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Origin and gas potential of pyrobitumen in the Upper Proterozoic strata from the Middle Paleo-Uplift of the Sichuan Basin, China

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Abstract

Recently a sizable quantity of natural gas has been discovered in Upper Proterozoic and Lower Paleozoic strata from the Middle Paleo-Uplift area of the Sichuan Basin. It has been assumed that the natural gas originated from Cambrian–Silurian black mudstones which are widely distributed in the Southern Depression. In the present paper, we report that pyrobitumen occurring widely in the Upper Proterozoic carbonate strata from the uplift could be another potential gas source. The pyrobitumen has a very high maturity with measured random reflectance values ranging from 3.2% to 4.9%. Easy Ro modeling has shown that the Cambrian dark shale from the Southern Depression matured to the peak stage of oil generation during the late Silurian. Combining this with burial history of the uplift, it is inferred that the precursor of the reservoir pyrobitumen was a type of heavy bitumen derived from early biodegradation during the Devonian–Carboniferous of crude oils that migrated from the Southern Depression during the late Silurian. The pyrobitumen-containing carbonate strata have very little or no kerogen, with an average TOC of 0.67%, and the strata cover an area of 4×10^4 km² with an average thickness of about 100 m. It has been estimated that as much as 4.396×10^{12} m³ of gas have been generated from the pyrobitumen during its geological history. The main stage of gas generation from the pyrobitumen was during the period from the Jurassic to the early Cretaceous, after the formation of an effective cap rock. We believe that the pyrobitumen has made a very important contribution to the gas pools associated with the Middle Paleo-Uplift. © 2006 Elsevier B.V. All rights reserved.

Keywords: Sichuan Basin; Middle Paleo-Uplift; Pyrobitumen; Natural gas potential; Upper Proterozoic strata

1. Introduction

The Sichuan Basin is located in the middle of the China plate (Fig. 1). It is well known for its very rich natural gas resources (CPGC, 1989). Before 1990, commercial gas reservoirs were mainly found in Upper Paleozoic and Triassic strata, chiefly from the Eastern Folded Area and the Southern Depression (CPGC,

* Corresponding author. Tel.: +86 20 85290176. *E-mail address:* xmxiao@gig.ac.cn (X.M. Xiao). 1989). However, more recently, many natural gas reservoirs have been found in the Upper Proterozoic-Lower Paleozoic strata from the Middle Paleo-Uplift of the Sichuan Basin (Qiu et al., 1994; Dai et al., 1999; Hus, 1999; Luo, 2000; Wang et al., 2001; Yao et al., 2003). It has been widely accepted that a set of black mudstones of Cambrian–Silurian age in the Southern Depression and East Folded Area are the major source rocks for the gas reservoirs in the Upper Proterozoic– Lower Paleozoic strata of the uplift (CPGC, 1989; Dai et al., 2001). However, recent geological and geochemical

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Fig. 1. Sketch map showing a structural outline of the Sichuan Basin and location of the Middle Paleo-Uplift.

studies suggest there should be other sources for the gas pools in this area (Wang, 2000; Liu, 2000). Dai et al. (1999) first reported that the Upper Proterozoic carbonate reservoirs of the Weiyuan and Ziyang gasfields contain significant amounts of pyrobitumen. These new observations have raised the possibility that the pyrobitumen is another gas source for these Upper Proterozoic–Lower Paleozoic gas reservoirs. During 2000, a new borehole, named GK1, was drilled in the uplift (Fig. 1). It penetrated sediments from Upper Proterozoic to Jurassic, with a total thickness of about 5.8 km yielding a complete core sample profile, providing a rare opportunity to study the reservoir pyrobitumen and address this problem. The purpose of this paper is to investigate the geochemical and geological characteristics of this reservoir pyrobitumen, and discuss its



Fig. 2. Interpreted seismic section through the Gaositi structure showing the outline of the Middle Paleo-Uplift. The location is shown in Fig. 1.



Fig. 3. Sketch map showing a generalised stratigraphic column for the Middle Paleo-Uplift of the Sichuan Basin.

gas potential and possible role in the formation of Upper Proterozoic–Lower Paleozoic gas reservoirs in the uplift.

2. Geological setting

The well GK1 is located at the top of the Gaositi structure of the Middle Paleo-Uplift area (Fig. 1). The Gaositi structure is a paleo-anticline with Upper Proterozoic carbonates as the reservoir strata (Fig. 2). To the south of the Gaositi structure lies the Southern Depression, and to the east, the Eastern Folded Area.

The uplift contains predominantly Upper Proterozoic, early Palaeozoic and Mesozoic strata, with total sediment

thicknesses in the range of 5000–7000 m (Fig. 3). Ordovician–Carboniferous and Cretaceous–Tertiary strata are absent in most parts of the uplift, but Ordovician–Silurian strata are well developed in the Southern Depression (Fig. 2). The sedimentary facies changed from late Proterozoic–early Paleozoic marine carbonate platform to late Paleozoic alternating marine and continental sediments, early Triassic marine carbonate platform, late Triassic alternating marine and continental sediments, and Jurassic continental lacustrine and fluvial sediments (Fig. 3). It has been confirmed that there is a set of dark shales and dark argillaceous carbonates in Cambrian and Silurian strata both in the uplift and depression areas which act as gas source rocks (CPGC, 1989; Song, 1995).

3. Samples and method

The samples studied were mainly selected from wells GK1 in the Gaositi structure and PL1 in the slope area of the uplift, and from a few wells in the Ziyang and Weiyuan gasfields, including dark shales and carbonates with ages from late Proterozoic to Triassic (Table 1).

Pyrobitumen and other maceral reflectance measurements were performed on polished whole rock blocks using a Leica MPV 3 microscope/photometer system interfaced to a personal computer. Reflectances were measured at random grain orientations in oil medium using standard procedures (Xiao et al., 2001). Maceral/pyrobitumen reflectance data for individual samples were discarded if less than 15 separate readings were obtained. Pyrobitumen reflectances (B R_r) were converted to equivalent vitrinite reflectance (V R_r) values using the expression developed by Liu and Shi (1994) for early Palaeozoic carbonates of the Sichuan Basin, i.e.

$$VR_{\rm r}(\%) = [0.668 \times BR_{\rm r}(\%)] + 0.346 \tag{1}$$

Reflectances of "vitrinite-like" macerals (VLM R_r), were transformed into V R_r values using the calibration published by Xiao et al. (2000) for a suite of naturally matured lower Palaeozoic core samples from the Tarim Basin, i.e.

$$VR_{\rm r}(\%) = [1.26.\text{VLM}R_{\rm r}(\%)] + 0.21; \text{VLM}R_{\rm r} < 0.75\%$$
(2)

$$VR_{\rm r}(\%) = [0.28. \text{VLM}R_{\rm r}(\%)] + 1.03; \text{VLM}R_{\rm r}$$

= 0.75-1.50% (3)

$$VR_{\rm r}(\%) = [0.81.{\rm VLM}R_{\rm r}(\%)] + 0.18; {\rm VLM}R_{\rm r} > 1.50\%$$
(4)

4. Results and discussion

4.1. Occurrence and features of pyrobitumen

Two major definitions of bitumen can be found in the published literature. Organic geochemists define bitumen as the portion of organic matter that is soluble in organic solvents (Tissot and Welte, 1984). Organic petrologists define bitumen as solid organic matter filling voids and fractures in rocks and classify it based on reflectance, fluorescence intensity, and micro-solubility (Jacob, 1989). Pyrobitumen, as defined by Stasiuk (1997), is a high temperature form of solid bitumen (Jacob, 1989). Pyrobitumen is present in Upper Proterozoic carbonate samples from the studied wells. It can be classified into two types (Fig. 4) according to their optical properties and occurrence.

Table 1 Geological and geochemical characteristics of some Sichuan Basin samples

Borehole	Depth	Age ^a	Lithology	TOC	Rock-Eval		
	m			(%)	S ₁ (mg/g)	S ₂ (mg/g)	
GK1	4855	\in_1	Black silt shale	1.12	0.12	0.21	
GK1	4960	\in_1	Black silt shale	1.27	0.05	0.02	
GK1	4963	\in_1	Black shale	1.80	0.07	0.12	
GK1	4964	\in_1	Black silt shale	1.40	0.09	0.20	
GK1	4971	\in_1	Black shale	3.81	0.05	0.01	
GK1	4974	\in_1	Black shale	3.11	0.012	0.02	
GK1	4984	\in_1	Black silt shale	1.62	0.04	0.02	
GK1	4985	\in_1	Black silt shale	1.39	0.07	0.14	
GK1	4987	Z_2	Bitumen-bearing dolomite	1.43	0.09	0.07	
GK1	4989	Z_2	Bitumen-bearing dolomite	1.93	0.04	0.05	
GK1	5025	Z_2	Bitumen-bearing dolomite	0.48	0.05	0.05	
GK1	5150	Z_2	Bitumen-bearing dolomite	0.62	0.04	0.05	
GK1	5346	Z_2	Argillaceous dolomite	0.81	0.09	0.12	
GK1	5352	Z_2	Black silt shale	2.14	0.15	0.23	
ZY1	3461	€ı	Argillaceous limestone	0.14	0.01	0.01	
ZY1	3885	Z_2	Bitumen-bearing dolomite	0.23	0.01	0.01	
ZY1	4029	Z_2	Bitumen-bearing dolomite	1.90	0.08	0.04	
ZY4	4313	€ı	Bitumen-bearing dolomite	2.00	0.05	0.04	
WY106	2781	€1	Black shale	5.93	0.11	0.02	
WY117	2999	Z_2	Bitumen-bearing dolomite	0.53	0.05	0.02	
WY117	3025	Z_2	Bitumen-bearing dolomite	0.61	0.07	0.34	
WY117	3261	Z_2	Bitumen-bearing dolomite	0.50	0.05	0.03	
WY28	2975	Z_2	Black shale	3.91	0.08	0.02	
WY4	3366	Z_2	Bitumen-bearing dolomite	2.56	0.07	0.03	
PL1	5544	Z_2	Bitumen-bearing dolomite	3.14	0.02	0.02	

^a €₁—Lower Cambrian; Z₂—Upper Proterozoic.

Type I pyrobitumen: Occurs in voids, solution holes, solution caves and other cavities of carbonates, characterized by variable shape such as conglomerated, vein-like or spherulitic, with a coarse-grained mosaic texture, strong anisotropy and very high reflectance. The measured random reflectance values are as high as 3.2-4.9%, with equivalent VR_r values of 2.5-3.6% (Table 2). The pyrobitumen infillings are variable, ranging from slight infilling, semi-infilling to complete infilling. Vein-like pyrobitumen can be as long as 10-15 cm in solution caves in some dolomites. The host carbonates that are rich in this



Fig. 4. Photomicrographs of two types of pyrobitumen. Incident white light, oil immersion, ×450. A—Type I pyrobitumen, gray carbonate, 5006 m, Upper Proterozoic, well GK1; B—Type II pyrobitumen, dark carbonate, 5025 m, Upper Proterozoic, well GK1.

type of pyrobitumen are whitish-gray, indicating that these carbonates contain very little dispersed organic matter (kerogen) (Fig. 4A).

Type II pyrobitumen: Occurs in intergranular pores between carbonate mineral grains in the micrometer to submicrometer size range. It is usually inlaid around the cryptomere dolomite particles. Under high magnification (such as with a $100 \times$ objective), it displays a fine-grained mosaic texture with obvious anisotropy. The measured random reflectance values are in the range from 3.2% to 4.5%, with a similar maturity range to the Type I pyrobitumen. Because the interstitial infilling is very concentrated, this type of pyrobitumen is usually represented by black bitumenrich zones when observed macroscopically. The host carbonate is usually compact with dark and white 'granophyric' texture (Fig. 4B).

In the Upper Proterozoic carbonate strata, Type I pyrobitumen is dominant, although it is associated with Type II pyrobitumen in some samples.

4.2. Precursors and origin of pyrobitumen

As is well known, pyrobitumen can be derived from any one of the following precursors: crude oil, asphaltenes, heavy bitumen or tar mats (Stasiuk, 1997), and expelled from source rocks in a mobile form. There are two possible geological processes for the formation of reservoir solid bitumen. One is direct thermal cracking of petroleum hydrocarbons, usually occurring at a great burial depth with a geological temperature of 170–200 °C (Waples, 2002). Another way is by oxidation of crude oils with a combination of water washing and biodegradation processes which often occur near the earth's surface (Curiale, 1986). Evidence suggests that the reservoir pyrobitumen of the Middle Paleo-Uplift did not originate directly from the thermal cracking of crude oils, but from a type of heavy bitumen derived from the oxidation of crude oils during the Devonian–Carboniferous. The evidence can be summarized as follows.

(1) Based on the vitrinite reflectance calculated using the kinetic Easy Ro approach (Sweeney and Burnham, 1990) for the wells GK1 and YS1 (see the geothermal model in Section 4.3), representative for the uplift and the depression center, respectively (Fig. 1), the Cambrian dark shale was at a low mature or immature stage in the uplift, but it matured to the peak stage of oil generation in the Southern

Table 2 Measured reflectance values of some typical Sichuan Basin samples

Sample I	Depth	Age ^a	Lithology	VR _r	VLM		Type I pyrobitumen	
	(m)			(%)	VLMR _r ^b (%)	$EqVR_r^{c}$ (%)	BR_r^d (%)	$EqVR_r^e$ (%)
GK1-35	2231	T ₃	Coal	1.2				
GK1-36	2236	T_3	Coal	1.3				
GK1-37	3148	T_1	Coal	1.7				
GK1-38	4000	P_2	Coal	2.6				
GK1-39	4042	P_2	Coal	2.6				
GK1-40	4134	P_2	Coal	2.6				
GK1-7	4815	\in_1	Gray black argillaceous carbonate		3.7	3.2		
GK1-8	4816	\in_1	Gray black argillaceous carbonate		3.6	3.1		
GK1-10	4821	\in_1	Gray black silt mudstone		3.5	3.0	3.7	2.8
GK1-11	4855	\in_1	Gray black silt mudstone		3.8	3.3	3.9	3.0
GK1-13	4963	\in_1	Gray black silt mudstone		3.8	3.2	3.7	2.8
GK1-16	4975	\in_1	Gray black silt mudstone		3.9	3.3	3.6	2.7
GK1-19	4987	Z_2	Gray carbonate, with bitumen				3.9	2.9
GK1-20	4989	Z_2	Gray carbonate, with bitumen				4.0	3.0
GK1-56	5006	Z_2	Dark carbonate, with bitumen				3.9	3.0
GK1-25	5150	Z_2	Gray carbonate, with bitumen				4.3	3.2
GK1-27	5344	Z_2	Gray carbonate, with bitumen				3.9	2.9
GK1-28	5346	Z_2	Gray carbonate, with bitumen				3.8	2.9
GK1-57	5352	Z_2	Gray black silt mudstone				4.2	3.2
GK1-30	5352	Z_2	Gray black silt mudstone				4.1	3.1
GK1-32	5356	Z_2	Gray black silt mudstone				4.4	3.3
GK1-55	5359	Z_{2d}	Gray carbonate, with bitumen				4.0	3.0
ZY-1	3461	\in_1	Argillaceous limestone		3.10	2.69		
ZY-1	3885	Z_2	Bitumen-bearing dolomite				3.2	2.5
ZY-1	4029	Z_2	Bitumen-bearing dolomite				3.5	2.7
ZY-4	4313	\in_1	Bitumen-bearing dolomite				3.7	2.8
WY-106	2781	€1	Black shale		3.18	2.76		
WY-117	2999	Z_2	Bitumen-bearing dolomite				3.6	2.7
WY-117	3025	Z_2	Bitumen-bearing dolomite				3.5	2.7
WY-117	3261	Z_2	Bitumen-bearing dolomite				3.7	2.8
WY-28	2975	Z_2	Black shale		3.36	2.90		
WY-4	3366	$\overline{Z_2}$	Bitumen-bearing dolomite				3.8	2.9
PL1	5544	$\overline{Z_2}$	Bitumen-bearing dolomite				4.9	3.6

^a €₁—Lower Cambrian; Z₂—Upper Proterozoic.

^b Vitrinite like maceral reflectance.

^c VRo=0.81 VLMo+0.18 (Xiao et al., 2000).

^d Pyrobitumen reflectance.

e VRo=0.668BRo+0.346 (Liu and Shi, 1994).

Depression during the late Silurian (Figs. 5 and 6). The generated petroleum from this depression is inferred to have migrated upward into the Upper Proterozoic carbonate reservoirs of the paleo-uplift to form an oil pool (Fig. 3).

(2) During the Devonian–Carboniferous, the paleo-uplift was uplifted again, and the Ordovician–Silurian strata were eroded. In this period the Upper Proterozoic strata were uplifted close to the Earth's surface, within a depth range of only 500–900 m (Figs. 5 and 6). Under this depth of burial, the crude oils in the Upper Proterozoic reservoirs would have undergone the processes of weathering, biodegradation, water washing and oxidation, altering them into a type of heavy bitumen.

- (3) According to an investigation on extractable organic matter of some pyrobitumen from Upper Proterozoic carbonate reservoirs in the uplift made by Xu (1999) and Xu et al. (2000), the saturated hydrocarbon is characterized by an obvious "UCM" hump with rare alkanes, and strong biodegradation information is still evident.
- (4) Song (1996) proposed another model for the precursors of the pyrobitumen. The generation and entrapment of oil from the Cambrian shale in the uplift during Jurassic (Fig. 5) was followed by cracking of



Fig. 5. Burial history of sediments (upper) and thermal maturation history of the Cambrian strata after Easy Ro modeling (lower) according to the profile of the well GK1. The Cambrian strata were in the immature or lower mature stage before the Triassic. During the Carboniferous, the Upper Proterozoic carbonates were uplifted to within 600 m of the Earth's surface. The strata and eroded sediment thickness data are based on Hu et al. (2003). See details in text.

the oil to gas and pyrobitumen. However, from the geological background it is hard to explain a downward migration of oil from the Cambrian shale to the underlying Upper Proterozoic reservoir as impermeable sealing beds are well developed at the base of the Cambrian sequence in the uplift (Xiao and Liu, 2005).

Thus, the formation of the pyrobitumen included three stages:

Stage I: Formation of a paleo-oil pool formed in the uplift during the late Silurian.

- Stage II: Formation of heavy bitumen by biodegradation of crude oils during the Devonian– Carboniferous.
- Stage III: Formation of pyrobitumen by thermal maturation of the heavy bitumen during the Mesozoic.

Fig. 7 shows the relationship between sample depths and reflectance values of the organic matter from the well GK1. The equivalent vitrinite reflectance values of both vitrinite-like macerals and pyrobitumen in the Lower Paleozoic and Upper Proterozoic sediments are consistent with the changing trend of vitrinite reflectance values of Permian and Triassic sediments. This implies that the late



Fig. 6. Burial history of sediments (upper) and thermal maturation history of the Cambrian strata after Easy Ro modeling (lower) according to the profile of the well YS1. The Cambrian strata were at the mature stage during the Silurian. The strata and eroded sediment thickness data are based on Dai et al. (1999) and the paleogeothermal gradients and paleo-surface temperature are based on the well GK1. See details in text.

thermal maturation was so strong that the differences between indicated maturation levels of the pyrobitumens and kerogen from their host rocks are small.

4.3. Dating the main stage of gas generation of pyrobitumen

Dating the main stage of gas generation from pyrobitumen in the geological situation is a challenging scientific exercise as there is no available model that is widely accepted. Recently, the combination of organic reaction kinetics with thermal history has proved to be a useful tool to trace gas generation history (Tang et al., 2000; Cramer et al., 2001). For this purpose, a low mature solid bitumen from the uplift with the same origin as the target pyrobitumens should be selected for determination of the reaction kinetic parameters. However, as noted above, the pyrobitumen from the studied area is at very high maturation levels, and is therefore not suitable for the kinetic parameter calculation. For this reason a solid bitumen with $BR_r=0.03\%$ formed by biodegradation of marine sourced oils taken from the Wuerhe outcrop in the Jungeer Basin, northern west China (Xiao, 1992), was selected for the programmed-heating experiment of gas generation with two different heating rates. The hydrocarbon gas generation kinetics can be determined by the software Kinetics developed by Braun and Burnham (1998), based on the pyrolysis experimental data (Fig. 8). The resulting kinetic parameters were then applied to the geothermal history to model the gas generation for the well GK1.

The geothermal history for the modeling was set up according the following geological condition.

(1) Burial history

Fig. 5 presents the burial history of sediments from the well GK1. The strata and eroded sediment thickness data are based on Hu et al. (2003). According to the above discussion, the precursor of the pyrobitumen is a type of heavy bitumen which formed during the Carboniferous by biodegradation. Thus, the burial history of the pyrobitumen began after this period.

(2) Paleogeothermal gradients

The present geothermal gradient is 2.7 °C/100 m in the Middle Paleo-Uplift (CPGC, 1989). According to Liu (2000), the paleogeothermal gradients of the paleo-uplift varied from 2.9 to 3.1 °C/100 m during geological



Fig. 7. A comparison between the equivalent VR_r values of vitrinitelike macerals and pyrobitumens in the Upper Proterozoic—Cambrian sediments from the well GK1. The equivalent VR_r values of the pyrobitumens are very close to those of the vitrinite-like macerals.



Fig. 8. The discrete distribution of activation energy (upper) and fitting results of calculated data based on the kinetics parameters derived from experimental data for two different heating rates(lower) for the heavy bitumen from the Jungeer Basin. See details in text (Exp = experimented, Calc = calculated; Cum = cumulative).

history. To simplify the calculation, an average of 3.0 $^{\circ}\mathrm{C}/$ 100 m was used.

(3) Paleo-surface temperature

Although different paleo-surface temperatures have been used in the Sichuan Basin by different authors (Liu, 2000), an average of 20 °C is widely accepted.

Fig. 9 presents the modeling results for the reservoir heavy bitumen and pyrobitumen in the Upper Proterozoic strata from the well GK1. The results suggest that the heavy bitumen matured to the main stage of gas generation during the middle Jurassic to early Cretaceous, with C_{1-5} conversion ratio of 0.95–0.97 and gas production of 665–678 ml/g TOC at surface pressure and temperature conditions. This gas production is two times greater than that of coals at the same maturation level (Li et al., 2004).

4.4. Gas potential of the pyrobitumen-bearing strata

For the wells GK1, ZY1 and WY28, the pyrobitumen mainly occurs in the Upper Proterozoic. Table 3 presents



Fig. 9. Gas generation history of pyrobitumen from Upper Proterozoic strata from the well GK1 based on the kinetics modeling of the heavy bitumen from the Jungeer Basin in this study. The main stage of gas generation occurred during the Jurassic–early Cretaceous. See details in text.

the TOC and maceral analysis results for some of the Upper Proterozoic samples. According to this Table, it can be stated that

- The carbonates contain very little primary organic matter (kerogen). For the carbonate samples with TOC above 0.30%, the pyrobitumen would account for 95–100% of the total organic matter.
- (2) Not all the carbonate samples contain pyrobitumen. The pyrobitumen-containing carbonates have a higher TOC ranging from 0.25% to 3.1%, whereas those containing no pyrobitumen have a much lower TOC, usually less than 0.10%.
- (3) The dark shales contain mainly micrinite kerogen, and only a small amount of pyrobitumen. The micrinite was derived from amorphinite during thermal maturation (Xiao et al., 1991). This indicates that the original primary source organic matter of the shales was oil-prone.

Thus, in the Upper Proterozoic carbonates primary organic matter is practically absent, and the pyrobitumen contributes almost all of their organic matter content (TOC) in this area. The following equation was used to evaluate the gas potential of the pyrobitumen-containing carbonates:

$$Q = \text{TOC}_{\text{B}}.\text{Rr}.P \tag{5}$$

where Q-Gas (C₁₋₅) production per unit rock (ml/g rock),

 TOC_B —contribution of pyrobitumen to total TOC (%), Rr—conversion ratio of C_{1-5} production from pyrobitumen at some maturation level and

P—the maximum C_{1-5} production of pyrobitumen (ml/g TOC).

As systematic analytical data are only available for the well GK1 in this area, the following parameters are mainly based on data acquired from this well.

- TOC_B: an average TOC of 0.67% was obtained from eight Upper Proterozoic carbonate samples (Table 3). These carbonates are essentially free of primary organic matter based on their maceral analyses result. Thus, TOC=TOC_B=0.67%.
 - Rr: The equivalent VR_r of the reservoir pyrobitumen is in the range of 2.9–3.3%, close to the dead line of gas generation, with a C_{1-5} conversion ratio of 0.89–0.97. To simplify the calculation, an average of 0.93 was used.
 - *P*: According to the programmed-heating experimental data and kinetic modelling, the total gas production (C_{1-5}) of the heavy bitumen can reach 699 ml/g TOC.

Using the above equation and data, the gas production per unit (Q) of the Upper Proterozoic carbonate is 4.07 ml/g rock, i.e. 10.99 m³/m³ rock, where the density of carbonate was taken to be 2.7 g/cm³.

In the GK1 well area, the reservoir pyrobitumen only occurs in the upper section of the Upper Proterozoic strata with a thickness of 100 m. According to the geological background and available well data, these pyrobitumen-containing strata cover a wide area around Gaositi–Wei-yuan–Ziyang and well PL1. By using a distribution area of 4×10^4 km² and an average thickness of 100 m of the Upper Proterozoic carbonate reservoirs, the total amount of gas generated from the pyrobitumen in this set of strata could have reached $439,600 \times 10^8$ m³. Therefore, these reservoir pyrobitumen-containing carbonates are a great potential source of gas.

It should be mentioned that the main stage of gas generation of the pyrobitumen occurred during the Jurassic– early Cretaceous and the trapping structures formed during the Triassic (Li et al., 2001). Thus, the gases were generated under good conditions for their preservation. The geochemical data from the gas pools in the Middle Paleo-

Table 3 Whole rock TOC and kerogen maceral analyses for some Sichuan Basin samples

Well	Depth	Age	Lithology	Whole	Kerogen maceral analysis (%)			
	(m)				VLM	ILM	М	В
GK1	4855	€1	Black silt mudstone	1.12	2	0	89	9
	4963	€1	Black silt mudstone	1.80	3	0	90	7
	4971	\in_1	Black shale	3.81	1	0	98	1
	4987	Z_2	Gray carbonate, with bitumen	1.30	0	0	0	100
	4989	Z_2	Gray carbonate, with bitumen	1.63	0	0	0	100
	4993	Z_2	Gray carbonate, with bitumen	0.25	0	0	0	100
	5025	Z_2	Dark carbonate, with bitumen	0.48	0	0	0	100
	5028	Z_2	Gray carbonate, with bitumen	0.30	0	0	0	100
	5030	Z_2	Gray carbonate, with bitumen	0.47	0	0	0	100
	5150	Z_2	Gray carbonate, with bitumen	0.62	0	0	0	100
	5154	Z_2	Dark argillaceous carbonate, without bitumen	0.10	0	0	100	0
	5180	Z_2	Dark argillaceous carbonate, without bitumen	0.08	0	0	100	0
	5186	Z_2	Dark argillaceous carbonate, without bitumen	0.07	0	0	100	0
	5352	Z_2	Gray carbonate, with bitumen	0.38	0	0	0	100
	5357	Z_2	Dark argillaceous carbonate, without bitumen	0.08	0	0	100	0
	5361	Z_2	Dark argillaceous carbonate, without bitumen	0.18	5	0	95	0
ZY1	4017	Z_2	Dark carbonate, with bitumen	2.60	0	0	0	100
	4049	Z_2	Dark carbonate, with bitumen	1.90	0	0	0	100
WY-4	3366	Z_2	Bitumen bearing dolomite	2.56	0	0	0	100
PL1	5544	Z_2	Dark carbonate, with bitumen	3.14	0	0	0	100

VLM: Vitrinite-like maceral; ILM: Inertinite-like maceral; M: Micrinite; B: Pyrobitumen; €1-Lower Cambrian; Z2-Upper Proterozoic.

Uplift present strong evidence that the pyrobitumen made a contribution to gas pools. Table 4 presents the compositions and isotopic values of some natural gases from some gas pools in the Middle Paleo-Uplift. The gases are dominated by hydrocarbons, with a C₁ content of 82–95%, C₂ less than 0.4%, and normally without C₄ and heavier hydrocarbon components. The gases are very dry, with an dryness index (C₁/(C₁–C₅) over 0.996, and contain a normal content of N₂, in the range of 0.97–9.67%. The gases have a variable δ^{13} C₁ value, in the range from -32% to -38% (Hu et al., 2003; Xiao and Liu, 2005). These characteristics indicate that the gases have a very high maturity. As discussed above, the possible sources of the gases include the Cambrian kerogen and Upper Proterozoic pyrobitumen, both of which are at a high ma-

turation level, with an average equivalent VR_r of 3.0%, close to the gas generation death line (Heroux et al., 1979). Thus, the gases should have the same range of levels of maturation. Numerous data from Chinese gas fields have shown that a gas derived from a marine kerogen with VR_r of 3.0% will have a $\delta^{13}C_1$ value heavier than -32% (Xiao and Liu, 2005). Although the $\delta^{13}C_1$ values of the Weiyuan gas could be explained by a single source model of the Cambrian kerogen, the Ziyang and Gaositi gases should be attributed to a mixed origin from the kerogen and the oil (bitumen). From this analysis we conclude it is possible that some gas pools of commercial significance were formed mainly from the pyrobitumen. Therefore serious attention should be paid to the contribution of the gases generated from this source in the area.

Table 4

Compositions and	isotopic data of	gases fron	n gas pools in the	e Middle Paleo-Uplift of the	Sichuan Basin
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Gas pool	Well	Age ^a	Depth (m)	C1 (%)	C ₂ (%)	N ₂ (%)	$C_1/C_1 +$	$\delta^{13}C_1$ (‰)	Data source
Ziyang	Z-1	Z_2	3944-4044	93.59	0.12	1.22	0.999	-37.10	Hu et al. (2003)
, ,	Z-3	Z_2	3819-3920	92.2	0.35	0.97	0.996	-38.00	
	Z-6	Z_2	3912-4000	82.05	0.03	9.67	0.999	-35.50	
Gaositi	GK-1	Z_2	5422-5430	94.47	0.12	3.78	0.997	-35.78	Xiao and Liu (2005)
Weiyuan	W27	Z_2	2851-2950	87.07	0.09	6.02	0.999	-31.96	Hu et al. (2003)
·	W2	Z_2	2836-3005	85.07	0.11	8.33	0.999	-32.38	

^a Z₂₋ Upper Proterozoic.

5. Conclusion

According to results presented, the following conclusions can be drawn with regard to the pyrobitumen in the Upper Proterozoic carbonates in the Middle Paleo-Uplift of the Sichuan Basin, and its role in the generation of hydrocarbon gases.

- (1) There are two types of pyrobitumen. Type I occurs in cavities of carbonates and Type II in pores between carbonate mineral grains. The pyrobitumen reached a high maturity, with the measured random reflectance (BR_r) values ranging from 3.2% to 4.9%.
- (2) The precursor of the reservoir pyrobitumen is inferred to have been a type of heavy-bitumen which was derived from biodegradation of crude oils during the Devonian–Carboniferous.
- (3) The pyrobitumen-bearing carbonates are a very important source for gases in this area. A very large quantity of gas was generated from the pyrobitumen, mainly during Jurassic–early Cretaceous.

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