



The genesis and evolution of carbonate minerals in shale oil formations from Dongying depression, Bohai Bay Basin, China

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ABSTRACT

Shale oil is attracting increasing attention in the petroleum industry as an essential target of unconventional oil and gas exploration. Recent exploration efforts have indicated that laminar carbonates are closely related to the favorable depositional strata of shale oil. This research focused on inter-bedded layers of laminar carbonates as well as mudstone and shale in the Dongying depression of the Bohai Bay Basin in eastern China, and attempted to reveal the genesis and evolution of the carbonates by means of drill core observations, thin sections, high resolution scanning electron microscopic observations, and fluorometric, cathodoluminescence and geochemical analyses. The results demonstrated that: (1) the carbonates could be categorized into two groups, crystalline carbonates and micritic carbonates; (2) the crystalline carbonates were well crystallized and greenish under fluorescent light; and (3) the micritic carbonates were not well crystallized and had biotic textures. It was discovered that the crystalline carbonates were mainly developed in the shale layer and were formed from the late-filling effects of the diagenetic cracks along the shale fissility planes; shale layers with crystalline carbonates are usually favorable for shale oil reserves. The micritic carbonates were formed from direct deposition controlled by biological effects in the stratified flows of the seasonal lake basins and are unfavorable for shale oil reserves.

1. Introduction

With the recent progress of oil and gas exploration, highly promising unconventional oil and gas, especially shale oil, have become popular for exploration and have attracted substantial attention from governments and petroleum companies. Currently, the scale of the global reserves of unconventional oil and gas has become almost equivalent to that of conventional reserves (Zou et al., 2013). The unconventional oil plays account for roughly 50% of the world's oil production and some countries especially China. Derive almost all shale oil reserves from lacustrine systems (Merkel et al., 2016; Zou, 2017). The shale oil reservoirs developed in lacustrine systems mainly in semi-deep to deep environments (Zou et al., 2013; Jiang et al., 2014; Merkel et al., 2016; Moradi et al., 2016; He et al., 2017; Lv et al., 2017). Some existing reports have indicated that many laminar carbonates occur in semi-deep to deep environments of different lacustrine basins globally (Scherer et al., 2015; Li et al., 2016a; Merkel et al., 2016; Moradi et al., 2016; Xu et al., 2016; He et al., 2017), and are highly associated with

shale oil (Zhang et al., 2003; Hao et al., 2014; Hargrave et al., 2014; Kang et al., 2015; Liu et al., 2015; Wang et al., 2016; He et al., 2017). The main factors controlling on laminar carbonate development in lacustrine basin include the structural framework of basin and depositional environment conditions such as water depth, hydrodynamics and lake chemistry (Lerman et al., 1995; Martínek et al., 2006; Hargrave et al., 2014; Liu et al., 2015). However, meticulously analysis of the genesis for laminar carbonates in shale oil reservoir is rare. Some other studies have demonstrated that the abundance of carbonate laminae which occurred in lacustrine basins corresponds to the contents of total organic carbon (TOC) and shale oil reservoirs positively (Zhu et al., 2005; Liu et al., 2015; Wang et al., 2015; Wang et al., 2016). Generally, carbonate sedimentation occurs in relatively shallow water environments, and the layers with more carbonate sedimentation thus usually have lower organic content than deep-water mudrocks (Flügel, 2004; Abouelresh and Slatt, 2012). The positive correlation between the organic content and carbonate content in shale oil reservoir layers is difficult to comprehend. Therefore, a correct understanding of the

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genesis of the carbonate laminae interbedded within the organic-rich mudrock is crucial for revealing the relationship between the shale oil and the carbonate laminae mentioned above.

The Dongying depression is a third-level tectonic unit within the Bohai Bay Basin in Eastern China where shale is well developed in Paleogene lacustrine sediments. Recent studies have shown that the Paleogene Shahejie Formation in the Dongying depression is an important reservoir of shale oil (Feng et al., 2013; Cao et al., 2014; Hao et al., 2014; He et al., 2017). In recent years, some studies have demonstrated that there are substantial carbonate laminae interlaid within the organic-rich shale and mudstone sequence of the third and fourth members of the Shahejie Formation (E_{S_3} - E_{S_4}) in the Dongying depression (Feng et al., 2013; Cao et al., 2014; Hao et al., 2014; He et al., 2017). We conducted petrological and geochemical studies based on the drill core samples from the Dongying depression. This paper reports the categorization of the different types of carbonates in the shale oil layers, discusses the genesis and depositional models of the carbonates and further explores the links between the organic content, carbonate contents and the shale oil.

2. Regional geology

The Bohai Bay Basin is located in the eastern part of the Northern China Craton. It is a middle-Mesozoic continental fault basin (Cao et al., 2014) (Fig. 1A). The Jiyang sub-basin is a second-level tectonic unit developed in the central part of the Bohai Bay Basin that strikes NW. It is bordered by the Tan-Lu fault to the east, contacts the Chengning Uplift to the northwest in the form of a large basement fault, and is bounded by the Luxi Uplift Zone to the south (Fig. 1B). The Dongying depression is a NE-SW-striking half graben in the south of the Jiyang subbasin. It is bordered by the Chengjiazhuang Rise to the north, the Luxi Uplift to the south, the Qingtuozhi Rise to the east, and the Binxian Rise and the Qingcheng Rise to the west. The depression covers an area of $5.7 \times 10^3 \text{ km}^2$ and consists of the Lijing sag, the Boxin sag, the Niuzhuang sag, the Minfeng sag, the Northern faulted margin, the Central dispirc anticline and the Southern hinged margin. The Dongying depression is a typical half graben in Eastern China. The northern margin is a faulted margin, whereas the southern margin is a hinged margin. The northern faulted margin is made up of a group of normal faults and the Chennan major fault. A central fault zone is developed in the center of the depression. The Niuzhuang sag is developed in the north. The Lijing sag is developed in the south. The Boxin sag and Minfeng sag are developed in the eastern and western parts of the depression, respectively. The southern hinged margin contacts the Luxi Uplift and the Guangrao Rise in the form of an overlap unconformity (Fig. 2A, B) (Feng et al., 2013; Cao et al., 2014; Hao et al., 2014).

The evolution of the Dongying depression has been categorized into 4 episodes: the pre-rift, the rift, the fault, and the depression episodes (Feng et al., 2013). The pre-Mesozoic rocks at the bottom of the sequence are mainly pre-rift episode sediments and constitute the lower tectonic layer of the basin. The Mesozoic and Paleogene rocks in the middle constitute the rift-episode and fault-episode sediments and form the middle tectonic layer of the basin. The top of the basin is partly covered by Neogene depression-episode sediments and forms the upper tectonic layer of the basin. The features of the episodes are very well distinguished (Feng et al., 2013; Hao et al., 2014; Ma et al., 2016).

Dongying depression is infilled with the strata of Paleozoic (Pz), Mesozoic (Mz), and Cenozoic (Cz). The Cenozoic include Paleogene (E), Neogene (N) and Quaternary (Q) from bottom upward. The Paleogene and the Neogene were the major filling periods of the Dongying depression. Based on lithology and sedimentary environments, the Paleogene has been divided into the Kongdian (Ek) and the Shahejie (Es) Formations. The Shahejie Formation has been further divided from the bottom upward into four members called the fourth (E_{S_4}), the third (E_{S_3}), the second (E_{S_2}) and the first member (E_{S_1}). E_{S_3} and E_{S_4} are the major deposition layers for shale oil exploration. E_{S_4} has been sub-

divided into two sub-members, the lower sub-member of E_{S_4} (E_{S_4-2}) and the upper sub-member of E_{S_4} (E_{S_4-1}), and E_{S_3} has been sub-divided into three sub-members, the lower sub-member of E_{S_3} (E_{S_3-3}), the middle sub-member of E_{S_3} (E_{S_3-2}) and the upper sub-member of E_{S_3} (E_{S_3-1}) (Feng et al., 2013) (Fig. 3).

E_{S_4} was formed during the early fault episode. Because of tectonic movement, E_{S_4} is characterized by steep slopes and clear disconnected deposition among the different sags. Sedimentation mainly occurred in the deep part of the lacustrine basins under hot and dry paleo-climatic conditions. These sags were connected with each other in the late E_{S_4} , were relatively shallow, and were predominantly deposited with shallow lacustrine facies sediments that led to the formation of fossil-rich salt lacustrine facies consisting of gypsum dolomite, argillaceous dolomite, argillaceous limestone, calcareous mudstone, and shale (Feng et al., 2013; Hao et al., 2014) (Fig. 3). The depression was subject to strong faulting in the E_{S_3} period. The basement rocks quickly subsided during an extensional dynamic setting to form a relatively long-lasting deep-lake environment. Humid paleo-climatic conditions were indicated in the early E_{S_3} period, with all types of well-developed fossils, and resulted in the formation of deep-lake sediments that mainly consist of oil shale, calcareous mudstone and mudstone. The depositional features in the middle E_{S_3} and the late E_{S_3} are similar to those of the early E_{S_3} and are characterized by stable sedimentation of the lacustrine basin that resulted in the formation of calcareous mudstone and dark color shale. Some of the late sags show a gradual lithological transition from shale to mudstone (Feng et al., 2013; Hao et al., 2014) (Fig. 3). The carbonate interlayers were substantially developed in the mudstone and the shale layers of the E_{S_3-3} and the E_{S_4-1} . Such carbonate interlayers were the major focus of this research.

3. Sampling and experimental methods

3.1. Cores and thin sections

In the stratum of this study, sedimentary rock containing > 50% (by weight or volume) of particles < $62.5 \mu\text{m}$ in size is variously known as shale, siltstone, claystone, or mudstone and is cumulatively referred to as mudrocks (Ilgen et al., 2017). In this research, we use “shale” to refer to the visibly laminated fissile variety of this sedimentary rock and we use “mudstone” to refer to fine-grained massive sedimentary rock that mainly consists of clay minerals with a few detrital minerals.

The Niuye1 and Fanye1 wells were the subjects of drill core observations because the wells had intact cores that contained the carbonate interlayers in E_{S_3} - E_{S_4} (Fig. 1). Observations were conducted in the drill core store of the Shengli Oilfield Company of the SINOPEC Group. Based on the observations of the sampled layers, a total of 26 core samples were collected for systematic analysis from the two wells for the range from E_{S_3-3} to E_{S_4-1} (14 samples from well Niuye1 and 12 samples from well Fanye1, shown in Fig. 4) (Table 1). All samples consisted of mudstone and shale laminae and carbonate laminae and were selected for the preparation of cubes with a size of $5 \text{ cm} \times 3 \text{ cm} \times 1 \text{ cm}$. The cubes were polished and drilled with a microdrill bit with a diameter of 2 mm for sampling of the mudstone and shale laminae and carbonate laminae after $10 \times$ magnification using a RELION microdrill sampling (Bedford, MA, U.S.A.) (Fig. 5). Thin sections were prepared from the samples. Three pieces of sections were prepared at the same position in the same sample, one section for polarizing microscope and fluorescence microscope observation, the second for cathodoluminescence observation, and the third for scanning electron microscope (SEM) observation. Polarizing microscope observation and microdrill sampling were conducted at the State Key Laboratory of Geological Processes and Mineral Resources of the China University of Geosciences, Beijing. The fluorescence microscope observations were conducted at the Key Laboratory for Unconventional Oil and Gas of the Oil and Gas Resource Surveying Center of the China Geological Survey.

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