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Research paper

Pore network characteristics of lacustrine shales in the Dongpu Depression, Bohai Bay Basin, China, with implications for oil retention

Xinhe Shao[a,b,*], Xiongqi Pang[a,b,**], Hui Li[a,b], Tao Hu[a,b], Tianwu Xu[c], Yuan Xu[a,b,d], Boyuan Li[a,b]

[a] State Key Laboratory of Petroleum Resources and Prospecting, China University of Petroleum, Beijing 102249, China
[b] Basin and Reservoir Research Center, China University of Petroleum, Beijing 102249, China
[c] Zhongyuan Oilfield Branch, SINOPEC, Puyang, Henan 457001, China
[d] Department of Higher Education, Ministry of Education of the People's Republic of China, 100816, China

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ABSTRACT

Pore development and the location of retained hydrocarbons are key topics to understand the shale oil system. In an attempt to investigate pore network characteristics and oil retention in lacustrine shales, 24 samples from the Es_3 shales in the Dongpu Depression, Bohai Bay Basin were subjected to mineralogical, petrological, geochemistry and pore structure investigations using X-ray diffraction (XRD), thin section, field emission-scanning electronic microscope (FE-SEM), N_2 adsorption-desorption. After removing retained oil by solvent extraction, 12 selected samples were again analyzed by Rock-Eval pyrolysis and N_2 adsorption-desorption experiment.

According to the mineral compositions and petrological characteristics, six types of lithofacies are identified in the Es_3 shales in the Dongpu Depression: laminated calcareous shales, lenticular calcareous shales, laminated argillaceous shales, massive argillaceous shales, laminated siliceous shales, and massive siliceous shales. Geochemical characteristics of these shales are quite different, as calcareous/laminated shales have higher TOC contents than massive argillaceous and siliceous shales. Interparticle pores, intraparticle pores, organic matter pores and fracture pores are observed in the studied samples, while bitumens were found retaining in well-connected pore network. The N_2 adsorption-desorption experiments on samples before and after solvent extraction reveal the different pore structure characteristics in six lithofacies and the occurrence of oil in lacustrine shales. In general, pore volumes within pore diameter range of 1.7–10nm and 10–200nm of samples show increase after solvent extraction, indicating that large and small pores in shales both play important roles in oil retention. Oil retained in shales can be divided as free and heavy fractions, as from the pyrolysis results. In lacustrine shales, larger pores are main storage space for hydrocarbon. Larger pores in different lithologies have similar capacity for free oil retention, while those in calcareous and argillaceous shales are more efficient for heavy oil retention. For both free and heavy oil, small pores in argillaceous shales are efficient for their storage.

1. Introduction

Drilling techniques include horizontal wells and hydraulic stimulation have triggered the exploration and exploitation of hydrocarbon in shale plays. Great shale oil resources are found in marine shales, such as Barnett shale (Jarvie, 2012; Han et al., 2015), Bakken shale (Schmoker, 1996; Soeder, 2018), Woodford shale (Cardott, 2012) in the United States, and Duvernay Shale in Western Canada (Wang et al., 2018). Meanwhile, researchers have put a lot of efforts into understanding oil storage mechanisms in shales, which significantly promote the economic development of shale oil resources: petroleum adsorption on organic matter has been documented to be major for hydrocarbon retention in shales (Stainforth and Reinders, 1990; Pepper, 1991; Sandvik et al., 1992; Ritter and Grøver, 2005); Jarvie (2012) proposed that organic-lean facies and open-fracture network in shales reduce oil sorption by organic matter; Han et al. (2017) suggested that porous biogenic matrices provide space for the storage of shale oil. Things can be quite different when coming to lacustrine shales (Pang et al., 2018). Lacustrine sediments have greater heterogeneity than marine sediments (Katz, 1990; Xie et al., 2016), thus results in complexities of lacustrine shales in the aspects of mineral compositions, organic matter deposition, sedimentary fabric, pore network development, etc. Therefore, great challenges exist when studying oil retention in lacustrine shale systems and locating shale oil reserves.

* Corresponding author. State Key Laboratory of Petroleum Resources and Prospecting, China University of Petroleum, Beijing 102249, China.
** Corresponding author. State Key Laboratory of Petroleum Resources and Prospecting, China University of Petroleum, Beijing 102249, China.
E-mail addresses: 715213805@qq.com, sophiashao0212@yahoo.com (X. Shao), pangxq@cup.edu.cn (X. Pang).

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The Dongpu Depression in the Bohai Bay Basin is one of the most important target areas for lacustrine shale oil exploration in China because of its great shale oil prospectivity. It has been a petroliferous area for conventional oil and gas exploration for decades, while a daily shale oil output of ~200m^3 in 2010 from the Es_3 Member shale declared the huge potential for shale oil in the Dongpu Depression (Wang et al., 2015). However, due to the lack of understanding in pore structure and oil occurrence in lacustrine shales, the shale oil exploration in the Dongpu Depression still faces great financial risk. Therefore, understanding the pore system and oil retention in lacustrine shales is essential.

A suite of techniques have been proved to be effective in characterizing shale pore structures: mercury intrusion porosimetry (Mastalerz et al., 2013; Jiang et al., 2016), gas adsorption (Chen et al., 2016; Shao et al., 2017), nuclear magnetic resonance (Karimi et al., 2015; Zhou and Kang, 2016). Among these, N_2 adsorption is a most commonly applied experiment, and has been performed on extracted and unextracted shale samples to study the influence of residual bitumen on shale pore structures (Wei et al., 2014; DiStefano et al., 2016). Therefore, we expected that characteristics of oil retained in shales could be studied by conducting N_2 adsorption experiments on extracted and unextracted samples.

In this paper, we applied TOC measurement, Rock-Eval pyrolysis to analyze the geochemical properties of the lacustrine shales in the Dongpu Depression, while X-ray diffraction (XRD) analysis and thin section observation were used to study the petrology and pore structure was characterized by FE-SEM observation along with gas adsorption. By comparing the experimental results of unextracted and extracted samples, we investigated the relationship between pore development and hydrocarbon retention. The objectives of this study are to: 1) characterize the pore network of the lacustrine shales in the Dongpu Depression, Bohai Bay Basin; 2) determine the oil retention pattern in lacustrine shales. The results will be expected to aid in the oil exploration in lacustrine shales.

2. Geological background

The study area, the Dongpu Depression, lies in the south western Bohai Bay Basin, which is the largest petroliferous basin in China (Wang et al., 2015; Zuo et al., 2017). The Dongpu Depression, covering an area of ~5300km^2, is developed as an NNE-trending Cenozoic extensional rift basin, and can be subdivided into four tectonic units: Eastern Sag Belt, Central Uplift, Western Sag Belt and Westen Slope Belt from east to west (Jiang et al., 2008; Wang et al., 2015; Lyu and Jiang, 2017) (Fig. 1A). Since Yanshan movement, the depression underwent two main tectonic stages-rifting from Eocene to Oligocene and subsidence during Neogene-Quaternary, which can be further divided in to five stages: initial rifting, advanced rifting, post-rifting, rifting termination and subsidence (Li et al., 1995; Lyu and Jiang, 2017).

The Dongpu Depression is filled with Cenozoic sediments, which are controlled by the Lanliao fault in the east of the study area (Chen et al., 2013; Wang et al., 2015). The Paleogene sediments include the Kongdian Formation (Ek), Shahejie Formation (Es) and Dongying Formation (Ed), while the Guantao Formation (Hg), Minghuazhen Formation (Nm) and the Quaternary Pingyuan Formation (Qp) were deposited from the Neogene to the present. The Shahejie Formation can be divided into four members from bottom to top: Es_4, Es_3, Es_2 and Es_1 (Fig. 1B). Among them, we focused on the third member (Es_3) in this study, which is a most important target layer for shale oil exploration in the Dongpu Depression. In addition, this strata has always been recognized as the economically important source rock in the study area. Due to the sedimentary facies belonging to semi-deep and deep lacustrine, fan delta and turbidite, along with brackish water environment in the northern area changing to fresh water environment in the south, the lithofacies vary markedly in the study area. The Es_3 is further divided into three sub-members according to their lithologies, which are the upper, middle, lower third member (Es_3^U, Es_3^M, Es_3^L). The Es_3^L mainly consists of gray mudstones interbedded with siltstones, while gypsum and halite occur in the northern area of the Dongpu Depression. Gypsum and salt are important lithologies in the Es_3^M, whereas mudstones and calcareous siltstones became secondary components. The Es_3^U is dominated by evaporates, while dark mudstones and siltstones are present as well (Jiao et al., 2014; Wang et al., 2014a,b, 2015). The thickness of the Es_3 member changes markedly in the study area, as the Es_3^U, Es_3^M, Es_3^L are 350–750m, 500–1200m, and 300–800m thick, respectively (Wang et al., 2015; Lyu and Jiang, 2017).

3. Samples and experiments

The Es_3 shale samples were collected from drilling cores at the Zhongyuan Oil Field Company, and the sampling sites cover different sedimentary facies, depth, and water environments. A total of 24 samples were subjected to total organic carbon (TOC) determination, Rock-Eval pyrolysis, X-ray diffraction (XRD) analysis, thin section and scanning electronic microscopic (SEM) observation, low pressure N_2 adsorption-desorption experiment. Solvent extraction and Rock-Eval pyrolysis were performed on all samples and 12 representative ones were selected for a second round of low pressure N_2 adsorption-desorption experiment after extraction. Solvent extraction were performed using a mixture of chloroform and ethanol (98:2, v:v) by a Soxhlet Apparatus for 72 h.

3.1. Total organic carbon (TOC) determination and Rock-Eval pyrolysis

Samples for geochemistry experiments were crushed to 100 mesh size. Before subjected to TOC measurements, samples were treated with 10% (by volume) HCL at 60 °C to remove carbonate minerals and cleaned with distilled water to remove residual HCL, and then sample powders were dried at 50 °C for 10h. The determination of TOC content was conducted using a LECO CS-230 analyser. The Rock-Eval pyrolysis was performed with a Rock-Eval II instrument, and samples were heated to 600 °C in a helium atmosphere while three parameters, S_1, S_2 and Tmax, were generated in this process.

3.2. X-ray diffraction (XRD)

Samples were first ground to 300 mesh size using agate pestle and mortal, and then XRD analysis was performed using a Bruker D2 X-ray diffractometer with CuKα-radiation produced at a voltage of 40kV and a current of 40mA. Samples were scanned from 5° to 45° with a rate of 2° (2θ)/min. The relative contents of minerals were determined semi-quantitatively using Jade software.

3.3. Thin section and field-emission scanning electron microscope (FE-SEM)

Thin section observation was conducted using a Leica DMLP polarizing microscope, and all samples were grinded to 0.03mm thickness before observation. Procedures of sample preparation for FE-SEM observation include pre-polishment with silicon carbide paper on one surface of samples perpendicular to the bedding to reduce the surface roughness, and polishment with argon ions using a Gatan 691.CS to create a smooth surface. The observation was performed using a FEI Quanta 200F in high vacuum mode with an acceleration mode of 20kV. In addition, an energy dispersive X-ray spectroscopy (EDS) was used to determine the elemental composition of minerals.

3.4. Gas adsorption experiment

Low pressure N_2 adsorption-desorption measurements were conducted using a Micrometrics ASAP-2020 apparatus. Prior to the experiment, samples were crushed to a particle size of 3–4mm, and automatically degassed at ~110 °C under vacuum for about 14h to