Genetic types and distribution of shallow-buried natural gases

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Abstract: Great volumes of shallow-buried (<2,000 m) natural gases which are mainly composed of biogases and low-mature gases have been found in the Mesozoic-Cenozoic sedimentary basins in China. Many shallow gas reservoirs in China are characterized by coexistence of biogas and low-mature gas, so identifying the genetic types of shallow gases is important for exploration and development in sedimentary basins. In this paper, we study the gas geochemistry characteristics and distribution in different basins, and classify the shallow gas into two genetic types, biogas and low-mature gas. The biogases are subdivided further into two subtypes by their sources, the source rock-derived biogas and hydrocarbon-derived biogas. Based on the burial history of the source rocks, the source rock-derived biogases are divided into primary and secondary biogas. The former is generated from the source rocks in the primary burial stage, and the latter is from uplifted source rocks or those in a secondary burial stage. In addition, the identifying parameters of each type of shallow gas are given. Based on the analysis above, the distributions of each type of shallow gas are studied. The primary biogases generated from source rocks are mostly distributed in Quaternary basins or modern deltas. Most of them migrate in water-soluble or diffused mode, and their migration distance is short. Reservoir and caprock assemblages play an important role in primary biogas accumulation. The secondary biogases are distributed in a basin with secondary burial history. The oil-degraded biogases are distributed near heavy oil pools. The low-mature gases are widely distributed in shallow-buried reservoirs in the Meso-Cenozoic basins.

Key words: Primary biogas, secondary biogas, oil-degraded biogas, low-mature gas, genetic type, gas identification, gas accumulation, distribution

1 Introduction

In oil and gas exploration, the natural gas which occurs at depths of less than 2,000 meters is often known as shallow gas. It is widely distributed in the Meso-Cenozoic basins in China, such as the Qaidam, Junggar, Songliao, and Bohai Bay basins, and is mainly composed of biogas and low-mature gas. The biogas is also distributed in the sediments of deltas, such as the Yangtze River Delta. By the end of 2006, the proven geological reserves of biogas are estimated to be $2,860 \times 10^8$ m³, and the total geological reserves are estimated to be $38,629 \times 10^8$ m³, which means that the proven reserves account for only 7.4% of the estimated biogas resource (Zhang et al, 2005).

Rice and Claypool (1981) proposed that biogas was generated from organic matter in shallow-buried deposits in a reducing environment. They stated that it is rich in methane and there are two ways for biogas generation, acetic acid fermentation and CO₂ reduction. Pallasser (2000) found that

biogas can be generated from trapped oil by biodegradation. Moreover, the organic matter in shallow-buried sediments can also generate gas rich in methane before thermal degradation (Xu et al, 1990; Xu, 1994; Wang et al, 1988; 2003b). This is called low-mature gas. The biogases are formed by anaerobic bacteria, while the low-mature gases are generated in low temperature and low maturity conditions (Song and Xu, 2005).

Many shallow gas reservoirs in China are characterized by coexistence of biogas and low-mature gas with different genetic types. Examples include the gas reservoirs in the Liuquan structure of the Langgu Sag and in the Jinhua Sag, northern Jiangsu. Because different genetic types of shallow-buried gases have different accumulation processes, identifying the genetic types is significant for understanding the gas accumulation.

Based on the geochemical characteristics of shallow-buried gases in different basins, we define the genetic types of gases according to their source materials and develop an identification standard. The accumulation and distribution of the shallow gases is discussed with examples, which helps exploration for shallow gases in different basins.

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2 Genetic types of shallow-buried gases

There is not a uniform classification for biogases and low-mature gases. In foreign countries, little attention is focused on the genetic types of shallow-buried gases and research has concentrated mainly on coalbed methane (CBM). Based on the study of coal bed biogenic methane in the Powder River Basin, Flores et al (2008) suggested that biogases could be divided into CO₂ reduction biogas and acetic acid fermentation biogas. In China, Dai (1992) believed that the shallow-buried gases consisted of biogases and sub-biogases. Xu (1994) suggested that the shallow-buried gases had two types, biogases and bio-thermal-catalytic transitional gases. Li et al (2005a) divided the biogases into primary biogases and secondary biogases.

Based on the study in the Qaidam, Bohai Bay, Turpan-

Hami, and Yinggehai-Qiongnan basins, the shallow-buried gases are divided into biogases and low-mature gases, and then the biogases are subdivided into two types according to their sources, one is from source rocks, and the other is from hydrocarbon reservoirs. The biogases derived from source rocks are further classified into primary biogases and secondary biogases. Table 1 gives the identifying parameters for each type of the gases.

2.1 Primary biogas

The methane content of primary biogases is as high as 98%, the ethane and higher alkane (C₂₊) content is very low (about 0.5%), the C₁/C₁₋₅ ratio is greater than 0.99, and the nonhydrocarbon contents are less than 5% (Table 2). The $\delta^{13}\text{C}_1$ of the primary biogas is less than -55‰ (Rice and Claypool, 1981; Dai, 1992).

Table 1 Genetic types and identifying parameters of biogases and low-mature gases

Genetic types	Biogas			Low-mature gas
	Derived from source rocks		Derived from hydrocarbon reservoirs	
	Primary biogas	Secondary biogas	Oil-degraded biogas	
Generation process	Generated from immature source rocks in primary burial history	Generated from source rocks in uplift or secondary burial history	Oil biodegradation	Thermal degradation from source rocks in the low maturity stage
Gas composition	Mainly methane, little C ₂₊	Mainly methane, a little C ₂₊ , CO ₂ and N ₂	Mainly methane, little C ₂₊ , more N ₂ than other biogas	C ₁ /C ₁₋₅ between 0.7 and 0.89, a little nonhydrocarbon
Carbon isotope composition	$\delta^{13}\text{C}_1 < -55\text{‰}$, $\delta^{13}\text{C}_2 < -40\text{‰}$	$\delta^{13}\text{C}_1 \leq -50\text{‰}$, $\delta^{13}\text{C}_2 > -40\text{‰}$	$-50\text{‰} < \delta^{13}\text{C}_1 < -75\text{‰}$	$-45\text{‰} < \delta^{13}\text{C}_1 < -55\text{‰}$
Examples	Qaidam, Yangtze River Delta, Jiangsu Basin	Langgu Sag	West slope of Songliao Basin, Jiyang Depression	Turpan-Hami Basin, Liaohe Depression

Primary biogases are generated by acetic acid fermentation or CO₂ reduction. The latter is the main source of biogas. $\delta\text{D}_{\text{CH}_4}$ from acetic acid fermentation is less than -300‰, and $\delta\text{D}_{\text{CH}_4}$ from CO₂ reduction ranges between -300‰ and -160‰. Whiticar et al (1986) analyzed 500 natural gas samples from different basins all over the world and suggested that the $\delta\text{D}_{\text{CH}_4}$ value of natural gases derived from source rocks deposited in fresh-water environment is lower than that of natural gases derived from source rocks deposited in saline waters. Therefore, $\delta\text{D}_{\text{CH}_4}$ is not an effective indicator for biogas identification.

The primary biogases are distributed mainly in the Quaternary in the Qaidam Basin, Neocene-Quaternary in the Yinggehai-Qiongnan Basin, and the Yangtze River Delta. A few primary biogas reservoirs are found in the Tertiary.

2.2 Secondary biogas

Shi (2002) conducted biochemical experiments on source rocks sampled from the Mesozoic in the Liaohe Depression and from the Tertiary in the Langgu Sag, and demonstrated that the source rocks after thermal maturation could also generate biogas.

Secondary biogas is usually mixed with low-mature gas in a reservoir. The methane content in the secondary biogas is as high as 98%, while the heavy hydrocarbon contents are

less than 2% (C₁/C₁₋₅ ratio > 0.98) and the nonhydrocarbon contents are rather low. Carbon isotope compositions of methane in the secondary biogases are slightly heavier than those in the primary biogases. Meanwhile, the carbon isotope compositions of ethane and propane in the secondary biogas are also heavier than those in the primary biogas (Table 3).

The sedimentary basins in east China experienced rifting in the Tertiary, and were uplifted in the Himalayan movement. Before the movement, the basins were in the primary burial stage and some source rocks were in the biogas generation stage. After the movement, some source rocks returned to the bacterial activity zone so that secondary biogas was generated. The secondary biogases are mainly distributed in the Bohai Bay Basin and small Tertiary basins, such as the Baoshan and Qujing basins in the Yunnan Province.

2.3 Oil-degraded biogas

Anaerobic microorganisms generate biogas from oils in shallow-buried reservoirs (Zhu et al, 2005). In oil biodegradation, the oil loses its light hydrocarbon components, so that its density, viscosity and sulfur content increase (Zhu et al, 2007).

Because of the isotope fractionation which occurs during the oil biodegradation, the carbon isotope value of methane in the oil-degraded biogas is very low (-100‰ - -55‰), and

Table 2 Geochemical characteristics of primary biogas in China

Structural belt	Well No.	Strata	Depth, m	Gas composition, %			$\delta^{13}\text{C}_1$, ‰	C_1/C_{1+}
				CH_4	C_2H_6	Nonhydrocarbon		
Yangxin Sub-sag	Y1	Es ₁	1504	98.1	0	1.91	--	1
	Y101	Es ₁	1504	98.4	0.17	1.04	-60.6	0.9985
	Y15	Es ₁	1412	97.0	0.08	1.79	-60.9	0.9992
	Y16	Es ₁	1309	97.8	0.18	2.06	-56.5	0.9982
	Y21	Es ₁	1520	95.0	0	5.00	--	1
	Average			97.3	0.14	2.36	-59.3	0.9986
Qaidam Basin	TN	Q ₁₋₂	--	94.3	0	5.73	-69.9	1
	SB1	Q ₁₋₂	--	98.7	0.13	1.15	-66.8	0.9986
	SB2	Q ₁₋₂	--	98.4	0	1.6	-65.4	1
	YH	Q ₁₋₂	--	95.2	0.28	4.52	-65.0	0.9971
	TFS	Q ₁₋₂	--	99.9	0	0.15	-68.8	1
	Average			97.3	0.08	2.63	-67.18	0.9992
Southeast Littoral	J1	Q	--	96.2	--	3.12	-74.2	--
	L1	Q	--	94.2	--	2.08	--	--
	D1	Q	--	92.3	--	2.03	--	--
	T1	Q	--	93.9	--	2.04	--	--
	F6	Q	--	91.4	--	1.47	-84.5	--
	Average			93.6	--	2.15	-79.4	--

Table 3 Geochemical characteristics of Es₃ biogas reservoir in the Liuquan structure

Well No.	Strata	Depth, m	Gas composition, %				$\delta^{13}\text{C}_1$, ‰	C_1/C_{1+}
			CH_4	C_2H_6	N_2	CO_2		
G26	Es ₃	1530	98.5	1.09	0.32	0.1	-55.7	0.99
G131	Es ₃	1168	98.6	0.05	1.36	0.05	-57.2	0.99
G13	Es ₃	1612	97.2	1.62	0.63	0.19	-55.9	0.98
G131	Es ₃	1597	96.8	2.11	0.81	0.12	-54.3	--
Q36	Es ₃	1001	98.8	0.63	0.167	0.32	-51.1	0.99
XQ2	Es ₃	1135	99.0	0	1.17	--	-55.6	0.99
Average			98.2	0.91	0.74	0.15	-55.0	0.988

$\delta^{13}\text{C}$ of CO_2 is abnormally high (up to +2‰) (Table 4). Oil-degraded biogases are often distributed with heavy oils, and usually accumulate with other shallow-buried gases.

The oil-degraded biogases have been found in the west slope of the Songliao Basin, Bohai Bay and Junggar basins. The potential reserves of oil-degraded biogases in the Songliao Basin are estimated to be about $240 \times 10^8 \text{ m}^3$.

2.4 Low-mature gas

The low-mature gas is also called early thermal genetic gas or bio-thermal-catalytic transitional gas (Xu et al, 1990). Actually, it is formed after the biogas generation and before

the oil generation peak (Xu, 1994; 1999).

Low-mature gases are characterized by lower $\text{C}_1/\text{C}_{1-5}$ ratio (0.7-0.9) and heavier carbon isotope ($\delta^{13}\text{C}_1$ between -55‰ and -42‰) compared with those of biogases.

The identification of low-mature gas is a disputed issue. Galimov (1988) believed that R_o of the source rocks generating low-mature gas ranged from 0.5% to 0.7% based on the study of Siberian gas. Wang et al (2003b) concluded that the low-mature gases are generated from the source rocks with R_o from 0.4% to 0.55%, even up to 0.7%, and the burial depth of the source rocks is 1,000-2,500 m, even up to 3,000 m. Xu et al (2008) suggested that source rocks generated low-

Table 4 Geochemical characteristics of oil-degraded biogas in the Songliao Basin

Well	Gas composition, %				C_1/C_{1+}	$\delta^{13}C_1$, ‰	$\delta^{13}C_2$, ‰	$\delta^{13}C_{CO_2}$, ‰
	CH ₄	C ₂₊	CO ₂	N ₂				
D2-2	94.4	0.08	0.059	5.405	0.9992	-56.32	-44.2	-2.69
D5-3	93.93	0.06	0.062	5.898	0.9994	-57.55	-46.2	0.82
D6	90.63	0.03	0.099	9.162	0.9997	-59.06	-46.7	-1.89
D602	91.14	0.05	0.072	8.665	0.9995	-58.93	-47.6	0.95
D603	91.66	0.12	0.115	8.031	0.9987	-58.45	-51.3	-3.87
D610	91.26	0.06	0.087	8.511	0.9993	-58.75	-47.4	-6.9
D621	89.86	0.13	0.091	9.328	0.9986	-59.45	-47.3	-18.78
D2-2	94.4	0.078	0.059	5.405	0.9992	-56.32	-44.2	-2.69
D5-3	93.93	0.058	0.062	5.898	0.9994	-57.55	-46.2	0.82
D610	91.26	0.059	0.087	8.511	0.9994	-58.75	-47.4	-6.9
D602	91.14	0.051	0.072	8.665	0.9996	-58.93	-47.6	0.95
D6	90.63	0.032	0.099	9.162	0.9986	-59.06	-46.7	-1.89
D603	91.66	0.115	0.115	8.031	0.9987	-58.45	-51.3	-3.87

mature gas when R_o was less than 0.8%.

3 Distribution of various genetic types of shallow-buried gases

The distributions of shallow-buried gases are controlled by the sources. Biogas generation needs conditions suitable for microorganism metabolism, such as temperature between 35°C and 75°C and appropriate pH, Eh and salinity (Chen et al, 1994). The distribution of shallow-buried gases is also controlled by structural aspects, sedimentary facies and seal qualities.

3.1 Distribution of primary biogases

The primary biogases are mainly distributed in the Quaternary basins or modern delta deposits, such as the eastern Qaidam Basin and the Yangtze River Delta. The primary biogases migrate in water-soluble or diffused mode. The migration distance is short, so most of the biogas reservoirs are distributed near the source kitchens.

The primary biogas reservoirs have high porosity and permeability due to their weak diagenesis. The dynamic balance of charging and dissipating of biogases in the reservoirs plays an important role in the biogas accumulation. In general, the biogas charging quantity is closely controlled by gas generating intensity. If the biogas source is sufficient, the cap-rock controls the enrichment of biogas in the charging area.

Taking the Es_1 primary biogas reservoir in the Yangxin Sub-sag for example, a series of traps are developed on the slope and structural highs, just updip on the source kitchen, and are charged fully with the biogas (Fig. 1). Its source rocks are grey mudstones and dark-grey oil shales with R_o between 0.27% and 0.35%, and TOC contents between 0.22% and

11.80%. The source rocks have been in the biogas generation stage since Ng. The reservoirs are mainly bioclastic limestones and dolomites deposited in lakeshore or sand sheet facies with high porosity and permeability with a caprock of lacustrine mudstones.

Fig. 1 Es_1 primary biogas accumulation in the Yangxin Sub-sag

The biogases in the Yangxin Sub-sag migrated in water-soluble or diffused mode. The migration distance is short because the biogases diffuse easily in rocks with weak diagenesis. Biogases mostly accumulated in the central uplift near the sub-sags. In the central uplift, many traps developed, which were suitable for gas capturing (Fig. 1). Biogases from the west sub-sag accumulated in the eastern drape structure and finally structural-lithologic gas pools were formed. The biogases in the Yangxin Sub-sag were preserved because of good cap-rocks and a lack of faulting.

3.2 Distribution of secondary biogases

Study of secondary biogas has been mainly about whether the source rocks are able to generate biogas after thermal evolution, but the distribution of secondary biogas is less

studied.

The source rocks of the secondary biogas experienced thermal evolution, and were then uplifted to the bacterial activity zone. These source rocks are widely distributed in basins. During the Mesozoic-Tertiary, some basins were in a rifting stage and in deep-water environments, so that source rocks were deposited. In the Himalayan movement, most of the basins were uplifted, and the source rocks went back into the biological activity zone. Thus, the secondary biogases could be generated.

Faults are well developed in the rift basins. The faults not only provide good migration pathways for biogases when they are active, but also act as a good seal for the biogases when they are not active.

Taking the Es_3 secondary biogas in the Liuquan structure as an example, fault activities are significant in gas accumulation (Fig. 2).

Fig. 2 Burial history of Es_3 -Ed source rocks of the Q35 well

The source rocks of Liuquan biogases are mainly dark lacustrine mudstones with thicknesses of 1,100 m. Their TOC contents are 1.24%-1.98%, and R_o value is generally 0.5. The kerogen type is mainly II_1 - II_2 . The source rocks are less than 2,000 m deep because of tectonic movements, and are favorable for biogas generation. The reservoirs consist of delta, fan delta and sublacustrine fan sandstones. The porosity is from 11.2% to 32.6% with an average of 26.8%, and the permeability is 0.1-876 md with an average of 193.6 md. The lacustrine mudstones cover the reservoirs directly as

caprocks.

The Liuquan structure has experienced complex tectonic activities, and many faults are developed. The distributions of biogases are controlled by the source conditions and fault activities. The evolution of source rocks can be divided into three stages, I: primary subsidence stage, II: uplift stage, III: secondary subsidence stage (Fig. 2).

During the first two stages, almost all faults are active, and the sediments are strongly uplifted by the structural movement so biogases were not easily preserved. Taking the Q2 well as an example, the distance between the top of the Es_3 formation and the bottom of the Ng formation is only 100 m (Fig. 3). After Ng, the secondary biogases are easy to be preserved because of weak fault activities.

Therefore, secondary biogas accumulation occurs in basins or depressions with uplifting or secondary burial history, and is controlled by fault activities.

3.3 Distribution of oil-degraded biogas

Oil-degraded biogases accumulated near heavy oil pools. In the biogas generation, water activity is important for microorganism propagation so that the biogas reservoirs are often located near the oil-water contact. The biogas reserves are determined by the quantity of biodegraded oils.

Oil-degraded biogases are distributed in shallow reservoirs in the Songliao, Bohai Bay (Fig. 4) and Junggar basins, because many heavy oil reservoirs are distributed in these basins.

The Alaxin lithologic-structural gas reservoir is located in the west slope of the Songliao Basin. It is less than 700 m deep. The reserves of biogases are 1.74×10^9 m³, and the daily gas production is 0.115×10^6 m³. It is a shallow-buried, low abundance, high production small gas field. The gases are characterized by high methane content and relatively light $\delta^{13}C_1$ with the content from -83.88‰ to -58.25‰, showing microorganism degradation characteristics.

The sources of Alaxin biogases are oil pools (reserves are 3.15 million ton) around the biogases. The oil pools are commonly degraded and most of the n-alkane in oil has been degraded by microorganisms (Fig. 5). The Alaxin biogas reservoirs are grey green mudstones and siltstones deposited

Fig. 3 Oil and gas accumulation in the Liuquan structure in the Langgu Sag

Fig. 4 Relationship between the distribution of heavy oil and shallow gas in the Bohai Bay Basin

Fig. 5 Chromatogram of oil associated with biogas in the Alaxin oilfield

in the Saertu formation of beach bar facies. These reservoirs are thin in single layer with limited lateral distribution with mudstone cap rock.

The Alaxin structure was uplifted before the hydrocarbon generation stage of the Gulong Sub-sag. It is prior to the

capture of oil and gas. According to the research of Yang et al¹, the hydrocarbon accumulation can be divided into three stages, 56 Ma, 45 Ma, and 35 Ma, all after the oil and gas generation period of the Songliao Basin.

3.4 Distribution of low-mature gas

Xu et al (2008) proposed a R_o 0.8% upper limit for the source rocks that generate low-mature gas. This gives a great scope for exploration for low-mature gases. According to their calculation, the low-mature gas resources in the Turpan-Hami Basin are more than $1 \times 10^{11} \text{ m}^3$. During the low-maturity stage, there are varieties of catalysis in the gas generation process, such as biological catalysis and clay mineral catalysis (Liu and Xu, 1995; 1996). So maturity can not be used as the only indicator for the assessment of low-mature gas source rocks. At low temperature, the activation energy for low-mature gas generation of Mesozoic source rocks in the Turpan-Hami Basin is lower than that in other basins (Table 5). It is a special reason why low-mature gases are very rich in the Turpan-Hami Basin.

Low mature and immature source rocks are widely distributed in petroliferous basins. Peng et al² concluded from pyrolysis experiments that the activation energy of gas generation in humic source rocks is lower than that of sapropelic source rocks, which showed that the humic source rocks are more likely to generate low-mature gas.

4 Conclusions

The shallow gas in the Qaidam, Bohai Bay, Turpan-Hami and Yinggehai-Qiongnan basins is mainly composed

¹ Yang Y F, et al. Oil-degraded biogas in the Songliao Basin. Biogas and low-mature gas generation and accumulation mechanism conference. 2006

² Peng P A, et al. Generated dynamics and genetic identification of low-mature gas. Biogas and low-mature gas generation and accumulation mechanism conference. 2008

Table 5 Activation energy of gas production from source rocks in different basins

Source rocks	Activation energy, KJ/mol		Data source
	Methane	Heavy hydrocarbon	
Jurassic coal in the Kuche Sag	235	269	Li et al, 2003
Jurassic mudstone in the Kuche Sag	206	223	Li et al, 2003
Tertiary oil shale in the Dongying Sag	272	259	Wang et al, 2003a
Jurassic coal in the southwest Tarim Basin	227	281	Li et al, 2004
Jurassic mudstone in the southwest Tarim Basin	223	281	Li et al, 2004
Yancheng Formation kerogen in the Qiongdongnan Basin	231	--	Li et al, 2005b
Shanxi Formation coal in the Ordos Basin	273	--	Mi et al, 2005
Coal in the Turpan-Hami Basin	209	169	Xu et al, 2008
Mudstone in the Turpan-Hami Basin	191	147	Xu et al, 2008

of biogas and low-mature gas. The biogas is divided into two types according to their sources, one is from source rocks, and the other is from hydrocarbon reservoirs. The biogases derived from source rocks are classified further into primary biogas and secondary biogas. The former is generated from immature source rocks in primary burial stage, and the latter is generated from the source rocks in uplift or secondary burial stage.

The identifying parameters of shallow-buried gas are given. Choosing suitable identifying parameters combined with the analysis of accumulation conditions can distinguish the genetic types of shallow-buried gases effectively. The heavy hydrocarbon content of biogas is lower than that of low-mature gas. The former is lower than 0.5%, and the latter is about 2%-30%. The $\delta^{13}\text{C}_1$ of biogas is less than -55‰, but that of low-mature gas is about -50‰ to -42‰. The difference between biogas derived from source rocks and biogas derived from hydrocarbon reservoirs is mainly demonstrated in the $\delta^{13}\text{C}$ of CO_2 . The latter is much heavier than the former. The carbon isotope of heavy hydrocarbons can be used to identify the primary biogas and secondary biogas. The heavy hydrocarbons of secondary biogas show thermal genetic characteristics and their carbon isotope of C_{2+} is heavier than that of the primary biogas, generally $\delta^{13}\text{C}_{2+} > -40\text{‰}$.

The distribution of different types of shallow-buried gas is controlled by different factors. Primary biogas generated from source rocks is mostly distributed in Quaternary basins or modern deltas, and its accumulation is controlled by the properties of the reservoir and caprock assemblages. Secondary biogas is mainly found in a basin with secondary burial history, and the distribution is controlled by fault activity. The oil-degraded biogas is distributed near heavy oil pools, and the more the oil is biodegraded, the more biogas accumulated. The low-mature gases are widely distributed in shallow-buried reservoirs in many basins.

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