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Research papers

The effect of water saturation on methane breakthrough pressure: An experimental study on the Carboniferous shales from the eastern Qaidam Basin, China

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A B S T R A C T

Breakthrough pressure plays an important role in shale gas flow, mining, and caprock evaluation. A series of breakthrough experiments were conducted under different water saturation conditions for four shales taken from the Carboniferous Hoit Taria Formation in the eastern Qaidam Basin, China to investigate the influence of water saturation on breakthrough pressure. Relevant geochemical tests (mineral composition, clay content, total organic carbon, thermal maturity and vitrinite reflectance) and micro structural characteristics of micro pores were also conducted. Breakthrough pressures under at least five different water saturations (from 0 to 100%) were obtained and relationship between breakthrough pressure and water saturation was fitted for each sample.

We found that breakthrough pressure increases exponentially with water saturation. The decrease in effective pore diameter caused by both the bound water films and the swelling of the clay minerals resulted in the increase in the breakthrough pressure. After water saturation reached about 60%, breakthrough pressure increased rapidly from connectivity reduction, caused by the sealing off of smaller pores and partial water saturation of the macropores. By analyzing the correlation between breakthrough pressure and pore structure characteristics, breakthrough pressure is inversely related to porosity, and is primarily affected by macropores. Because macropores consist of many microfractures with lengths up to dozens of micrometers, they determine the porosity and then affect the connectivity of the rock. Correlation analysis between the mineral compositions and breakthrough pressure showed that TOC content exhibits a positive correlation with breakthrough pressure, but neither quartz content nor the clay mineral content exhibits a correlation. By combining this information with Field Emission Scanning Electron Microscope results, we found that microfractures are easily created where the TOC (total organic carbon) is concentrated, and these microfractures substantially contribute to the number of macropores. However, dissolution pores in the quartz and clay minerals are rare. Therefore, the correlation between TOC content and breakthrough pressure primarily results from the microfractures. This study provides practical information for further studies of shale gas migration as well as shale gas mining and caprock evaluation.

1. Introduction

Shale gas is sometimes referred to as an unconventional energy source (natural gas—mainly methane (CH_4)) mined from low-permeability, organic-rich shale (Chapiro and Bruining, 2015; Dong et al., 2015). Global shale gas resources are abundant and could be as high as 208 trillion cubic meters according to the International Energy Agency (Bocora, 2012). In recent years, shale gas resources have attracted attention in many countries. Petroleum geoscientists have responded to severe energy shortages and the rapid growth of energy prices, with increased exploration of shale gas over the past decade (Chapiro and Bruining, 2015; Wang et al., 2016). Shale gas is abundant and it is regarded as an important additional, but unconventional, energy source in Australia (Cook et al., 2013). The United States has also been involved in shale gas development and has even started a 'shale gas revolution' (LaShelle, 2015; Curtis, 2002). The Chinese government also began to switch energy supplies from conventional energy to unconventional oil and gas resources such as shale gas. Shale gas is the third largest source of natural gas in China (Zhong et al., 2015). With the emergence of horizontal well and hydraulic fracturing technology,

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shale gas production has been promoted significantly on a global scale (Chapiro and Bruining, 2015; Romero-Sarmiento et al., 2013).

With low porosity and permeability, shales are not only source rocks but also reservoir rocks and caprocks (Al-Bazali et al., 2005). Capillary pressure ($P_{c,e}$ and $P_{c,b}$) is closely related to the formation of gas reservoir, gas flow, and gas exploration. Capillary pressure (P_c) plays a very important role in the multiphase (water-gas-rock) flow mechanism in low-permeability media (Rezaeyan et al., 2015; Xu et al., 2015; Boulin et al., 2013). Capillary pressure in low-permeability porous media can be described by the Laplace equation:

$$P_c = P_g - P_w = \frac{2\sigma\cos\theta}{r}, \quad (1)$$

where P_g and P_w are pressure of the gas phase and the water phase, respectively, σ is the interfacial tension, θ is the contact angle, and r is the radius of the largest pore throat in the porous media. In physical experiments, definitions of capillary pressure in the different point during the displacement process are slightly different. The pressure at which the gas phase starts to enter the rock core from one side and water is expelled is defined as the capillary entry pressure ($P_{c,e}$) (Rezaeyan et al., 2015; Boulin et al., 2013). When the gas phase breaches the rock and gas is detected at the outlet, the gas pressure is defined as the breakthrough pressure ($P_{c,b}$). Capillary breakthrough pressure refers to the critical capillary resistance pressure that the non-wetting phase (gas) has to overcome to penetrate into a pore throat and pass through the wetting phase (water) (Hildenbrand et al., 2002; Li et al., 2005; Aggelopoulos et al., 2010). However, many researchers found that it was difficult to precisely distinguish between the capillary entry pressure and the breakthrough pressure in low-permeability porous media using displacement experiments and they considered the two pressures to be almost equal (Rezaeyan et al., 2015; Boulin et al., 2013). Only when the pressure of the gas generated from organic matter is higher than $P_{c,e}$ (almost equivalent to $P_{c,b}$) (Hildenbrand et al., 2002; Rezaeyan et al., 2015), can shale gas overcome the flow resisting force originating from the capillary effects and continuous flow pathways formatted in the rock (Xu et al., 2015). As a result, shale gas migrates and accumulates along the pathways to form the gas reservoir. This process determines the hydrocarbon expulsion amount. In the exploitation of low porosity and permeability reservoirs, only when the gas pressure is higher than the $P_{c,e}$ ($P_{c,b}$) of the formation fracture network, can free gas in the formation fracture networks be transported to the wellbore under the pressure difference (Zhang et al., 2015). The capillary pressure generated by the interfacial tension of the liquid-solid-gas interface causes a decline in gas reservoir permeability and results in the water lock effect that greatly reduces natural gas production (Shang et al., 2008). However, when shales act as caprocks, the capillary breakthrough pressure must exceed the pressure difference between the gas phase (mainly CH_4) and the wetting phase (water) to avoid gas leakage (Kima and Santamarina, 2013; Li et al., 2015; Liu and Birkholzer, 2012). Therefore, breakthrough pressure can be regarded as a comprehensive index for estimating shale gas potential (Li et al., 2015).

Many experiments have been conducted on capillary breakthrough pressure. Li et al. (2015) found that it is very difficult for gas to break through fully saturated shale and a long time is needed for the breakthrough process, which can be divided into three stages. However, only two samples were conducted breakthrough pressure experiments and relationship between water saturation and breakthrough pressure hasn't been studied. Hildenbrand et al. (2004) conducted gas breakthrough experiments on intermediate to low-permeability sedimentary rocks using nitrogen (N_2), carbon dioxide (CO_2), and CH_4, and they attributed the systematic differences during the gas breakthrough process to differences in wettability and interfacial tension. Using residual capillary pressure (RCP) experiments, Hildenbrand et al. (2002) found that capillary breakthrough pressure measured by the RCP method was lower than the results from drainage experiments because re-imbibition was impeded. Using both step-by-step (SBS) and RCP methods, Rezaeyan et al. (2015) found, that capillary breakthrough pressure increased with decreasing temperature, with a clear increase in breakthrough time. By comparing experimental investigations, Boulin et al. (2013) found that, although the SBS method is time-consuming, it is the most representative approach of in situ hydrocarbon migration through caprocks.

Breakthrough experiments have been investigated on dry or fully water-saturated cores by previous researchers. However, naturally-occurring shales in the depths of the earth are partially water-saturated (Haghighi and Ahmad, 2013; Li et al., 2016), so breakthrough experiments on partially water-saturated cores are needed for improving our understanding of breakthrough pressures.

A previous study has already investigated the effects of pore structure and mineral composition on breakthrough pressure (Li et al., 2015). However, only two dry and fully water-saturated samples were used in the breakthrough experiments, and attention was not paid to the relationship between water saturation and breakthrough pressure. Thus, in this study, four shale samples were chosen from the Chaiye2 borehole, located in the eastern Qaidam Basin, to investigate how factors, especially water saturation influence breakthrough pressure. The water saturation process for shale is difficult and time-consuming because of the low permeability and porosity, and the shale may be crushed under the high confining pressure. Moreover, specific water saturation is not easy to obtain. Therefore, we designed a device to saturate the shale with water that guarantees the shale can be saturated completely without being crushed. As part of this procedure, weighed the shale every half an hour to get representative water saturation data. The mineral composition was measured using the X-ray diffraction (XRD) technique, and pore size distributions were analyzed by N_2 and CO_2 adsorption and high pressure mercury intrusion methods. During our breakthrough experiments, each shale core was saturated to at least five different levels of water saturation. After the breakthrough experiments, the effects of water saturation, microstructural characteristics, and main mineral compositions on breakthrough pressure were analyzed. In addition to these parameters, gas composition, temperature and ion compositions of underground water also have an effect on breakthrough pressure, but these are not the main focus of this study and will be emphasized in the future research. The results of this study provides useful information on shale gas migration, which could influence further shale gas mining and caprock evaluation.

2. Experimental materials and apparatus

2.1. Shale samples

As one of the three largest oil and gas bearing basins in the northwest of China, the Qaidam Basin offers opportunities for exploitation (Li et al., 2014; C.L. Liu et al., 2012; Ma et al., 2012). Carboniferous deposits are widely developed in the eastern Qaidam Basin, and geologists have found oil sands in the Carboniferous deposits in recent years (Xu et al., 2014), which indicates the Carboniferous deposits have hydrocarbon-production potential and facilitate the migration of oil and gas. In addition, geoscientists have found that the Carboniferous deposits were not only unmetamorphosed, but also formed in shore-platform facies under warm and wet climate conditions (Li et al., 2014, 2015; C.L. Liu et al.,