

Invited review

Insights on the gas permeability change in porous shale

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Abstract: Due to abundant nanoscale pores developed in shale, gas flow in shale presents a complex dynamic process. This paper summarized the effects from effective stress increase, shale matrix shrinkage, gas slippage and Knudsen diffusion on the gas permeability change in shale during shale gas recovery. With the reduce in gas pressure, effective stress increase leads to the decline of the permeability in an exponential form; the permeability increases due to the shale matrix shrinkage induced by gas desorption; appearances of gas slippage and Knudsen diffusion cause an additional increase in the gas permeability particularly in small pores at low pressures. In addition, some reported models evaluating the shale permeability were reviewed preliminarily. Models considering these four effects may be potentially effective to evaluate the gas permeability change in shale.

Keywords: Shale gas, permeability, effective stress, matrix shrinkage, gas slippage, Knudsen diffusion.

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1. Introduction

Shale gas (CH₄), primarily in adsorbed, free and dissolve states, accumulates in carbon-rich mudstone and shale reservoirs (Curtis, 2002). During the shale gas recovery, free gas was extracted at the early stage. With continuous decrease in gas pressure, adsorbed gas will be released gradually from the adsorption layer, and then transports into the natural pore-fractures. Subsequently, the desorbed gas moves into macroscopic fractures induced by hydraulic fracturing and eventually flows into well borehole. Multi-scale flow of shale gas from matrix pore to the borehole shows different flow regimes (Javadpour et al., 2007).

Intrinsic permeability of shale is extremely low due to a small seepage space developed in shale. Javadpour et al. (2007) analyzed 152 cores from North America and presented that 90% shales have permeability of less than $150 \times 10^{-6} \mu\text{m}^2$ and pore diameter of shales mainly ranges from 4-200 nm. Other researches (Ambrose et al., 2010; Sondergeld et al., 2010) reported that shale pore diameter is mainly of 8-100 nm in North America. Gas-bearing shales from China have pore diameter of 5-300nm, mainly ranging from 80-200 nm (Zou et al., 2010; 2011). Because that the mean free path of

gas molecules is close to pore size, the nanoscale effect cannot be ignored as gas flows in nanopores. Therefore, the Darcy law reflecting continuum flow is no longer applicable for shale gas due to the significant influence of molecular diffusion and gas slippage. To date, it has been generally accepted that gas flow regimes can be subdivided into continuum flow ($K_n < 0.001$), slip flow ($0.001 < K_n < 0.1$), transition flow ($0.1 < K_n < 10$) and free molecular flow ($K_n > 10$) based on the Knudsen number (K_n , i.e., the ratio of the mean free path of molecule to pore diameter) (Yao et al., 2013; Kim et al., 2015; Kazemi and Takbiri-Borujeni, 2015), as shown in Fig. 1. As $K_n > 1.0$, Knudsen diffusion was not negligible. Theoretical analyses (Yao et al., 2013; Wu et al., 2016) showed that shale gas flow in the nanometer (1-1000 nm) pores is mainly characterized by slip and transition flows at the pressures of 0-10 MPa; and it occurs free molecular flow at low pressures of < 1 MPa in micropores (< 2 nm diameter).

Resulting from abundant nanopores, varied mineralogical compositions, low porosity and permeability, complex microstructure and rich organics (Li et al. 2017a), it is very difficult in understanding the permeability change in shale during the gas recovery. Rock deformation and changes of temperature and stress fields appear in shale reservoir and cau-

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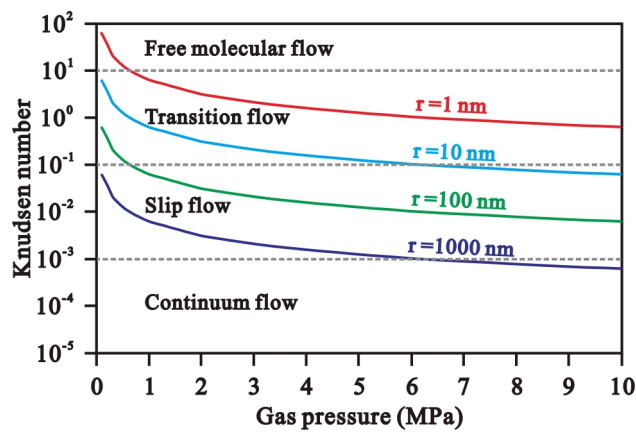


Fig. 1. Variation of Knudsen number (K_n) with gas pressure at different pore radius (r) (after Yao et al. (2013)).

se a remarkable permeability change with the gas pressure depletion, which will significantly influences the recovery of shale gas. In recent years, gas flow in porous shale has been studied intensively, and has becoming an importantly scientific issue (Javadpour, 2009; Cai et al., 2012, 2017; Zhang et al., 2015a; Peng et al., 2015). In this paper, several possible factors impacting the gas permeability change in shales were summarized. Further, reported models evaluating the permeability change were reviewed preliminarily.

2. Influence factors of gas permeability change

Microscopic seepage of shale gas are influenced by gas pressure, and pore-fracture system (pore structure, pore size distribution) etc. (Naraghi and Javadpour, 2015; Peng et al., 2015; Guo et al., 2015). During the shale gas exploitation, there is a change in the gas pressure and shale pore structure, which causing the gas permeability change. Moreover, a series of geological phenomena impacting on the permeability change occur in shale reservoir, which including the effective stress, gas slippage, matrix shrinkage, Knudsen diffusion and surface diffusion etc. (Reyes and Osisanya, 2002; Javadpour, 2009; Yao et al., 2013; Mehmani et al., 2013; Singh and Javadpour, 2016). The effective stress (i.e., the stress acting on the rock framework, and is approximately equal to the confining stress minus fluid pressure) increase leads to the decline of the permeability. The permeability increases due to the matrix shrinkage induced by gas desorption. Appearance of gas slippage and Knudsen diffusion causes an additional increase in the gas permeability. For the effect of surface diffusion of adsorbed gas on the gas permeability, it has not been determined today. Zhang et al. (2015a) reported that surface diffusion cannot be ignored; while Wang et al. (2015) thought that a weak effect of the surface diffusion can be negligible in reservoir conditions.

2.1 Effective stress

The increase in effective stress causes the compression and even closure of shale pore-fractures, which inducing a decrease in inherent permeability of shale. Previous experi-

ments showed that the permeability decrease in an exponential form with the increase in the effective stress (Mckee et al., 1988; Reyes et al., 2002; Chalmers et al., 2012; Ghanizadeh et al., 2014a; 2014b; Chen et al., 2015; Zhang et al., 2015a; 2015b; 2016). Change of rock effective stress can be caused by internal pressure (i.e., gas pressure) and external pressure (i.e., confining stress) variations. The effect of effective stress on shale permeability is closely related to shale pore-fracture system (Chalmers et al., 2012). In the study by Li et al. (2014), they reported that the coal with a lower permeability will have a stronger sensitivity of the permeability to the effective stress. Zhang et al. (2015b) investigated the impacts of nanopore structure and elastic properties on shale permeability, and showed that (a) pore compressibility increased with a decreasing pore aspect ratio and Youngs modulus; (b) the permeability of micro-fractures in shale were more sensitive to effective stress than hydraulic fractures. Zhang et al. (2016) reported the negative effect of effective stress on shale permeability, and indicated that shale has higher stress sensitive coefficients and lower porosity sensitive exponents than those of sandstone.

2.2 Matrix shrinkage

Gas absorbed content accounts for 20-85% in shale (Curtis, 2002). During gas pressure depletion, inherent permeability of shale also changes due to the matrix shrinkage induced by gas desorption (Peng et al., 2015). The matrix swelling/shrinkage induced by gas adsorption/desorption is a complex process. Experiments on coals have been conducted by many researchers (Levine, 1996; Karacan, 2007; Day et al., 2008; Pan et al., 2010). The strains occur in coals as the matrix swelling or shrinkage (Harpalani and Chen, 1995; Gilman and Beckie, 2000; Cui et al., 2004; Shi and Durucan, 2004). The Langmuir-like equation, which proposed by Levine (1996), was often used to describe the coal volume strain. Chen et al. (2015) researched the CH_4 adsorption swelling of shale, showing that swelling deformation is a function of gas pressure and can be described by the Langmuir-like equation. Lyu et al. (2015) summarized the H_2O adsorption swelling of shale, and presented the volume swelling of shale is negatively related to the initial water content and the logarithm of confining stress, but is positively proportional to clay content. Lu et al. (2016) tested the shale swelling in supercritical CO_2 , and found that the swelling initially increases but subsequently decreases with increasing pressure; maximum swelling decreases with the increase in measured temperature.

2.3 Gas slippage and Knudsen diffusion

Gas slippage impacts gas flow behavior in lowly permeable porous media, such as shale, and causes a gas permeability increment compared to liquid. Previous research has shown that gas slippage is associated with the mean free path of gas molecules and the mean radius of pores within porous media (Klinkenberg, 1941). Commonly, a linear relationship between the gas permeability and the reciprocal of the mean gas pressure is considered to be a response to the gas slippage

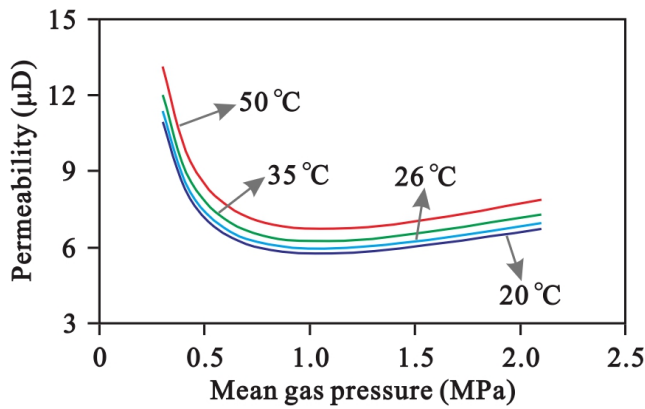


Fig. 2. Gas (CO_2) permeability changes at different experimental temperatures (Li et al., 2015).

and is mathematically expressed by the Klinkenberg equation. Gas slippage causes that the apparent gas permeability is greater than liquid permeability (Klinkenberg, 1941; Li et al., 2014; Firouzi et al., 2014). The slippage effect is mainly affected by pore size and gas pressure but is not obviously influenced by the temperature (Fig. 2). Generally, the smaller (lower) the pore diameter (gas pressure and density) is, the more obvious the gas slippage occurs. As the diameter of porous media is smaller up to a level of the free path of gas molecule, Knudsen diffusion appears in pores due to the frequent collision of gas molecule with pore wall.

During shale gas recovery, effects of gas slippage and Knudsen diffusion enhanced the gas permeability of shale. The effects of gas slippage and Knudsen diffusion on the permeability are remarkable in small pores at low pressures. Javadpour (2009) reported that the effects of gas slippage and Knudsen diffusion on the permeability are gradually significant during the decreasing processes of gas pressure (10-0 MPa) and pore diameter (100-0 nm).

3. Evaluation models of gas permeability change in shale

Models about the shale gas flow are mainly focused on numerical simulations considering various transmission mechanisms of gas. Beskok and Karniadakis (1999) derived a gas flow equation (classic permeability model) which considers continuum, slip, transition and diffusion flows, but ignored the influences of effective stress and matrix shrinkage on the permeability. Based on the study by Beskok and Karniadakis (1999), Xiong et al. (2012) considered the effect of gas adsorption and surface diffusion. Javadpour (2009) established an apparent permeability model by a linear superposition of Knudsen diffusion and gas slippage. But this model did not consider the influence of effective stress and gas adsorption. On the basis of Javadpour (2009), Azom and Javadpour (2012) took the effect of real gas on the permeability into consideration; Darabi et al. (2012) thought over the impact of pore wall roughness on Knudsen diffusion. Subsequently Wu et al. (2015) defined the weighting coefficient of Knudsen diffusion and slip flows based on the molecular collision theo-

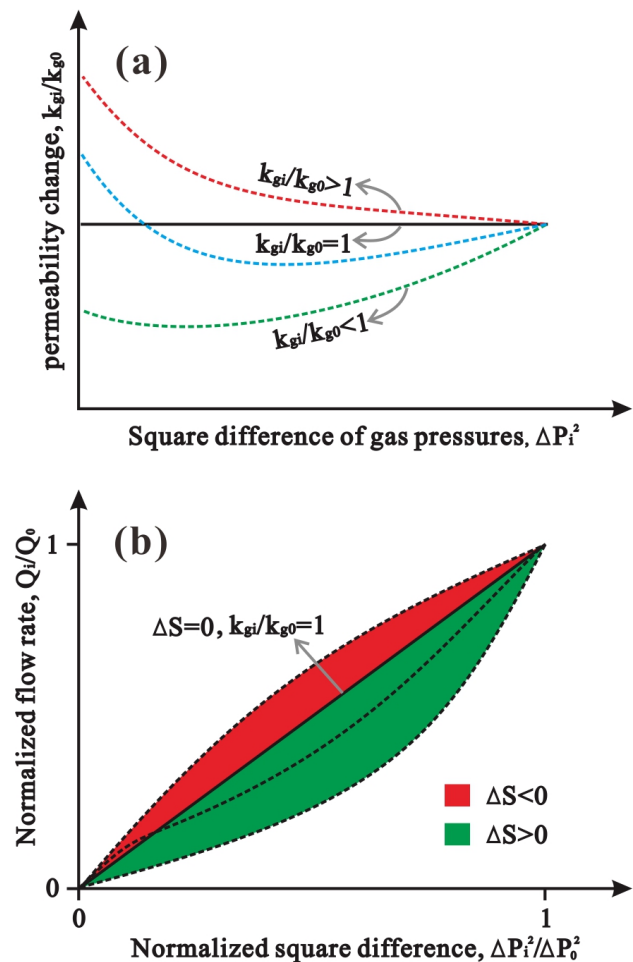


Fig. 3. General model of changes in the permeability (a) and flow rate (b) (Li et al., 2017b). In this figure, the negative (positive) ΔS value reflects the increase (decrease) in the cumulative gas flow rate compared to the condition of $k_{gi}/k_{g0} = 1$ during the depressurization process. Overall, ΔS varies from -0.5 to 0.5. During pressure decrease, as $\Delta S = -0.5$, the gas flow rate is always the highest; as $\Delta S = 0$, the gas permeability is constant; $\Delta S = 0.5$, reservoir is impermeable.

ry. Mehmani et al. (2013) established a gas permeability model for micro-nanometer pore of shale matrix. This model considered gas slippage and Knudsen diffusion, but ignored the influences of effective stress and matrix shrinkage on the permeability. Wu et al. (2016) built a gas transport model in the organic nano-pore of shale by comprehensively considering the influences of gas slippage, Knudsen diffusion, effective stress and surface diffusion except for the matrix shrinkage.

Recently, Li et al. (2013, 2014, 2015, 2017b) analyzed the effects of effective stress, gas slippage, matrix shrinkage and Knudsen diffusion on the gas permeability change in coals. They also proposed a prediction model of gas permeability change based on the effects of effective stress, gas slippage and matrix shrinkage (Li et al., 2013). And then, the model was improved by comprehensively considering the effects of effective stress, gas slippage, matrix shrinkage and Knudsen diffusion (Li et al., 2015), which has a good match with experimental data. A new method was presented to evaluate the dynamic gas flow process (Li et al., 2017b), as shown in Fig. 3. Moreover, there is a certain relationship between the

gas permeability changes and material composition, pore-crack system of coals (Li et al., 2017b). However, the prediction of gas permeability change in shale and the coupling relationship of the permeability change with shale reservoir characteristics have been not studied systematically. These results may be potentially useful to analyze shale.

4. Conclusions

In this paper, we presented several possible factors impacting the gas permeability change in shale. Further, some reported models evaluating the permeability were reviewed preliminarily. The gas permeability of shale varies due to the comprehensive effects of effective stress increase, matrix shrinkage, gas slippage, Knudsen diffusion and surface diffusion etc. during shale gas recovery. According to recent studies, it is concluded that gas permeability change is the comprehensive result of the effective stress increase, shale matrix shrinkage, gas slippage and Knudsen diffusion as the gas pressure decreases. First, with the pressure decrease, increased effective stress and matrix shrinkage occur in shale, causing shale deformation accompanied with varied shale permeability. Second, the effects of gas slippage and Knudsen diffusion on the permeability change gradually become significant. In addition, model established considering these four effects may be potentially effective to evaluate the gas permeability change in shale.

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References

- Ambrose, R.J., Hartman, R.C., Diaz-Campos, M., et al. New pore-scale considerations for shale gas in place calculations. Paper SPE131772 Presented at the SPE Unconventional Gas Conference, Pittsburgh, Pennsylvania, USA, 23-25 February, 2010.
- Azom, P.N., Javadpour, F. Dual-continuum modeling of shale and tight gas reservoirs. Paper SPE 159584 Presented at the SPE Annual Technical Conference and Exhibition, San Antonio, Texas, USA, 8-10 October, 2012.
- Beskok, A., Karniadakis, G.E. A model for flows in channels, pipes, and ducts at micro and nano scales. *Microscale Thermophys. Eng.* 1999, 3(1): 43-77.
- Cai, J., Ghanbarian, B., Xu, P., et al. Virtual special issue: Advanced theoretical and numerical approaches and applications to enhanced gas recovery. *J. Nat. Gas. Sci. Eng.* 2017, 37: 579-583.
- Cai, J., Yu, B. Advances in studies of spontaneous imbibition in porous media. *Adv. Mech.* 2012, 42(6): 735-754.
- Chalmers, G.R.L., Ross, D.J.K., Bustin, R.M. Geological controls on matrix permeability of Devonian gas shales in the Horn River and Liard basins, northeastern British Columbia, Canada. *Int. J. Coal Geol.* 2012, 103(23): 120-131.
- Chen, D., Pan, Z., Ye, Z. Dependence of gas shale fracture permeability on effective stress and reservoir pressure: Model match and insights. *Fuel* 2015, 139: 383-392.
- Cui, X., Bustin, R.M., Dipple, G. Selective transport of CO₂, CH₄, and N₂ in coals: Insights from modeling of experimental gas adsorption data. *Fuel* 2004, 83(3): 293-303.
- Curtis, J.B. Fractured shale-gas system. *AAPG Bull.* 2002, 86(11): 1921-1938.
- Darabi, H., Eftehad, A., Javadpour, F., et al. Gas flow in ultra-tight shale strata. *J. Fluid Mech.* 2012, 710(12): 641-658.
- Day, S., Fry, R., Sakurovs, R. Swelling of Australian coals in supercritical CO₂. *Int. J. Coal Geol.* 2008, 74(1): 41-52.
- Firouzi, M., Alnoaimi, K., Kovscek, A., et al. Klinkenberg effect on predicting and measuring helium permeability in gas shales. *Int. J. Coal Geol.* 2014, 123(2): 62-68.
- Ghanizadeh, A., Amann-Hildenbrand, A., Gasparik, M., et al. Experimental study of fluid transport processes in the matrix system of the European organic-rich shales: II. Posidonia Shale (Lower Toarcian, northern Germany). *Int. J. Coal Geol.* 2014a, 123(2): 20-33.
- Ghanizadeh, A., Gasparik, M., Amann-Hildenbrand, A., et al. Experimental study of fluid transport processes in the matrix system of the European organic-rich shales: I. Scandinavian Alum Shale. *Mar. Petrol. Geol.* 2014b, 51(51): 79-99.
- Gilman, A., Beckie, R. Flow of coal-bed methane to a gallery. *Transport Porous Med.* 2000, 41(1): 1-16.
- Guo, C., Xu, J., Wu, K., et al. Study on gas flow through nano pores of shale gas reservoirs. *Fuel* 2015, 143: 107-117.
- Harpalani, S., Chen, G. Influence of gas production induced volumetric strain on permeability of coal. *Int. J. Geotech. Geol. Eng.* 1995, 15(4): 303-325.
- Javadpour, F. Nanopores and apparent permeability of gas flow in mudrocks (shales and siltstone). *J. Can. Petrol. Technol.* 2009, 48(8): 16-21.
- Javadpour, F., Fisher, D., Unsworth, M. Nanoscale gas flow in shale gas sediments. *J. Can. Petrol. Technol.* 2007, 46(10): 07-10-06.
- Karacan, C.Ö. Swelling-induced volumetric strains internal to a stressed coal associated with CO₂ sorption. *Int. J. Coal Geol.* 2007, 72(3-4): 209-220.
- Kazemi, M., Takbiri-Borujeni, A. An analytical model for shale gas permeability. *Int. J. Coal Geol.* 2015, 146: 188-197.
- Kim, C., Jang, H., Lee, J. Experimental investigation on the characteristics of gas diffusion in shale gas reservoir using porosity and permeability of nanopore scale. *J. Petrol. Sci. Eng.* 2015, 133: 226-237.
- Klinkenberg, L. The permeability of porous media to liquids and gases. Paper API41200 Presented at the Drilling and

- Production Practice, New York, 1 January, 1941.
- Levine, J.R. Model study of the influence of matrix shrinkage on absolute permeability of coalbed reservoirs. *Geol. Soc. Special Pub.* 1996, 197-212.
- Li, J., Liu, D., Lu, S., et al. Evaluation and modeling of the CO₂ permeability variation by coupling effective pore size evolution in anthracite coal. *Energ. Fuel.* 2015, 29(2): 717-723.
- Li, J., Liu, D., Yao, Y., et al. Controls of CO₂ permeability change in different rank coals during pressure depletion: an experimental study. *Energ. Fuel.* 2014, 28(2): 987-996.
- Li, J., Liu, D., Yao, Y., et al. Evaluation and modeling of gas permeability changes in anthracite coals. *Fuel* 2013, 111: 606-612.
- Li, J., Lu, S., Cai, Y., et al. Impact of coal ranks on dynamic gas flow: An experimental investigation. *Fuel* 2017b, 194: 17-26.
- Li, J., Zhang, P., Lu, S., et al. Microstructural characterization of the clay-rich oil shales by nuclear magnetic resonance (NMR). *J. Nanosci. Nanotechnol.* 2017a, 17(9): 7026-7034.
- Lu, Y., Ao, X., Tang, J., et al. Swelling of shale in supercritical carbon dioxide. *J. Nat. Gas Sci. Eng.* 2016, 30(4): 268-275.
- Lyu, Q., Ranjith, P.G., Long, X., et al. A review of shale swelling by water adsorption. *J. Nat. Gas Sci. Eng.* 2015, 27: 1421-1431.
- Mckee, C., Bumb, A., Koenig, R. Stress-dependent permeability and porosity of coal and other geologic formations. *SPE Form. Eval.* 1988, 3(1):81-91.
- Mehmani, A., Prodanovi, M., Javadpour, F. Multiscale, multiphysics network modeling of shale matrix gas flows. *Transport Porous Med.* 2013, 99(2): 377-390.
- Naraghi, M.E., Javadpour, F. A stochastic permeability model for the shale-gas systems. *Int. J. Coal Geol.* 2015, 140: 111-124.
- Pan, Z., Connell, L.D., Camilleri, M., et al. Effects of matrix moisture on gas diffusion and flow in coal. *Fuel* 2010, 89(11): 3207-3217.
- Peng, Y., Liu, J., Pan, Z., et al. A sequential model of shale gas transport under the influence of fully coupled multiple processes. *J. Nat. Gas Sci. Eng.* 2015, 27: 808-821.
- Reyes, L., Osisanya, S.O. Empirical correlation of effective stress dependent shale rock properties. *J. Can. Petrol. Technol.* 2002, 27(12): 47-53.
- Shi, J.Q., Durucan, S. Drawdown induced changes in permeability of coalbeds: A new interpretation of the reservoir response to primary recovery. *Transport Porous Med.* 2004, 56(1): 1-16.
- Singh, H., Javadpour, F. Langmuir slip-Langmuir sorption permeability model of shale. *Fuel* 2016, 164: 28-37.
- Sondergeld, C.H., Ambrose, R.J., Rai, C.S., et al. Microstructural studies of gas shales. Paper SPE131771 Presented at the SPE Unconventional Gas Conference, Pittsburgh, Pennsylvania, USA, 23-25 February, 2010.
- Wu, K., Chen, Z., Li, X., et al. A model for multiple transport mechanisms through nanopores of shale gas reservoirs with real gas effect adsorption-mechanic coupling. *Int. J. Heat Mass Tran.* 2016, 93: 408-426.
- Wu, K., Li, X., Wang, C., et al. A model for gas transport in microfractures of shale and tight gas reservoirs. *AIChE J.* 2015, 61(6): 2079-2088.
- Xiong, X., Devegoda, D., Villazon, G.G.M., et al. A fully-coupled free and adsorptive phase transport model for shale gas reservoirs including non-darcy flow effects. Paper SPE159758 Presented at the SPE Annual Technical Conference and Exhibition, San Antonio, Texas, USA, 8-10 October, 2012.
- Yao, J., Sun, H., Huang, Z., et al. Key mechanical problems in the development of shale gas reservoirs. *Sci. Sin. Phys. Mech. Astron.* 2013, 43(12): 1527-1547 (in Chinese).
- Zhang, L., Li, D., Lu, D., et al. A new formulation of apparent permeability for gas transport in shale. *J. Nat. Gas Sci. Eng.* 2015a, 23: 221-226.
- Zhang, R., Ning, Z., Yang, F., et al. Impacts of nanopore structure and elastic properties on stress-dependent permeability of gas shales. *J. Nat. Gas Sci. Eng.* 2015b, 26: 1663-1672.
- Zhang, R., Ning, Z., Yang, F., et al. A laboratory study of the porosity-permeability relationships of shale and sandstone under effective stress. *Int. J. Rock. Mech. Min.* 2016, 81: 19-27.
- Zou, C., Dong, D., Wang, S., et al. Geological characteristics and resource potential of shale gas in China. *Petrol. Explor. Dev.* 2010, 37(6): 641-653.
- Zou, C., Zhu, R., Bai, B., et al. First discovery of nano-pore throat in oil and gas reservoir in China and its scientific value. *Acta Petrol. Sin.* 2011, 27(6): 1857-1864.