



Research paper

Estimation of marine shale methane adsorption capacity based on experimental investigations of Lower Silurian Longmaxi formation in the Upper Yangtze Platform, south China



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ABSTRACT

Methane adsorption experiments were performed on eight Lower Silurian Longmaxi moisture-equilibrated shale samples from the Upper Yangtze Platform, south China, at pressures up to 12 MPa, at 20 °C, 40 °C, 60 °C, 80 °C and 100 °C to investigate the effects upon methane adsorption capacity and to estimate the variation of methane adsorption capacity as a function of depth and burial history. The methane adsorption capacity of the Longmaxi shale shows a significant positive correlation with total organic carbon (TOC) content, which is accompanied by an increase in BET (Brunauer Emmett Teller) surface area and total pore volume. A quadratic relationship was observed between clay content and methane adsorption capacity. There is a threshold clay content (42.3–44.4%) in this trend. Methane adsorption capacity declines with increasing clay content below the threshold, and later increases with increasing clay content. The Langmuir pressure decreases exponentially with the reciprocal of temperature and the Langmuir volume decreases linearly with temperature. The isosteric heat of adsorption and standard entropy for the Longmaxi shale ranges from 10.34 to 11.67 kJ/mol and from –57.04 to –61.32 J/mol/K, respectively, which are clustered around kerogen type I. Using these relationships a computational scheme was developed to calculate methane adsorption capacity as a function of TOC content, temperature and pressure based on the Langmuir equation. This algorithm was applied to estimate methane adsorption capacity of the Longmaxi shale as a function of depth. Due to the dominating effect of pressure methane adsorption capacity increases initially with depth, through a maximum and then decreases as a result of increasing temperature at a greater depth. The maximum gas adsorption capacity is typically located at a depth range between 800 and 1350 m. With TOC content increasing, the curves shift continuously towards higher adsorption capacity. Furthermore, the maximum methane adsorption capacity moves higher and shifts to a deeper depth as TOC content increases. Ultimately, four principal dynamic evolutionary stages of methane adsorption capacity of the Longmaxi shale over geologic time as a function of burial history were reconstructed.

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1. Introduction

Shale gas is one of the most promising unconventional gas systems (Wang et al., 2014; Weijermars, 2015). 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; Jarvie et al., 2007; Hill et al., 2007; Strapoć et al., 2010; Hao et al., 2013). Natural gas storage in shale differs significantly from conventional gas reservoirs and may be composed of free compressed gas, adsorption gas and dissolved gas (Curtis, 2002; Ross and Bustin, 2007; Gasparik et al., 2012, 2014; Zhang et al., 2012; Hao et al., 2013; Rexer et al., 2013). Among these storage types, adsorption gas accounts for between 20% and 85% of the total gas in five US shale formations (Antrim shale, Ohio shale, New Albany shale, Barnett shale and Lewis shale) (Curtis,

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2002; Montgomery et al., 2005). In general, the free gas is primarily responsible for the gas-in-place of shale gas plays, whereas the adsorption gas partially determines the longevity of shale gas producing wells (Tan et al., 2014). Understanding the relative proportions of gas stored in these different forms is critical to an accurate assessment of shale gas resources and design of effective production strategies (Chalmers and Bustin, 2008; Ross and Bustin, 2008, 2009; Zhang et al., 2012). 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). The 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). Some researchers have recently studied shale gas adsorption characteristics and their geological controls, such as organic matter character (type, abundance, and maturity), composition of inorganic minerals (especially clay minerals), micro and nano pore structures, moisture content, and pressure and temperature regimes (e.g. Jarvie et al., 2007; Ross and Bustin, 2008; Strąpoć et al., 2010; Gasparik et al., 2012; Zhang et al., 2012; Hao et al., 2013; Rexer et al., 2013; Ji et al., 2014; Tan et al., 2014).

Organic matter present in these gas shales is generally thought to be the principal contributor to the gas adsorption capacity. A generally positive linear correlation between methane adsorption capacity and TOC (total organic carbon) content of organic shales was reported in previous studies (Jarvie et al., 2007; Ross and Bustin, 2007, 2008, 2009; Chalmers and Bustin, 2008; Strąpoć et al., 2010; Weniger et al., 2010; Gasparik et al., 2012, 2014; Zhang et al., 2012; Hao et al., 2013; Ji et al., 2014; Tan et al., 2014). With increasing thermal maturation, the TOC-normalized methane adsorption capacities increase (Zhang et al., 2012; Ross and Bustin, 2009; Tan et al., 2014). This may be caused by the increase in organopores and surface areas resulting from the thermal transformation of kerogen into hydrocarbons (Jarvie et al., 2007; Loucks et al., 2009; Slatt and O'Brien, 2011; Mastalerz et al., 2013; Milliken et al., 2013), and the increase in aromatization (Zhang et al., 2012) and pore surface homogeneity (Ross and Bustin, 2009) with increasing maturity. The methane adsorption capacities of varied kerogen were shown in the order: type I < type II < type III kerogen (Chalmers and Bustin, 2008; Zhang et al., 2012) and this was contributed to (1) the higher adsorption capacity of vitrinite compared to other maceral composition and (2) a product of increasing kerogen aromaticity (Zhang et al., 2012). In addition to organic matter, clay minerals have also been reported to participate in the methane adsorption process in clay-rich shales due to their internal crystal layers or porous structures (Ross and Bustin, 2008, 2009; Gasparik et al., 2012; Ji et al., 2012). However, some researchers argued that the contribution of clay minerals to methane adsorption capacity in shale is drastically reduced in the presence of moisture due to the high affinity of water to the hydrophilic clay surface that blocks the access of gas molecules to the adsorption sites by filling pore throats or occupying adsorption sites (Ross and Bustin, 2007, 2008, 2009; Zhang et al., 2012). Additionally, the adsorption energy for methane on clay minerals is far less than that of organic matter (Zhang et al., 2012; Ji et al., 2012). As a consequence, clay minerals may play a relatively less significant role in gas adsorption within organic shale. The complex pore structure also has an important influence on gas adsorption capacity of organic shales (Ross and Bustin, 2009; Chalmers et al., 2012; Hao et al., 2013). The gas adsorption capacities for the Devonian–Mississippian shale in the Western Canada sedimentary basin and the Cretaceous shale in northeastern British Columbia increase with increasing micropore volume and microporous area, respectively, suggesting that gas adsorption is mainly associated with

micropores (Ross and Bustin, 2009). This may be attributed to large internal surface area and greater adsorption energy of <2 nm pores compared with that of larger pores (Dubinin, 1975; Ross and Bustin, 2009).

Obviously, gas adsorption in organic shale is a complex multi-parameter process, which has been extensively studied. However, the estimation of adsorption gas under representative temperature and pressure condition was rarely studied (Khosrokhavar et al., 2014), in particular, the adsorption gas evolution of organic shale throughout geological history has not been extensively discussed. We have recently reported the estimation algorithm of methane adsorption capacity in lacustrine shale in the Ordos basin, China (Ji et al., 2014). In this study, a series of methane adsorption isotherms for marine shale samples were measured at temperatures between 20 and 100 °C and pressures up to 12 MPa under moisture-equilibrated conditions to (1) elucidate the most significant effects on methane adsorption capacity in marine shales and (2) outline how the methane adsorption capacity can be estimated as a function of depth and burial history of the Silurian Longmaxi shale in the Upper Yangtze Platform, south China.

2. Samples and experimental methods

2.1. Geological background and samples

The Upper Yangtze Platform is located in the western portion of the Yangtze Platform (Fig. 1A). The Lower Silurian shale, widely developed in the Upper Yangtze Platform, south China, has recently been selected as the key target for shale gas exploration and evaluation in the region (Wang et al., 2009; Pu et al., 2010; Nie and Zhang, 2012; Dong et al., 2012; Guo and Zhang, 2014). The gas-in-place of the lower Silurian shale has been estimated at 9.7 trillion cubic meters (EIA, 2011). The Silurian shales were deposited in a restricted marine basin environment (Tan et al., 2014). During the Early Silurian, a major transgression occurred towards the southwest resulting in widespread marine deposition of black shales in the Upper Yangtze Platform. The Lower Silurian strata experienced deep burial during the Early Mesozoic. This was followed by intense uplifting from the Late Mesozoic to the Cenozoic, which has greatly changed the accumulation and preservation conditions for shale gas (Zhu et al., 2006; Ma et al., 2008; Tan et al., 2015). At present, the burial depth of the Lower Silurian shale varies by location in the Upper Yangtze region, and is mostly within a range of 2000–7000 m in the Sichuan Basin (Zhou et al., 2014). In some strongly uplifted areas, the shales occur as outcrops. This indicates that the Lower Paleozoic shales in the Upper Yangtze region have undergone a complicated tectonic evolution which is a vital factor influencing the gas content in the shales. These tectonic movements were characterized by intensive uplift, erosion of overburden rocks, and faulting. Free gas stored in macropores and fractures in the organic-rich shales may be completely lost during the uplift and erosion phase, resulting in undersaturation (Hao et al., 2013).

The lower section of the Longmaxi formation mainly is comprised of dark gray–black arenaceous shale, carbonaceous shale and graptolitic shale interbedded with bioclastic limestone. The upper section is mainly comprised of greyish–yellowish green shale and arenaceous shale interbedded with siltstone and marlstone. High TOC sections mainly developed in the bottom part of the Longmaxi Formation. The TOC contents decrease and the color of the shale turns lighter upward with an increase in the calcareous and silt content (Dai et al., 2014). The Longmaxi shale is currently highly overmature, generating mainly dry gas and secondary gas (Tan et al., 2015). Most of the organic matter is amorphous, indicating an aquatic origin. The sapropelinite maceral accounts for

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