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Distribution and geological significance of 17α(H)-diahopanes from different hydrocarbon source rocks of Yanchang Formation in Ordos Basin

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Based on GC-MS testing data of many saturated hydrocarbon samples, 17α(H)-C_{30} diahopanes (C_{30}^*) are extensively distributed in the lacustrine hydrocarbon source rocks of the Yanchang Formation in Ordos Basin, but show remarkable differences in relative abundance among various source rocks. Generally, Chang 7 high-quality source rock (oil shale) developed in deep lake anoxic environment shows lower C_{30}^* content, whereas Chang 6—9 dark mudstone developed in shallow to semi-deep lake, sub-oxidizing environment shows relatively high to high C_{30}^* value. Particularly, Chang 7 and Chang 9 black mudstones in Zhidan region in the northeast of the lake basin show extremely high C_{30}^* value. A comparative analysis was made based on lithology, organic types and various geochemical parameters indicative of redox environment, and the results indicate that environmental factors such as redox settings and lithology are key factors that control the C_{30}^* relative abundance, while organic types and maturity may be minor factors. High to extremely high C_{30}^* values are indicative of sub-oxidizing environment of fresh-brackish water and shallow to semi-deep lake. Therefore, research on C_{30}^* relative content and distribution in lacustrine hydrocarbon source rocks in the Yanchang Formation, especially on the difference in C_{30}^* between Chang 7 high-quality source rocks (oil shale) and Chang 6—9_1 source rocks (dark mudstone), will provide an important approach for classification of Mesozoic lacustrine crudes and detailed oil-source correlation in the basin.

diahopane, neohopane, sedimentary environment, Yanchang Formation, Ordos Basin

Diahopanes refer to a class of biomarkers with carbon ring framework identical to that of regular hopane, with methane side chain carbon position being distinct from that of regular hopane. Multiple homologues occur in hydrocarbon generation rocks or crudes, and 17α(H)-diahopane series are such homologues. Similar to regular hopane, 17α(H)-diahopane series generally show major peak at C_{30}(C_{30}^*). Moldowan et al. determined in the 1990s the structure of C_{30} diahopanes[1], and held that C_{30} diahopanes are consistent with the so-called Ts pseudo-homologous serial compounds (C_{29}—C_{34}) detected by Summons et al.[2] in the middle Proterozoic crudes in McArthur Basin in Australia. C_{30}^* is characterized by high thermal stability[3−5], and its precursors could possibly be bacterial hopanoids, which were regarded as being resulted from rearrangement of hopanes with functional groups on D rings[6]. Currently, many scholars have explored the formation conditions and geochemical attributes of diahopanes, and the majority held that sub-oxidizing environment, acidic media and catalysis facilitated by clay minerals are favorable for

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formation of C_{30}^*[1,6−11], while some scholars considered that catalysis by clay minerals under appropriate alkaline conditions is helpful to formation of C_{30}^*[12]. Bacterial hopanoids are generally regarded as biological precursors for diahopanes. However, as C_{30}^* can be detected frequently in coal measures, terrigenous hydrocarbon source rocks and terrigenous crudes, C_{30}^* is also regarded possibly as a kind of terrigenous biomarker[7], while some scholars proposed that red algae could possibly be the biogenic source of diahopanes[13]. As a result, arguments continue regarding precursors for diahopanes. As diahopane parameters show geochemical attributes like maturity[5] and sedimentary-organic facies[11], the relative abundance of diahopanes can be used for oil-source correlation[12] and regarded as a parameter for maturity[5,11]. Generally speaking, lithology, biogenic sources, redox attributes, sedimentary and diagenetic media conditions, thermal maturity of hydrocarbon generation rocks are important factors that affect the relative abundance of diahopanes. Nevertheless, the origin for diahopanes of high content remains a puzzle[9]. One possible explanation is that the key influencing factors could vary with regard to different geological conditions and study objects, so individual dedicated research is required.

In Zhidan region in Ordos Basin, exceptionally abundant diahopanes were first detected in Chang 9_1 black mud shale, which is a kind of lacustrine hydrocarbon source rock of the Yanchang Formation[14], and were subsequently detected in Chang 7 black mudstone. Further research indicates that remarkable differences in C_{30}^* content occurs among different reservoirs and different types of hydrocarbon source rocks. Furthermore, differences in C_{30}^* relative abundance occur among hydrocarbon source rocks of the same type and in the same reservoir group, for example, among Chang 7 oil shales. Therefore, further study is needed on the geological factors that affect the relative abundance of C_{30}^* in various reservoirs and various types of hydrocarbon source rocks by referring to previous research results, in order to discover the C_{30}^* distribution patterns and key control factors, which would be significant to the detailed correlation between Mesozoic oil and source rocks in the basin and to the accurate determination of dominant oil source rocks.

1 Geological background

In the Late Triassic, mainly affected by regional structural events including collision and collaging between North China Block and Yangtze Block and accelerated uplift of Qinling Mountains, large continental lake, i.e., the present day Ordos Basin, was formed in Shaanxi-Gansu-Ningxia area in the southwest of North China Block. Based on the characteristics of the Late Triassic sedimentary evolution (cycle) of continental lake basin and lithological combination in Ordos Basin and given the convenience for application in current exploration, the Upper Triassic Yanchang Formation can be divided into 10 reservoirs (Chang 10—1 from bottom to top in vertical direction). Here Chang 7 reservoir is a sedimentary stratum deposited at the maximum lake flooding stage, consisting of black mudstone of semi-deep lake facies, and large scale organic-rich high-grade hydrocarbon source rocks (oil shale) developed in the deep lake to semi-deep lake at the middle of the lake basin[15]. The stage for deposition of Chang 9 member underwent smaller scale lake flooding. Other reservoirs are typically composed of shallow water facies sandstone- mudstone combinations, as black fine-grained sediments of deep water facies, generally of smaller thickness below 5—10 m, only occur at the center or local depressions of the lake basin. Based on de- positional environment, lithology and organic precursor type, the hydrocarbon generation rocks of the Yanchang Formation can be divided into oil shale, black mudstone, argillaceous mudstone and carbonaceous mudstone. Here oil shale is generally developed in deep lake and semi-deep lake (Chang 7), reducing-anoxic environment, and shows high organic abundance and organic types mainly of I—II_1, which are similar to those for black mudstone[15], and hence is the dominant Mesozoic hydrocarbon source rock in the basin. Black mudstone is generally developed in semi-deep lake/shallow lake, sub-oxidizing environment, shows high clay content and high organic abundance, $\delta^{13}C_{PDB}$ of kerogen at about –29‰, high hydrogen index (generally more than 250 mg/g for most samples), organic types mainly of I—II_1, and hence is the important Mesozoic hydrocarbon source rocks. Argillaceous mudstone and carbonaceous mudstone (enriched in higher plant fossils) are both developed in shallow water and oxidizing environment, where higher plant input is remarkable, organic abundance is low for argillaceous mudstone and relatively high for carbonaceous mudstone, kerogen is relatively enriched in ^{13}C generally with $\delta^{13}C_{PDB}$>–26‰, hydrogen index is small and generally smaller than 200 mg/g, organic precursor types are mainly II_2—III, and thus argillaceous mudstone and carbonaceous mudstone belong to general hydrocarbon generation rocks.

For this study, 78 drill core samples of hydrocarbon generation rocks of the Yanchang Formation were collected. The samples were then rinsed with clean water, dried and crushed. Trichlormethane solvent after secondary distillation was used to extract bitumen "A", and then saturated hydrocarbon samples were acquired after column chromatographic separation and GC-MS analysis was performed respectively at State Key Laboratory of Organic Geochemistry of Guangzhou Institute of Geochemistry, Experimental Research Center of Petroleum Geology under PetroChina Institute of Exploration, Development and Research and Chemical Engineering College of Petroleum University.

2 Difference in 17α(H)-C_{30} diahopanes among hydrocarbon generation rocks of Yanchang Formation

As revealed by test data (Figure 1), hydrocarbon generation rocks of Yanchang Formation show significant variations in the relative abundance of C_{30}^*. Generally speaking, Chang 7 oil shale and Chang 6, Chang 4+5 carbonaceous mudstone show low to relatively low C_{30}^*, while the lacustrine dark-colored mudstone of various reservoirs show high to extremely high C_{30}^*.

Figure 1 shows that in hydrocarbon generation rocks, the relative abundance of C_{30}^* is positively correlated with those of other compounds, particularly rearranged steranes and terpanes-neohopane series and diasterane series. Samples of Chang 9_1 black mudstone and part of Chang 7 black mudstone, with exceptionally high C_{30}^*, show high diasteranes content, high Ts abundance, low regular hopanes abundance, etc. Samples of Chang 6, Chang 7, Chang 8 black mudstone, with high C_{30}^*, show abundant diasteranes, Ts and C_{29}Ts. Samples of Chang 7 oil shale, with low to relatively high C_{30}^*, show low to relatively low diasteranes content and abundant C_{30} hopanes.

3 Major factors affecting the relative abundance of C_{30}^*

3.1 Biogenic components

Generally, the abundance of hopanoid precursors, i.e., biogenic components for C_{30}^*, would affect the relative abundance of C_{30}^*. Based on this, some scholars deemed that the C_{30}^* relative abundance would possibly be related to terrigenous plants[7], red algae[13], and so on. As revealed by past research results[14,15], Chang 7 oil shale and Chang 9_1 black mudstone show similar organic precursor types, i.e., sapropelic-humic-sapropelic type, while Chang 7 black mudstone shows precursor-type-diagnostic geochemical indices like kerogen maceral, carbon isotope, primitive steranes distribution (Figure 1), etc., which are very close to those for Chang 7 oil shale and Chang 9_1 black mudstone. However, significant differences in C_{30}^* relative abundance occur among Chang 7, Chang 9_1 black mudstone and Chang 7 oil shale with similar precursor types (Figures 1 and 2). Figure 3 further demonstrates the remarkable differences in C_{30}^* relative abundance in Chang 7 oil shale among different bore holes; for example, Chang 7 oil shale from Ning 36 well and Zheng 8 well shows low C_{30}^* relative abundance (C_{30}^*/C_{30} hopane), while Chang 7 oil shale from Li 57 well shows obviously higher C_{30}^* relative abundance, Chang 7 oil shale from Li 68 well shows even higher C_{30}^* relative abundance. However, Chang 7 oil shale, a kind of hydrocarbon generation rock with distinct precursor types, and Chang 4+5 (Chang 6) carbonaceous mudstone with remarkable input of higher plants, all show low C_{30}^* relative abundance (Figure 1). Based on the difference in C_{30}^* relative abundance among hydrocarbon generation rocks with similar precursor types in Yanchang Formation and the similarity in C_{30}^* relative abundance among hydrocarbon generation rocks with distinct precursor types, the influence posed by biogenic components upon the C_{30}^* relative abundance remains to be unclear, and could be minor in significance as compared to other factors.

3.2 Lithology and rock fabrics

Chang 7 and Chang 9 black mudstone and Chang 7 oil shale with similar precursor types show remarkable differences in lithology, rock fabrics, organic enrichment and mode of occurrence, framboidal pyrite abundance, and clay minerals content. Therefore, correlation in C_{30}^* relative abundance between these two types of hydrocarbon source rocks can reflect the influence posed by rock fabrics of hydrocarbon source rocks on C_{30}^* relative abundance. As shown in Figure 4, samples with high C_{30}^* relative abundance (C_{30}^*/C_{30} hopane>1) are all black mudstone with TOC varying in the range of 2.9%—5.3%, while for Chang 7 oil shale with higher

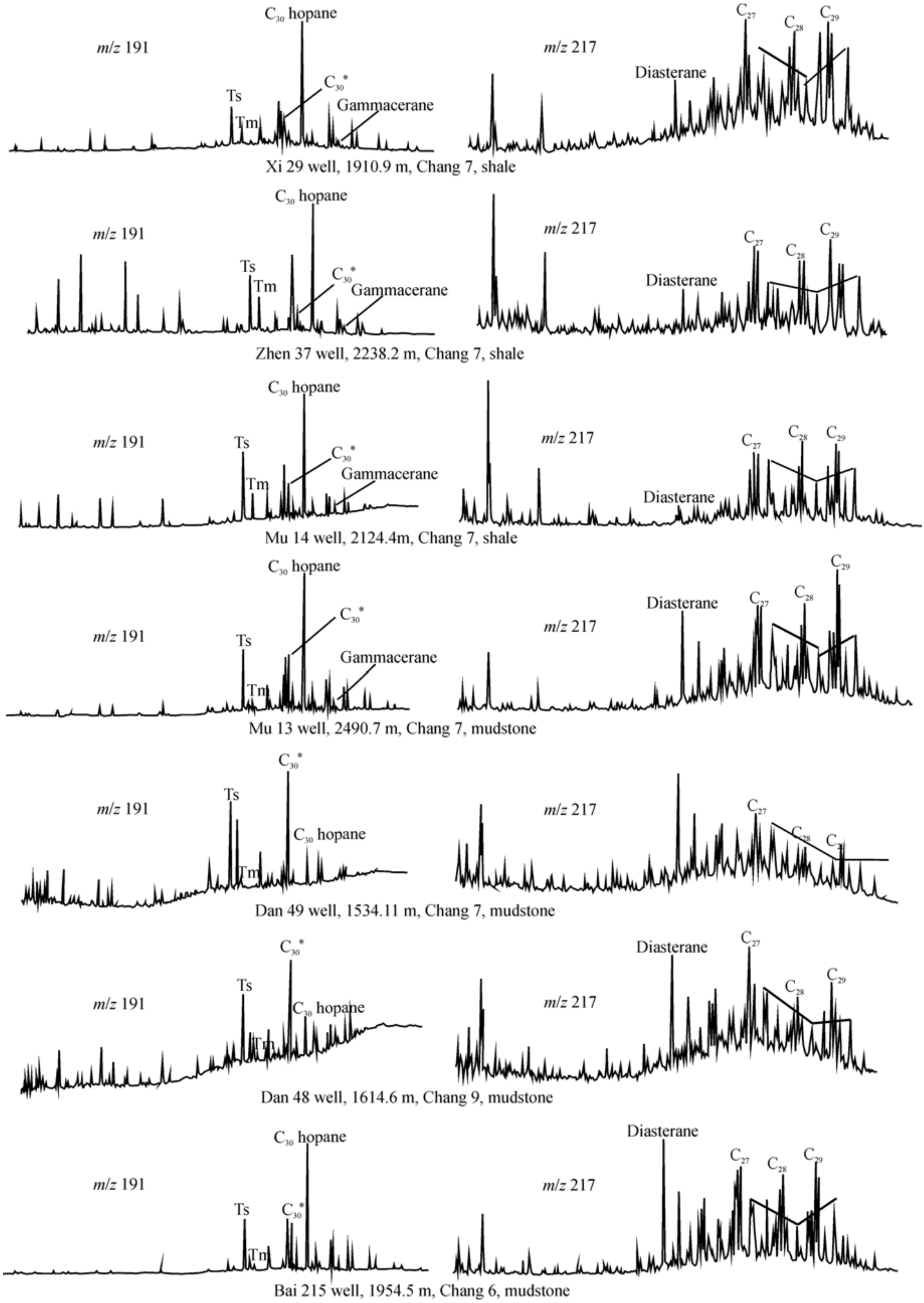

Figure 1 (*to be continued*)

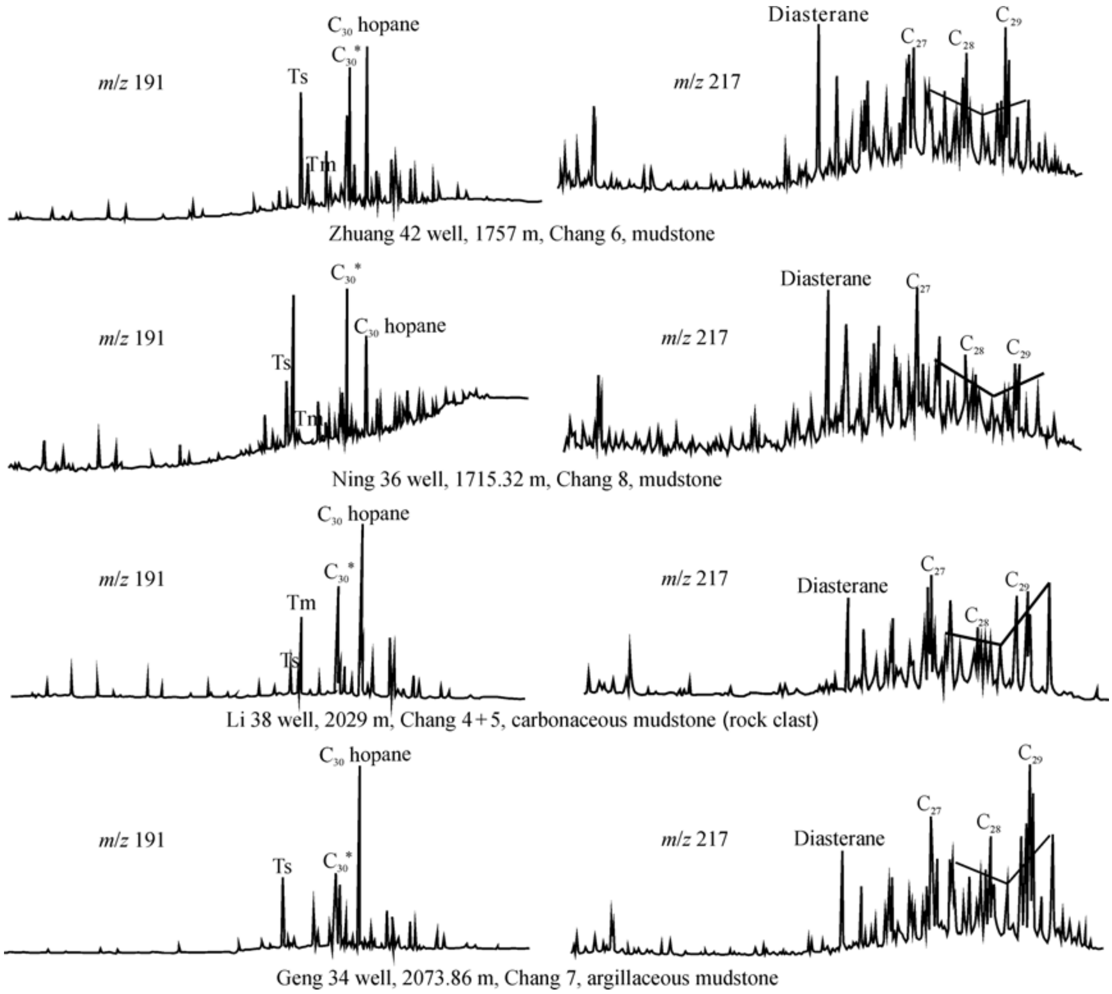

Figure 1 Diagrams showing distribution of steranes and terpanes in major lacustrine hydrocarbon source rocks of the Yanchang Formation.

organic abundance, no samples show very high C_{30}^* relative abundance, since samples with low C_{30}^* relative abundance (C_{30}^*/C_{30} hopane < 0.2) all belong to Chang 7 oil shale (Figure 2). In general, with increase in TOC, the C_{30}^* relative abundance of Chang 7 hydrocarbon generation rocks shows a trend of variation from dramatic increase to gradual decrease. From the correlation in C_{30}^* relative abundance among samples of hydrocarbon source rocks with different organic abundances taken from Chang 7 oil shale at the same bore hole (e.g., Li 57 well, see Figure 4), it can be seen that when TOC changes from 5% to 6%, C_{30}^*/C_{30} ratio shows remarkable decrease, when TOC>6%, C_{30}^* relative abundance shows little change with increase in TOC. This may be resulted from influence posed by the difference in sedimentation environment (redox attribute) on the one hand, and could also be quite possibly related to the remarkable difference in lithology and rock fabrics on the other hand. Numerous samples were observed under electron microscope, and the results indicate that Chang 7 oil shale with TOC>6% show mainly laminated organics and abundant framboidal pyrite[16] (Figure 5), while black mudstone with 3%－6% TOC shows mainly scattered organics and less abundant framboidal pyrite but abundant clay minerals (Figure 6). Here 5%－6% TOC corresponds to the zone where organic occurrence in rocks changes from scattered distribution to laminated distribution. Obviously, in this zone characterized by remarkable change in organic occurrence mode and corresponding decrease in clay minerals content, catalysis facilitated by clay minerals will also change significantly from intense to weak as a response. As a result, the difference in lithology and rock fabrics as well as the difference in induced catalysis facilitated by clay minerals may be the internal factors that result in variation of C_{30}^* relative abundance with TOC as shown in Figure 4.

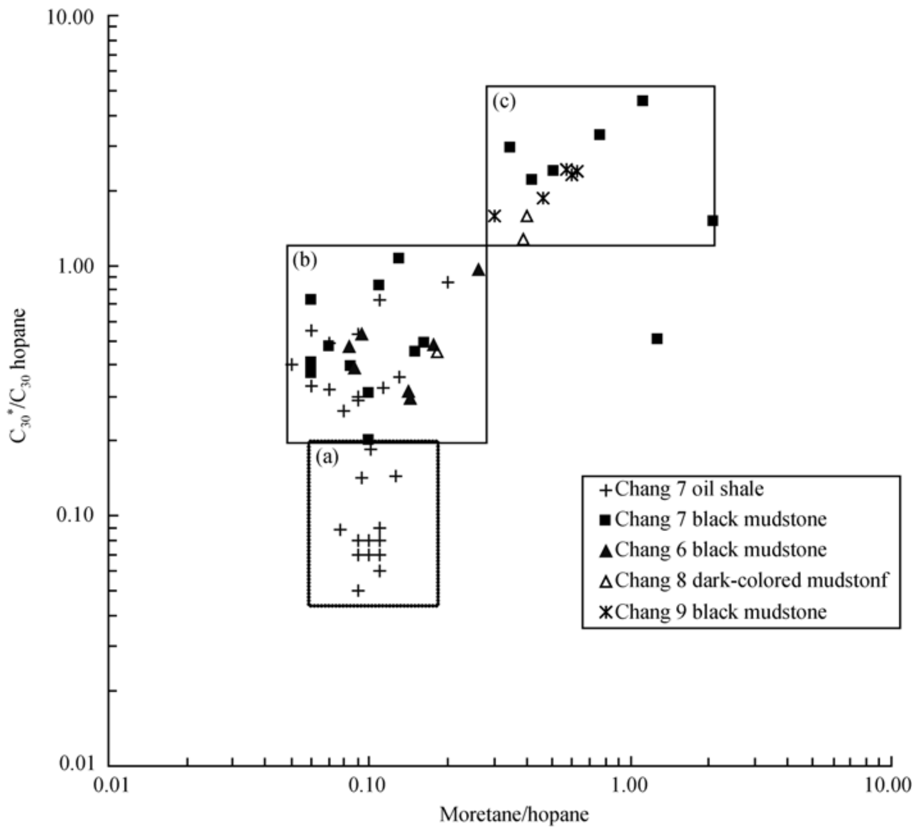

Figure 2 C_{30}^{*}/C_{30} hopane-C_{30} moretane/C_{30} hopane diagram for hydrocarbon source rocks of the Yanchang Formation.

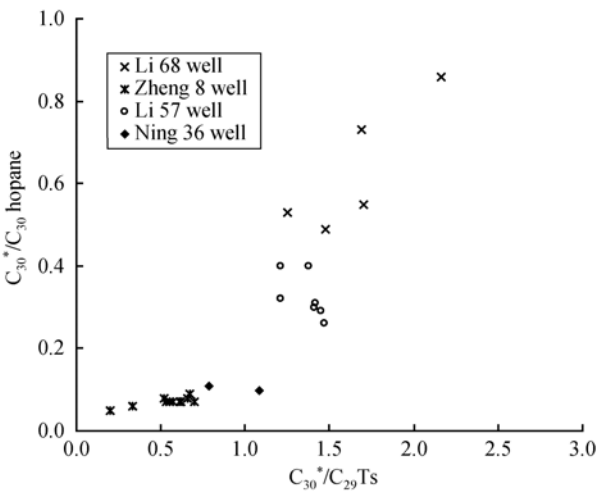

Figure 3 Comparison in C_{30}^{*} relative abundance in Chang 7 oil shale among different boreholes.

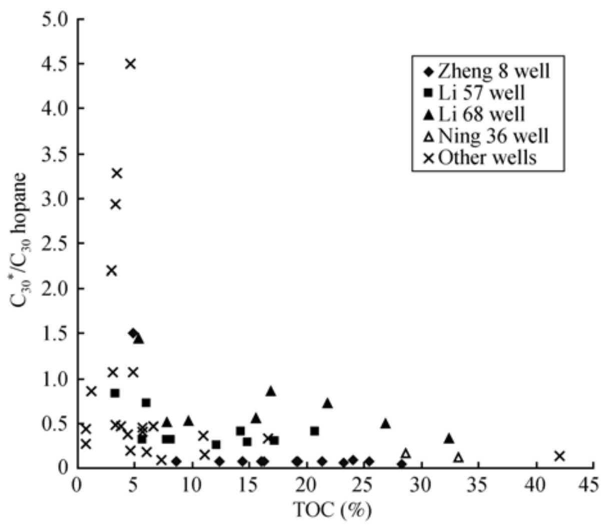

Figure 4 C_{30}^{*}/C_{30} hopane vs. TOC diagram for Chang 7 hydrocarbon generation rocks.

These discussions help explain that the differences in organic enrichment and rock fabrics among hydrocarbon generation rocks may induce differences in catalysis to organics (molecules) as facilitated by clay minerals in rocks, and further result in differences in C_{30}^{*} relative abundance. Higher clay minerals content and scattered distribution of organics seem to be more favorable for formation of high C_{30}^{*}. This is also supported by the fact that C_{30}^{*} of carbonaceous mudstone is remarkably lower than that of argillaceous mudstone.

3.3 Deposition and diagenetic environment (redox attribute)

Based on analysis of formation mechanism, the redox

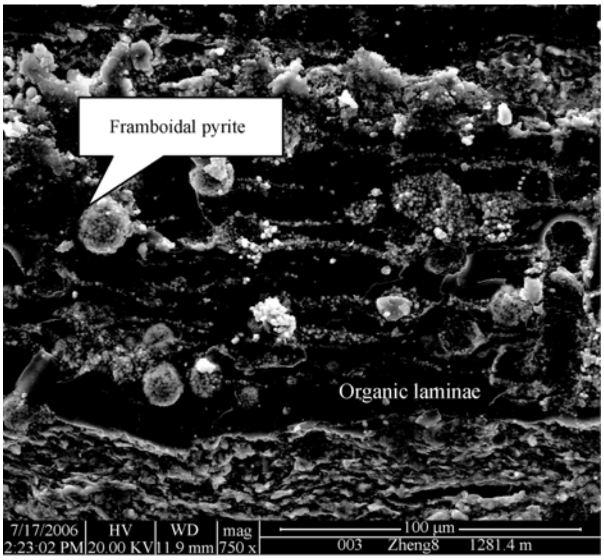

Figure 5 SEM photograph for a sample from Zheng 8 well, Chang 7 oil shale (TOC: 11.72%).

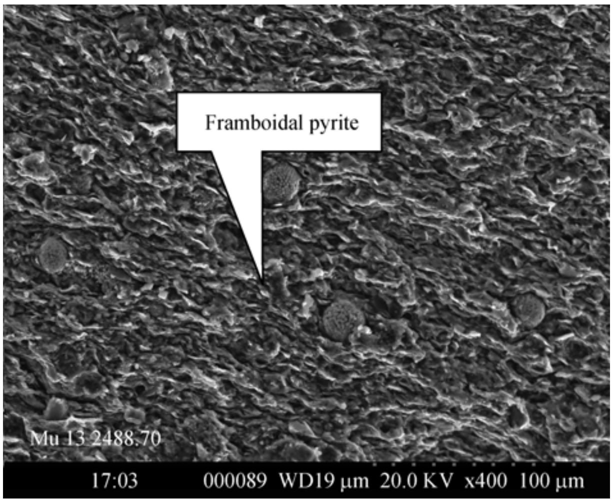

Figure 6 SEM photograph for a sample from Mu 13 well, Chang 7 black mudstone (TOC: 5.29%).

attributes for deposition and diagenetic environment are the important factors that affect C_{30}^* relative abundance in hydrocarbon generation rocks[1,6−12]. As reflected by the geological and geochemical data concerning hydrocarbon generation rocks of the Yanchang Formation (Table 1), Chang 6—9 black mudstone and Chang 7 oil shale, even with remarkable differences in C_{30}^* relative abundance, show similar organic precursor types (I—II_1), similar sedimentary water type (freshwater to slightly saline water), and similar balanced preponderance of both pristane and phytane, low gammacerane index, low Sr/Ba ratio[14,16]), but maximum differences in rock fabrics and redox environment resulted from different sedimentary facies zones. Generally speaking, Chang 7 oil shale mainly shows balanced preponderance of both pristane and phytane, but part of the samples may be affected by strong hydrocarbon expulsion and correspondingly show phytane preponderance, high to very high S^{2-} content, high U/Th, V/Sc, V/(V+Ni) ratios, etc., which are indicative of the deep lake to semi-deep lake, anoxic-weakly anoxic attributes of the sedimentary-diagenetic environment for Chang 7 oil shale, and this type of high-grade hydrocarbon source rocks generally show low to high C_{30}^*(C_{30}^*/C_{30} hopane varies from 0.05—0.86). Chang 6—9 black mudstone also shows mainly balanced preponderance of both pristane and phytane, but remarkably lower S^{2-} content, U/Th, V/Sc and V/(V+Ni) ratios than Chang 7 oil shale, which suggests that its development environment is less anoxic than that for Chang 7 oil shale, and is actually sub-oxidizing environment in semi-deep lake-shallow facies zone. This type of rocks show high to very high C_{30}^*. As a result, the redox attribute of depositional environment remarkably influences the C_{30}^* relative abundance in lacustrine hydrocarbon source rocks of the Yanchang Formation.

As for the lacustrine hydrocarbon source rocks, i.e., oil shale, black mudstone, of the Yanchang Formation, C_{30}^* relative abundance is a good environmental index, as high to very high C_{30}^* signifies shallow water and sub-oxidizing environment, while low C_{30}^* signifies anoxic sedimentary environment. Correspondingly, it can be inferred that the difference in C_{30}^*/C_{30} hopane ratio for Chang 7 oil shale among different bore holes as shown in Figure. 3 may mainly reflect the difference in anoxic degree, and hence the sedimentary-diagenetic environment for Chang 7 oil shale at Zheng 8 well and Ning 36 well (with low C_{30}^*/C_{30} hopane ratio) appears to be more anoxic than those at Li 57 well and Li 68 well (with high C_{30}^*/C_{30} hopane ratio), and appears to have higher water depth for sedimentation than the latter.

3.4 Thermal maturity

The thermal maturity of hydrocarbon generation rocks is also one of the factors that affect rearrangement of hopanoid (steroid) compounds. Our test data reveal that samples of Chang 7, Chang 9_1 hydrocarbon source rocks with high to very high C_{30}^* also show high diasteranes content, suggesting that C_{30}^* and diasteranes are similar in formation conditions and mechanisms. Generally, higher maturity is favorable for rearrangement of steranes, and formation of high diasteranes content[17].

Table 1 Part of geochemical parameters for lacustrine hydrocarbon generation rocks of Yanchang Formation[a)]

Horizon	Lithology	C_{30}^*	C_{30}^*/C_{30} hopane	Deposition facies	Organic type	Gammacerane/C_{30} hopane	Pr/Ph	U/Th	V/Sc	S^{2-} (%)
Chang 6	dark-colored mudstone	high	0.29－0.97	semi-deep lake	II_1	0.04－0.16	0.6－1.8	0.22－3.20	6.08－18.56	0.17－5.63
Chang 7	black mudstone	high-extremely high	0.40－4.51	semi-deep lake-shallow lake	I－II_1	0.05－0.18	0.8－1.5	0.52－1.19	6.65－11.67	0.33－2.84
	oil shale	low-high	0.05－0.86	deep lake-semi-deep lake	I－II_1	0.02－0.15	0.6－1.3	0.97－12.66	10.33－46.51	2.66－18.26
Chang 8	dark-colored mudstone	high-extremely high	0.45－1.58	shallow lake	II_1－II_2	0.09－0.46	>1.5	0.29－0.63	4.64－15.77	0.15－0.75
Chang 9_1	black mudstone	high-extremely high	1.57－2.39	semi-deep lake-shallow lake	I－II_1	0.20－0.37	1.4－1.55	0.24－0.35	5.45－7.38	0.89－2.97
Chang 6－9	argillaceous mudstone	low-high	0.12－0.56	shallow lake	II_2－III	0.03－0.23	>1.5	0.20－0.27	6.24－6.90	0.02－0.23

a) Classification plan based on C_{30}^* relative abundance: C_{30}^*/C_{30} hopane<0.2, low; C_{30}^*/C_{30} hopane＝0.2－1.0, high; C_{30}^*/C_{30} hopane >1.0, extremly high.

Similarly, it can be inferred that high maturity should also be favorable for formation of diahopanes. Geological investigation reveals that Chang 7 black mudstone and oil shale show similar maturity, and both have evolved into maturity stage, but show considerable difference in C_{30}^*/C_{30} hopane ratio. Moreover, samples taken from the same hydrocarbon source rock at the same bore hole also show remarkable difference in C_{30}^*/C_{30} hopane ratio (Figure 3), signifying that even with close maturity, other factors can affect C_{30}^* relative abundance more evidently. Therefore, attention shall be paid when C_{30}^* relative abundance is regarded as a maturity parameter.

4 Geological significance of C_{30}^* difference among lacustrine hydrocarbon source rocks of the Yanchang Formation

Because the Mesozoic crudes in the Ordos Basin generally show multiple distribution horizons, large distribution scope, singular genetic type, and all belong to lacustrine oil type[18－22], and hydrocarbon generation rocks of I－II_1 types were developed in different scales in Chang 4+5－Chang 9 members of the Yanchang Formation, detailed division of and correlation among lacustrine oil types, as well as accurate determination of dominant hydrocarbon source rocks and their relationships with reservoir groups in various regions and various reservoirs, are issues awaiting urgent solutions in petroleum exploration. The discussion as formulated in this study could provide an effective approach for final solutions to these issues.

As discussed above, among the multiple factors that affect C_{30}^* relative abundance, sedimentary facies zone (redox attribute) and rock fabrics (organic occurrence state and clay minerals content) prove to be major geological factors that control C_{30}^* relative abundance in terpane compounds in lacustrine hydrocarbon source rocks of Yanchang Formation. Obviously, sedimentary facies zone controls the lithology of hydrocarbon source rocks and redox attribute of their sedimentary environment; for example, Chang 7 oil shale was developed at the deep lake to semi-deep lake facies zone in the middle of the lake basin, and Chang 7 black mudstone was developed at the semi-deep lake to shallow lake facies zone. Therefore, C_{30}^* relative abundance can be an important index for recognition of hydrocarbon source rocks of Yanchang Formation, which provides an effective approach for division of crude groups and detailed correlation between crude-source rocks.

C_{30}^*/C_{30} hopane ratio and its positively-correlated parameters, such as C_{30} moretane/C_{30} hopane (Figure 2) can be used to formulate diagrams, for recognition of lithologic types of hydrocarbon source rocks of Yanchang Formation and detailed correlation between crude-source rocks.

As shown in C_{30}^*/C_{30} hopane-C_{30} moretane/C_{30} hopane diagram (Figure 2), C_{29}Ts/C_{29} norhopane-C_{30}^*/C_{29}Ts diagram (Figure 7), even though different hydrocarbon source rocks demonstrate some degree of overlap, sample points still show some rule of distribution: sample points distributed in the zone defined by low C_{30}^*/C_{30} hopane (<0.2) and low C_{30} moretane/C_{30} hopane (<0.2) (Figure 2 (a)) all belong to Chang 7 oil shale; samples of Chang 9_1 black mudstone in Zhidan

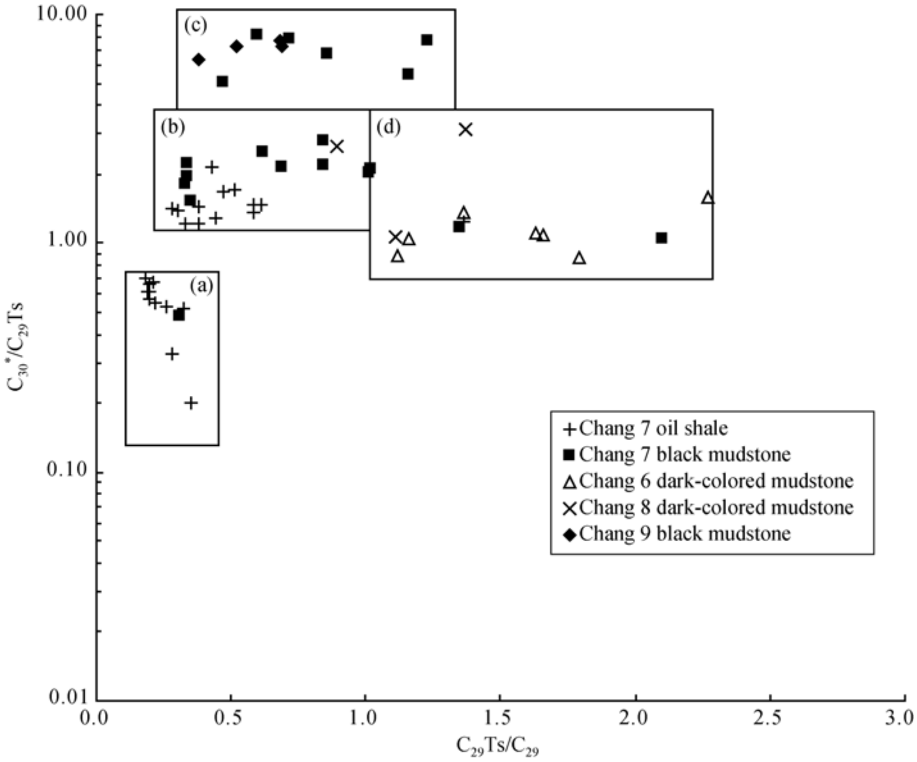

Figure 7 $C_{29}Ts/C_{29}$ norhopane-$C_{30}^*/C_{29}Ts$ diagram for hydrocarbon source rocks of the Yanchang Formation.

region are all distributed in the zone defined by exceptionally high C_{30}^*/C_{30} hopane (>1.0) and high C_{30} moretane/C_{30} hopane (>0.2) (Figure 2(c)). In the $C_{29}Ts/C_{29}$ norhopane-$C_{30}^*/C_{29}Ts$ diagram, Chang 7 oil shale and Chang 6 black mudstone, Chang 9 black mudstone can be easily distinguished from each other, and samples of Chang 7 oil shale are distributed in a relatively independent zone (Figure 7(a)). Of course, more parameters and similar diagrams with similar geological significance can be further grouped.

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