Journal of Natural Gas Science and Engineering 55 (2018) 191–205

Contents lists available at ScienceDirect

Journal of Natural Gas Science and Engineering

journal homepage: www.elsevier.com/locate/jngse

Porosity-preserving mechanisms of marine shale in Lower Cambrian of Sichuan Basin, South China

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ARTICLE INFO

Keywords:
Lower cambrian
Marine shale
Pore preserving mechanism
Connectivity
Thermal maturity
Sealing ability

ABSTRACT

Porosity preservation is of great significance in the capacity of gas shale reservoir. Whether pore space can be preserved at the later diagenetic stage controls the capacity of shale reservoir. In this study, porosity-preserving mechanisms of marine shale from Lower Cambrian, both inside and at the margin of Sichuan Basin, South China, were investigated via a combination of the X-ray diffraction (XRD), organic geochemistry analysis, gas adsorption analysis and focused ion beam milling-scanning electron microscopy (FIB-SEM) in both two-dimension (2-D) and three-dimension (3-D). The results showed that there was little difference in mineralogical compositions between samples of Qiongzhusi shales and Niutitang shales that brittle minerals and clay were predominate components. The mean value of the total organic carbon (TOC) of Qiongzhusi shales was 1.44 wt% lower than that of Niutitang shales (4.37 wt%), whereas Qiongzhusi shales were at relatively lower thermal maturity stage with equal-vitrinite reflectance (R_o) ranging from 2.66% to 2.8% compared with Niutitang shales (3.06% < R_o < 3.59%). Nitrogen (N_2) and Carbon dioxide (CO_2) adsorption revealed that micropore-mesopore (0.4–60 nm) volumes (PV) of Qiongzhusi shale samples (averaging 0.0305 ml/g) were higher than those of Niutitang shale samples (averaging 0.0251 ml/g), whereas the mean micropore-mesopore surface area (PSA) of Qiongzhusi samples (20.77 m^2/g) was relatively lower than that of Niutitang samples (26.71 m^2/g). FIB-SEM (2-D) observation manifested three distinctive pore types in terms of pores of rigid framework, pores associated with clay minerals and organic matter (OM)-hosted pores. FIB-SEM (3-D) reconstruction illustrated that pore networks possessed appropriate connectivity in the upside of Qiongzhusi shale samples.

These observations suggest that brittle mineral grains can function as rigid frameworks or grain supporters due to high brittleness which preserves ductile matrix relating pores (OM-hosted pores and clay associated pores) from collapse by sharing the effective stress; in addition, either excessive OM abundance (TOC > 5.0 wt%) or over thermal maturity (R_o > 3.3%) is detrimental to pore preservation; moreover, poor sealing ability of shale systems might deteriorate pore preserving ability through losing overpressure environment or stress supporting mechanism.

1. Introduction

In order to meet the rapidly increasing demand of energy consuming, shales, which were regarded as both sources of and seals of hydrocarbons in the past, now convert to economic hydrocarbon reservoirs (Curtis et al., 2010). Owing to the breakthrough of drilling completion technologies (the horizontal drilling and the multi-stage hydraulic fracturing), the low porosity/low permeability of gas-bearing shales can be spurred and industrial gas flow and long duration of gas production can be achieved (Curtis, 2002; Hill et al., 2007). It is reported that shale gas, which stemmed from OM and stayed within shale systems, generated through either biogenic or thermogenic progress (Hao et al., 2013). In the United States, the production of natural gas increased by nearly 48% from 2005 to 2016 largely depending on the rapid development of shale gas exploration (EIA, 2017). While, in China, the Upper Yangtze Platform is currently deemed as the main

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https://doi.org/10.1016/j.jngse.2018.05.002
Received 20 November 2017; Received in revised form 3 April 2018; Accepted 4 May 2018
Available online 08 May 2018
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target area for conducting shale gas exploration and development, especially inside and at the margin of Sichuan Basin (Liu et al., 2015; Wang et al., 2016). Vigorous exploration and active development have been operated on the two main marine shale-gas-producing series in both Longmaxi shales of Lower Silurian and Qiongzhusi shales (Niutitang shales) of Lower Cambrian (Sun et al., 2017; Yang et al., 2017a, 2017b).

Although the resource of shale gas has become a favorable alternative and the exploration of these self-sourced reservoirs has spread worldwide, shales still remain poorly understood as reservoirs which partly lie with the complicated microstructure compared with conventional ones (Curtis et al., 2010). It is reported that natural gas within shales performs as three main occurrences: free phase, adsorbed phase and little dissolved state partly hinging on pore properties in nanoscale, which are of great importance to gas storages and permeability of hydrocarbons (Chalmers et al., 2012a, 2012b; Zhang et al., 2012).

In order to figure out pore properties qualitatively and quantitatively, intensive researches have been conducted on the analysis of pore microstructure. The pore properties can be represented as many parameters including pore width, pore volume, pore surface area and spatial distribution (Curtis, 2002; Loucks and Ruppel, 2007; Loucks et al., 2009, 2012). Also pore types can be classified into three categories based on various diameters: macropore (> 50 nm), meso-pore (2 nm–50 nm) and micropore (< 2 nm), according to the definition provided by the International Union of Pure and Applied Chemistry (Sing et al., 1985). Generally, macropores provide storage spaces and pathways for free gas (measured by high-pressure Hg analysis quantitatively), whereas adsorption and desorption occur in micropores (tested by CO_2 adsorption analysis quantitatively). Mesopores act as a transitional type which can be documented by N_2 adsorption analysis quantitatively (Clarkson et al., 2013; Tian et al., 2013). With these physical methods harnessed, pore features such as pore volume, pore surface area and pore size distribution can be obtained but in indirect ways.

In 2007, scanning electron microscopy (SEM) combined with Ar-ion polishing initially allowed nano-scale pores in shales to be observed directly in 2-D imaging (Loucks and Ruppel, 2007), which facilitated pore classification in origin. Loucks et al. (2012) proposed a pore category consisting of three major matrix-related types in terms of interparticle pores observed between particles and crystals, intra-particle pores developing within particles and OM pores which were interparticle ones but associated with OM. In addition, Loucks and Reed (2014) defined depositional and migrated OM (applied by this paper), as well as OM associated pores, by identifying sequence between mineral cementation and OM migration through a field-emission scanning electron microscope (FE-SEM). Moreover, 3-D reconstructions of marine shale samples from nine different formations (Barnett, Eagle Ford, Fayetteville, Floyd, Haynesville, Horn River, Kimmeridge, Marcellus and Woodford) were generated from focus ion beam (FIB) and SEM imaging which reinforced the facts that shales could vary dramatically either between plays or within the same one and that their nano-pore constructions were highly complicated and heterogeneous (Curtis et al., 2012).

Attributes of shale reservoirs, in terms of storage capacities, gas occurrences and flow pathways, are largely administrated by their nano-pore constructions. Several factors may lead to their differences mentioned above, including TOC, kerogen types, mineralogical composition and thermal maturity (Fishman et al., 2012; Mastalerz et al., 2013; Ma et al., 2015). Unlike the marine gas shales in the United States, the marine shales in the Upper Yangtze Platform, China, possess very high maturity. Especially in Sichuan Basin, marine shales have experienced multiple tectonics and deep burial, which are featured by over-thermal maturity with R_o = 2.0%–4.0% (Huang et al., 1974; Liu et al., 2015; Wang et al., 2016; Zhou et al., 2016). Whether pore space can be preserved or not at the later diagenetic stage hinges on the porosity-preserving mechanisms to a great extent. Hence, researches on the preserving mechanisms of pore space will be of great importance to the evaluation and prediction of sweet spots in South China.

In order to ascertain porosity-preserving mechanisms of marine shales, Lower Cambrian shales in different regions with various productions were selected to conduct comparable analyses in this paper. Methods, in terms of the X-ray diffraction (XRD), chemistry geology measurements, FIB-SEM 2-D imaging, FIB-SEM 3-D reconstruction and N_2/CO_2 adsorption, were integrated with each other synergistically. In details, we document and illustrate pore structure (size, distribution, arrangements, and origins of the pores) of gas shale. The controls of organic matter richness, thermal maturity and mineralogy on porosity were examined. Besides, the influence of sealing ability on pore preservation was analysis. The main work and results obtained from this paper are summarized as follows: 1) both prevalent mineral components and organic geological parameters were analyzed and compared among samples; 2) pore properties such as categories, size arrangements, volumes, surface areas and origins were documented; 3) the connectivity of pore networks was made a contrast across different shale samples; 4) samples (#5, #7, #9 and #11) selected from the bottoms of both Qiongzhusi Formation and Niutitang Formation with feeble pore properties were analyzed. Finally, mechanisms of pore preservation were discussed and concluded.

2. Geological setting and samples

Sichuan Basin, a superimposed basin with a diamond shape, is subordinate to the Upper Yangtze Block which locates in the south of China (Fig. 1a). The basin has experienced three important evolutionary stages including 1) rifting stage inside of the carton and at its margin in the period of Neoproterozoic, 2) depression stage in the craton during Sinian—Late Triassic phase and 3) the evolutionary stage of foreland basin at Late Triassic—Cretaceous period (Zhu et al., 2007). Marine sedimentary developed in Sinian—medium Triassic strata with a gross thickness of 4000–7000 m (Guo et al., 1996; Li et al., 2015). These marine sequences have experienced complex tectonic movements in terms of Tongwan movement, Caledonian movement and Indosinian movement etc., which consequently resulted into multiphase palaeo-uplifts and multiple regional unconformities (Fig. 1b) (Huang et al., 1974).

Qiongzhusi Formation in southwestern district (locating in the southwest of Sichuan Basin) and Niutitang Formation in the northeast of Chongqing Province (siting at the margin of Sichuan Basin in northeast) were both deposited during Early Cambrian (Fig. 1a and b), named differently by locations of target areas. Regional unconformity developed between the bottom of Lower Cambrian and the roof of Upper Sinian either inside or at the margin of Sichuan Basin (Fig. 1b) (Wei et al., 2015; Ma et al., 2015). Also depositing in marine abyssal continental-shelf environment, both Qiongzhusi shales and Niutitang shales were dominated by type I kerogen deriving from lower grade aquatic lives (Dai et al., 2014). Intensive tectonics, in terms of uplifts, denudations and deformations, brought about complex burial histories and various thermal evolution progresses in different regions. Compared with shales of Lower Cambrian series in southwestern district of Sichuan basin, thermal maturity degrees might be higher at the margin of northeastern Sichuan Basin corresponding to the deeper ancient burial depth (Zhou et al., 2016; Wang et al., 2016).

In this paper, 11 samples were selected out of 120 core plugs sophisticatedly to conduct comparable analysis, which were all collected from successfully drilled wells in two regions of Sichuan Basin with Well JY1 in southwestern district of Sichuan Basin and Well YC2, Well YC3 and Well YC1 in the northeast of Chongqing Province (at the margin of Sichuan Basin) respectively (Fig. 1).

3. Experimental procedures

A Bruker D8 DISCOVER diffractometer was used to measure bulk

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