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Origin of authigenic quartz in organic-rich shales of the Wufeng and Longmaxi Formations in the Sichuan Basin, South China: Implications for pore evolution

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ABSTRACT

Primary sedimentary composition of shales and subsequent diagenetic modifications are key factors in controlling the pore development and preservation in shale gas reservoirs. Thin section examination, X-ray diffraction analysis, scanning electron microscopy (SEM) determination with cathodoluminescence and geochemical analyses were combined to investigate Wufeng and Longmaxi shales of the Sichuan Basin. These experimental methods allowed us to focus on the origin of authigenic quartz in organic-rich shales and evaluate its importance in the evolution of shale gas reservoirs. The siliceous shales in Wufeng and Longmaxi Formations contain up to 60% microcrystalline aggregates of quartz, which are more abundant than silt-size detrital quartz to suggest an authigenic origin. Most authigenic quartz originates from biogenic siliceous dissolution and re-precipitate. Therefore, high excess silica concentration indicates a higher surface productivity with more organic matter (OM) deposition. The abundance of OM pores is positively correlated with the excess-Si content ($R^2 = 0.77$). We suggest that the migrated OM filled in authigenic microcrystalline quartz aggregates is the principal matrix for OM pore development. Authigenic microcrystalline quartz aggregates filled in primary pore space and destroyed primary interparticle porosity. However these authigenic quartz can restrain compaction and preserve the internal pore structure as a rigid framework. Pore space is abundant enough in microcrystalline quartz aggregates for oil and bitumen filling during the oil window. Migrated OM filled the associated pore network in authigenic microcrystalline quartz aggregates would more likely to generate a better three-dimensional OM pore network through secondary cracking and provides favorable reservoir spaces for shale gas.

1. Introduction

Shale gas has a great resource potential to attract exploration in many countries. Shale deposition and diagenesis have sparked an increasing interest to the sedimentary geologists and petroleum geologists over the past decade (Schieber et al., 2000; Macquaker et al., 2007, 2010; Ross and Bustin, 2009; Thyberg et al., 2010; Aplin and Macquaker, 2011; Zeng et al., 2011; Milliken et al., 2012; Zou et al., 2012; Jiang et al., 2013; Hart et al., 2013; Lazar et al., 2015; Jin et al., 2016). The primary sedimentary composition of shales and subsequent diagenetic modifications are key factors controlling the pore development and preservation in shale gas reservoirs. (Aplin and Macquaker, 2011; Fu et al., 2011; Milliken et al., 2012; Loucks et al., 2012; Liang et al., 2012; Macquaker et al., 2014; Wang et al., 2015; Baruch et al., 2015; Zhao et al., 2016a). Quartz is an important component of the organic-rich shale, and its content not only controls the physical properties of shales, but also influences the hydraulic fracturing process (Jarvie et al., 2007; Ding et al., 2013; Zeng et al., 2013). Authigenic quartz precipitated during

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the diagenetic process is a major component of several organic-rich shale (e.g., Barnett, Woodford, Muskwa, Marcellus) and is possibly far more abundant than detrital silt-size quartz (Loucks and Ruppel, 2007; Milliken et al., 2012; Hart et al., 2013). The source material and formation mechanism of authigenic quartz in shale are a complicated subject for geologists to tackle (Schieber et al., 2000; Peltonen et al., 2009; Milliken et al., 2012).

The origin of authigenic quartz has yet to be completely understood. Siliceous ooze (i.e. diatoms, radiolarians, silicoflagellates, and sponges) with an opal mineralogy will undergo a transformation to opal CT and to cryptocrystalline quartz during diagenetic process (Kastner et al., 1977; Schieber et al., 2000). This dissolution-reprecipitate process may simply transform the siliceous particles in place or re-precipitate dissolved silica in pores (Loucks and Ruppel, 2007). The conversion of smectite to illite will release an amount of silica, which may be further precipitated as quartz (Hower et al., 1976; Foster and Custard, 1980; Bjørlykke, 1998; Van der Kamp, 2008; Peltonen et al., 2009). Other sources of authigenic quartz may come from pressure solution of detrital quartz, sericitization of illite, illitization and chloritization of kaolinite, as well as the alteration of detrital feldspars in shale (Cyziene et al., 2006). However, authigenic quartz is difficult to observe in shale, and there are few reports focusing on its petrographic evidence (Peltonen et al., 2009). The presence of authigenic quartz may be an important factor controlling the physical properties of shale.

Organic matter (OM) pores are mainly of nano-scale, and have been recognized as an important component of the pore system in shales (Loucks et al., 2009, 2012; Milner et al., 2010; Curtis et al., 2012; Pommer and Milliken, 2015). Current understanding indicates that the abundance of OM pores is related to the thermal maturity and types of OM (Loucks et al., 2009; Ambrose et al., 2010; Passey et al., 2010; Fishman et al., 2012; Pommer and Milliken, 2015). During different thermal evolution stages, OM pores can develop not only in kerogen and solid bitumen, but also in pyrobitumen (Loucks et al., 2009; Bernard et al., 2012a, 2012b; Schieber, 2013; Reed et al., 2014). Therefore, OM pore networks are related to the occurrence of different OM types (e.g., kerogen and bitumen) (Bernard et al., 2012a,b; Loucks and Reed, 2014), which are controlled by the interaction of the OM and minerals. There is a close relation between authigenic quartz and OM. However, the physical interaction of authigenic quartz with OM is still poorly understood and needs further research.

Organic-rich shale layers of the Wufeng and Longmaxi Formations are key targets for shale gas exploration and development in China. The objectives of this paper are to (1) identify the origin of authigenic quartz and its diagenetic evolution, and (2) decipher the role of authigenic quartz on the evolution of shale pores. The research will bear conclusions of great theoretical significances on understanding the role of diagenesis in shale reservoirs development, but it also brings important practical concepts for predicting favorable reservoirs for shale gas.

2. Geological setting

From the Late Ordovician to Early Silurian, the Caledonian orogeny reached a maximum intensity, compressing the Yangtze Platform and developing the Chuanzhong uplift in the northwestern Yangtze Platform, the Qiangzhong uplift in the southern and the Jiangnan-Xuefeng uplift in the southeastern areas (Liang et al., 2009). Due to the compression and development of several uplifts around the Sichuan Basin, the marine area was reduced to the northeast, east and southeast Sichuan Basin with a relative rise in sea-level (Su et al., 2007; Huang et al., 2011). During the same time interval, a relatively low energy and anoxic conditions prevailed in the southeastern Sichuan Basin, which led to the deposition of a thick layer of organic-rich shale (Chen et al., 2004; Zhao et al., 2016b) (Fig. 1). A series of thin-layered siliceous shales containing abundant graptolites was deposited over a large area during the Late Ordovician period under the influence of tectonic movements and transgressions (Chen et al., 2000, 2005; Su et al., 2007). The top of the Wufeng Formation (Guanyinqiao Member) was usually deposited as a thin shell-rich marl and limey mudstone during a rapidly falling sea-level attributed to the Hirnantian glaciation (Rong et al., 2002; Chen et al., 2004, 2006) (Fig. 2). During the deposition of the Early Silurian Longmaxi Formation, tidal flat and lagoon facies were principally developed at the margin of the uplifts and most of the Sichuan Basin was characterized by a restricted shelf (Guo and Zhang, 2014; Zhao et al., 2016c). The Lower Longmaxi Formation is composed of graptolites-bearing black organic-rich shales (Fig. 2).

This study focus on two wells: well JY2 in the Fuling shale gas field and well YZ1 in Shizhu area (Fig. 1). The present burial depth of the base of the Wufeng Formation increases gradually from 2575.03 m at the JY2 site to 4517.79 m at the YZ1 site (Fig. 3). The lithofacies and lithofacies association of Wufeng and Longmaxi shale are similar for both wells. The Wufeng Formation contains mainly siliceous shale, argillaceous shale and thin-layered limey mudstone on top. The base of the Lower Longmaxi Formation is composed of siliceous shale with a thickness ranging from 11.2 m in YZ2 to 25.7 m in JY2. The middle of the Lower Longmaxi Formation consists of silty and argillaceous shales. The thickness of the silty shale varies from 1 m to 12 m. The summit of the Lower Longmaxi Formation is defined principally by argillaceous shale.

3. Sampling and methodology

Samples collected from the core provided by the two wells (JY2 and YZ1) are used to study the Wufeng and Longmaxi shales (Fig. 1). A total of 33 core samples (14 from JY2 and 19 from YZ1) were collected from both formations for petrological, TOC, and geochemical analyses. There are 12 siliceous shale samples (5 from JY2 and 7 from YZ1), 13 silty shale samples (5 from JY2 and 8 from YZ1) and 8 argillaceous shale samples (4 from JY2 and 4 from YZ1). The depth of sampling varies from 2478.45 to 2573.8 m in the JY2 well, and from 4517.72 to 4460.10 m in the YZ1 well (Fig. 3).

Polished thin sections, cut perpendicular to the bedding, X-ray Diffraction (XRD) analysis, scanning electron microscopy (SEM), and the TOC contents were carried out at the State Key Laboratory of Petroleum Resources and Prospecting, China University of Petroleum (Beijing). Thin polished sections were analyzed for petrographic composition and texture. The mineralogical composition of the bulk powder samples (less than 200 mesh) was determined by X-ray Diffraction (XRD) using a Bruker D2 PHASER diffraction. Ion-milling provided a flat sample surface for imaging with focused ion beam scanning electron microscopy (FEI Helios Nanolab 650). All samples were examined using both secondary electron (SE) and backscattered electron (BSE) images, and additional observations using cathodoluminescence (Mono CL detector) and energy disperse spectroscopy (EDS) were carried out for the identification and characterization of authigenic minerals. TOC contents were measured via IR spectroscopy using a CS Mat 230, after treating each sample with hydrochloric acid (1:9 HCl: water) at $60 \pm 5\,^{\circ}C$ to remove carbonate minerals.

The Large Area Mapping can stitch numerous SEM images and provide a large field of view with the same resolution as an individual image, which is effective to investigate pore types and their quantitative distributions. Five areas were randomly selected on each Ar-milled sample for Large Area Mapping processing. Each area ($100 \times 100\ \mu m^2$) contains 100 images taken at 14000×