Graptolite-derived organic matter in the Wufeng–Longmaxi Formations (Upper Ordovician–Lower Silurian) of southeastern Chongqing, China: Implications for gas shale evaluation

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A B S T R A C T

The organic-rich shales of the Wufeng–Longmaxi Formations (Ordovician–Silurian) are one of the most important hydrocarbon source rocks and shale gas exploration targets in China. Graptolites are abundant in these shales, but their contribution to the dispersed organic matter (OM) has been overlooked. This lack of attention could seriously affect the evaluation of OM type, OM maturity, hydrocarbon generation potential, reservoir characteristics and gas content of these shales. Optical microscope and focused ion beam-scanning electron microscope (FIB-SEM) techniques were employed to describe the optical characteristics and micro-nanopores of the graptolites, to determine the contribution of the graptolites and to evaluate the thermal maturity of the Wufeng–Longmaxi Formations. Non-granular graptolites, which show a stronger anisotropy and higher reflectance under polarized light than the granular graptolites, were primarily observed in these sediments. Graptolites account for 20–93% of the dispersed OM in the Wufeng–Longmaxi Formations, and the level of their contribution has been attributed to the radiation of graptolites at the late Ordovician (Kaptian) and the recovery of graptolites at the early Silurian (Rhuddanian). The bireflectance values of the non-granular graptolites fall between 2.22 and 4.32%, indicating a behavior similar to biaxial anorthacitic vitrinite. The equivalent vitrinite reflectance (EqVRo) values of the sediments in the Wufeng–Longmaxi Formations, calculated from the mean maximum reflectance of non-granular graptolite, fall in the range of 3.08–4.29%, and these values have a precision that is greater than in previous studies on the Wufeng–Longmaxi Formations. Abundant organic pores were observed in the non-granular graptolites, and they are aligned parallel to the fusellar layers of the graptolites. This indicates that the fusellar layers of non-granular graptolites controlled the development and distribution of organic pores. Such abundant organic pores played a significant role in the accumulation of shale gas in the Wufeng–Longmaxi Formations of the Upper Yangtze Platform.

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1. Introduction

The shales of the Wufeng–Longmaxi Formations (Upper Ordovician–Lower Silurian) contain abundant organic matter (OM) with total organic carbon (TOC) values mainly ranging from 2 to 5%. They have been regarded as one of the most important source rocks in southwestern China and were recognized as the main exploration target for shale gas in China (Liang et al., 2009; Dong et al., 2010; Zou et al., 2010, 2011, 2012; Guo and Zhang, 2014). The Wufeng–Longmaxi shales are also characterized by their widespread occurrence, large thickness (30–130 m) (Fig. 1), high thermal maturity, strong gas generation intensity, abundant organic pores and favorable mineral composition (Dong et al., 2010; Zou et al., 2011, 2012; Tian et al., 2013; Dai et al., 2014; Ma et al., 2015), all of which factors are beneficial for the formation and accumulation of shale gas. To date, the Chongqing area has become the leading area for exploration and development of shale gas in China, and in March 2014, the first large shale gas field was discovered by China Petroleum & Chemical Corporation (SINOPEC) in the Fuling area, Chongqing (Fig. 2a). The proven geological reserves and economically recoverable reserves of this field are estimated to be 106.75 and 13.74 billion m³, respectively (Zhang and Lu, 2015). This example indicates that the shales of the Wufeng–Longmaxi Formations are now realistic targets for shale gas exploration in China.

It is worth noting that many paleontologists have observed an abundance of graptolites in the Wufeng–Longmaxi shales (Chen et al., 2000, 2004, 2005). Based on the observation of cores and corresponding polished rocks, numerous graptolites were observed in our samples (Fig. 3), however, their contribution to the dispersed OM has been overlooked. There was a major mass extinction at the end of the Ordovician period (Hirnantian), and this was the oldest and second largest
of the five great extinctions in the Phanerozoic (Sheehan, 2001; Chen et al., 2005). However, there was a small rebound of marine animal diversity during the late Ordovician (Kaptian) before the end-Ordovician mass extinction, and there was a recovery of marine fauna during the early Silurian (Rhuddanian) after the end-Ordovician mass extinction (Sepkoski, 1995; Chen et al., 2005; Schmitz et al., 2008). Previous work on the graptolites in this area has focused on their implications for biostatigraphy, paleoecology, and sedimentary environment (Chen et al., 2000, 2004, 2005). As shown by the large number of graptolites in Fig. 3, it is important to determine their contribution, which will be beneficial to the evaluation of OM type, OM maturity, hydrocarbon generation potential, reservoir characteristics, and gas contents of the shales.

Thermal maturity is one of the most important factors when assessing hydrocarbon source rocks, and is also one of the most important indices for the evaluation of shale gas (Curtis, 2002; Jarvie et al., 2007; Kinley et al., 2008). Vitrinite reflectance was thought to be the most reliable maturity parameter, however, it cannot be measured in pre-Devonian source rocks because the higher plant precursors to vitrinite were not yet evolved. The thermal maturity of the Wufeng–Longmaxi Formations was therefore generally determined on the basis of the bitumen or vitrinite-like maceral that was measured in previous studies may actually be graptolites, because graptolites can be easily mistaken for bitumen or vitrinite-like macerals due to a lack of understanding of the optical characteristics of graptolites.

With the development of shale gas, the microstructural features of shales have been widely studied in recent years. The OM in the shales not only provides a strong potential for the adsorption of gas, but also provides space for free gas due to abundant organic pores (Loucks et al., 2009; Curtis et al., 2012; Fishman et al., 2012). Curtis et al. (2012) pointed out that the development of organic pores can be anticipated to vary in relation to OM contents, thermal maturity and maceral type. In recent years, the relationship between OM contents, thermal maturity and organic pores have been widely studied (Loucks et al., 2009, 2012; Curtis et al., 2012; Milliken et al., 2013), but comparatively little attention has been paid to the relationship between OM type (especially individual maceral types, e.g., graptolite) and organic pores. Numerous organic pores have been observed in the Wufeng–Longmaxi shales of the Sichuan Basin. The contribution of these organic pores to the total porosity has been estimated to range from 31.3 to 70% (Tian et al., 2013; Cao et al., 2015). However, we still have insufficient understanding of the organic pores in the graptolites. In this study, the Wufeng–Longmaxi sediments from the southeastern Chongqing (Figs. 1 and 2b) were sampled in order to determine the optical characteristics and micro-nanopores of the graptolites, and to assess the contribution of the graptolites and the thermal maturity of the Wufeng–Longmaxi Formations.

2. Geological setting

The investigated region is located in southeastern Chongqing, Upper Yangtze Platform (Fig. 2). This area has been subjected to intense tectonic activity, and large thicknesses of strata have been removed by erosion. From northwest to southeast, the structural pattern changes from trough-like folds to partition-style folds in southeastern Chongqing (Zhai, 1987). During the early-middle Ordovician, the study area was isolated due to uplift, and covered by a broad epeiric sea that provided a low-energy, anoxic environment. During the late Ordovician–early Silurian, two global transgressions affected the Yangtze Platform, leading respectively to the deposition of the Wufeng and Longmaxi shales (Mu et al., 2011). The Wufeng–Longmaxi Formations are composed mainly of black shales and cherts, and they yield an abundant and diverse graptolite fauna. So far, more than 10 graptolite zones (or subzones) have been recognized in the Wufeng–Longmaxi Formations and these have been correlated precisely with biozones worldwide (Chen et al., 2000).

3. Sampling and methods

The Wufeng–Longmaxi samples for this study were collected from Wells XY5, YC4, YC8 and the Dingshi outcrop (Fig. 2b). Samples chosen on the basis of TOC were used to the preparation of polished blocks. To prepare a polished petrographic mount, a representative sample was first crushed to < 1 mm with a minimum of fines, subsampled and dried at 50 ℃. The sample was then set under vacuum and/or pressure in 25 mm square rubber molds with a cold setting epoxy resin. After the resin was properly cured, the sample was ground on a rotating metallographic polishing/grinder with various grades of silicon carbide paper using water as a lubricant. The sample was then polished using diamond and colloidal silica compounds on rotating laps to give a flat, highly polished surface suitable for microscopic examination.

The maceral characteristics and compositions were analyzed on a Leica microscope with a 50× oil-immersion objective using reflected light. Graptolite reflectance were measured in oil immersion under

<table>
<thead>
<tr>
<th>Period</th>
<th>Stage</th>
<th>Formation</th>
<th>Depth (m)</th>
<th>lithology</th>
<th>Sample position</th>
</tr>
</thead>
<tbody>
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<td>Rhuddanian</td>
<td>Longmaxi</td>
<td>Q21</td>
<td></td>
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</tr>
<tr>
<td></td>
<td></td>
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<td>Q2</td>
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<td>Q0</td>
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<tr>
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<td>Wufeng</td>
<td>Q21</td>
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Fig. 1. The stratigraphic column and the sample position of the Wufeng–Longmaxi Formations in well XY5.
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