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RESEARCH ARTICLE

Application of quantitative grain fluorescence techniques to study subtle oil migration pathway of lithological reservoir

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Abstract This paper analyzes the quantitative grain fluorescence (QGF) and quantitative grain fluorescence on extract (QGF-E) properties of 101 rock samples by using quantitative grain fluorescence techniques. The samples are collected from five wells in tight sandstone and thin siltstone in the third sector of the Shahejie Formation in the Niuzhuang sag of the Dongying depression. It was observed that both the tight sandstone and thin siltstone show relatively high fluorescence intensity of hydrocarbon, which suggests that they are possibly good subtle oil-migration pathways in the present or geological time. These thin subtle oil-migration pathways afford important clues for the researches on hydrocarbon accumulation in lithological reservoirs in the middle and lower of Es_3 in deep sag zone, which has the hydrocarbon source from the upper of Es_4 when there is no apparent fault playing oil migration conduits to connect lithologic traps and deep source rocks. This study shows good prospect of QGF techniques in discriminating oil migration pathways and paleo-oil zones. The results of this study may be of great significance to the researches on hydrocarbon accumulation mechanism of subtle reservoirs in the Dongying depression and other areas.

Keywords QGF, QGF-E, subtle conduit, lithological pool, Dongying depression

1 Introduction

The oil and gas exploration in the continental basin of China has been at the important stage of searching for stratigraphic and lithological reservoirs which account for 42% of residual oil reserves, and has been the major target for increasing oil reserves in almost all the mature basins in China (Jia and Chi, 2004), especially in the Songliao and Bohai Bay basins (Guan and Li, 2003; Shan et al., 2005; Zhao et al., 2005; Feng et al., 2006). Three accumulation models for lithological reservoirs have been established in the published work. The hydrocarbon in the first model is generated in shallow strata and migrated to lithological reservoirs in deep strata along faults and fractures, such as the hydrocarbon in the Fuyang oilfield in the Songliao basin (Meng et al., 2004). The hydrocarbon in the second model is generated in source rock in deep sags and migrated to lithological reservoirs in the same strata by capillary pressure, stratigraphic pressure along pores and fractures through short migrating pathways (Zhang et al., 2000; Chen et al., 2004, 2006), such as the hydrocarbon in the Es_3^m in the Niuzhuang sag in the Jiyang superdepression, Putaohua oilfield in the Songliao basin, and in the west Liaohe basin. The hydrocarbon in the second model is generated in deep strata migrated to lithological reservoirs in shallow strata by abnormal stratigraphic pressure and buoyancy drive along faults that connect source rock and reservoirs, such as in Es_3^u and Es_2 in the Jiyang superdepression and in the Heidimiao strata in the North Songliao basin.

For a long time, the accumulation model for lithological reservoirs in the Es_3^m of the Niuzhuang sag in the Jiyang superdepression has been considered as the second model showing in the above models (Wang and Guan, 1999), and the Es_3 is regarded as the source rock for lithological reservoirs in the Es_3^m (Zhang, 1989). However, recent research work shows that the oil source rocks might be very complicated. Li et al. (2006) analyzed the oil in the reservoirs and the results of Pr/Ph(< 1). The high content of steranes and gamacerane as well as low tetracyclic/tricyclic terpane ratio are similar to the features of the source rock of Es_4^u in the Dongying depression. The high 4-methyl sterane shows the contribution of source rock of Es_3^l in the Dongying depression (Jiang and Li, 2005). These new searching results suggest that oil in the lithological reservoir in the Es_3^m in the Niuzhuang sag might originate from deep strata (Zhuo, 2005). However, hydrocarbon migration pathway or conduits have not been

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found. There are two hypotheses for the issue. One possible migration pathway is a large fault connecting source rock in the Es_4^u and reservoir in the Es_3^m and transiting hydrocarbon by abnormal stratigraphic pressure and buoyancy driving to form stratigraphic reservoirs. The other possible migration pathway is subtle conduits such as thin sandstones, small-scale fractures, and kerogen organic networks. Because the Niuzhuang deep sag is located in the intensive rifting stage in the Es_3^m, the faults developed within the sag zone has small-scale characteristics of only 1.5–6.0 km in striking in length, 30–50 m of fault fall. For the lithological reservoirs in the Es_3^m in the Niuzhuang deep sag, the urgent issue is how hydrocarbon is generated in the Es_4^u source rock passing through the several hundred meters strata to Es_3^m without fault playing an important role in the hydrocarbon migration. Subtle migration pathway is believed to be important in transiting hydrocarbons. However, the new challenge is how to detect whether these subtle conduits make effective transiting migration pathways. In this presentation, thin sandstone and liquid sample inclusions are selected to detect QGF-E and QGF to analyze oil-bearing property by QGF techniques.

2 Brief on quantitative grain fluorescence (QGF)

Quantitative grain fluorescence (QGF) and QGF-E (quantitative grain fluorescence on extract) are applied to analyze hydrocarbon migration pathways and paleo-oil zones (Liu et al., 2003, 2005; Jiang et al., 2006). A highly sensitive spectrophotometer is used in this method to analyze 1 g clean, dry, whole quartz grains in bulk volume (QGF) and the solvent extract (QGF-E) after a pre-cleaning procedure removes surface contaminants. Short ultraviolet (UV) excitation wavelengths of 254 and 260 nm for QGF and QGF-E are used, respectively, to record a continuous emission spectrum between 300 and 600 nm. The method is non-destructive and provides relatively rapid and cost-effective techniques for screening petroleum exploration wells for hydrocarbon charge history compared with existing fluorescence techniques (Liu et al., 2003, 2005). Fluorescence spectra of three kinds of petroleum, a condensate liquid, a light (42.5° API) and a heavy oil (25° API), respectively, and two oil fractions (tetra-aromatics and polars) prepared using liquid chromatography, were obtained as films deposited on an aluminum plate without any solvent dilution using a 254 nm excitation wavelength. The tetra-aromatic hydrocarbon and the polar fractions have spectra skewed towards the short wavelength and have distinct spectral maxima at around 360–380 nm. The tetra-aromatic fraction also has a secondary peak around 320 nm. The asphaltenes yield a rather symmetrical and broad spectrum with a spectral peak around 420 nm. The condensate and light oils are characterized by asymmetric fluorescence spectra skewed towards the shorter wavelengths between 300 and 600 nm with maximal intensities between 375 and 475 nm. The heavy oil has a broad spectral peak around 475 nm and shoulder between 475 and 600 nm. The spectra for the condensate, light and heavy oils shift progressively to longer wavelength. The tetra-aromatics and polar compounds have maximal intensities at 470 and 550 nm respectively, and secondary peaks at around 375 nm. The intensity of QGF samples reflects the concentration of crude oil or organic matter. Analysis and statistics of 182 samples in 13 wells in the Jiyang superdepression shows that the value of QGF-E ranges from 40 to 100 in oil-bearing layer, and ranges from 4 to 21 in water-bearing layer and the value of QGF is generally higher than 4 pc in oil-bearing layer, and less than 4 pc in water-bearing layer (Liu, 2002).

3 Samples and experiment

3.1 Samples

Many tight and thin sandstone layers developed in the Es_3^m and Es_3^l in sag zones in the Dongying depression,

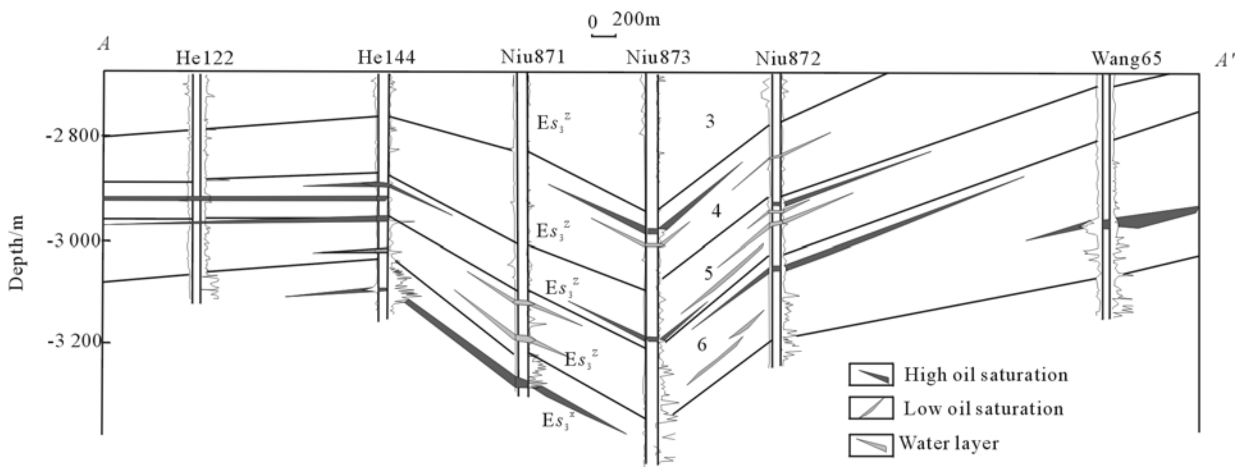

Fig. 1 Reservoir section of the Well He 122-Well Wang 65

and sandstones alternately deposit with mudstone in the sedimentary environment because of frequent fluctuating lake level. These thin sandstone layers are turbidites and are characterized by high mud content and carbonate content near the interface of sandstone and mudstone, which results in bad physical property in sandstone and low abundance of oil and

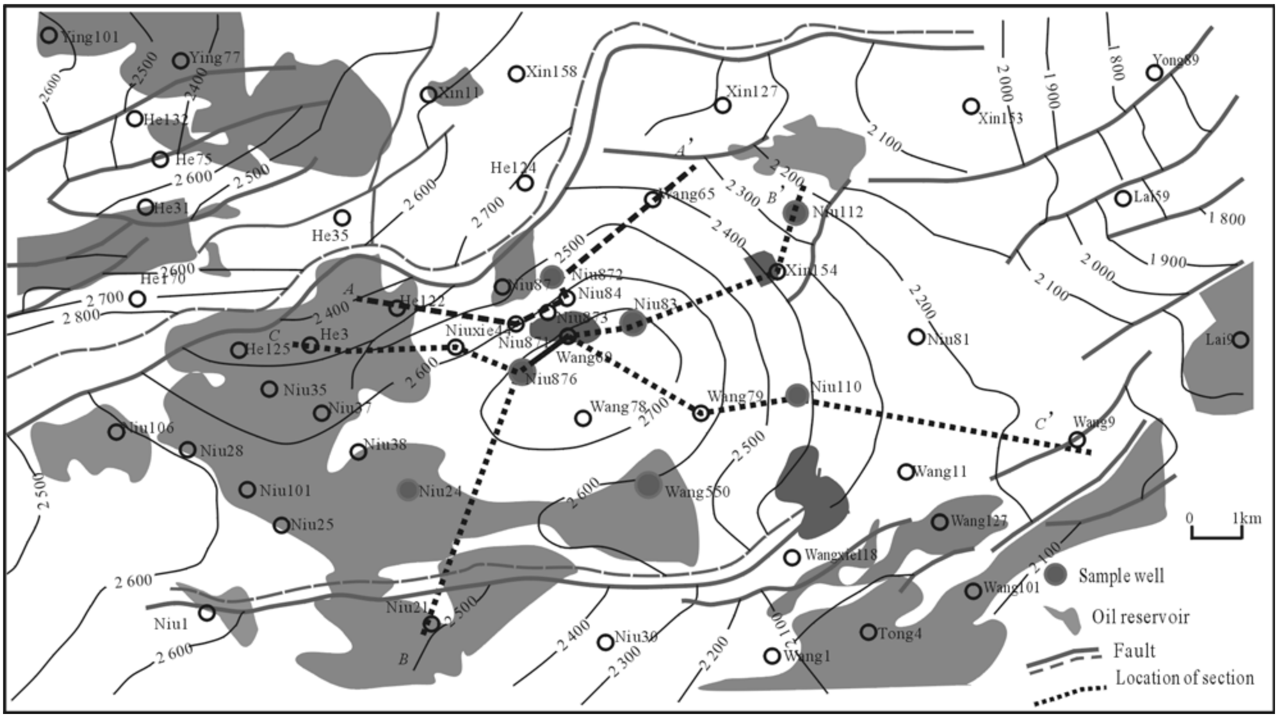

Fig. 2 Petroleum exploration sketch and sample location of the Niuzhuang sag, Dongying depression

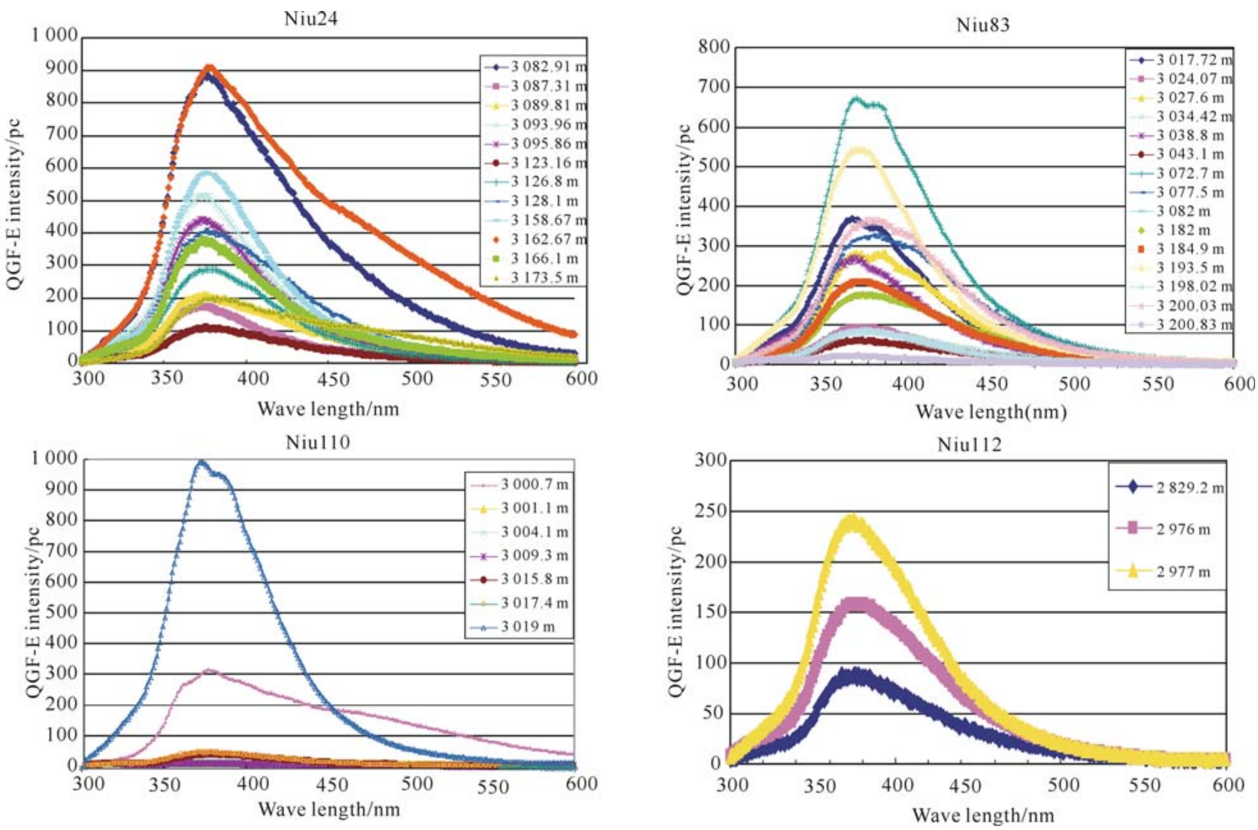

Fig. 3 QGF-E spectrogram of the reservoir grains from the Niuzhuang sag

gas. The statistics of the correlation of scale of sandstone and oil filing degree show that the sandstone is a non-effective oil layer when its thickness is less than 2 m, in areas less than 0.3 km^2 and with hydrocarbon filling degree less than 25%. Take the east-west pool section of Well He 122-Well Wang 65 (Fig. 1, location shown in Fig. 2) as an example. Dozens of thin sandstone layers can be detected by general seismic section and this value would be increased with the seismic solution. It is apparently shown in this figure that the filing degree of thick and wide sandstone will be higher, such as the sixth sandstone in the Es_3^m in the Well Niu 871, the fourth sandstone in the Es_3^m in the Well Niu873, and the fifth sandstone in the Es_3^m in the Well Niu 872. The filling degree of relative thin sandstone would be relatively lower, such as the sandstone located in the middle of the Wells Niu 872 and Niu 873. The filling degree will be far lower in the thinnest sandstone, such as the fourth sandstone in the Es_3^m in the Well Niu 872, and the fifth sandstone in the Es_3^m in the Well Niu 873. If these thin sandstones are not effective oil-bearing layers, can they be a hydrocarbon migration pathway or connect small fault or fracture for the hydrocarbon accumulation of the upper turbidite sandstones?

One hundred and one samples of the Wells Niu 24, Niu 83, Niu 876, Niu 110 and Niu 112 in the Niuzhuang sag in the Dongying depression are analyzed, the location of samples is shown in Fig. 2. These samples are almost from tight and thin siltstone in the Es_3^m and Es_3^l.

3.2 Method of experiment

Core and cutting samples are disaggregated into individual grains by physical crushing and gentle agitation. The grains are then sieved for selecting the sand fraction (63l–1 mm). The QGF cleaning procedure includes cleaning the grains in dichloromethane (DCM) in an ultrasound bath for 10 min to remove soluble hydrocarbons or additives. Samples are oven-dried at 60°C. The sample is then digested in 10% H_2O_2 at room temperature for 1 h with the initial and final 10 min in an ultrasonic bath to degrade and remove reactive organic compounds and clay particles adhering on the grain surfaces. The sand grains are decanted and then digested in 3.6% hydrochloric acid (1501 M HCl) at room temperature, with periodic agitation, to remove carbonate minerals and coatings that might produce mineral fluorescence, and iron oxides and hydroxides, which may coat grain surfaces. The remaining grains, mainly consisting of quartz and feldspar, are washed again with high-performance liquid chromatography (HPLC) grade DCM in an ultrasonic bath for 10 min to extract hydrocarbon compounds adhering on the grain surface. The

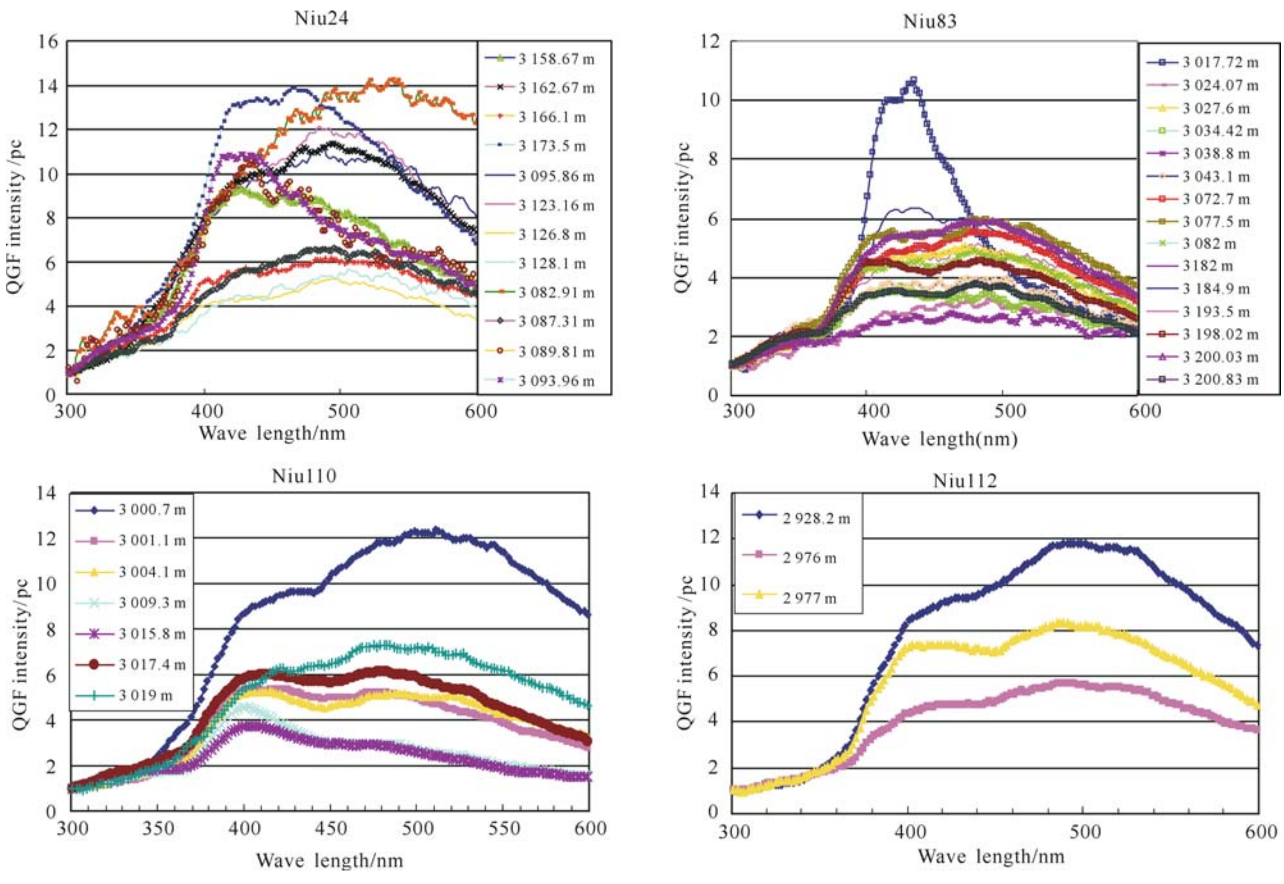

Fig. 4 QGF spectrogram of the reservoir grains from the Niuzhuang sag

solvent is preserved for QGF-E analysis, whereas the quartz grains are dried for QGF analysis.

3.3 Results of experiment

3.3.1 QGF-E fingerprint property of adsorption hydrocarbon in reservoir

Quantitative grain fluorescence on extract (QGF-E) fluorescence fingerprint property of adsorption hydrocarbon in sandstone of the Wells Niu 24, Niu 83, Niu 110 and Niu 112 in the Niuzhuang sag in the Dongying depression is shown in Fig. 3. The analysis results show that almost all the samples in the siltstone of the Well Niu 24 has high QGF-E intensity and all the values of intensity are higher than 40–100 pc, with some samples even reaching 909.6 pc, and the spectral ranges from 350 to 450 nm. Almost all the samples in the siltstone of the Well Niu83 also have high QGF-E intensity except for three samples with low intensity because it is silty mudstone. The QGF-E intensity reaches 178.7 pc in the section of 3,373.69 m of the Well Niu 876. These wells with high QGF-E intensity suggest high oil-bearing property. However, less than 40 pc QGF-E intensity is detected in several samples in the siltstone in the Wells Niu 110 and Niu 112 through one point reaching 900 pc, the relatively low QGF-E intensity suggests low oil-bearing property.

3.3.2 QGF fingerprint property of inclusion hydrocarbon in reservoir

Quantitative grain fluorescence (QGF) fluorescence fingerprint properties of inclusion hydrocarbon in sandstone of the Wells Niu 24, Niu 83, Niu 110 and Niu 112 in the Niuzhuang sag in the Dongying depression are shown in Fig. 4. The research of Jiang et al. (2006) showed that paleo-oil layers have characteristic spectra range peak from 375 to 475 nm,

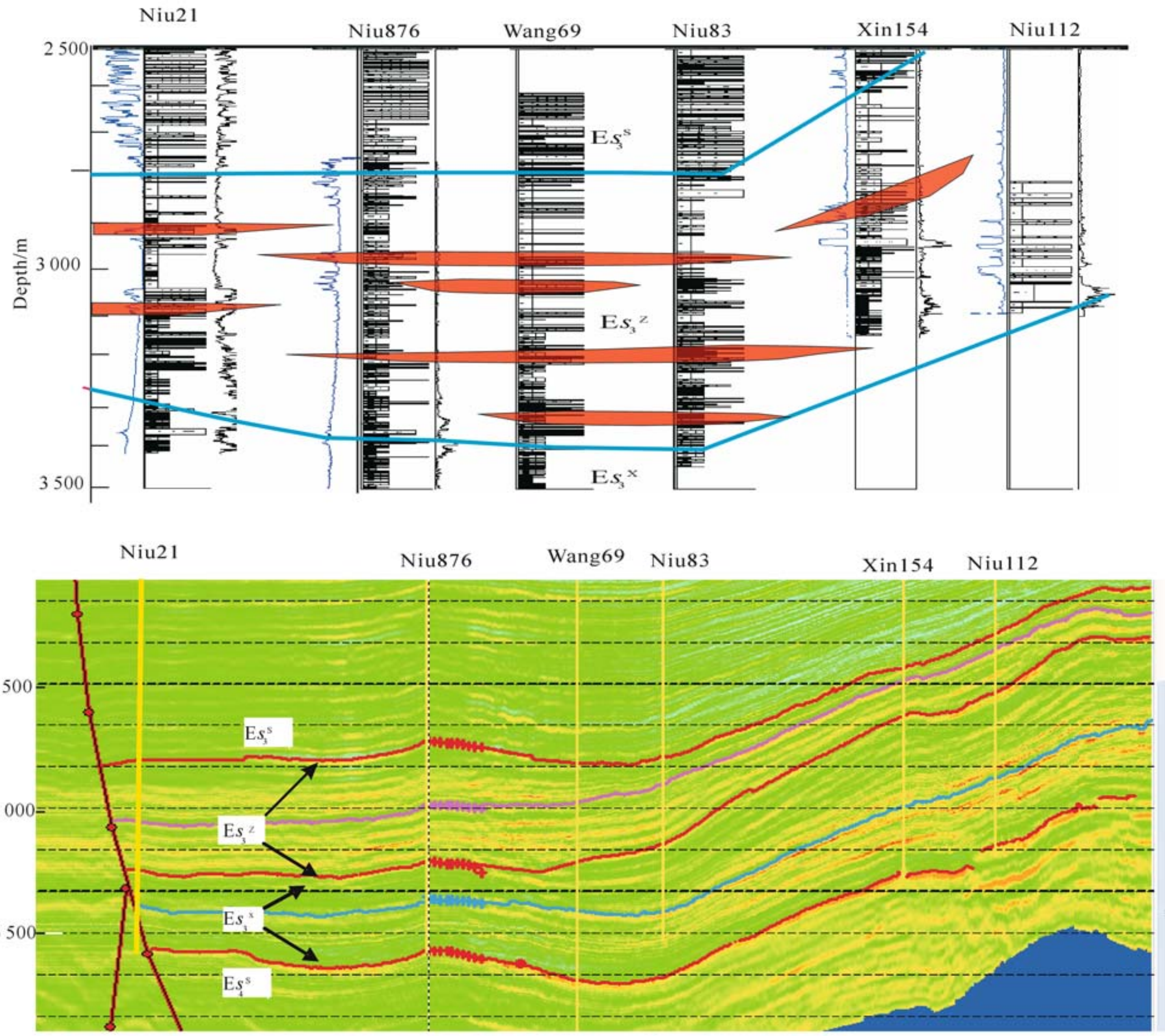

Fig. 5 Reservoir section and seismic interpretation section of the Well Niu 21-Well Niu 112

but most of the water layer has gentle fluorescence spectra and apparently lower fluorescence in the water layer than that in the oil layer. Almost all of the QGF intensity are higher than 4 pc in the samples of siltstone in the Wells Niu 24, Niu 83 and Niu 876, with some even higher than 14 pc in the Well Niu 876, showing high oil-bearing level. However, the QGF curve is gentle in the samples in the Wells Niu 110 and Niu 112 through having a QGF intensity higher than 4 pc, suggesting low oil-bearing level.

4 Discussion

The analysis on QGF-E and QGF of tight and thin siltstone in the Wells Niu 24, Niu 83, Niu 876, Wang 550 and Niu 872 (the spectral properties of the latter two are well shown in the paper of Li et al. (2006)) shows high fluorescence intensity detected in most samples except for a few samples with low value suggesting water-bearing property. This result may indicate that hydrocarbon had transited or migrated in these samples, suggesting these tight and thin siltstones could be good hydrocarbon migration pathway and conduits at present or in the geological time. A northeast-southwest seismic reflection section and oil pool section of the Wells Niu 27-Niiu 876-Wang 69-Niu 83-Xin 154-Niu 112 in the Niuzhuang sag zone is shown in Fig. 5 (location shown in Fig. 2 *B-B′*). A northeast-southwest seismic reflecting section and oil pool section of Wells Niuxie 44-Niu 876-Wang 69-Wang 79-Niu 110 in the Niuzhuang sag is shown in Fig. 6 (location shown in Fig. 2 *C-C′*). Lithological reservoirs of the Es_3^m and Es_3^l are discovered in the area of the Wells Niu 876, Wang 69 and Niu 83. The scale of lithological reservoirs in the Es_3^m is larger than the lithological reservoirs in the Es_3^l. The horizontal and longitudinal continuities of sandstones in the Es_3^m genetic turbidities are better than that of sandstone in the Es_3^l for hydrocarbon accumulation. The seismic reflection section shown in Figs. 5 and 6 also displays the horizontal and longitudinal continuity of sandstones in the Es_3^m. The different distribution properties of sandstone in the different sedimentary stages result in many thin sandstone layers in multi-stages superposed in a longitudinal section. Sandstones in the upper Es_3^m of Well Niu 876 and in the upper Es_3^m of Well Wang 69 in the seismic reflection section in Fig. 5 show many thin sandstone layers. These sandstones developed in the deep sag zone where no fault distinguished by seismic reflection section distributed near the sandstone. If

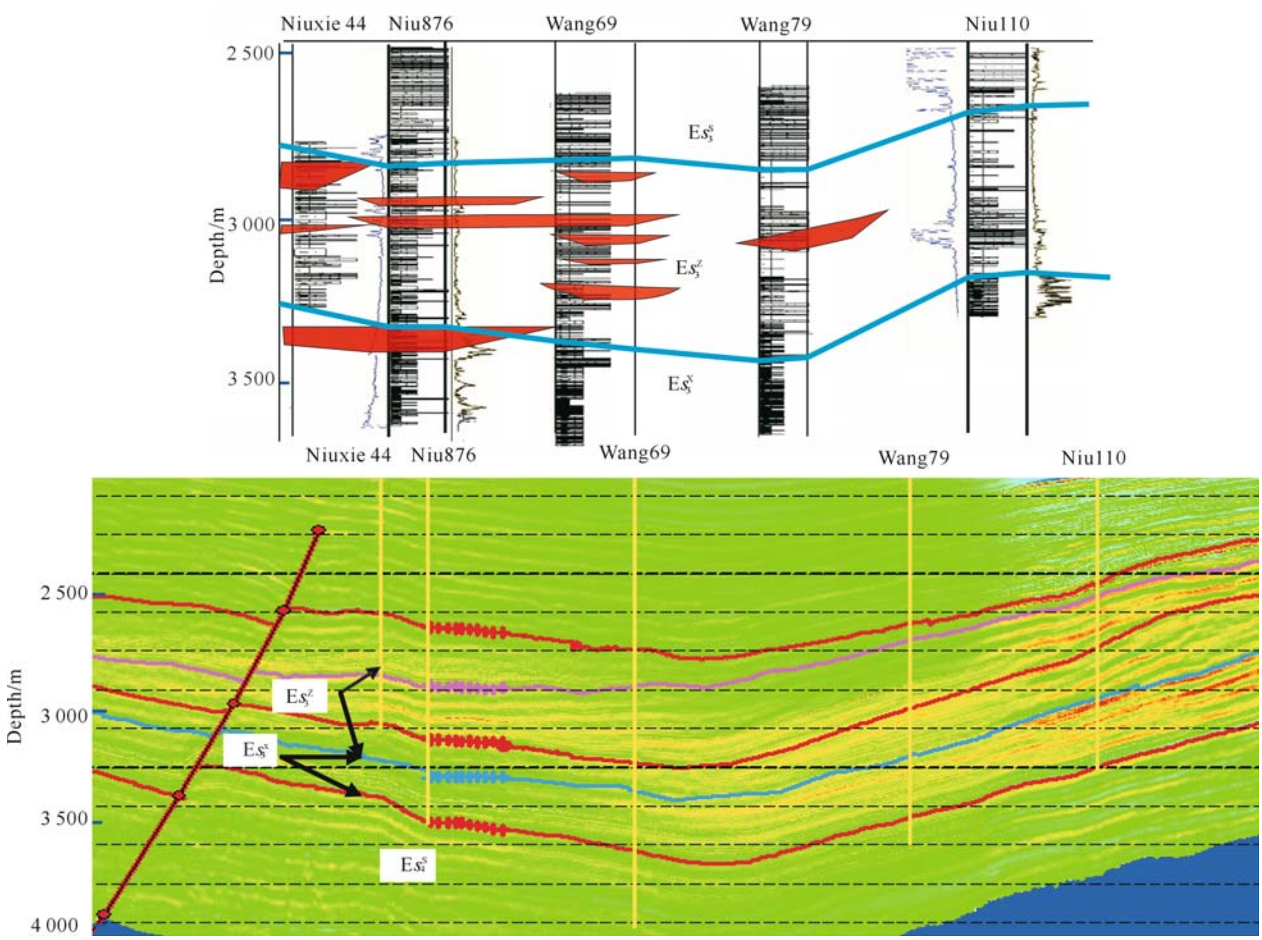

Fig. 6 Reservoir section and seismic interpretation section of the Well NiuXie 44-Well Niu 110

hydrocarbon migrated from source rock in the Es_4^u and Es_3^l to the upper lithological reservoirs in Es_3^m, the possible migration pathways and migration process may be hydrocarbon generated in the Es_4^u and Es_3^l migrating upward through organic network by buoyancy, and when hydrocarbon migrates in thin sandstone layers in the Es_3^m, some will accumulate in the well-developed sandstones with relative high porosity and permeability. Lithological reservoirs developed in the Es_3^m in the area of Wells Niu 876, Wang 69, Wang 68 (nearby Wang 79) and Niu 83 in the deep sag zone shown in the section of Figs. 5 and 6. SEM and core analyses show the sandstone in the wells Niu110 and Niu112 having high-physical property with an average porosity and permeability of the section of 3 000.5–3 022.4 m, being 19.6% and 96 mD, respectively, showing these sandstones are good reservoirs. No lithological reservoirs are discovered in the Wells Niu 110 and Niu 112 in the Es_3^m. The possible reason is that Well Niu 110 is not located in the range of effective source rock of Es_3^m, and no fault or sandstone connecting deep source rock in the Es_3^l and Es_4^u and lithological trap.

5 Conclusions

(1) Both the tight sandstone and thin siltstone show generally high fluorescence abundance of hydrocarbon and suggest that they are possibly good subtle oil-migration pathways at present or in geological time.

(2) Subtle oil-migration pathways afford important clues for the research of hydrocarbon accumulation in lithological pools in the middle and lower of the Es_3 in deep sag zone

(3) Good prospects of QGF techniques have important significance in discriminating subtle oil migration pathways and paleo-oil zones and in studying the hydrocarbon accumulation mechanism of subtle reservoirs.

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