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Impact of total organic carbon and specific surface area on the adsorption capacity in Horn River shale

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ABSTRACT

Among unconventional gas reservoirs, shale gas has become an increasingly important source of natural gas supply in North America and around the world. Horizontal drilling and multi-stage hydraulic fracturing are the two key technologies for the economic development of shale gas reservoirs. Generally, natural gas in shale reservoirs is stored as free gas state in both larger mineral pores and natural fractures, as well as adsorbed gas state within organic matter and clay minerals. Since a considerable fraction of the gas in place (GIP) is in the adsorbed state, investigation of gas adsorption can provide critical insights into the evaluation of resources volume in shale. In this study, several experiments are carried out on fourteen shale core samples obtained in a vertical well drilled in Horn River Basin, Canada. The total organic carbon (TOC) content, specific surface area (SSA), and CH₄ adsorption isotherm are measured for the samples. In order to measure the volume of adsorbed gas at reservoir pressure, the high pressure Belsorp-HP equipment applying volumetric method is used. As results, it is observed that adsorption capacity is a function of not only the TOC but also the SSA. When the samples have similar TOC, adsorption capacity becomes higher as SSA increases and Langmuir volume increases too. We also consider the relationship between SSA and adsorption capacity for each kerogen quality that it shows a linear trend. Also, we attempt to find a correlation of TOC and SSA to the adsorption capacity by categorizing experimental results. Finally, gas in place calculation is carried out for the free gas and adsorbed gas in Horn River shale, respectively.

1. Introduction

Shale gas and coal-bed methane (CBM) are important unconventional gas reservoirs in which much of the gas is stored in an adsorbed state (Bustin, 2005a). Shale is a fine-grained sedimentary rock formed from the compaction of silt and clay-sized mineral particles. It is a member of the group of sedimentary rocks known as mudstones and has a laminated structure composed of many thin layers. Despite the very low permeability of shale, hydraulic fracturing makes commercial extraction of shale gas viable. Hydraulic fracture growth is mostly affected by the horizontal principal stress, and shale laminations restrict the hydraulic fracture height (Miskimins and Barree, 2003; Ferri et al., 2012).

From a geological viewpoint, shale includes organic matter when it is deposited, which leads to gas formation by biogenic and/or thermogenic processes, with the shale acting as both a source rock and a reservoir (Hill et al., 2007; Strapoc et al., 2010). Natural gas in shale exists in three forms: (1) free gas in larger mineral pores and natural

fractures, (2) sorbed gas adhered onto the organic matter, and (3) gas dissolved in oil and water (Zhang et al., 2012; Leahy-Dios et al., 2011; Clarkson and Hagshenas, 2013). In general, in the current industry standard calculations, gas dissolved in oil and water is not considered (Ambrose et al., 2010).

Because a considerable fraction of the gas in place may be in the adsorbed state, investigations of gas adsorption provide critical insights into the evaluation of the volume of resources in shale. Among several theories for the adsorption onto rock surfaces, Langmuir isotherm theory is widely accepted in the petroleum industry. The Langmuir equation is presented as Eq. (1) (Langmuir, 1918):

$$G_c = \frac{V_L P}{P + P_L} \quad (1)$$

where G_c is the adsorption volume of the gas at any pressure, V_L is the maximum volume of gas that can be adsorbed onto shale at an infinite pressure, and P_L is the pressure at which one-half of the Langmuir volume can be adsorbed (Fig. 1).

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Nomenclature

TOC	total organic carbon, %
SSA	specific surface area, m ² /g
BET	Brunauer-Emmett-Teller
T	temperature, K
P	pressure, MPa
n	adsorption mole
T_e	equilibrium temperature, K
P_e	equilibrium pressure, MPa
T_i	initial temperature, K
P_i	initial pressure, MPa
Z	non-ideal gas deviation factor
R	gas constant
SW	sample mass, g
G_c	adsorbed gas content of shale, SCF/ton
V_L	langmuir volume, SCF/ton

P_L	langmuir pressure, psi
$V(k)$	total adsorption volume at (k) th step, cm ³ /g
V_s	volume of reference cell
V_d	volume of sample cell – volume of sample
AGIP	adsorbed gas in place, BCF
GIP	gas in place, BCF
G_{st}	total gas storage capacity, SCF
G_f	free gas storage capacity, SCF
G_a	adsorbed gas storage capacity, SCF
A	area, acre
h	reservoir thickness, ft
ϕ	porosity
S_g	gas saturation
B_g	gas volume factor ($\frac{Z \times T_{res} \times P_{sc}}{P_{res} \times T_{sc}}$), ft ³ /SCF
ρ	density, g/cc

In previous studies, the adsorbed capacity of CH₄ has been reported to be correlated most strongly with its TOC (Manger et al., 1991; Lu et al., 1995; Ross and Bustin, 2007). The TOC represents the amount of organic carbon in a geological formation, particularly the source rock for a petroleum play and no unit exists to quantify it (EIA, 2013). Chalmers and Bustin (2007) reported a strong positive relationship between the micropore volume, methane sorption capacity, and organic matter content. Beaton et al. (2010) measured the methane adsorption isotherms of several shale samples from the Duvernay, Muskwa, Banff, and Exshaw formations. Nuttall et al. (2005) reported the methane and carbon dioxide adsorption isotherms of New Albany shales and Ohio shales extracted from various depths. They observed a correlation between the percentage of TOC and depth. A good shale play or a sweet spot must have a TOC greater than 5% (Ouafeul and Aliouane, 2015). Yu et al. (2014) used laboratory measurements to analyze the gas adsorption of four samples from the lower Marcellus shale. The relation between the TOC and gas storage capacity at a reference pressure of 5000 psi is shown in Fig. 2, which reveals a good linear relation between the two. Lu et al. (1995) described a laboratory apparatus and experimental methodology for accurately measuring gas adsorption in a Devonian shale under high pressure and over a wide range of temperatures. An approximately linear correlation between the adsorption capacities and the TOC of the shale samples was observed, suggesting that adsorption onto organic matter is a major mechanism in adsorbed gas storage.

The significance of shale mineral composition and pore structure has also been reported. Pore size distributions and surface-area

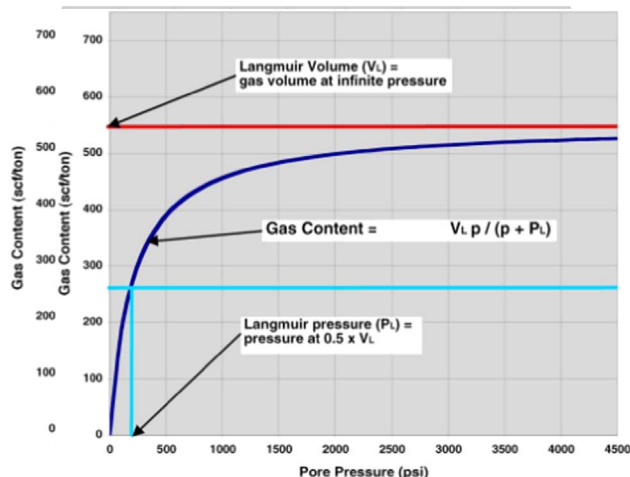


Fig. 1. Langmuir isotherm (Lewis et al., 2004).

heterogeneities at the nanometer scale make predicting the adsorbed gas capacity of shales on the basis of the TOC alone difficult (Ross and Bustin, 2009). The SSA, which is defined as the surface area per gram of solid, plays an important role in determining the adsorption capacity. A rougher solid surface and smaller pore size can increase the SSA (Solar et al., 2010) and a rough solid surface with many nanometer-scale cavities can adsorb gas more strongly than a polished surface (Rouquerol et al., 2013; Solar et al., 2010). The SSA can be calculated using the Brunauer–Emmett–Teller (BET) method (Brunauer et al., 1938).

Ross and Bustin (2007) also demonstrated that the organic matter mainly controls gas adsorption. TOC-lean shales have a low adsorbed gas capacity because minimal adsorption sites are available to methane. Most pores in shales are located in organic matter that is nanoporous material composed primarily of micropores, with a pore length less than 2 nm, and mesopores, with a pore length between 2 and 50 nm (Kang et al., 2011; Reed and Loucks, 2007; Loucks et al., 2009; Ambrose et al., 2010; Zhang et al., 2012). Recent work on siliceous mudstones from the Barnett shale shows that the pores are dominantly nanopores and most pores are in grains of organic matter as intraparticle pores (Loucks et al., 2009; Milliken et al., 2012). Ross and Bustin (2009) have also reported that microporosity associated with the organic fraction controls the CH₄ sorption and that the thermal maturation process affects both the volume of micropores and SSA. Generally, microporosity is positively correlated with the TOC in shales (Chalmers and Bustin, 2007) and with larger SSAs and higher adsorption energies. Therefore, thermally mature shales tend to exhibit larger micropore volumes and SSAs than immature shales, resulting in higher adsorption capacities (Zhang et al., 2012). These mechanisms

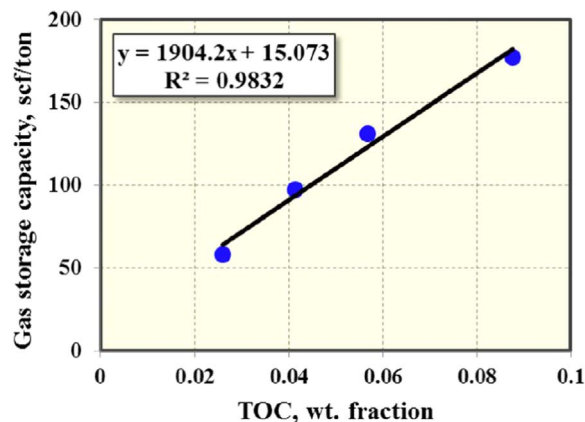


Fig. 2. Relationship between gas storage capacity and TOC (Yu et al., 2014).

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