Available online 19 June 2015

*Keywords:*
Shale gas
Nanopore
Flow regime
Knudsen diffusion
Fick diffusion

ABSTRACT

The aim of this study is to investigate the diffusion characteristics of nanoscale gas flow in a shale gas reservoir. An experimental apparatus was designed and set up to measure porosity and permeability at the nanopore scale. The measured properties have been used to determine a diffusion coefficient by classification standard of gas flow regime. To investigate the impact of pressure and pore size, the analysis of diffusion flow was conducted using Knudsen and Fick diffusion coefficients. From the results, it was revealed that Knudsen diffusion coefficient gradually increased with the growth of pressure and pore radius and Fick diffusion coefficient was dependent on the gas molecular diameter and temperature. It was also found that Knudsen diffusion coefficient was equal to Fick diffusion coefficient if pore radius is too small. The study implied that the characteristics of gas diffusion should be implemented by using the diffusion coefficient theory based on gas flow regime in shale gas reservoirs.



## 1. Introduction

Shale gas boom in the United States has opened a new frontier for the world energy market. Shale gas is natural gas trapped within shale formations underground. Gas production from shale formation is one of the main fields focused on the global natural gas development industry (Rutqvist et al., 2013; Zhao et al., 2013). In general, conventional analysis models have used to measure the fine grained, organic rich shale properties. However, the methods yield erroneous data in characterizing the pore system of shale formation. Cui et al. (2009) described that the conventional methods are not practical because it is time consuming and the equipment performance is too inappropriate to measure small pressure changes or flow rates in tight rocks such as shale or coal.

In order to measure the shale matrix permeability, Luffel et al. (1993) developed GRI method using crushed samples. Based on the study, crushing samples nearly destroys microfractures in shale so it is effective to measure the shale permeability in nanopores. Luffel et al. (1993) validated the test results of GRI method by comparing with simulation model and it presented that the pressure transient corresponded to simulation results practically. Cui et al. (2009) introduced the method to obtain permeability and diffusivity from crushed shales by considering adsorption. They examined analytically the relative error of method on pressure difference and it was found that pressure change in the experiment causes a significant error for the property measurement. Tinni et al. (2012) examined which experimental factors affect the crushed samples permeability. In the experiment, particle size, pore pressure, gas species, and initial pressure condition were changed to analyze the property variation of crushed samples. From the results, Tinni et al. (2012) proposed that the pore pressure and particle size have a significant effect for the permeability measurement.

Darcy equation, used to investigate gas flow in rocks with micropores, cannot be used for shale matrix due to nanopores in shale (Cui et al., 2009; Swami et al., 2012; Javadpour et al., 2007). Roy et al. (2003) designed 2-D model to investigate gas flow in nanopores. The model was based on Knudsen diffusion and Navier–Stoke equation. Roy et al. (2003) also conducted the validation by comparing the model results with that of membrane experiment and it has an error within 5%. Based on the previous study, Javadpour et al. (2007) established a mathematical model which illustrates shale gas diffusion. The model was designed by using Gaussian distribution function. Javadpour et al. (2007) also excludes the viscous effect to consider gas diffusion in shale matrix. The diffusion model was validated by gas desorption data and it presented that the model corresponded with the experiment data practically. After that, Freeman et al. (2011) conducted the analysis of gas flow using DGM (dusty-gas model). The DGM was used to investigate the effects of Knudsen diffusion on gas composition in shale gas reservoir systems. From the literature survey, it was revealed that the previous studies did not consider the type

* Corresponding author. Fax: +82 62 530 1729.
*E-mail address:* jhwan@jnu.ac.kr (J. Lee).

http://dx.doi.org/10.1016/j.petrol.2015.06.008
0920-4105/

**Nomenclature**

| | |
|---|---|
| $c_g$ | gas compressibility, $Pa^{-1}$ |
| $D_k$ | Knudsen diffusion coefficient, $cm^2/s$ |
| $D_{fick}$ | Fick diffusion coefficient, $cm^2/s$ |
| $D_{transition}$ | transition diffusion coefficient, $cm^2/s$ |
| $F_R$ | gas residual ratio of remaining gas over total gas to be taken up by sample or $1-F_U$ ($F_U$, gas uptake ratio) |
| $f_0$ | the $y$-intercept of the straight line |
| $J$ | total mass flux, $kg/s/m^2$ |
| $J_a$ | mas flux of gas diffusion, $kg/s/m^2$ |
| $J_D$ | mas flux of gas flow, $kg/s/m^2$ |
| $K$ | apparent transport coefficient, $m^2/s$ |
| $K_a$ | partial derivative of adsorbate density with respect to gas density |
| $K_c$ | gas capacity ratio of void volume of cells over total pore volume of sample |
| $Kn$ | Knudsen number |
| $k$ | permeability, $m^2$ |
| $k_b$ | Boltzmann constant, $1.38\times10^{-23}$ J/K |
| $M$ | sample mass, g |
| $M_g$ | gas molar mass, kg/kmol |
| $p$ | pressure, MPa |
| $p_0$ | initial pressure in sample cell, MPa |
| $p_{r0}$ | initial pressure in reference cell, MPa |
| $p_e$ | equilibrium pressure in reference and sample cells, MPa |
| $\nabla p$ | pressure gradient, MPa |
| $R$ | gas constant, J/mol/K |
| $R_a$ | radius of crushed samples, mm |
| $r$ | radial distance from the center of spherical sample, cm |
| $r_A$ | radius of gas molecular, cm |
| $r_{pore}$ | pore radius, nm |
| $s_1$ | slope of the straight line part of $\ln(F_R)$ versus time at late-time |
| $T$ | temperature, K |
| $t$ | time, s |
| $V_b$ | bulk volume of samples (including pore space), $cm^3$ |
| $V_c$ | total volume of open space in reference and sample cells ($V_r+V_s-V_b$), $cm^3$ |
| $V_r$ | reference cell volume, $cm^3$ |
| $V_s$ | sample cell volume, $cm^3$ |
| $z$ | gas compressibility factor (=1.0 for ideal gas) |
| $z_0$ | gas compressibility factor in sample cell |
| $z_{r0}$ | gas compressibility factor in reference cell |
| $z_e$ | gas compressibility factor in reference and sample cells |
| $\lambda$ | gas phase molecular mean free path, nm |
| $\mu$ | viscosity, Pa/s |
| $\mu_B$ | viscosity of fluid in the pore of shale, Pa/s |
| $\alpha_n$ | the $n$th roots of the transcendental Eq. (8) |
| $\rho$ | gas density, mol $/m^3$ |
| $\rho_0$ | initial gas density in sample cell, $mol/m^3$ |
| $\rho_b$ | sample bulk density, $g/cm^3$ |
| $\rho_{c0}$ | average initial gas density in void volume of reference and sample cells, mol $/m^3$ |
| $\rho_{r0}$ | initial gas density in reference cell, $mol/m^3$ |
| $\phi$ | porosity of porous media, fraction |

of diffusion. The studies have just researched the gas flow using Knudsen diffusion or Fick diffusion model (Javadpour et al., 2007; Shabro et al., 2009; Sigal and Qin, 2008). From the results, Wang et al. (2013) conducted the investigation of gas flow ability in the shale by considering Knudsen and Fick diffusion. Wang et al. (2013) suggested that the determination of flow regimes should be implemented by $Kn$ primarily, and then taken the diffusion coefficient of system from the flow regime. In order to examine gas diffusion in shale matrix it is essential to determine the shale properties at nanopore scale. However, the previous studies used the core experiment such as pressure pulse decay (PPD) method to determine the low permeability in the shale gas reservoirs. It cannot be the representative of nanopores in shale matrix due to the microfracture in core samples. Thus, the permeability obtained from the PPD method is not valid for the characterization of gas diffusion in nanopores of shale matrix (Kamath et al., 1992; Luffel et al., 1993).

This study presents a new apparatus to measure the porosity and permeability of shale matrix at nanopore scale. Knudsen and Fick diffusion model are determined by the measured properties and the diffusion coefficient is used to analyze gas diffusion in the shale matrix. Finally, the impact of the pore radius and pressure are investigated to verify the gas flow on reservoir conditions.

## 2. Theoretical background

### 2.1. Permeability measurement of crushed sample

GRI technique was introduced by Luffel et al. (1993) to measure the permeability of shale using crushed sample. Luffel et al. (1993) described that this method is rapid to run and crushed samples rarely have micro-fractures which affect the permeability. It means that the method is practical to measure the shale permeability at nanoscale. GRI technique only used helium, a non-adsorbate gas, to measure the permeability. However, methane which is major component of shale gas has the adsorption effect. It means that the measurement by methane results in erroneous permeability and needs to be corrected for adsorption in the measurement. To solve this problem, Cui et al. (2009) revised the technique to correct the impact of gas adsorption on permeability measurements. Fig. 1 presents a schematic sketch of a pycnometer apparatus to apply the corrected GRI technique. This study proposed early and late time method for determining permeability. The early time method can only calculate the permeability by using the initial pressure data. However, the data quality may be poor due to the kinetic gas expansion from the reference cell into the sample cell. The expansion causes the temperature change in the pycnometer system,

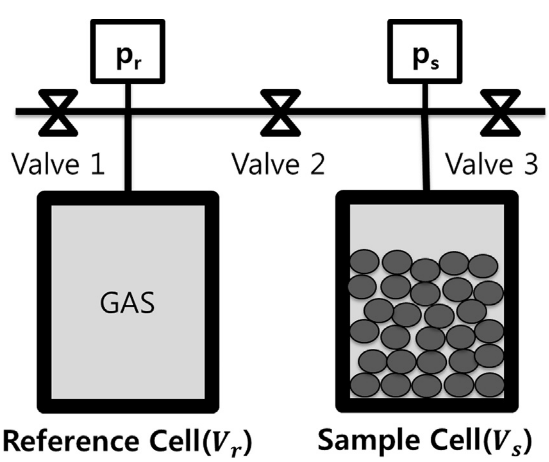


**Fig. 1.** A schematic sketch of a pycnometer apparatus (Cui et al., 2009).