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Abnormal Concentration and Origin of Heavy Hydrocarbon in Upper Permian Coal Seams from Enhong Syncline, Yunnan, China

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ABSTRACT: Coalbed gas (CBG) in Enhong (恩洪) syncline, eastern Yunnan (云南), China, is characterized by high concentration of heavy hydrocarbon with the highest content of ethane, which is more than 30%. Some previous investigators paid much attention to the abnormal concentration of heavy hydrocarbon in the CBG of Enhong, but few have researched on its origin. This article describes the characteristics of abnormal high concentration of heavy hydrocarbon in Enhong syncline and analyzes its reason from the aspects of origin and evolution of heavy hydrocarbon by carbon isotope, coal petrography, and coal rank. Features of gas carbon isotope composition display that there is no inorganic gas or oil components in the CBG, which is classified to thermogenetic gas produced by humic material, with characteristic of secondary biogenic gas in shallow coal seam. The concentration of heavy hydrocarbon in Enhong syncline increases with the increase of vitrinite, vitrinite/inertinite ratio, and hydrogen/carbon ratio and decreases with the increase of inertinite, so hydrogen-rich vitrinite may be a very important factor resulting in the abnormal concentration of heavy hydrocarbon. It also increases with the increase of degree of coalification of coking to lean coals during which the peak of heavy hydrocarbon generation is reached. Therefore, we think that high concentration of heavy hydrocarbon originated from the coupling effect of higher content of the hydrogen-rich vitrinite in the coal and the coal rank of coking to lean coals.

KEY WORDS: coalbed gas, heavy hydrocarbon, abnormal concentration, gas isotope, hydrogen-rich vitrinite.

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INTRODUCTION

The molecular compositions of coalbed gas (CBG) mainly are CH_4, heavy hydrocarbon (C_{2+}), N_2, and CO_2, which can be classified into dry gas (concentration of C_{2+}<5%) and wet gas (concentration of C_{2+}>5%). CBG in China is generally characterized by dry gas. However, in some cases, the concentration of C_{2+} is up to 5% to 25% and even greater than the concentration of methane (Wu, 1994), which we call abnormal concentration of C_{2+}, such as in some parts of eastern Yunnan Province, western Guizhou Province, Chongqing City, central Jiangxi Province, southern Jiangsu Province, northern Zhejiang Province, and western Liaoning Province (He and Qin, 2007; Zhang et al., 2002). In the Enhong syncline of eastern Yunnan Province, the concentration of ethane in CBG varies from 4.38% to 33.90%, with an average value of 16%, and that of propane ranges from 0.7% to 5.88%, generally less than 3% (Wu et al., 2003).

Discussion on the origin of abnormal high concentration of heavy hydrocarbon in CBG is very valuable to understand the CBG source, to optimize CBG utilization, and to prevent gas disasters in coalmines. Previous interpretations have suggested the gas-generating parent material, exogenous oil and gas mixture, contact metamorphism, and coalification stage (Qin et al., 1998; Rice, 1993; Yu and Li, 1981).

Wu (1994) suggested that hydrocarbon displacement effects (larger molecules take up the available pore to accumulate and make the smaller ones migrate), different adsorption of coal to various gas components (adsorption ability of coal to C_{2+} is larger than CH_4), and molecular sieving effects of micropore in coal (larger molecules are controlled in the pores for their sizes and smaller molecules are easier to migrate) could account for the abnormal composition. However, although abnormal concentration of heavy coalbed hydrocarbons in Enhong syncline has received growing attention for many years, detailed studies on its origin have been very limited thus far.

Enhong syncline is located in eastern Yunnan Province, South China, where the Upper Permian coal-bearing strata are included in the Xuanwei Formation (Fig. 1). The thickness of the strata varies from 205 to 335 m, averaging 250 m, and total thickness of the coal seams ranges from 15.99 to 67.68 m, with an average of 18.04 m. Lithologies of lower Xuanwei Formation are gray siltstone and fine sandstone with wave bedding, those of middle section mainly are shallow sandy mudstone with horizontal bedding, and those of upper section are green gray sandy mudstone, fine sandstone, siltstone, and coal. Enhong syncline is a large synclinorium whose main axis extends in a near NNE to SN direction, with numerous secondary folds separated by the major faults and cut by numerous associated and/or induced fractures (Fig. 1).

EXPERIMENTS AND ANALYTICAL METHODS

Data in this article come from coalmines and CBG wells in the Enhong syncline. Basic information on the coals in this area was derived from data from 1 208 boreholes drilled in the coal geological exploration. Coal samples for the basic experiments are from boreholes with monolayer coal thickness equal to or greater than 0.6 m. Petrographic, proximate, and ultimate analyses of the coal samples and molecule composition and gas content of gas samples are carried out in laboratories of exploration teams. Table 1 summarizes the 1 208 borehole data and Tables 2 and 3 show the statistics of 118 borehole data of gases containing abnormal heavy hydrocarbon.

Data in Table 4 were derived from CBG in SJ-01 well. Three gas samples were taken from each sample desorption canister for compositional analysis. The gas content measurements were executed following the Xi'an Branch's specification for gas content measuring and referring to the direct method of American Mining Bureau's standards. The analysis of gas composition follows the National Standard GB/T 13610-1996. Proximate analysis follows the National Standard GB/T 212-1991. The ultimate analysis measuring follows the National Standard GB/T 476-2001. The maceral composition determination follows the National Standard GB/T 8899-1998. The vitrinite reflectance determination follows the National Standard GB/T 6948-1998.

RESULTS

Proximate Analysis

Table 1 shows the organic composition, proximate, and ultimate data of the coal seams studied. The ash yields of the samples vary from 4.76% to 49.59%, with an average value of 22.81%, which suggest medium-high ash coals. The ash yields in the middle part of the formation are lower than in the top and bottom parts (coal seams below No. 21 and above No. 7-1). No. 9 coal seam has the lowest ash yield, with an average value of 16%.

Total sulfur content of the samples varies from 0.06% to 28.00%, with an average of 1.88%, which indicates very low to high sulfur coals. In vertical profile, total sulfur decreases significantly from base to top, whereas, in horizontal distribution, the northwestern part of the syncline is obviously higher than the southeastern part. Volatile yield of the samples varies from 18.84% to 24.83%, with an average of 21.11% and increases from bottom to top in vertical profile and from southeast to northwest in horizontal distribution.

Petrographic Analysis

Lithotypes of the coal seams in Enhong syncline are dominated by clarain and durain. The main maceral component is vitrinite and ranges from 50.3% to 97.8%, averaging 74%. The content of inertinite varies between 1.0% and 41.4%, averaging 18%. Liptinite content is very low, usually less than 1%. Inorganic components are dominated by clay minerals (12%) and secondly by quartz (1%–20%) and sulfide (0–3.6%).

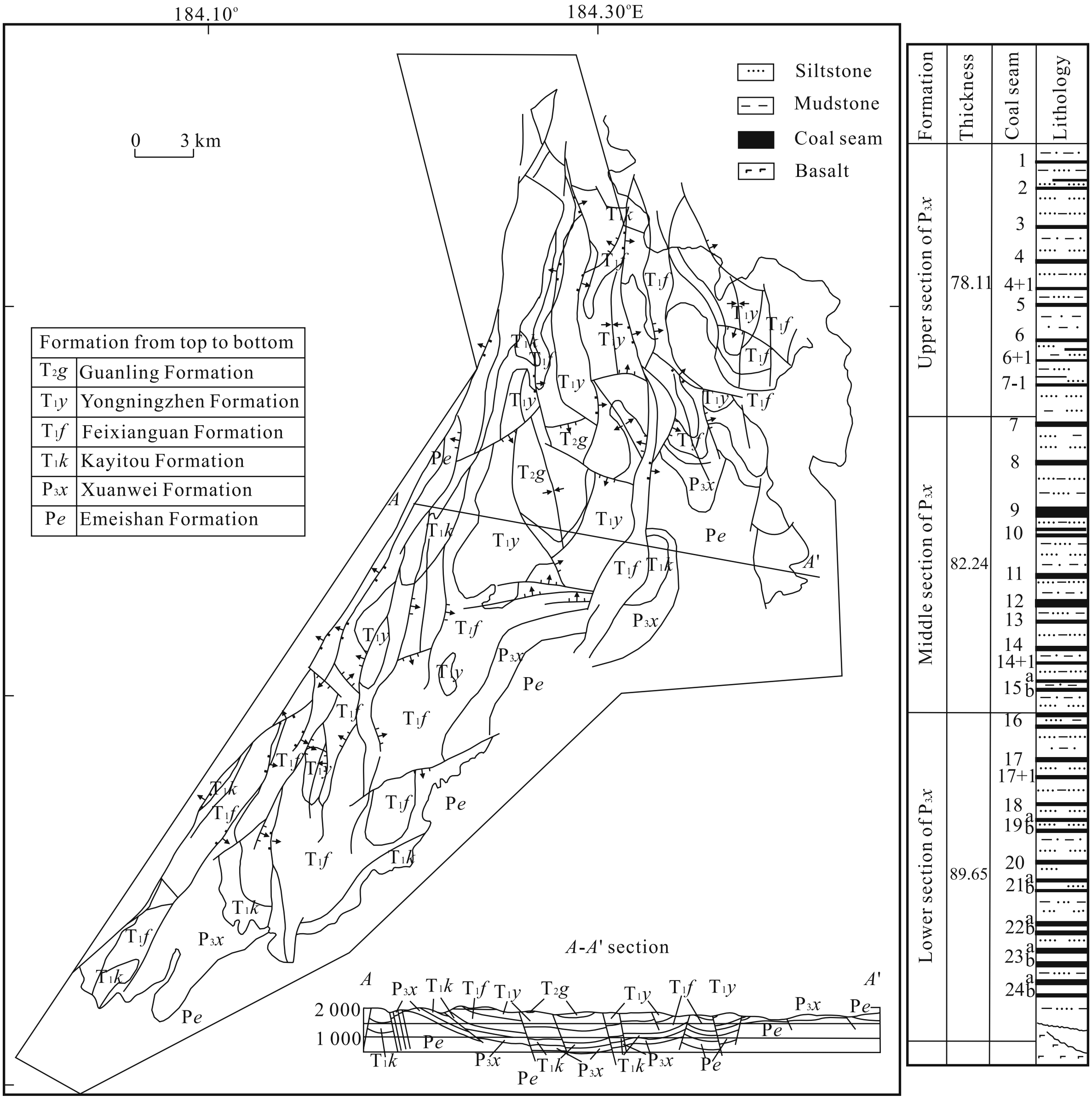

Figure 1. Structure outline, section map and stratigraphic column of Enhong syncline.

Table 1 Organic composition, proximate and ultimate analyses of coal seam studied

Coal seam	Organic matter (%)			*MM* (%)	Proximate analysis (%)				$S_{t, d}$ (%)	Ultimate analysis (%)				$R_{o, max}$ (%)
	V	*I*	*E*		M_{ad}	A_d	V_{daf}	*FC*		C_{daf}	H_{daf}	N_{daf}	$(O+S)_{daf}$	
1	66.1	33.6	0.3	14.6										1.28
2	60.1	39.9		20.2										1.35
3	66.6	32.6	0.8	24.7										1.36
4	58.2	41.8		30.8										1.35
6+1	67.7	32.2	0.1	21.9										1.42
7-1	76.0	23.9		23.3	0.6	24.3	24.8	56.6	0.2	88.8	4.9	2.8	3.1	1.48
7	72.5	27.5		20.1	0.8	21.9	22.6	60.0	0.2	89.5	5.0	2.0	3.5	1.50
9	78.7	20.7	0.6	10.7	0.9	16.0	22.1	64.8	0.2	98.9	5.0	1.9	3.7	1.42
11	77.2	22.6	0.2	13.8	0.8	22.3	21.3	61.5	0.2	89.6	4.9	1.9	3.6	1.40
13	69.5	29.9	0.5	24.3	0.8	22.9	23.7	58.3	0.2	89.5	5.0	1.8	3.5	1.54
14	73.2	26.5	0.3	15.4	0.9	22.8	20.5	60.8	0.3	89.4	4.8	1.5	4.2	1.38
14+1	73.0	27		17.5										1.48
15a	77.0	23		21.0	0.9	21.4	20.6	61.9	1.4	90.0	4.7	1.8	3.2	1.43
15b	77.8	22.2		18.1										1.55
16	74.8	25.1	0.1	15.6	0.8	20.6	19.9	63.1	1.3	89.7	4.8	1.6	3.6	1.55
17	80.3	19.5	0.3	17.6	0.8	22.5	19.4	62.0	1.0	89.7	4.8	1.7	3.3	1.48
17+1	77.1	22.9		20.5										1.70
18	76.8	68.2		22.7										1.68
19a	81.6	18.4		20.9	0.8	21.1	18.8	63.5	3.7	89.0	4.7	1.7	3.5	1.52
19b	79.8	20.2		14.5										1.64
21a	80.6	19.4		14.9	0.8	25.5	19.3	59.6	4.3	88.7	4.7	1.5	3.5	1.64
21b	81.5	18.5		16.7										1.66
23	82.2	17.8		19.3	0.8	26.2	20.3	58.4	5.2	88.9	4.7	1.4	4.4	1.69
24	79.8	19.2		16.4	0.9	29.1	21.2	55.6	6.5	88.7	4.7	1.1	3.4	1.58

V. vitrinite content (%); *I*. inertinite content (%); *E*. liptinite content (%); *MM*. mineral matter content; M_{ad}. moisture content (air dry base); A_d. ash yield (dry base); V_{daf}. volatile yield (dry ash-free base); *FC*. fixed carbon; $S_{t, d}$. total sulfur (air dry base); $R_{o, max}$. maximum vitrinite reflectance.

Coal Rank

Coal rank changes regularly in Enhong syncline with a coal rank increase from northwest to southeast. In vertical profile, the deeper the coal seam buried, the higher coal rank is.

Content and Concentration of Heavy Hydrocarbon in CBG

Characteristics of content and concentration of heavy hydrocarbon in CBG, in Enhong syncline, are summarized according to desorption data of 118 coal cores, about 224 samples (Table 2). The mean values of the concentration of heavy hydrocarbon in these samples vary from 1.94% to 14.95%. The minimum mean value is from No. 4+1 coal seam, whereas the maximum is from No. 15 coal seam. The maximum values of heavy hydrocarbon's concentration of these samples vary from 2.90% to 36.98%, the minimum and maximum of which are from Nos. 3 and 13 coal seams, respectively. Coal seams whose concentration is larger than 30% are Nos. 6, 7, 9, 13, and 23 coal seams. The mean value of heavy hydrocarbon's content of these samples varies from 0.10 to 1.03 m^3/t and shows the minimum value in No. 4+1 coal seam and the maximum value in No. 15 coal seam. The maximum value of heavy hydrocarbon's content varies from 0.16 to

Table 2 Composition and gas content of coal seams

Coal seam	Sample quantity	Composition content (%)					Gas content (m^3/t, daf)		
		CH_4	Heavy hydrocarbon		N_2	CO_2	Heavy hydrocarbon		CH_4
			Mean	Max			Mean	Max	
3	2	91.37	2.22	2.90	4.52	1.90	0.14	0.18	6.24
4	4	63.33	5.31	10.06	30.07	1.30			
4+1	2	54.98	1.94	3.04	37.64	5.45	0.10	0.16	5.25
5	4	72.04	2.45	5.09	23.62	1.90	0.14	0.30	5.89
6	7	64.43	8.75	30.72	24.89	2.25	0.58	2.04	6.63
7	27	70.64	9.47	30.04	17.82	2.08	0.90	2.86	9.53
8	8	64.25	6.24	10.71	27.27	2.55	0.38	0.66	6.15
9	50	62.58	12.11	34.60	21.14	5.15	0.85	2.44	7.04
10	3	75.12	6.64	10.48	16.23	2.01			
11	3	54.09	7.70	17.72	37.46	1.12	0.53	1.22	6.90
12	18	63.75	10.51	20.64	24.10	1.75	0.74	1.45	7.03
13	9	65.41	12.96	36.98	19.39	2.51	0.83	2.36	6.39
14	6	68.62	7.49	11.68	20.67	3.22	0.25	0.39	3.34
15	16	64.78	14.95	27.49	15.47	3.12	1.03	1.90	6.91
16	23	67.20	11.54	29.65	18.62	2.30	0.85	2.18	7.34
17+1	1	86.36	8.30	8.30	4.11	1.23	0.91	0.91	10.98
18	4	62.26	5.87	12.02	26.03	3.29	0.22	0.45	3.73
19	5	78.69	11.81	29.43	8.72	1.94	0.91	2.26	7.66
20	1	71.32	12.37	12.37	15.03	1.28			
21	13	70.01	8.92	25.51	19.81	1.23	0.58	1.66	6.52
22	1	71.85	14.75	14.75	12.45	0.95			
23	17	67.19	11.95	31.01	17.97	3.07	0.93	2.42	7.81

CH_4. methane; N_2. nitrogen; CO_2. carbon dioxide; daf. dry, ash-free base.

2.86 m^3/t, the minimum and maximum of which are from Nos. 4+1 and 7 coal seams, respectively.

Minefields appearing abnormal heavy hydrocarbon in Enhong syncline include Laoshuzhuo minefield, Zhongduannanbu minefield, Zhengji coalmine, Bumu coalmine, Dahe coalmine, Daping exploration area, Wudeli minefield, and Shidongshan exploration area (Fig. 2). Concentrations of C_{2+} are between 2.92% and 34.6% in Laoshuzhuo minefield, with an average of 18.04% (Table 3); 1.06% and 30.72% in Shidongshan exploration area, with an average of 10.99%; 0.75% and 36.98% in Daping exploration area, with an average of 10.79%; 0.30% and 25.51% in Bumu coalmine, with an average of 10.37%; 0.12% and 24.99% in Dahe coalmine, with an average of 9.94%; 0.25% and 27.34% in Zhongduannanbu minefield, with an

Table 3 Composition and gas content in mine fields

Mine field	Gas content (m^3/t)	Heavy hydrocarbon concentration (%)		
		Min	Mean	Max
Laoshuzhuo	9.33	2.92	18.04	34.60
Shidongshan		1.06	10.99	30.72
Daping	5.34	0.75	10.79	36.98
Bumu	9.10	0.30	10.37	25.51
Dahe	6.64	0.12	9.94	24.99
Zhongduannanbu	6.34	0.25	8.42	27.34
Zhengji	7.35	0.26	4.90	12.05
Wudeli		0.83	4.95	11.46

average of 8.42%; 0.26% and 12.05% in Zhengji coalmine, with an average of 4.90%.

Coal seams with high abnormal heavy hydrocarbon are (Fig. 3) Nos. 5, 4+1, 6, 7, 7-1, 8, 8+1, 9, 11, 12, 13, 14, 15, 15a, 15b, 16, 17+1, 18, 19, 19a, 19b, 21, 23, and 23b coal seams, which indicate that abnormal high heavy hydrocarbon is not limited to few coal seams. Among them, No. 9 coal seam has the most samples that contain abnormal concentrations of heavy hydrocarbon, and the concentrations are also high. The No. 7 coal seam takes second place, and the maximum value happened in No. 13 coal seam.

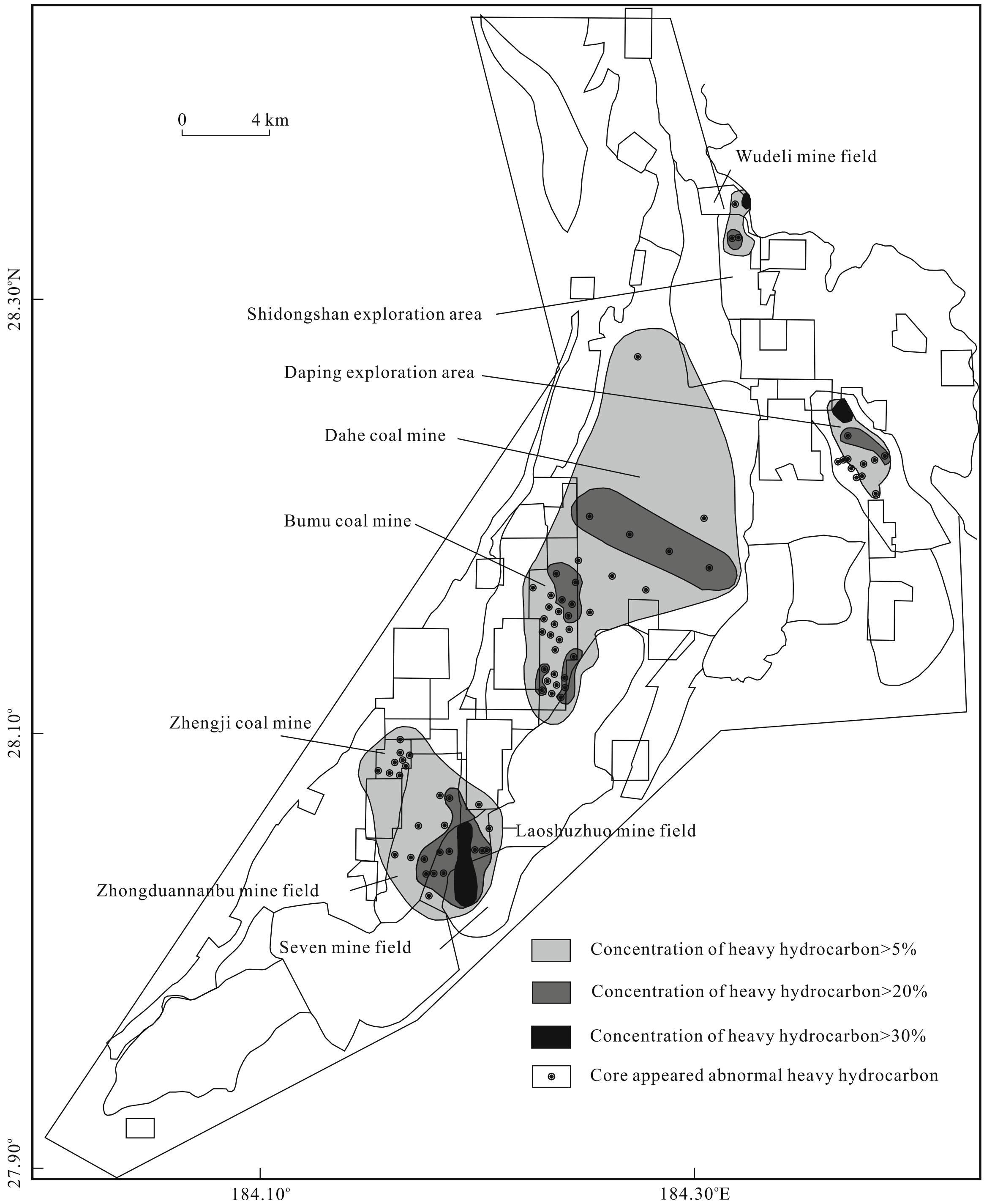

Figure 2. Distribution chart of abnormal concentration of heavy hydrocarbon in Enhong syncline.

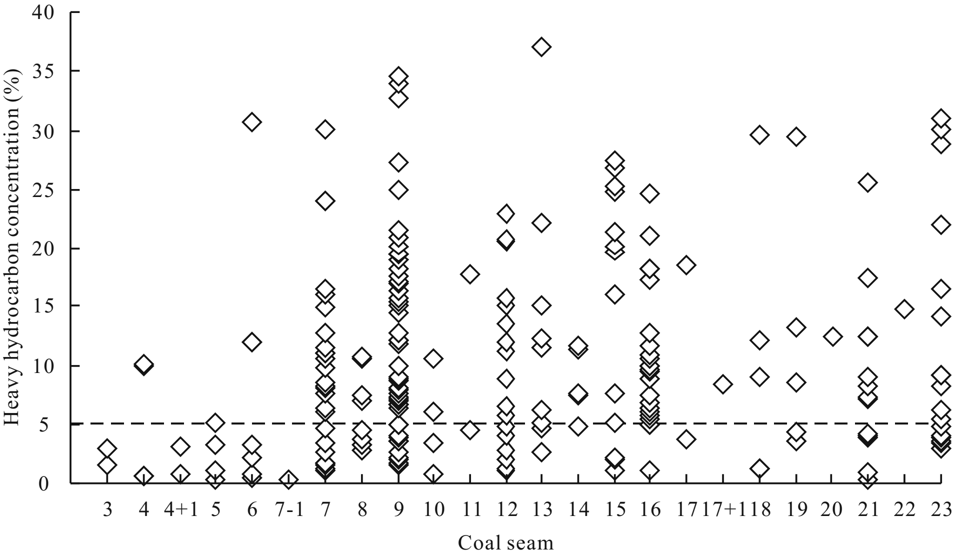

Figure 3. Relationship between concentration of heavy hydrocarbon and number of coal seam.

DISCUSSION

The reason for abnormal high concentration of heavy hydrocarbon can be discussed from the aspects of origin and evolution of heavy hydrocarbons. The abnormal concentrations may originate from the coal seam itself due to its gas-generating parent material or from exogenous reservoirs outside the coal seam, which could be organic gas (petroliferous gas) or inorganic gas. Evolution is reflected in the generation of heavy hydrocarbon changing orderly along with the rising of coal rank. The reason of abnormal high heavy hydrocarbon in Enhong syncline will be analyzed from the aspects of carbon isotope, coal petrography, and coal rank.

Carbon Isotope

Research shows that the carbon isotope composition of CBG is inherited from the characteristics of the parent material. It is also related to the degree of thermal evolution of the organic matter, biological actions, exchange equilibrium effects between CH_4 and CO_2, and fractionation effects in the process of desorption-diffusion-migration (Shen et al., 2007; Gao et al., 2002; Zhang and Tao, 2000; Qin et al., 1998; Dai et al., 1986; Qi, 1985). Carbon isotopes have become an indispensable part to study the origin of CBG. Characteristics of carbon isotopes of CBG in the Enhong syncline are (Table 4): $\delta^{13}C_1$ is between -50.1‰ and -47.0‰, with an average of -48.43‰; $\delta^{13}C_2$ is between -25.9‰ and -24.5‰, with an average of -25.1‰; $\delta^{13}C_3$ is between -22.3‰ and -16.9‰, with an average of -19.6‰; and $\delta^{13}CO_2$ is between -10.8‰ and -2.5‰, with an average of -6.53‰.

The carbon isotope distribution pattern of alkane gases can be divided into three types by Dai et al.: organic genetic alkane gas characterized by normal carbon isotopic distribution, inorganic alkane gas characterized by negative carbon isotopic distribution, and secondary modified gas characterized by reversal trend of alkane gas carbon isotope (Dai et al., 2008). Carbon isotope in Enhong syncline belongs to the normal carbon isotopic distribution ($\delta^{13}C_1<\delta^{13}C_2<{}^{13}C_3$), which indicates that heavy hydrocarbon in it is organic gas and not inorganic gas.

Organic gas can be divided into coal-type gas (gas generated by humic organic matter) and oil-type gas (gas generated by sapropelic organic matter) according to parent material. Many scholars think that $\delta^{13}C_2$ is a very important indicator to recognize the origin of organic gas. Wang thought that $\delta^{13}C_2$ higher than -29‰ is a mark of coal-type gas (Wang, 1994). Dai et al. indicated that natural gas has $\delta^{13}C_2$ higher than -27.5‰ and $\delta^{13}C_3$ higher than -25.5‰ is coal-type gas (Dai et al., 2002). Fu et al. summarized predecessors' discriminant indexes: when $R_{o,\ max}$ is between 0.5% and 2.5%, gas whose $\delta^{13}C_1$ is greater than -30‰ is classified as coal-type gas, whereas, if lighter than -30‰, it is classified as oil-type gas; gas whose $\delta^{13}C_2$ and $\delta^{13}C_3$ greater than -25.1‰ and -23.2‰, respectively, is coal type, and gas whose

$\delta^{13}C_2$ and $\delta^{13}C_3$ lighter than -28.8‰ and -25.5‰, respectively, is oil type (Fu et al., 2007). $\delta^{13}C_2$ of CBG in Enhong syncline is between -25.9‰ and -24.5‰, with an average of -25.1‰; $\delta^{13}C_3$ is between -22.3‰ and -16.9‰, with an average of -19.6‰. Judging from $\delta^{13}C_2$ and $\delta^{13}C_3$, gas in Enhong syncline belongs to coal type. However, $\delta^{13}C_1$ varying from -50.1‰ to -47.0‰, with an average of -48.43‰, is obviously lighter compared with $\delta^{13}C_1$ of thermogenic coal gas (>-30‰). From these results, it can be supposed that CBG in Enhong syncline is coal-type gas, and there is no oil-type gas coming from reservoir outside the coal seam. The reason why $\delta^{13}C_1$ becomes obviously lighter may be that the coal has been influenced by microorganism.

Coal-type gas can be divided into thermogenetic gas and biogenic gas. $\delta^{13}C_1$ of biogenic gas is usually about -55‰ to -90‰, and $\delta^{13}C_1$ of thermogenetic gas is usually greater than -50‰ (Fu et al., 2007). $\delta^{13}C_1$ in Enhong syncline is between -50.1‰ and -47.0‰, with an average of -48.43‰, greater than -55‰, which meets the standards of thermogenetic gas. In natural gas research, it is generally acknowledged that the origin of CO_2 could be divided into biogenic (organic) or abiogenic (inorganic) origin. $\delta^{13}C_{CO2}$ is also a very important indicator that can reflect the origin of CBG. Dai et al.'s (1993) research showed that $\delta^{13}C_{CO2}$ of organic CO_2 is usually between -39‰ and -8‰. Kotarba's study showed that $\delta^{13}C_{CO2}$ produced by pyrolysis of humic organic is usually between -25‰ and -5‰ (Kotarba, 2001). $\delta^{13}C_{CO2}$ of CBG in Enhong syncline is between -10.89‰ and -2.59‰, with an average of -6.54‰, which can be ascribed to the thermogenic gas. The reasons of some $\delta^{13}C_{CO2}$ being greater than -5‰ will be discussed later.

Table 4 Molecular and isotopic composition of coalbed gases

Sample	Coal	Molecular composition (vol.%)				Gas indices		C isotope $\delta^{13}C_{PDB}$ (‰)			
number	seam	CH_4	C_{2+}	N_2	CO_2	Wetness	CDMI	C_1	CO_2	C_2	C_3
9-1	9	91.50	6.24	0.51	1.75	6.38	1.88				
9-2	9	91.97	5.09	0.91	2.02	5.25	2.15				
9-3	9	89.85	7.18	0.63	2.34	7.40	2.54	-47.0	-2.5		-22.3
9-4	9	89.69	7.69	0.65	1.98	7.89	2.16				
9-5	9	90.90	7.37	0.60	1.13	7.50	1.23				
14+15-1	14+15	94.27	4.91	0.48	0.34	4.95	0.36				
16-1	16	94.92	4.13	0.31	0.64	4.17	0.67				
16-2	16	95.28	3.96	0.25	0.51	3.99	0.54	-47.5	-10.8	-24.9	
16-3	16	94.28	4.12	0.74	0.86	4.19	0.90				
16-4	16	95.36	3.19	0.62	0.82	3.24	0.86				
21-1	21	93.34	3.92	2.18	0.57	4.03	0.61				
21-2	21	95.34	2.81	0.86	0.99	2.86	1.02	-50.1	-7.7	-25.9	
21-3	21	95.91	2.47	0.82	0.80	2.51	0.83				
23-1	23	96.37	2.22	0.80	0.61	2.25	0.62				
23-2	23	96.23	2.32	0.61	0.84	2.36	0.86				
23-3	23	96.09	2.64	0.76	0.51	2.67	0.53				
23-4	23	96.12	2.36	0.52	0.99	2.40	1.02				
23-5	23	96.67	1.94	0.51	0.89	1.96	0.91	-49.1	-5.1	-24.5	-16.9
23-6	23	96.65	1.96	0.63	0.76	1.99	0.78				
23-7	23	97.07	1.92	0.37	0.63	1.94	0.65				
23-8	23	93.55	1.87	3.51	1.08	1.96	1.14				

Wetness=$100\times(1-(CH_4/(CH_4+C_{2+}))$; CDMI=$[CO_2/(CH_4+CO_2)]\times100\%$.

Concentrations of CO_2 and $\delta^{13}C_{CO2}$ in Enhong syncline are positively correlation (Fig. 4). Both of them are the highest and two to four times higher than other coal seams in No. 9 coal seam. Origin of CO_2 can be known by discussing the relationship between CDMI value (also called CO_2-CH_4 coefficient, which is $CO_2/(CO_2+CH_4)\times100\%$) and $\delta^{13}C_{CO2}$. Figure 5 shows that CO_2 of CBG in Enhong syncline belongs to category of thermogenic gas produced by humic material, except CO_2 in No. 9 coal seam, which is biogenic gas. The reason may be that No. 9 coal seam is easier affected by microorganisms due to its shallow burial depth. That may also be the reason why $\delta^{13}C_{CO2}$ in No. 9 coal seam is greater than 5‰. $\delta^{13}C_{CO2}$ became greater because of the reducing action of microorganisms,

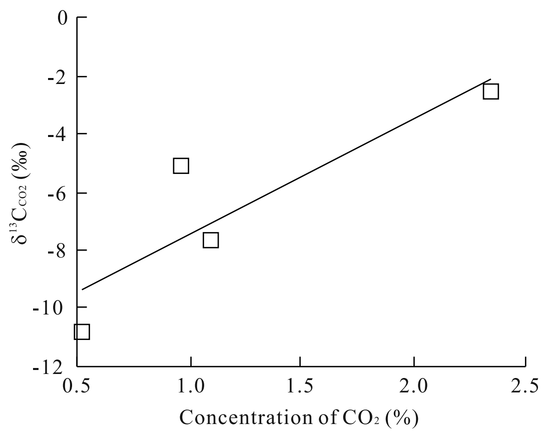

Figure 4. Relationship between $\delta^{13}C_{CO2}$ and CO_2.

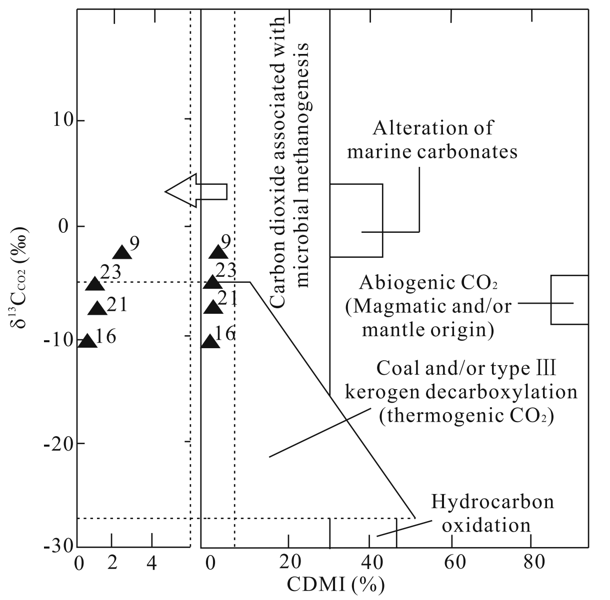

Figure 5. Relationship between CDMI and $\delta^{13}C_{CO2}$ (according to Kotarba, 2001).

which is in agreement with Fig. 4 showing that No. 9 coal seam contains associated CO_2 of microorganism methane.

According to the analysis above, it is thought that CBG in Enhong syncline is organic thermogenic gas. Shallow coal seam is affected by microorganism, making $\delta^{13}C_1$ in it lighter and $\delta^{13}C_{CO2}$ heavier, with characteristic of secondary biogenic gas.

Coal Maceral Composition

The ability of coals to generate hydrocarbons strongly depends on their maceral composition, and as a general rule, liptinite-rich coals are oil-prone, whereas vitrinite-rich coals are gas-prone (Alsaab et al., 2007; Petersen and Nytoft, 2006). Predecessors had different opinions on relationship on maceral and concentration of heavy hydrocarbon. Some authors consider that a high content of heavy hydrocarbons is related to higher liptinite content (Alsaab et al., 2008). Some authors (Killops et al., 1998; Bertrand, 1984) did not observe any clear relationship between the liptinite content and the capability for oil generation and found that coal poor in liptinite may possess the capacity to generate oil. Some other authors (Suggate, 2002; Wilkins and George, 2002; Killops et al., 2001; Rice, 1993; Bertrand, 1984) consider that some vitrinite group macerals, such as desmocollinite, are more and more recognized to have the potential for oil generation.

Concentrations of heavy hydrocarbon have a close connection with contents of vitrinite and inertinite in Enhong syncline. The concentrations increase with the increase of vitrinite and decrease with the increase of inertinite (Fig. 6a). The relationship between concentration of heavy hydrocarbon and the content of liptinite is not obvious, with a weakly positive correlation (Fig. 6b).

Inertinite is carbon-rich and oxygen-rich, whereas vitrinite is carbon-rich and hydrogen-rich. Correlations between hydrogen in vitrinite and the concentration of heavy hydrocarbons will be investigated by separately discussing the relationships between heavy hydrocarbons and vitrinite/inertinite ratio (*V*/*I*) and the hydrogen/carbon ratio (*H*/*C*). Figure 7 shows that the concentration of heavy hydrocarbons increases with the increase of *V*/*I* and *H*/*C*. From the above analysis, the heavy hydrocarbon in Enhong syncline has a positive correlation to hydrogen-rich degree of vitrinite, demonstrating that to a large extent heavy hydrocarbon comes from hydrogen-rich vitrinite.

Hydrogen-rich vitrinite as an important parent material of coal-formed oil has attracted many people's attention (Petersen and Rosenberg, 1998; Cheng and Zhang, 1994; Bertrand, 1984). The main contributors for oil generation by coal in the typical coal-formed oil basin Tuha basin are desmocollinites (which belong to hydrogen-rich vitrinite) and suberinite (Cheng and Zhang, 1994). Although the hydrocarbon generation potential of vitrinite is lower than that of liptinite, quantity can compensate quality. For most humic coals, the hydrocarbon potential depends not only on the content of liptinite plus sapropelinite but also on the type and content of vitrinite, which may be the more important factor, especially the content of hydrogen-rich vitrinite (Li et al., 1997). The coal maceral composition in Enhong syncline is dominated by vitrinite, with little liptinite, and heavy hydrocarbon has a strong positive correlation with the extent of hydrogen-rich vitrinite, so the parent material is a very important factor to explain the origin of abnormal concentration of heavy hydrocarbons in Enhong syncline, especially the effect of vitrinite and its submacerals on the generation of heavy hydrocarbon.

Degree of Coalification

Thermal simulation experiment of lignite indicates that the peak stage of heavy hydrocarbon generation is in the middle coalification bituminous stage, especially in the fat coal to coking coal stage in which concentration of heavy hydrocarbons could reach 10% (Fu et al., 2007; Petersen, 2006). $R_{o,\max}$ of coal seams in Enhong syncline is between 1.14% and 1.88%, equal to the coking coal to lean coal stage. Figure 8 shows that the concentration of heavy hydrocarbons increases with increasing $R_{o,\max}$. $R_{o,\max}$ of coal seams is between 1.34% and 1.88% in Laoshuzhuo minefield in which concentration of heavy hydrocarbon is the most abnormal. This relationship indicates that coalification may be one of the factors resulting in abnormal concentration of heavy hydrocarbon.

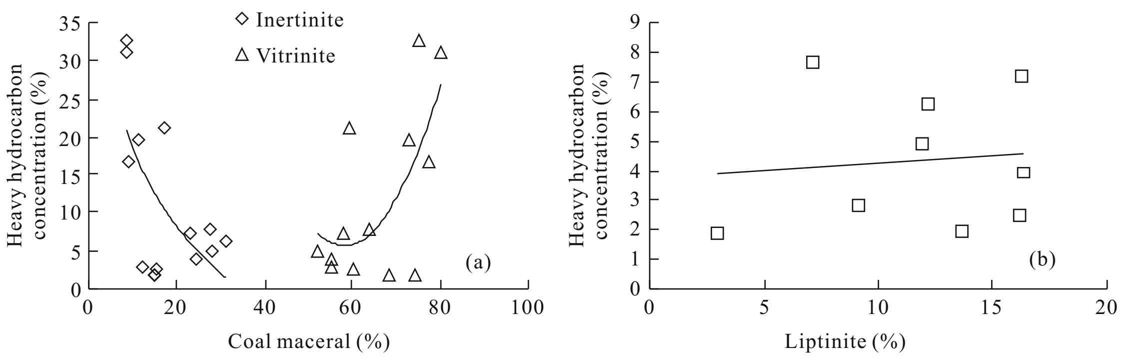

Figure 6. Relationship between concentration of heavy hydrocarbon and maceral content.

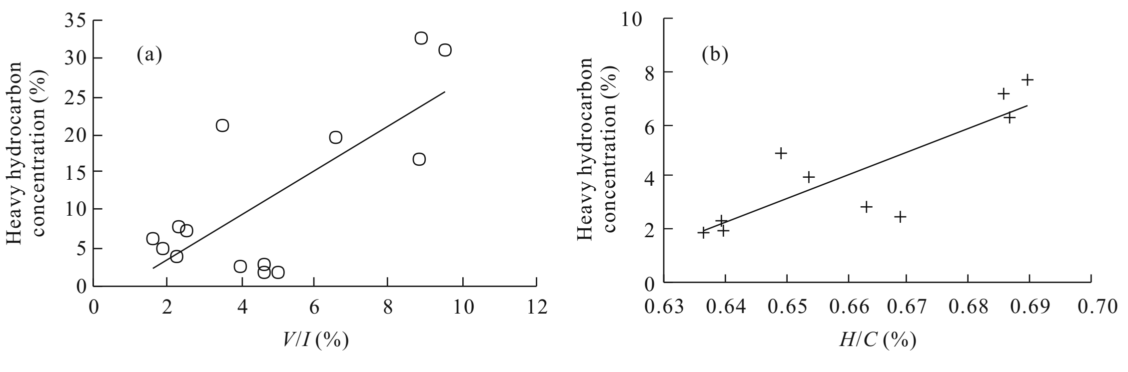

Figure 7. Relationship between concentration of heavy hydrocarbon and *V*/*I* and *H*/*C*.

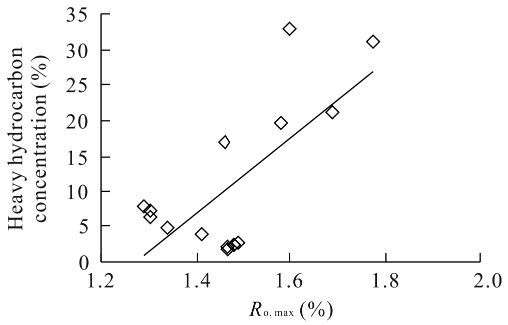

Figure 8. Relationship between concentration of heavy hydrocarbon and $R_{o, max}$.

CONCLUSIONS

This article describes the characteristic of abnormal high concentration of heavy hydrocarbons in Enhong syncline and analyzed its reasons from the aspects of origin and evolution of heavy hydrocarbon by carbon isotope, coal petrography, and coal rank.

1. Carbon isotopes of methane, ethane, and propane of coal gas in Enhong syncline have a normal carbon isotopic distribution, which displays the characteristics of organic gas. According to the characteristic of concentration of CO_2 and carbon isotope and the relationships among them, coal gas in Enhong syncline is classified as thermogenetic gas produced by humic material, with characteristic of secondary biogenic gas in shallow coal seam.

2. The concentration of heavy hydrocarbons in Enhong syncline increases with the increase of vitrinite and decreases with the increase of inertinite and also increased with the increase of V/I and H/C, so hydrogen-rich vitrinite may be a very important factor resulting in the abnormal concentration of heavy hydrocarbon.

3. The degree of coalification of coal in Enhong syncline is in the coking coal to lean coal stage in which abundant heavy hydrocarbons are generated. Concentration of heavy hydrocarbon increases with the increase of $R_{o, max}$. Therefore, coalification may be one of the factors that resulted in abnormal concentration of heavy hydrocarbon.

In conclusion, high concentration of heavy hydrocarbons originates from the coupling effect of higher contents of hydrogen-rich vitrinite in the coal and the coal rank of coking to lean coals during which the peak of heavy hydrocarbon generation is reached.

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