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Thermal Effect on the Distribution of Regular Sterane and Geological Significance

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Abstract

In order to study thermal effect on distribution characteristics of regular sterane in humic coals, a suit of 25 coals derived from Kuqa Depression in Tarim Basin and Ordos Basin are studied to investigate the distribution of regular sterane in different maturity, which mainly discusses the thermal effect. The results show that the $C_{27}aaa/C_{29}aaa$ regular sterane ratio has a linear relationship with R_o (t_{max}) and aromatic hydrocarbon maturity parameters in the coals from mature to high-mature stage. It suggests that the distribution of the C_{27}, C_{29} regular steranes will fail to reflect original source input in high evolutionary stage. On the basis of thermal simulation experiment results, it further proves that the distribution patterns result from the demethylation effect of C_{29} sterane in the mature to high mature evolution stage and the $C_{27}aaa/C_{29}aaa$ regular sterane increases with increasing maturity. The breakage of C-C key in branched chain from C_{27}, C_{29} regular steranes to make $C_{27}aaa/C_{29}aaa$ regular sterane ratio invariance at high and over mature stage. Therefore, the ratio can be used to distinguish the maturity in high and over mature stage.

Keywords

Sterane, Maturity, Pyrolysis Experiment, Geochemistry Characteristic, Oil-Rock Correlation

1. Introduction

Sterane refers to the compounds which derive from the animals, plants and other organisms for complex sterol mixture of coal, hydrocarbon source rocks and crude oil species [1]. Because of its special structure containing rich geochemical information, it is often used to study oil source correlation, sedimentary environment, thermal evolution degree, the biodegradation and geological evolution history [2] [3] [4] [5] [6]. So far, these geochemical properties of sterane compounds had been researched by many domestic and foreign scholars. The research has shown that the carbon number distribution of regular sterane is mainly affected by organic matter input, indicating that regular sterane compound could act as powerful source parameters. C_{27} regular sterane indicates the source of aquatic algae, while C_{29} regular sterane suggests the source of higher plants. So the relative percentage content of C_{27}, C_{28} and C_{29} regular sterane can be frequently used to judge source input and oil source correlation [1] [7] [8]. If the compound of C_{27} and C_{29} regular sterane appears on a double peak in the chromatograms, this can be speculated that the Organic matter is not only derived from continental deposits but also from Marine deposits [9].

In 1975, Alomon studied the influence of clay minerals for oil and gas formation mechanism using pyrolysis experiment [10]. Meanwhile, the relationship of sterane and thermal evolution was studied in the Paris Basin shale, and the maturity parameters of $C_{29}20S/(20S + 20R)$ and C_{29}-$\beta\beta/(\alpha\alpha + \beta\beta)$ regular sterane were first put forward by Mackenzie in 1980 [11]. Because the R configuration on sterane C-20 was considered that it exists only in the precursor of living creatures' body. With the buried depth and thermal evolution degree increasing, the R configuration gradually converts a mixture of R and S configuration until to balance. After that, the phenomenon of the C_{29} regular sterane maturity parameter reversing was found in high maturity of hydrocarbon source rocks [12] [13]. In addition, immature hydrocarbon source rocks have done the pyrolysis experiments in the conditions of 320˚C pyrolysis water using 72 h. The distribution of regular steranes showed great changes under pyrolysis experiments, compared with immature hydrocarbon source rocks. The relative content of the C_{27} regular sterane was found an increase in pyrolysis experiments with the maturity increasing. That is to say, when reaching high and over matured stage, the relatively percentage of C_{27}, C_{28} and C_{29} regular sterane can indicate homogenization. The result shows that the C_{27}, C_{28} and C_{29} regular sterane are uneffective to determine source type [14] [15] [16].

Although the sterane compounds have been researched widely, the relationships between the regular sterane and the thermal evolution degree have rarely been studied in nature profile. Hence, the present paper comprises a detailed study of 25 coal samples from the Tarim Basin and Ordos Basin. Through the study of the pyrolysis analysis, total organic carbon (*TOC*) analysis, Biomarker compounds and vitrinite reflectance and whole rock maceral analysis, the aim of this research is to explore the thermal effect on the regular sterane in hydrocarbon source rocks. In addition, it is significant for oil-source correlation and the identification of primary hydrocarbon source rocks in high-over mature, which provides the theoretical foundation for the hydrocarbon generation mechanism in coal.

2. Experimental and Samples

2.1. Samples

A total of 25 coal samples were collected from the Ordos Basin and Tarim Basin.

Ten coal samples are selected from C_2y, C_2j, C_2b, P_1t and P_1s of Carboniferous to Permian in Ordos Basin, which mainly develops alluvial fan, delta, marshes and tidal flat sedimentary environment [17] [18] [19] [20]. Another coal samples are from T_3h, T_3t, J_1y, J_2k and J_2q of Triassic to Jurassic, where sedimentary environment is fan delta system [19] [20]. Nineteen samples were selected for vitrinite reflectance and whole rock maceral analysis especially for this research. All coal samples were crushed into fine powder and analyzed for their contents of total organic carbon (TOC) and rock pyrolysis.

2.2. Analytical Methods

The measurement of reflectance and maceral were determined by Leica MPV-SP photomicroscope. Before the measurement, the samples were crushed to a maximum size of 1-mm. While at least 30 points were counted in every sample, then the type index (T_i) is used to calculate kerogen type indicators.

$$\text{TI} = \left(\text{sapropelic} \times 100 + \text{exinite} \times 50 + \text{vitrinite} \times (-75) + \text{inertinite} \times (-100)\right) / 100$$

Fortotal organic carbon (TOC) and rock pyrolysis, all the coal samples need to be crush into fine powder and analyzed. However, before TOC determinations, all samples were removed by HCl treatment. The total carbon content was determined by LECO CS-2000 induction furnace. Pyrolysis was determined by Rock Eval VI made in China.

Bitumen "A" was extracted by the Soxhlet extraction for 3 days with a dichloromethane/methanol mixture (93:7 v/v). Then, the extracted bitumens were fractionated into saturated, aromatic hydrocarbons, NSOs (nitrogen, sulfur and oxygen) and asphaltenes using open column chromatography filled silica gel and the alumina. The study mainly analyzes the saturate and aromatic hydrocarbons fraction. The model of gas chromatography-mass spectrometry is HP-GC 6980/5873 MSD. And the HP-6890 GC is equipped with a HP-5MS fused silica capillary column (30 m × 0.25 mm × 0.125 μm). Helium is used as the carrier gas with a rate of 1.0 ml/min. The injector temperature is 300˚C. The GC temperature is programmed to start at 50˚C for 1 min; the temperature will increase to 310˚C at a rate of 3˚C/min with a final hold of 18 min. The scanning range is 50 - 550 amu.

3. Results and Discussion

3.1. Basic Geochemical Characteristics

The results of rock pyrolysis and R_o determination showed that the maturity range of coal sample from the kuqa Depression in Tarim Basiniswide. t_{max} value range from 430˚C to 529˚C. R_o ranges from 0.51% to 1.63% with an average value of 0.72%. In contrast the maturity of the coal samples from the Ordos Basin is relatively high. The average of R_o is 0.99%. In the whole rock maceral, T_i is an index to judge the type of parent material. The T_i index can be calculated using the relative percentage content of vitrinite, exinite, inertinite and so on. T_i values in the selected coal sample is −68.48 - −37.88, which reveals that all coal samples are III kerogen (Table 1).

Table 1. The basic geochemical data of source rocks in the study area.

area	Well number	position	lithology			TOC%	R_o %	maceral					$T_i/1$	type
				t_{max} (˚C)	$S_1 + S_2$ / ($mg \cdot g^{-1}$)			*a*	*b*	*c*	*d*	*e*		
Kuqa	KC13	T_3t	coal	432	119.6	59.45	0.55	0.12	0.4	6.4	74.4	1.6	−53.78	III
	KC8	T_3h^4	coal	437	227.97	67.19	0.65	0.12	0	3.2	79.6	0.4	−58.38	III
	KC28	T_3t^3	coal	454	30.77	40.85	0.7	0.12	0	0.4	52.4	16.8	−55.78	III
	KC41	J_1y^1	coal	439	15.54	48.5	0.69	0.17	0	0.4	74	1.6	−56.73	III
	KC52	J_1y^1	coal	441	19.41	68.68		0.17	1.0	1.0	80.8	0	−59.18	III
	KC76	J_2k^2	coal	439	38.47	67.13	0.67	0.12	0.4	1.2	85.6	1.2	−64.38	III
	KC84	J_2k^2	coal	440	30.5	43.82								
	KC85	J_2k^2	coal	440	13.04	51.93	0.51	0.12	0	0.4	64.4	0	−47.98	III
	KC89	J_2q^1	coal	430	22.39	78.01	0.58	0.12	0.4	1.6	67.6	12.8	−62.28	III
	KC92	J_2q^2	coal	435	16.36	52.08								
	KP19	J_1y^1	coal	529	14.74	46.38								
	KP21	J_1y^1	coal	524	20.14	60.49	1.63	0.12	0	0	41.6	6.8	−37.88	III
	KP33	J_1y^2	coal	507	34.14	83.16	1.49	0.12	0	0	82	6	−67.38	III
	KP37	J_2k^2	coal	484	40.2	80.7								
	KP38	J_2k^2	coal	507	19.48	46.4	1.21	0.12	0	0	78	10	−68.38	III
Ordos Basin	CC-10	C_2b	coal	517	1.16	33.18	1.63	0.22	0	0	56.8	0	−42.38	III
	SU-8	C_2b	coal	456	141.79	58.43	0.88	0.12	0	0.8	84.8	2.4	−65.48	III
	WD-40	C_2j	coal	499	0.65	39.94								
	WD-9	P_1t	coal	463	115.51	63.03	0.87	0.55	0	0.4	76.8	7.2	−64.05	III
	BD-13	P_1t	coal	435	24.14	38.48	0.66	0.12	0	0.8	84.8	2.4	−65.48	III
	WD-3	P_1s	coal	441	115.51	63.03	0.66	0	0	0	68.8	12	−63.48	III
	S-7	P_1s	coal	478	69.45	33.05	1.15	0.12	1.6	0.8	71.2	16.8	−68.48	III
	SU-5	P_1s	coal	450	209.63	63.14	0.86	0.22	1.4	1.0	80	7.2	−65.43	III
	LL-2	P_1S	coal	472	89.13	54.54	1.13	0.5	0.6	0.8	74.2	13	−67.3	III
	BD-2	P_1s	coal	432	105.72	43.88	0.68	0.12	0	0.8	84.8	2.4	−65.48	III

Note: $T_i = (100a + 75b + 50c + (-75)d + (-100)e)/100$, *a*, *b*, *c*, *d* and *e* respectively represent nonfixiform organic matter, alginate, exinite, vitrinite and inertinite.

3.2. The Distribution of Sterane Compounds

In this study, the complete distribution of sterane series compounds was detected in the coal sample from Kuqa Depression and Ordos Basin. When the t_{max} value is about 440˚C (R_o = 0.7%), the distribution of C_{27} and C_{29} regular sterane is consistent with the typical coal measures hydrocarbon source rocks with C_{29} regular sterane accounting for absolute advantage. It presents asymmetrical "V" type or the "L" type (**Figure 1(a)**). With the thermal evolution degree increasing, the typical coal sample of C_{29} regular sterane relative content advantage has disappeared. On the contrary, the relative content of C_{27} regular sterane has a tendency of increasing (**Figure 1(b)**). The distributions of different maturity sterane on the same section area in Ordos basin revealed that the relative content of C_{27}, C_{29} regular sterane are consistent with typical coal measures hydrocarbon source rock under the condition of low maturity (t_{max} value 441˚C, R_o values 0.66%). In other words, the C_{29} regular sterane has absolute predominance. However, with the increase of maturity, when the t_{max} value reaches 499˚C, C_{27} regular sterane predominates, which is similar to **Figure 2(b)**. This phenomenon is consistent with previous studies, when thermal evolution degree is higher; the C_{27} regular sterane relative content will gradually increase with the increasing maturation [21]. Although the samples from the same section, the distribution of C_{27} sterane and C_{29} sterane is different in different maturity. However, the distribution of C_{27} sterane and C_{29} steraneis consistency are in different parts of the section under the condition of similar maturity. Compared C_{27} sterane and C_{29} sterane, C_{28} sterane has no obvious change in the same profile under the condition of different maturity. That is to say, the distribution of sterane cannot be used to distinguish the hydrocarbon source rocks in the same area under the condition of different maturity.

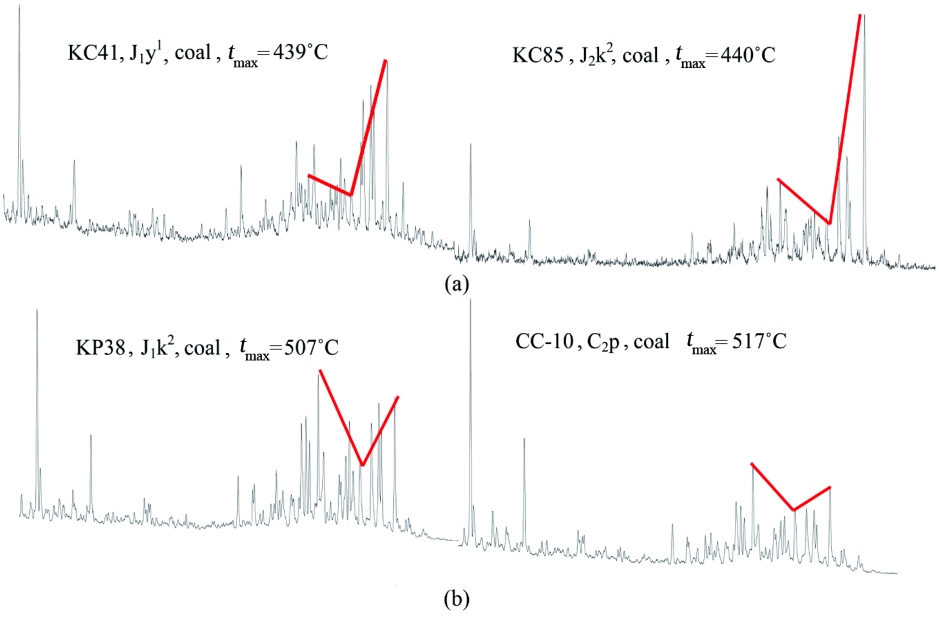


Figure 1. The distributions of sterane in different mature coal from Kuga sag and Ordos Basin.

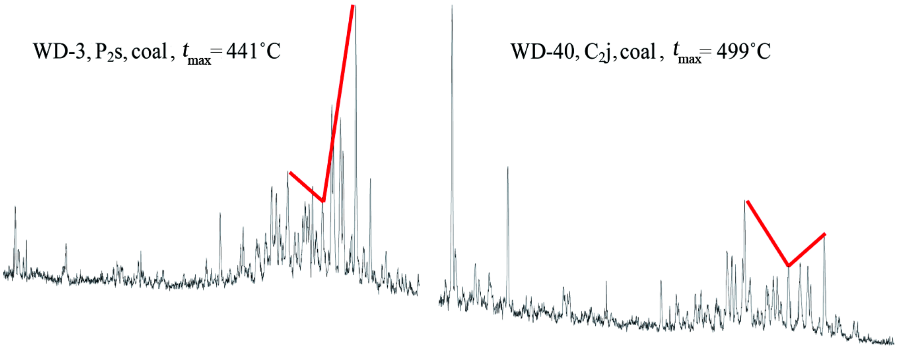


Figure 2. The distributions of sterane in different mature coal from the same profile Ordos Basin.

3.3. The Relative Content Change of Sterane Compounds

In general, aquatic organisms are rich in $C_{27}\alpha\alpha\alpha$20R sterane, while terrigenous higher plants are rich in $C_{29}\alpha\alpha\alpha$20R sterane. That view has been generally accepted [22]. The relationship between the relative content C_{27}, C_{28}, C_{29} regular sterane and the vitrinite reflectance R_o (%) in Kuqa Depression in Tarim Basin and Ordos Basin reveals that the thermal action has influence to the C_{27}, C_{28} and C_{29} regular sterane. When the R_o (%) value is less than 0.7%, the coal samples are still accounts for absolute advantage for C_{29} regular sterane. With the higher evolution degree, the relative content of C_{29} regular sterane gradually diminishes, while the C_{27} regular sterane increases. This leads to "invert" phenomenon that the content of C_{27} regular sterane is greater than the content of C_{29} regular sterane with the R_o (%) values increasing continually. In the stage of mature to high mature, the relative content of C_{28} regular sterane seems to be no obvious change. Thus, in the high and over mature evolutionary stages, when using the C_{27}, C_{28} and C_{29} regular sterane to judge parent material types needs to be careful. Otherwise, the conclusions from the distribution of regular sterane may be inconsistent with the practical point in the natural profile.

3.4. The Relationship between the Sterane Compound and the Maturity Parameter R_o and t_{max}

Previous studies illustrated the change rule of the C_{27}, C_{28} and C_{29} regulars terane compounds with the increasing maturity in simulation experiments. The change is quite outstanding especially in high or over maturity [14] [15] [16]. To further exploring the relationship between the sterane C_{27}, C_{28} and C_{29} regular sterane and the thermal action, the relationship between C_{27}/C_{29} regular sterane ratio and maturity parameters of R_o (%) and t_{max} (˚C) were discussed firstly (**Figure 3**). When R_o value is 0.7% - 1.5%, the C_{27}/C_{29} regular sterane ratio has a good linear relationship with R_o in the Kuqa Depression and the Ordos Basin (**Figure 4(a)**). Mean while, the C_{27}/C_{29} regular sterane ratio and t_{max} (<500˚C) is a linear relationship, which is similar to the R_o (%). That may be the thermal effect on the demethylation of C_{29} sterane obviously in the evolution stage of mature to high-mature [17] [23].

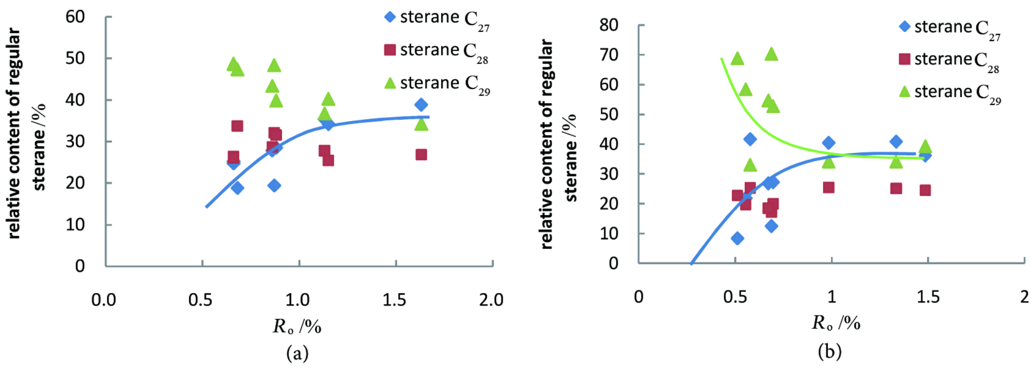


Figure 3. The plot relative content of regular sterane vs R_o (%) from Kuga depression and Ordos Basin.

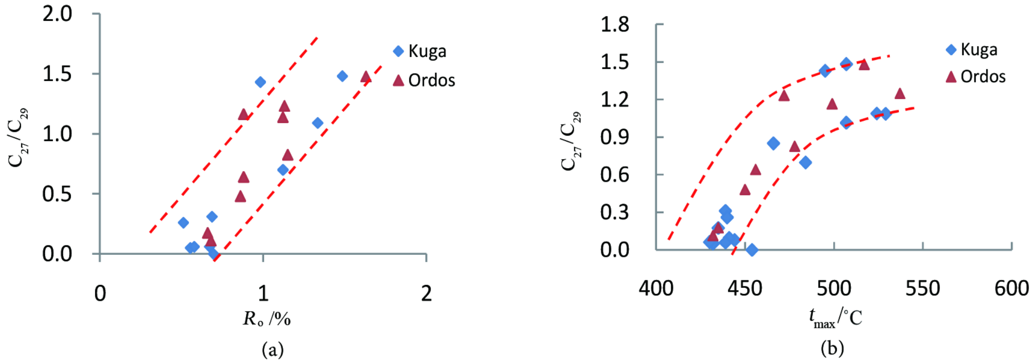
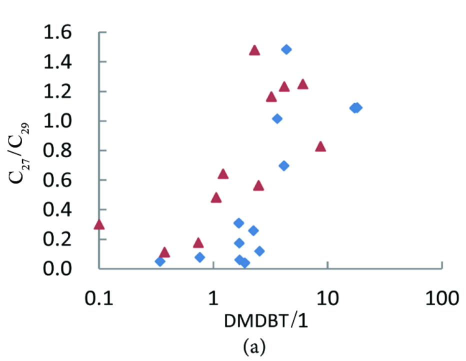


Figure 4. The plot sterane C_{27}/C_{29} vs R_o and t_{max} of coal from Kuga sag and Ordos Basin.

3.5. The Relationship between C_{27}/C_{29} Ratio and Aromatics Maturity

The coal samples are at low mature to over mature stage with t_{max} ranging from 432˚C to 529˚C. The maturity parameters of saturated hydrocarbons have reached equilibrium value; so it cannot effectively represent the maturity of samples. However, compared with the maturity parameters of saturated hydrocarbon, the maturity parameters of aromatic hydrocarbon could judge the high maturity samples. In addition to the commonly used methyl phenanthrene index MPI [24] [25], Chakhmakhchev *et al.* put forward dimethyl-benzothiophene maturity parameters. The works of the relationship between R_o and a dimethyl-benzothiophene ratio parameter have done a lot of. And the conversion formula of R_o and dimethyl-benzothiophene ratio has been put forward [26] [27]. The ratio of three methylnaphthalene TMNr = 2,3-TMN/(1,2,5-TMN + 2,3,6-TMN) were put forward [28]. And when the ratio of TMNr is greater than 0.6, it represents the high maturity [29]. In the aromatic compounds, many evaluation parameters can be used to evaluate the maturity. This present paper chose naphthalene, fluorene series compounds for research.

The C_{27}/C_{29} regular sterane ratios and aromatic maturity parameters DMDBT and TMNr also have a good linear relationship. And the relationship is similar to t_{max} and R_o (**Figure 5**). That is to say the thermal of hydrocarbon source rocks has distinct effect on the relative concentration distribution of regular sterane C_{27}, C_{28} and C_{29} regular sterane. So, in the same research area, the C_{27}/C_{29} regular sterane ratio can be used to determine hydrocarbon source rock maturity, especially in mature-high and over mature stage of evolution.

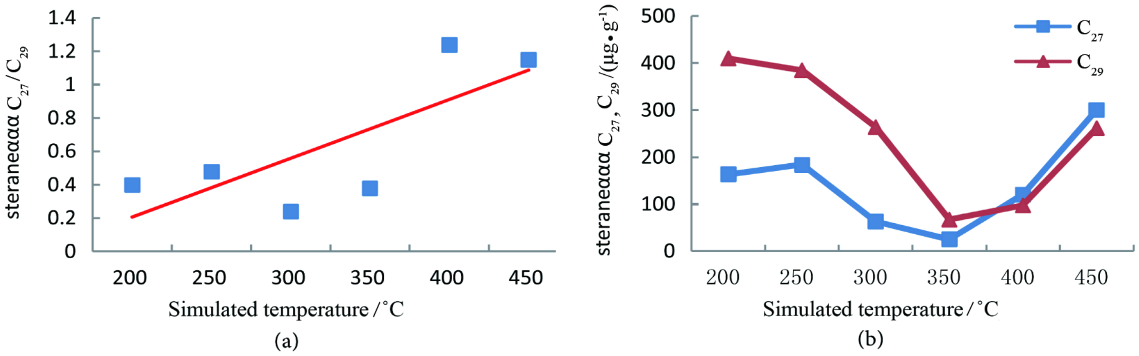


(a) (b)

Figure 5. The plot C_{27}/C_{29} sterane and aromatic maturity parameters from Kuga sag and Ordos Basin.

3.6. The Change of Sterane in Thermal Simulation Experiment

In order to demonstrate the thermal effect on sterane distribution rules, the thermal simulation test of a coal sample had been done. Six temperature points, 200˚C, 250˚C, 300˚C, 350˚C, 400˚C, 450˚C, were chosen in the process of experiment. The results show that C_{27}/C_{29} regular sterane ratio increases with the simulative temperature rising (**Figure 6**). This further elucidated that the influence of the distribution of C_{27} and C_{29} regular sterane in the sample is objective, especially, in the high-mature stage (thermal simulative temperature >350˚C). The relationship of the absolute concentration of *aaa*C_{27}, C_{29} regular sterane and pyrolysis temperature are shown in **Figure 6(b)**. The absolute concentration of C_{29} regular sterane decrease is faster than C_{27} sterane in low mature stage (thermal simulative temperature <350˚C). But when the temperature is greater than 350˚C, the C_{27} regular sterane is increasing faster than C_{29} regular sterane. This phenomenon proves that the C_{27}/C_{29} regular sterane ratio and the degree of thermal evolution have a certain correlation.



(a) (b)

Figure 6. The variation of ratios C_{27}/C_{29} sterane and C_{27}, C_{29} sterane absolute concentration with simulated temperature.

Structures determine properties in organic chemistry. It is because of the diversity of steranes compounds configuration change. Therefore, steranes compounds have high practical value in researching sources, thermal evolution and sedimentary environment of organic matter. In general, the longer chain of carbon alkane molecules is, the more will be easily broken. So the naphthene is steadier than side chain alkanes, and the more branched alkanes is prone to cracking reaction [30]. The C_{29} regular sterane adds an ethyl in C-24 place compared with the C_{27} regular sterane. Owing to hydrocarbons with different carbon number needing different chemical bond dissociation energy to fracture, it leads to the variation of C_{27}/C_{29} regular sterane ratio with the increasing thermal maturity (**Table 2**).

Table 2. Bond dissociation energy related with the structure of sedimentary organic matter (Hou Dujie, *et al.* 2011) [31].

C-C key	CH_3-CH_3	C_2H_5-CH_3	C_3H_7-CH_3	C_4H_9-CH_3	C_3H_7-C_2H_3
Bond dissociation energy (kJ/mol)	369	356	348	345	335

4. Conclusions

1) The steranes, especially C_{27}, C_{29} regular sterane compounds, are mainly controlled by source of parent material factor in evolution of hydrocarbon source rocks in the lower stage of development. The relative content of sterane compounds is very different in terrestrial higher plants and low aquatic algae parent material sources, so it can be served as an indicator of good source material. But on the high evolutionary stage, the relative content distribution of C_{27}, C_{28} and C_{29} regular sterane tends to be "homogenization". In high evolution stage, therefore, it needs to be cautious to use the parameters of the relative content to determine parent material types and oil-source correlation.

2) In order to describe the relationship of the relative content of regular sterane and the thermal action, the pyrolysis experiment has been performed to show that the C_{27}/C_{29} regular sterane ratio has a good of correlation with the maturity parameter like R_o, t_{max}, aromatic compounds DMDBT and TMNr. The results are similar to natural geological section, which suggests that the distribution of regular sterane will be effected by the thermal evolution.

3) To understand the characteristics of thermal evolution, the formation mechanism of the C_{27}/C_{29} regular sterane ratio with evolution degree changes is discussed by molecular structure stability. Due to the structural differences of C_{27} and C_{29} regular sterane, the demethylation effect is more apparent in C_{29} sterane than C_{27} sterane at mature to high mature evolution stage. It is the reason why the C_{27}/C_{29} regular sterane ratio rise with thermal evolution degree increasing; however, when the thermal evolution levels continue to rise, the breakage of C-C

key in branched chain makes C_{27}/C_{29} regular sterane ratio invariance at high and over mature stage. Therefore, the C_{27}/C_{29} regular sterane ratio may be used to distinguish hydrocarbon source rock maturity in the similar sedimentary system at high and over mature stage.

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