Marine and Petroleum Geology 80 (2017) 349–357

Contents lists available at ScienceDirect

Marine and Petroleum Geology

journal homepage: www.elsevier.com/locate/marpetgeo

Research paper

Smectite-illitization difference of source rocks developed in saline and fresh water environments and its influence on hydrocarbon generation: A study from the Shahejie Formation, Dongying Depression, China

Yingli Li [a], Jingong Cai [a,*], Xuejun Wang [b], Yunqing Hao [b], Qing Liu [b]

[a] State Key Laboratory of Marine Geology, Tongji University, Shanghai 200092, China
[b] Geological Scientific Research Institute, Shengli Oil Field Company, Sinopec, Dongying 257015, China

ARTICLE INFO

Article history:
Received 17 September 2016
Accepted 5 December 2016
Available online 9 December 2016

Keywords:
Dongying Depression
Source rocks
Smectite illitization
Interlayer organic matter
Hydrocarbon generation

ABSTRACT

Samples of argillaceous source rocks from three sub-members of the Shahejie Formation (Es) in the Dongying Depression, China, were collected to investigate the differences in hydrocarbon generation among the sub-members, which developed in fresh (Es_3^2, Es_3^3) and saline (Es_4^1) water environments. Pyrolysis, XRD and thermo-XRD analyses were used to compare the characteristics of organic matter (OM), clay minerals and OM occurrences. Total organic carbon and hydrocarbon potential proxies suggest that the samples from Es_3^3 were much better than the other two intervals, which agrees with previous studies. The characteristics of clay minerals suggest that the samples from Es_4^1 have the most abundant illite, with a maximum illite percentage in mixed-layer illite-smectite (I–Sm), and the best crystallinity of I–Sm with a main stacking mode of R1.5. However, the stacking modes of I–Sm in Es_3^2 and Es_3^3 were primarily R0 and R1, respectively, and the crystallinity was relatively poor. Thus, the smectite illitization process was faster in Es_4^1 than in the other two intervals, and a saline environment was a primary cause for the acceleration of the process. Moreover, OM occurrence indicates that the samples from Es_4^1 had the lowest amount of interlayer OM, whereas Es_3^3 had the largest amount. Therefore, the rapid illitization in Es_4^1 caused abundant interlayer OM to be desorbed and discharged, which in turn caused the amount of residual interlayer OM in Es_4^1 to be less than that in the other two intervals. Thus, the source rocks of Es_4^1 made a more significant contribution to hydrocarbon generation than those of the other two units. In conclusion, the inconsistent illitization among these intervals was a major cause of the differences in hydrocarbon generation.

1. Introduction

The Dongying Depression belongs to a third-class negative structural element of China's Bohai Bay Basin and developed as a typical fault-depression basin during the Late Jurassic to Early Cenozoic (Guo et al., 2001). As the most important oil-bearing basin in East China, its source rocks mainly developed in the Paleogene (Chen and Zha, 2005; Zhu and Jin, 2003). Dark mudstones, which are 1500 m thick and comprise three sub-members of the Shahejie Formation (Es), including the upper part of the Fourth Member (Es_4^1), the lower part of the Third Member (Es_3^3), and the middle part of the Third Member (Es_3^2), were the main source rocks that had been discovered there (Li et al., 2000). Studies show that Es_3^3 primarily consists of brackish to fresh-water lacustrine sediments, whereas Es_4^1 contains brackish to saline lacustrine sediments (Li and Xiao, 1988; Wang and Qian, 1992).

According to the potential resource evaluation of the three units from Jiang et al. (2007a, 2007b), the hydrocarbon expulsion quantity of Es_3^3 comprises 27% of the entire Depression, whereas that of Es_4^1 comprises 73%. Based on the different molecular organic geochemical features of the three intervals, the proven reserves of Es_4^1 account for 85.1% of the total quantity in the Niuzhuang Sag (a secondary structural unit of the Dongying Depression), whereas those of Es_3^3 account for 14.9% (Zhang et al., 2014). Although Es_4^1

* Corresponding author. Tongji University, School of Ocean and Earth Science, Room 613, Siping Road 1239, 200092, Shanghai, China.
E-mail address: jgcai@tongji.edu.cn (J. Cai).

http://dx.doi.org/10.1016/j.marpetgeo.2016.12.004

underlies Es_3^3, slight difference in vitrinite reflectance exists between these two intervals (0.5%–1.30% for Es_4^1 and 0.56%–1.01% for Es_3^3) (Zhang et al., 2008). However, conventional organic geochemical proxies indicate that the source rocks of Es_3^3 are superior to those of Es_4^1 with respect to the total organic carbon (TOC), organic matter (OM) type and hydrocarbon potential (Zhang et al., 2003, 2009; Zhu et al., 2003). This finding is inconsistent with the petroleum resource quantity and proven reserves, which further inspires us to explore other indicators that have synergistic relationships with hydrocarbon generation.

Source rocks are mainly composed of OM and inorganic minerals, which can combine with each other and change with diagenesis. Studies have indicated that more than 80% of OM could be combined with clay minerals and become protected in the form of organo-clay complexes (Cai, 2004; Guan and Xu, 1998; Kennedy et al., 2002). This OM can be adsorbed as organic irons and molecules in pore spaces, along the external surfaces of clay crystals, and in interlayer spaces (Garfinkel-Shweky and Yariv, 1997; Salmon et al., 2000; Schulten et al., 1996). Compared to illite, smectite has high expansibility, large internal and external specific surface areas, and high cation exchange capacity. Smectite can adsorb certain amounts of OM within its interlayer space and then form clay-organic complexes (Cai et al., 2007; Guan and Xu, 1998; Kennedy et al., 2002). The interlayer spaces of illite, however, contain only K^+ and can adsorb little OM. Hence, the amount of OM that is adsorbed by clay minerals could be affected by smectite illitization during diagenesis. Meanwhile, the illitization rate could be affected by the solution chemistry (Herbert et al., 2004, 2016). Simulation experiments suggest that the presence of K^+, Na^+, Ca^{2+} and Mg^{2+} induce different effects on the rate of smectite illitization (Huang et al., 1993; Roberson and Lahann, 1981).

Differences in the hydrocarbon generation within different intervals of the Dongying Depression have been investigated by considering organo-clay interactions. In this study, samples were selected from the Es_3^2, Es_3^3, and Es_4^1 sub-members, and pyrolysis and XRD analyses were used to study the diagenetic organic-clay interactions in argillaceous source rocks from the Dongying Depression, Bohai Bay Basin, China (Fig. 1).

2. Geologic setting

The Dongying Depression, which has an area of 5850 km^2, is located in the southeastern Bohai Bay Basin (Fig. 1). This depression is bounded by the Chenjiazhuang Uplift to the north, the Qingtuozi and Guangrao Uplifts to the east, the Luxi Uplift to the south, and the Qingcheng-Linjia-Binxian Uplifts to the west (Fig. 1). The Cenozoic deposits in the study area consist of continental siliciclastic rocks from the Kongdian, Shahejie, Dongying, Guantao, Minghuazhen, and Pingyuan Formations (Fig. 1, Table 1). This study primarily focused on the Shahejie Formation. Traditionally, three sub-members of the Shahejie Formation, namely, the upper part of the Fourth Member (Es_4^1), the lower part of the Third Member (Es_3^3), and the middle part of the Third Member (Es_3^2), have been suggested to be the primary source rocks in this basin (Zhang et al., 2009). Es_4^1 contains brackish to saline lacustrine sediments. The lithology includes gray to dark-gray mudstones, siltstones and oil shales with gypsum interbeds in the lower part. Es_3^3 is primarily composed of brackish to fresh-water lacustrine sediments. This interval is dominated by dark-gray mudstones, oil shales and turbidite sandstone. Es_3^2 contains freshwater lacustrine to prodelta sediments. The lithology is mainly composed of dark-gray sandstones and siltstones (Table 1, Fig. 1).

3. Materials and methods

3.1. Materials

Nineteen mudstone samples were collected from seven wells in the Paleogene Shahejie Formation at depths of 2245–3492 m. Six samples from Es_3^2 are dominated by gray to dark-gray mudstones and calcareous mudstones. Five samples were collected from the Es_3^3 interval, which comprises lacustrine oil shales, calcareous mudstones and lamellar mudstones. Eight samples from Es_4^1 consist of gray to dark-gray mudstones, argillaceous limestone and gypsiferous mudstones (Table 2).

The rocks were first ground into powder by using a jaw crusher and hammer mill. The extraction of <2 μm clay fractions was conducted in accordance with Ding et al. (2013). The <2 μm clay fractions were prepared for XRD analysis.

3.2. Methods

3.2.1. Pyrolysis

A whole-rock sample of 50 mg was pyrolyzed by using a Rock-Eval 6 pyrolysis apparatus (Vinci Technologies, France). The crushed samples were first subjected to a 3 min isotherm at 300 °C, at which free hydrocarbons were volatilized (peak S1, mg/g of rock). Then, a programmed pyrolysis at 50 °C/min to 600 °C was

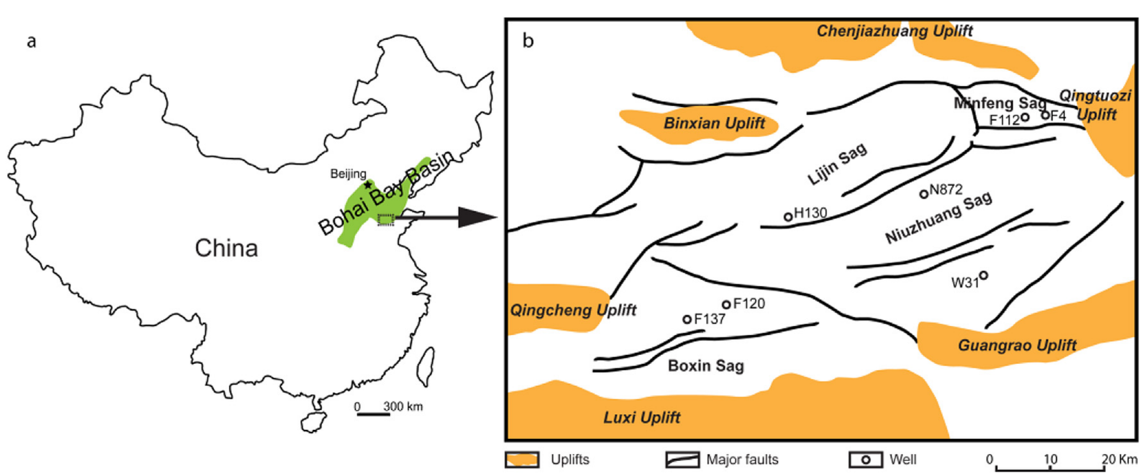

Fig. 1. Location map. (a) Study area in the Dongying Depression, Bohai Bay Basin, China. The dotted box denotes the Dongying Depression. (b) Structural map of the Dongying Depression with the locations of the sampled wells, modified after Zhang et al. (2009).