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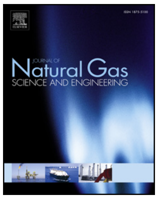

Natural gas potential of Carboniferous and Permian transitional shales in central Hunan, South China

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ABSTRACT

The Carboniferous Ceshui formation and Permian Longtan and Dalong formations were deposited in transitional settings preserved in what is now central Hunan Province, South China, as they are potential natural gas plays. In this study, we analysed the total organic carbon (TOC), vitrinite reflectance (Ro), kerogen type, mineralogy, porosity, permeability, and methane adsorption of representative shale samples from these rock units. Our results indicate that TOC content can be as high as 9.2%, with a mean ($\bar{x}$) of 3.5%. The Permian shale formations were deposited in more strongly reducing environments than the Carboniferous Ceshui shale. The kerogen composition of the Carboniferous Ceshui shale is dominated by Type III, while both of the Permian shales contain primarily Type II kerogens; Ro values range from 1.1% to 2.4% ($\bar{x} = 1.6\%$). The organic matter in all the studied shales is in the wet gas window of thermal maturity and is relatively less mature than Lower Palaeozoic marine shales in south China. Mineral compositions are dominated by quartz ($\bar{x} = 53.8\%$) and clay ($\bar{x} = 35.6\%$), suggesting a high brittleness index. Porosity ranges from 0.5% to 14% ($\bar{x} = 6.4\%$), while permeability varies from 0.0026 micro Darcy (mD) to 0.0640 mD ($\bar{x} = 0.0130$ mD). The gas adsorption capacity varies from 1.24 to 4.53 cm^3/g ($\bar{x} = 2.40\ cm^3/g$). Relatively less mature shale samples (Ro < 1.5%) have low methane adsorption capacities, regardless of their TOC values. However, the methane adsorption capacity of more mature (Ro > 1.5%) shales samples exhibit a positive correlation with TOC content.

1. Introduction

The remarkable success of shale gas development in North America has triggered a flourishing of shale gas exploration and increased the number of investigations into the gas potential of shales worldwide (e.g., Bowker, 2007; Jarvie et al., 2007; Tan et al., 2014a,b,2015; Tang et al., 2014). To enhance domestic energy supply via shale gas exploitation, the Chinese government has already set ambitious plans. Geological surveys and exploration activities are underway across the country (Tan et al., 2013). To date, China has begun developing several shale gas fields in the Sichuan Basin (e.g., the Fuling, Weiyuan, and Changning shale gas blocks).

In China, three types of organic rich shale are widely distributed in sedimentary basins, comprising marine shales, marine-lacustrine transitional shales (hereafter referred to as "transitional shales"), and lacustrine shales (Bu et al., 2015; Tan et al., 2015). Marine shales in the Cambrian and Silurian starta and transitional shales in the Carboniferous and Permian strata are widely distributed cross South China (e.g. Tan et al., 2015; Zou et al., 2010). Although natural gas has been successfully produced from the marine shales, transitional shales have not been yet successfully developed (Dong et al., 2016). In the central part of Hunan, Carboniferous and Permian transitional shales occur in thick and laterally extensive beds (Bao et al., 2016; Gu et al., 2015; Jing et al., 2013; Xu et al., 2015). Previous studies have shown that Carboniferous and Permian paralic coal-bearing strata constitute source rocks for conventional petroleum fields in this region (Wang et al.,

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2010a; Zhan et al., 2006; Zhou and Guo, 2014; Zhu and Yi, 2012). These transitional shales have been more recently identified as promising targets of shale gas exploration (Bao et al., 2016; Gu et al., 2015; Liang et al., 2014a,b; Luo et al., 2012). National petroleum companies, including Sinopec, have already shown strong interest in developing natural gas resources from transitional shale intervals. Therefore, there is an urgent need to characterize these shale intervals and evaluate their reservoir potential.

It has been recognized that no two shale gas systems are exactly alike, and thus exploitation strategies differ from one system to the next (Tan et al., 2015). However, prolific shale plays are commonly distinguished using certain minimal technical thresholds. These include, but are not limited to: shale lateral extent and effective thickness, organic matter richness, thermal maturity, mineralogy, porosity, permeability, adsorptive capacity, and gas-in-place (GIP) (Bowker, 2007; Jarvie et al., 2007; Tan et al., 2013, 2015). These indices have been widely applied to evaluate the reservoir potential of marine shales in South China. However, for transitional shales, numerous issues remain concerning the composition and source(s) of organic material, depositional environment, thermal maturity, petrophysical properties, and methane adsorption capacity. Additionally, the correlations among organic materials, mineralogy, and depositional conditions, as well as the influences of porosity and permeability, and the relative effects of total organic carbon (TOC) and thermal maturity on methane adsorption have not been clarified. This study aims to investigate these problems through a systematic characterization of transitional shales from the Carboniferous Ceshui formation and the Permian Longtan and Dalong formations in the central region of Hunan. We comprehensively analysed the TOC content, organic matter type, thermal maturity, mineral composition, reservoir physical properties, and gas adsorption capacity of the shales. We then compared our results with those reported for prolific shale plays in China and the United States. Lastly, we discuss the correlations among the selected reservoir characterization parameters.

2. Materials and methods

2.1. Geological setting and shale deposition in central Hunan

The study area is located in the Middle Yangtze Region and in the north of the South China fold system (Fig. 1a). It lies east of the Hengshan Uplift, and west of the Xuefeng Uplift. This region is tectonically composed of five subunits. From north to south they are the Lianyuan Depression, Longshan Uplift, Shaoyang Depression, Guandimiao Uplift, and the Lingling Depression (Fig. 1b). Regional faults extend primarily in the NE-NNE direction, and include the Chengbu-Xinhua fault, Qiyang Arc fault, Miluo-Shaoyang fault, Xinshao-Xinning fault, and Zhuzhou-Shuangpai fault (Fig. 2). Additionally, thrust nappe and gravity gliding structures have been formed by multiple tectonic events. The Lianyuan and Shaoyang depression began forming during the Ordovician at the onset of the Caledonian orogeny. The Qiyang arc was dominantly formed through structural deformation caused by the Triassic Indosinian orogeny (Li et al., 2013; Wang et al., 2010b).

Shales are well developed in certain formations of the study area. Sedimentary strata are characterized by carbonates interbedded with clastics deposited in the Late Palaeozoic to Middle Triassic. The region has experienced four primary sedimentary cycles from the Devonian to the Permian (Jing et al., 2013; Xu et al., 2015). The first sedimentary cycle occurred between the Tiaomajian and Qiziqiao periods during the Devonian when a transgression initiated during a large-scale geological extension and was followed by a regression generated by the Liujiang orogeny. This cycle resulted in the deposition of the Shetianqiao and Qiziqiao marine shales. The second transgression started in the Mississippian (early Carboniferous), but ended as a full-scale regression in the mid-Mississippian with the deposition of the Ceshui formation as transitional beds formed under the alternating influences of shallow marine and shoreline environments (Fig. 1c) (Shao et al., 1992). The third cycle was primarily controlled by the Dongwu tectonic movement and is the largest transgression that occurred during the development of the mid-to late Carboniferous paraplatform. However, this transgression was terminated by a full-scale regression during the early Lopingian (late Permian) (Fig. 1c), and coastal marsh shales were subsequently deposited (Gu et al., 2015; Ji et al., 2011). The fourth cycle persisted for a shorter time (i.e., only during the Lopingian), and resulted in the deposition of the Dalong formation (Fig. 1c). During this time, siliciclastic rocks, siliceous limestone, and shales were deposited in littoral-bathyal-abyssal facies (Fig. 1c) (Feng et al., 1993).

2.2. Samples

Fresh transitional shale samples are exposed in some outcrops of the study area. A total of 96 representative samples were collected from the Shimingqiao (SMQ), Qixingjie (QXJ), Duanpoqiao (DPQ), Tantou (TT), Doulishan (DLS), Nantang (NT), Jilong (JL), Jingzhushan (JZS), Zhaoyang (ZY), Liangshuijing (LSJ), Xiandong (XD), Lumaojiang (LMJ), and Douling (DL) sections (Fig. 1b). Generally, shale samples were collected from at least 1 m deep, and every 10 m from the bottom to top of the exposed sections.

2.3. Methods

The selected samples were analysed for TOC, vitrinite reflectance (Ro), kerogen type, mineralogy, porosity, permeability, and methane adsorption.

TOC contents were measured using a Leco carbon-sulphur analyser and reported as the weight percentage (wt%) of the total rock material. Samples were crushed into a powder < 200 mesh, and 1–2 g samples were pyrolysed to 600 °C. Thin-sections of 42 samples were prepared for investigation mineral components and structural fabrics. The samples were examined using a Zeiss Axiophot Electronic Microscope equipped with a Carl Zeiss Axiocam digital camera and Axiovision 2.0 software. This system was capable of taking high-resolution photomicrographs under magnification of 10x, 20x, 30x, 40x, and 50x. Kerogen type was analysed by transmitted light microscopy. Thermal maturity, represented by Ro, was determined using a MVP-3 microscope photomultiplier.

A total of 25 samples were analysed with a D/max-2600 X-ray diffractometer (XRD) to quantify the principal mineralogical constituents. The diffraction data were recorded from 4° to $75°2\theta$ with a step width of 0.02°, and a counting time of 4 s per step. Experimental conditions were set to 40 kV and 30 mA. The measured data were analysed qualitatively using EVA (Bruker) software, and quantitatively using AutoQuant software.

High-resolution scanning electron microscopy (SEM) analysis was performed on representative samples with different TOC contents and lithological types. Small subsamples 0.3–0.5 cm thick, 0.5–1.0 cm wide, and > 2 cm long were cut and prepared. The subsamples were dried in an oven at 40 °C for 24 h to remove moisture. Analysis was conducted using a TESCAN VEGA scanning probe microscope, and images were obtained under high vacuum at 20 kV acceleration voltage.

Porosity was measured with an ULTRAPORE-200A helium porosimetre. Permeability was tested using an ULTRA-PERMTM200 permeametre. Measurements were performed at room temperature and normal pressure (~ 23 °C, 102 kPa) and 50% humidity.

Methane adsorption isotherms were measured for selected moisture-equilibrated samples at 40 °C. The experimental procedure was: 1) degas the sample, 2) conduct leak tests, 3) determine the void volume as well as the sample volume using helium expansion, 4) evacuate for 60 min at 1 MPa to remove helium, and 5) perform the methane adsorption measurement (Tan et al., 2014a). The Langmuir isotherm was applied to model gas adsorption capacity. The equation used is: $V = V_L P/(P_L + P)$ (Pan et al., 2015, and references therein), where V is