



Research paper

Water activity characteristics of deep brittle shale from Southwest China

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ABSTRACT

Understanding the effect of water activity on shale is of major importance for the exploration and development of shale gas reservoirs, especially in the drilling and completion processes. Deep brittle shale samples from the lower Silurian Longmaxi Formation located in Southwest China were characterized using X-ray diffraction (XRD), swelling ratio tests, computerized tomography (CT), Field Emission Scanning Electron Microscope (FE-SEM) and uniaxial compressive strength (UCS) tests. Because of the physical characteristics of the samples, the relationship between water activity and swelling ratio could be approximated by the Frenkel–Halsey–Hill (FHH) equation. The relationships between clay minerals, UCS and water activity were investigated. Based on the physical description of the internal structure, the impact of water activity on micro-fractures was also discussed. The results showed that multilayer adsorption was the main contributor to the swelling of the deep brittle shale samples. The swelling ratio of brittle shale samples was described by the water activity of external environment via a logarithmic expression of the FHH equation, with fractal dimensions ranging from 2.71 to 2.75. The mineralogical composition was a controlling factor on water activity, as shown by the positive correlation between water activity and the total content of clay minerals (TCCM), especially the amount of montmorillonite in illite/smectite mixed layer clay. When soaking in a solution with a water activity above the critical water activity, the brittle samples developed micro-fractures because the clay swelled, acting as cementing material in the weakened planes. The environmental water activity was used to evaluate mechanical properties of the hydrated brittle shale. Brittle shale samples with larger water activity had a higher critical water activity. Water activity analysis led to a better understanding of the characteristics of the brittle shale and the performance of the downhole fluid.

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1. Introduction

Shale gas has recently received attention in the field of energy production (Loucks and Ruppel, 2007; Gasparik et al., 2012). As an effective way to cope with energy shortages and energy security, as previously proven by the USA during the past few decades, shale gas exploration is a viable option in China, especially in the southwest Sichuan Basin (Chen et al., 2011; Wang et al., 2013). Compared with the Mesozoic and Upper Paleozoic erathem as the target stratum for the USA's shale gas industry, the Lower Paleozoic erathem for the Sichuan Basin is deeper (Zhang et al., 2009). To reduce the cost of shale gas production, China still requires fundamental research on the physicochemical properties of shale at increasing depths (Chen et al., 2011; Wang et al., 2013).

As a type of mud rock, shale is a fine-grained rock mainly composed of clay mineral flakes (Blatt and Tracy, 1996). Clay minerals form in the presence of water (Ehlmann et al., 2011). The system, consisting of clay minerals and water, has a significant effect on the physicochemical properties of the shale, which has been reported and observed by

using modern technologies. Water content plays an important role in the mechanical properties of shale, especially the uniaxial compressive strength (UCS) (Hsu and Nelson, 2002; Vales et al., 2004; Erguler and Ulusay, 2009). It has also been shown to be related to the meso-scale damage characteristics of shale hydration when studying the internal morphology by computerized tomography (CT) (Shi et al., 2012; Ma and Chen, 2014). Clay minerals can contain and adsorb water because of their internal structure and type, which can be detected by X-ray diffraction (XRD) analysis (Ballard et al., 1994; Josh et al., 2012), and also has been proved to be a controlling factor on bedding and deformability, such as swelling (Wright, 2001; Fouche et al., 2004). However, compared with water content, water activity has a closer relationship with the physicochemical properties of the material (Troller and Christian, 1978; Rockland and Nishi, 1980). Because water content cannot reflect the influence of water on the material, water activity, as a description of energy state, can precisely reveal the relationship between water and the material. Water activity is a criterion used to measure the amount of energy that would be needed for the water to be removed from the material. The effect of environmental water activity on the mechanical properties of a shale, such as the reduction of the compressive strength caused by high water activity, has been observed by several researchers

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in laboratory experiments and field cases (Chenevert, 1970; Chen et al., 2003; Al-Bazali et al., 2008), but to date, no report on the water activity of a shale and its impact on the shale physical and mechanical properties has been published. This study focused on shale characteristics from a water activity perspective.

The major goals of this paper are to investigate the water activity characteristics of deep brittle shale from the Sichuan Basin in China. An equation of a similar form to the Frenkel–Halsey–Hill (FHH) equation, an isotherm equation containing the fractal dimension and describing the surface coverage as a function of the equilibrium pressure, is applied to investigate the relationship between water activity of the external environment and the swelling ratio of the brittle shale. The relationship between the mineralogical composition and water activity of the brittle shale will also be investigated. Furthermore, based on the physical significance of water activity, the effects of the water activity in the external environment on the micro-fracture mechanisms and mechanical properties of brittle shale will also be discussed.

2. Experimental procedures

2.1. Materials

Cores obtained from the Sichuan Basin located in Southwest China were used in this study (Fig. 1). The cores consisted of black shale and were obtained from the lower Silurian Longmaxi Formation (Fig. 2). Geological evidence indicates that the Longmaxi Formation formed in a shelf facies sedimentary environment (Wang et al., 2013). The stratigraphy of the lower part of the Longmaxi Formation consists of silty mudstone, silt and carbonaceous mudstone and carbon mudstone. With TOC (total organic carbon) $\geq 0.57\%$, it has a

cumulative thickness of 153 m. For the formation, the type of organic matter is I–II₁, a favorable type for shale gas generation with an R_0 (vitrinite reflectance) value $\geq 2.0\%$, the formation was in the shale gas generation stage. Test samples from the formation were cut from preserved cores obtained from depths ranging from approximately 4350 m to 4370 m and 2377 m to 2415 m, respectively. More detailed information on the stratigraphy, geology and petroleum potential of these cores can be obtained from Wang et al. (2013) and the references therein.

The Lower Cambrian Niutitang Formation may also be a shale gas reservoir (Fig. 2). The strata were below the Longmaxi Formation and formed in a similar sedimentary environment (Nie et al., 2011). The stratigraphy of the upper part of the formation consisted of gray silty mudstone, siltstone and limestone, whereas the lower part of the formation consisted of black shale (Yang et al., 2014). Relevant detailed geological information can be found in Nie et al. (2011) and Wang et al. (2013). Because of the geological correlation between the Longmaxi and the Niutitang Formations, the latter was used as a comparison to the former.

2.2. Equipment

The samples were analyzed experimentally using XRD analysis, water activity tests, swelling ratio tests, CT analysis, Field Emission Scanning Electron Microscope (FE-SEM) analysis and UCS tests (Chalmers et al., 2012).

XRD analysis was performed with a MiniFlex II Desktop X-ray diffraction instrument from Rigaku Corporation in Japan. The sample preparation, experiment operation and quantitative analysis were performed according to the U.S. Geological Survey Open-File Report

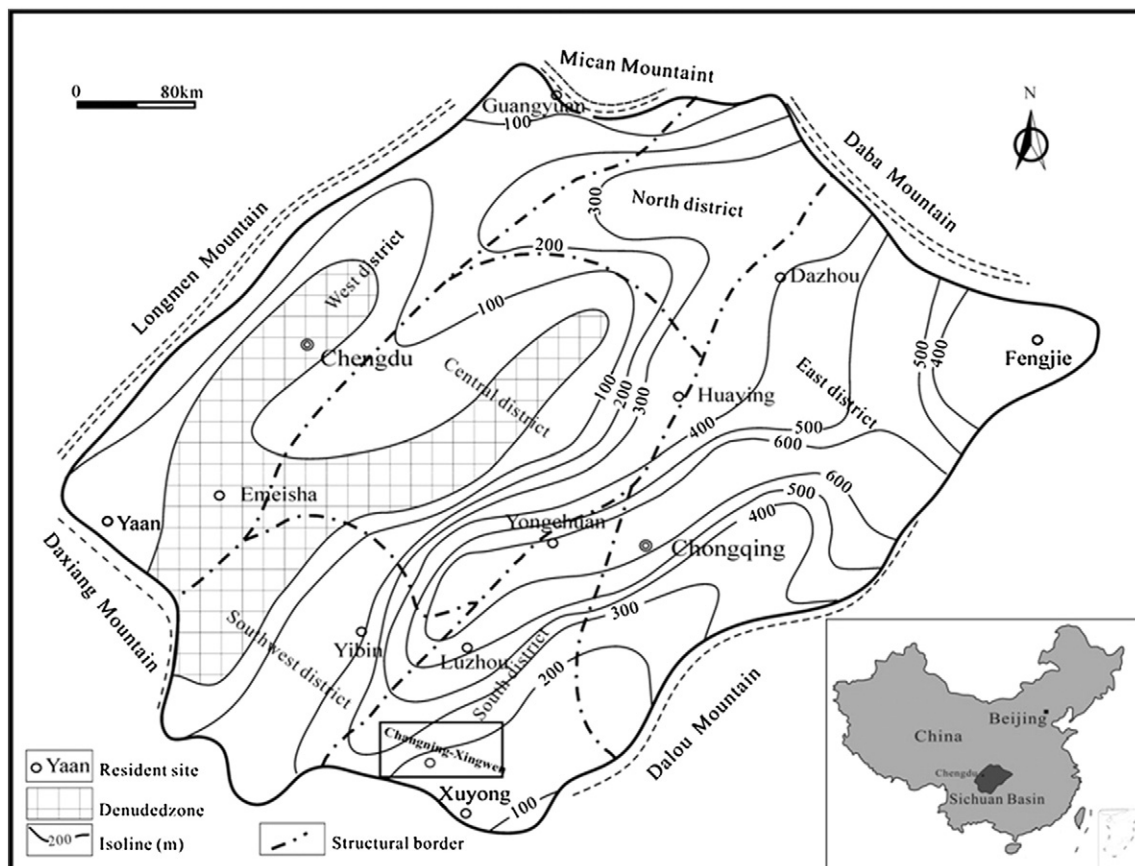


Fig. 1. Isopach map of the Longmaxi Formation in the southern Sichuan Basin (Chen et al., 2011).

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