



Geological controls and estimation algorithms of lacustrine shale gas adsorption capacity: A case study of the Triassic strata in the southeastern Ordos Basin, China



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ABSTRACT

High-pressure methane adsorption experiments on a series of Triassic lacustrine shale moisture-equilibrated samples from the southeastern Ordos Basin, China, were conducted at pressure up to 20 MPa, two of which were measured at 30 °C, 40 °C, 50 °C, 60 °C, and 70 °C, and seven were performed under reservoir temperature (from 48 °C to 62 °C) to investigate the effect of organic matter content, maturity, mineralogical compositions and reservoir conditions (temperature and pressure) on the methane sorption capacity. The total organic carbon contents (TOCs) range from 0.91 wt.% to 6.11 wt.%. The thermal maturities, as inferred from Rock-Eval Analysis, range from low mature to high mature. The minerals of the shale samples are dominated by clays (36–57 wt.%) and quartz (19–44 wt.%). For the entire shale samples the dominant clay minerals are mixed-layer illite/smectite with some illite and no smectite exists, corresponding to a stage of late diagenesis. The studied samples have N₂ BET surface areas ranging between 1.47 and 9.21 m²/g and pore volumes of 0.013–0.034 cm³/g.

The methane sorption capacities of moisture-equilibrated shale samples show a positive correlation with TOC contents and BET surface areas. No relationship was observed between the clay contents and methane sorption capacities, indicating that clay minerals do not significantly contribute to methane sorption capacity in these organic shales. The Langmuir pressure (P_L) increases exponentially with temperature and the Langmuir volume (V_L) decreases linearly with temperature. A computational scheme has been developed to calculate the methane sorption capacity of shales as a function of TOC content, temperature and pressure based on Langmuir sorption isotherm function. Using this algorithm methane sorption capacity of organic shales as function of depth can be obtained. Due to the predominating effect of pressure the methane sorption capacity increases with depth initially, through a maximum and then decreases due to the influence of increasing temperature at a greater depth. The maximum gas sorption capacity typically occurs at a depth range between 400 and 900 m. With TOC content increasing, the maximum methane sorption capacities of organic shales and the corresponding depths increase.

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1. Introduction

Over the past decades, the combination of horizontal drilling and hydraulic fracturing has allowed access to large volumes of shale gas (Hao et al., 2013; Rexer et al., 2013), which were previously thought to be either impossible or uneconomical to produce. Gas shales have been recognized as natural gas reservoirs with enormous shale gas potential in the United States and Canada (Chalmers and Bustin, 2008b; Jarvie et al., 2007; Loucks et al., 2009; Montgomery et al., 2005; Ross and Bustin, 2008). Many other countries, such as China, India, Poland,

South Africa, Australia, Ukraine, and United Kingdom, launched into evaluating their shale gas resources to cope with the growing energy demand (Wang et al., 2014).

Shale gas is an unconventional gas system in which the shale is both the source of, and the reservoir for, natural gas, which is derived from the organic matter within the shale through biogenic and/or thermogenic processes (Curtis, 2002; Hill et al., 2007; Jarvie et al., 2007; Strapoć et al., 2010). Natural gas stored in shale reservoirs may be composed of: (1) free compressed gas in pores and fractures, (2) adsorbed gas on organic matter and inorganic mineral surfaces, and (3) dissolved gas in oil and water (Curtis, 2002; Gasparik et al., 2012, 2013; Hao et al., 2013; Rexer et al., 2013; Ross and Bustin, 2007; Zhang et al., 2012). Understanding the relative proportions of gas stored in these different forms is critical to an accurate assessment of shale gas resources and

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design of effective production strategies (Chalmers and Bustin, 2008a,b; Ross and Bustin, 2008, 2009; Zhang et al., 2012). Among these storage types, adsorbed gas accounts for between 20% and 85% of the total gas in five US shale formations (Curtis, 2002; Montgomery et al., 2005).

Adsorption is the process of molecule accumulation on the surface of a material as a consequence of surface energy minimization (Gregg and Sing, 1982; Zhang et al., 2012). Adsorption process in unconventional reservoirs is generally identified as physisorption that is a weak, reversible attraction due to Van der Waals forces and is associated with a small heat of adsorption (Ross and Bustin, 2007). Generally, the methane sorption capacity of organic shales is controlled by (1) characteristics of organic matters (type, abundance, and maturity), (2) composition of inorganic minerals (especially clay minerals), (3) micro-nano pore structures, (4) moisture content, and (5) pressure and temperature regimes (Gasparik et al., 2012; Hao et al., 2013).

Organic matter present in these gas shales is generally thought to be the principal contributor to the gas adsorption capacity. In the literature, several authors have found a general positive linear correlation between the methane sorption capacity and TOC content of organic shales (e.g. Chalmers and Bustin, 2007b, 2008a; Gasparik et al., 2013; Hao et al., 2013; Jarvie et al., 2007; Lu et al., 1995; Ross and Bustin, 2007, 2008, 2009; Strapoć et al., 2010; Zhang et al., 2012). With the increasing maturation, the TOC-normalized methane sorption capacities increase (Ross and Bustin, 2009; Zhang et al., 2012), this may be caused by the increase in organopores and surface areas from the thermal transformation of kerogen into hydrocarbon (Jarvie et al., 2007; Loucks et al., 2009; Mastalerz et al., 2013; Slatt and O'Brien, 2011), and the increase in aromatization (Zhang et al., 2012) and pore surface homogeneity (Laxminarayana and Crossdale, 1999; Ross and Bustin, 2009) with increasing maturity. However, recent several studies have shown that the evolution of organic porosity of shale does not follow a monotonous trend with increasing maturity, but instead follows a nonlinear, trough-shaped path (e.g. Gasparik et al., 2013; Mastalerz et al., 2013). The methane sorption capacities of varied kerogen were shown in the order: type I < type II < type III kerogen (Chalmers and Bustin, 2008a; Zhang et al., 2012) and this was contributed to (1) the higher sorption capacity of vitrinite compared to other maceral composition and (2) a product of increasing kerogen aromaticity (Zhang et al., 2012). Additionally, Liu and Wilcox (2012a,b) via molecular simulations found that the surface heterogeneity plays an important role in gas adsorption capacity due to the fact that various surface functional groups have different sorption energy, such as the oxygen-containing functional groups increase the adsorbed CO₂ density in the micropore structure.

In addition to organic matter, clay minerals have also been reported to participate in the methane sorption process in clay-rich shales due to their internal crystal layers or porous structures (Aringheri, 2004; Gasparik et al., 2012; Ji et al., 2012; Lu et al., 1995; Ross and Bustin, 2008, 2009; Schettler and Parmoly, 1990, 1991). Pores of 1–2 nm radius between crystal layers of clay minerals (mainly kaolinite, illite, and smectite) provide the adsorption sites for CH₄ and other gasses due to the large surface area (Cheng and Huang, 2004). Recent studies have also shown that diagenesis has erased the control of primary texture on bulk rock properties in the Barnett Shale, and primary detrital composition exerts its control on bulk rock properties only through its effect on the diagenetic pathway (Milliken et al., 2012a). However, some researchers argued that the contribution of clay minerals to the methane sorption capacity in the shales is irrelevant in the presence of moisture due to the high affinity of water to the hydrophilic clay surfaces that block the access of gas molecules to the sorption sites by filling pore throats or occupying sorption sites (Busch et al., 2003; Clarkson and Bustin, 1996, 1999, 2000; Joubert et al., 1974; Krooss et al., 2002; Ross and Bustin, 2007, 2008, 2009; Zhang et al., 2012). Additionally, the sorption energy for methane on clay minerals is far less than that of organic matter (Ji et al., 2012; Lu et al., 1995; Zhang et al., 2012). As a consequence, clay minerals may play a relatively less significant role in gas adsorption within organic shales.

The complex pore structures, such as geometry, pore sizes, PSD (pore size distribution) and connectivity, also have an important influence on gas sorption capacity of organic shales (Chalmers et al., 2012; Hao et al., 2013; Liu and Wilcox, 2012a,b; Mosher et al., 2013; Ross and Bustin, 2009). Ross and Bustin (2009) showed that the gas sorption capacities for the Devonian–Mississippian organic rich shales in the Western Canada sedimentary basin increase with increasing micropore volumes, and the gas sorption capacities for the Cretaceous organic rich shales in northeastern British Columbia display an increasing trend with microporous area, suggesting that gas adsorption is mainly associated with micropores. This may be attributed to large internal surface areas and greater adsorption energies of <2 nm pores compared with that of larger pores of solid with similar compositions (Dubinin, 1975; Ross and Bustin, 2009). Generally, larger pores have lower adsorbed gas density compared to smaller pores (Liu and Wilcox, 2012a,b; Mosher et al., 2013; Rexer et al., 2013). The change in adsorbed phase density is consistent with molecular simulations of CH₄ or CO₂ on porous carbon system such as coal and the kerogen organic matrix of gas shales (Liu and Wilcox, 2012a,b; Mosher et al., 2013), which also show that gas densities near pore walls are higher than in the pore center due to the overlapping potentials associated the strong gas–wall and gas–gas interactions.

Clearly, gas adsorption in organic shales is a complex multi-parameter process and elucidates and quantifies the effect of individual parameters which requires reliable experimental data at well-defined conditions. In this paper, a series of methane sorption isotherms for moisture-equilibrated organic shale samples have been measured at temperatures between 30 and 70 °C and pressure up to 20 MPa. The main objectives of the study were to (1) elucidate the most primary geological controls on gas sorption capacity in organic shales under reservoir conditions; (2) develop an accessible computational scheme of gas adsorption affected by combined individual variables, and (3) predict methane sorption capacity of organic shales under geological reservoir temperature and pressure conditions. Our studies are of great significance for the assessment and exploitation of natural gas in the shale reservoirs.

2. Samples and experimental methods

2.1. Samples

The Ordos Basin is an important hydrocarbon-containing basin situated in the central part of the North China Plate. It is a multicycle cratonic basin developed on the Archean granulites and lower Proterozoic greenschists of the North China block. The development of the Ordos basin during the Paleozoic–Mesozoic can be divided into three evolutionary stages: Cambrian–Early Ordovician cratonic basin with divergent margins; Middle Ordovician–Middle Triassic cratonic basin with convergent margins; and Late Triassic–Early Cretaceous intraplate remnant cratonic basin (Yang et al., 2005). Although the margins of the North China block–Ordos Basin experienced intense tectonic movements throughout its evolution, its interior has not been strongly folded and deformed because of the rigid basement. With less influence by later tectonic movements, the Paleozoic–Mesozoic strata in the principal part of the basin dip gently to the west with an angle of less than 1°. Primarily, three suites of effective source rocks were developed vertically in ascending order: lower Paleozoic marine carbonatite-type source rock, Upper Paleozoic Carboniferous–Permian coal and dark argillite-type gas source rock, and Mesozoic–Triassic Yanchang formation lacustrine dark argillite-type source rock.

The suit of studied shale samples originated from Upper Triassic Yanchang Formation lacustrine black shales in the southeastern Ordos Basin, China (Fig. 1). The Yanchang Formation extends over a wide area in Ordos Basin and offers the source rock for many commercial oil fields. From the Early Triassic to the Middle Cretaceous, the basin experienced continuous subsidence and three episodes of weak uplifts.

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