

Geochemistry and origin of gas pools in the Gaoqing-Pingnan fault zone, Jiyang Depression

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Abstract In the surroundings of the Gaoqing-Pingnan fault zone are developed quite a number of gas reservoirs. Based on gas compositions, they can be divided into two groups, i.e., CO₂ and CH₄. Their composition and isotope geochemistry were dealt with in this study. The CO₂ contents range from 60.72%–99.99%, the $\delta^{13}\text{C}_{\text{CO}_2}$ values from -3.41‰– -9.8‰, and the ³He/⁴He ratios from 4.35×10^{-6} – 6.35×10^{-6} (i.e. R/Ra=4.45–4.35). Based on the data on composition and isotope geochemistry, deep geological background, deep faults and volcanic rocks, it is shown that CO₂, distributed in the Gaoqing area, mostly originated from mantle-source inorganic matter which is associated with magmatic rocks. The favorable tectonic environment for the formation of CO₂ reservoirs is the rift, which is related to great fault-magmatic activity, the formation of CO₂ gas pools and their space-time correlation to the most recent magmatic activities. Hydrocarbon gas pools occur in the Huagou area. The CH₄ contents are within the range of 88.83%–99.12%, and the $\delta^{13}\text{C}_{\text{CH}_4}$ values, -44.7‰– -54.39‰. This indicates that the hydrocarbon gas resulted from the decomposition of oil-type gas at high temperatures. Volcanic rocks in the CO₂ gas pool- and CH₄ gas pool-distributed areas show significant differences in Fe₂O₃ and FeO contents. This has proven that the hydrocarbon gas may have resulted from various chemical reactions. Magmatic activities are the primary reason for the distribution of CO₂ and CH₄ gas pools in the Gaoqing-Pingnan fault zone.

Key words gas pool; geochemistry; magmatic activity; mantle origin; Jiyang depression

1 Introduction

The Dongying depression of the Jiyang sub-basin lies in the southeastern part of the Bohai Bay Basin, which is an intracontinental extensional basin formed on the basis of the Proterozoic craton with Paleozoic cover rocks as a result of interaction between the Eurasian, Kula-Pacific and India plates since the Triassic period. The Gaoqing-Pingnan fault zone located in the west of the Dongying depression strikes north-eastwards and extends as long as 60 km from south to north. It can be divided into 3 segments: Gaoqing segment, Pingnan segment and Bingnan segment; the Gaoqing segment strikes west-eastwards, extending as long as 34 km; the Pingnan segment is NNE-striking, extending as long as 14 km; and the Bingnan segment is NE-striking, extending as long as 12 km. Their movement took place in different periods. The movement of the Gaoqing segment started at Early Cenozoic, and lasted till Pliocene. The movement of the Pingnan and Bingnan segments started at Middle Jurassic and ended at Early Pliocene.

According to its extending depth and detachment structures, the Gaoqing-Pingnan fault belongs to a deep-large fault which reaches the basement (Guo Dong et al., 2006). The deep fault directly links up the magma chamber and gas source, and controls the development of second-ordered tectonic units and synsedimentary faults. The synsedimentary faults are the conduit of magma volatiles ascending from depth to shallow level.

In the Gaoqing, Huagou and Pingnan segments, the accumulation of natural gases is related to the distribution of deep faults. Faults mainly controlled the zonation of gas pools and became a fault trap under appropriate conditions and further became the host of gas pools. There are two different types of gas reservoirs distributed along the Gaoqing-Pingnan fault zone. Hydrocarbon gas pools occur on the southern side, and non-hydrocarbon gas pools on the northern side (Fig. 1). Many researchers described the geochemical characteristics, distribution regularities, origin and reservoir-forming models, and suggested that CO₂ was derived from a mantle source (Dai Jinxing et al., 1995; Zheng Leping et al., 1995, 1997,

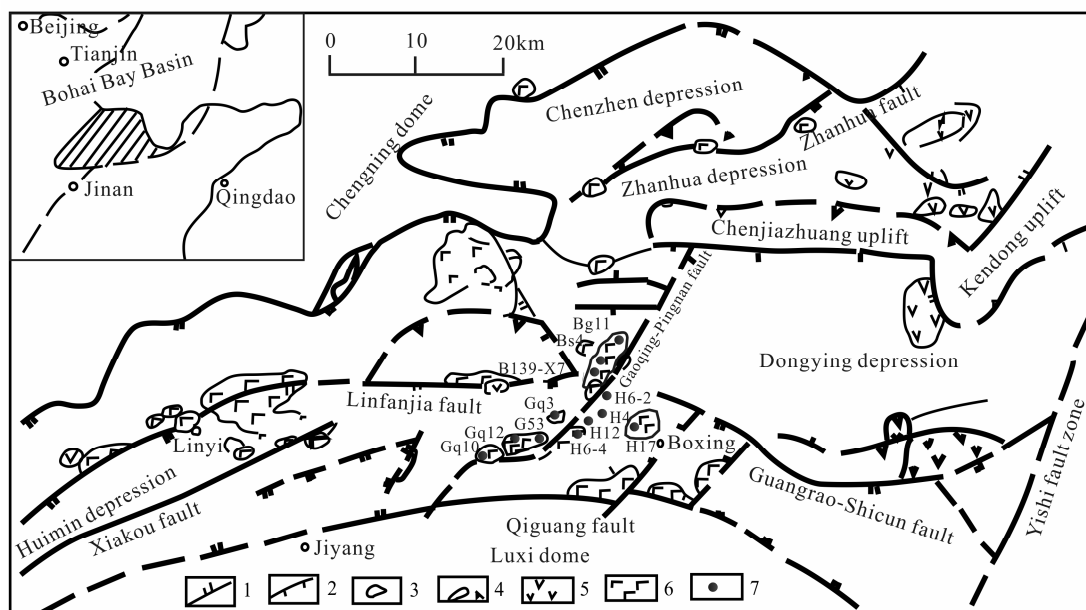


Fig. 1. Geological map showing the distribution of gas pools along the Gaoqing-Pingnan fault zone, Jiyang depression. 1. Deep fault; 2. fault; 3. gas pool; 4. Mesozoic intrusive rock; 5. effusive rock; 6. alkali basalt; 7. well.

2001; Hou Guiting et al., 1996; Liao Yongsheng et al., 2001; Shen Weizhou et al., 1998). In this paper, we used the available chemistry and isotope data in combination with the geological background to constrain the origin of natural gases in the Gaoqing-Pingnan fault zone.

2 Geological setting

Our study area is distributed mainly in 4 regions with 12 wells. Wells Bs-4 and B139-x7 are both located in the Pingfangwang Economic Development Region. The two wells are of bio-reef facies deposition because of loose limestones, well developed cracks and good permeability due to intensive weathering of Es₄ limestones. The gas-bearing formations are distributed mainly in reef limestones in the upper Es₄ and two sets of hydrocarbon-bearing formations in the middle Es₄. Well Bs-4 is located in the gas cap zone of reef limestones. Well B139-x7 is located near the gas-oil contact boundary of reef limestones. They are both located within the oil domain of the middle Es₄. The trap types of gas-bearing reservoirs include anticline traps which are controlled by both fault system and lithology. The strata from Es₂ to Es₃ are regional cap rocks.

Gas pools Gq-10 and Gq-12 lie in the upper part of the southwestern segment of the Gaoqing-Pingnan fault zone in the Qingcheng salient. The gas-bearing reservoirs belong to Mesozoic erosion-surface tectonic-lithological traps formed in the Qingcheng

salient. The gas-bearing reservoirs are composed of thin-layered siltstones of the Guantao Formation. The cap rocks are lilac, brownish-yellow and grayish-green mudstones overlying the Guantao Formation sandstones.

Both gas pools G-53 and G-3 are located in the west of the Qingcheng salient. The gas-bearing reservoirs consist dominantly of the Guantao Formation fine siltstones. The main types of deposition include flood plain, interchannel and channel marginal deposition because of loose rocks, favorable physical properties and minor carbonate. The cap rocks were composed of mudstones at the top of gas-reservoirs.

Gas pool H-6 is located in the Huagou fault zone. The gas-bearing reservoir is made up of Upper Guantao Formation siltstones which are characterized as being loose in lithology as well as by good physical properties, minor carbonate and relatively high argillaceous material. The depositional environments include flood plain, interchannel and channel marginal environments. The cap-rocks of the gas-reservoirs are composed of mudstones lying between the Upper Guantao Formation and the Lower Minhuazhen Formation.

Gas pool H-17 is located in the east of down-throw block of the Gaoqing-Pingnan fault. Gas-bearing formations are in the fan-shaped sand bodies at the bottom of Es₃. The cap rock is mudstone which is 100 m thick in the mid-Es₃.

3 Samples and methodology

All samples are pure gas phases which were collected by means of 1000 cc stainless steel cylinders sealed with high-pressure valves. The cylinders were directly connected to the gas well heads by copper pipes in commercial hydrocarbon production fields. The pipe lines were first flushed for 5–10 minutes to avoid air contamination before the samples were connected to the cylinders.

The carbon isotope ratios of hydrocarbons C₁, C₂, C₃, C₄, and those of CO₂ were measured by Gas Chromatography-Micro-Combustion-Isotope Ratio Mass Spectrometry (GC-C-IRMS) at the Stable Isotope Laboratory of the Geological Science Center

in the Shengli oilfield. The major gas compounds, hydrocarbons and non-hydrocarbons, were measured by gas chromatography on a Varian chromatograph. Hydrocarbon contents (including CH₄, C₂H₆, C₃H₈, and C₄H₁₀) were measured on a gas chromatograph. CO₂ contents were analyzed by GC-C-IRMS. The analytical error is ±2.5%.

4 Results

The abundances of the major gas species and the δ¹³C values of the sampled natural gases are given in Tables 1 and 2. The principal components of natural

Table 1. Gas composition in Pingfangwang, Gaoqing and Huagou gas fields

No	Situation (m)	Member	Component (%)						Source
			N ₂	CO ₂	CH ₄	C ₂ H ₆	C ₃ H ₈	C ₄ H ₁₀	
Bs4	1510.0–1568.0	Es4	1.39	60.72	32.62	2.3	2.03	0.58	a
B139-x7	1890.0–1898.0	Es4		94.13	3.53	0.85	0.86	0.38	
Bg11	1980.2–2250.0	Es4	0.30	97.32	1.31	0.34	0.37	0.35	b
Gq10	824.3–838.9	Ng		99.99	0				a
Gq12	820.0–850.0	Ng		99.91	0.08				
Gq3	833.4–834.8	Ng	2.06	97.87	0.07				c
Gq53	811.4–818.0	Ng		99.96	0.04				
H6-4	790.0–830.0	Ng	1.11	1.93	96.5	0.43	0		a
H6-2	743.8–783.4	Ng	0.93	1.92	96.7	0.43	0		
H12	804.8–813.6	Ng	0.27	0.04	99.12	0.40	0.05	0.02	
H4	1276.1–1282.0	Es1	7.90	0.08	88.83	0.99	0.86	0.88	c
H17	1965.1–1980.0	Es3	2.05	89.70	7.47	0.20	0.31	0.27	

Note: a. This work; b. Zheng Leping et al. (1995); c. Huang Gaojian et al. (2002).

Table 2. The carbon isotope characteristics of Pingfangwang, Gaoqing and Huagou gas fields

No	Member	δ ¹³ C _{PDB} (‰)					Source
		CO ₂	C ₁	C ₂	C ₃	C ₄	
Bs4	Es4	-9.8	-49.4	-32.4	-28.9	-28.2	a
B139-x7	Es4	-6.1	-45.8	-30.1	-27.4	-26.8	
Bg11	Es4	-5.9	-47.55				d
Gq10	Ng	-5.2					a
Gq12	Ng	-7.7					
Gq3	Ng	-4.41	-35.00				b
Gq53	Ng	-6.8					a
H6-4	Ng	-8.3	-44.7	-25.2			
H6-2	Ng	-8.6	-44.7	-24.8			
H17	Es3	-3.41	-54.39	-33.16	-31.25		b

Note: a. This work; b. Zheng Leping et al. (1995); d. Shen Weizhou et al. (1998).

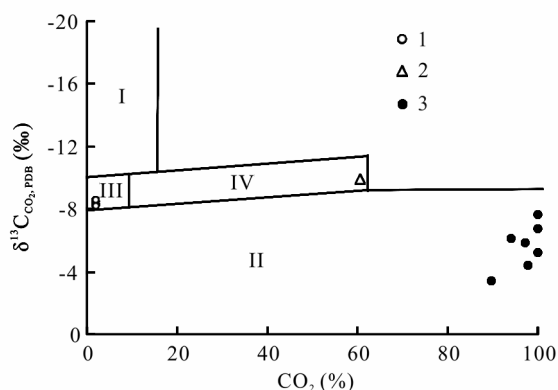


Fig. 2. The correlation between CO_2 and $\delta^{13}\text{C}_{\text{CO}_2}$ in the Gaoqing-Pingnan fault zone (modified after Dai Jinxing et al., 2001b). I. Organic; II. inorganic; III. organic and inorganic mixture; IV. inorganic and organic mixture. 1. H6-2, H6-4; 2. Bs-4; 3. CO_2 gas pool.

gases in this study are CO_2 and hydrocarbons. Almost pure CO_2 gases were found in the north of the Gaoqing-Pingnan fault zone. The CO_2 contents range from 60.72%–99.99% in volume, except for Bs-4—a which is a mixture of CO_2 and CH_4 . In the other wells the CO_2 contents are higher than 94%. The gas samples taken from the southern side of the fault zone were dominated by methane, while those from the northern side have higher concentrations of CO_2 . Methane contents are higher than 88.83% except for H-17. $\delta^{13}\text{C}_{\text{CO}_2}$ values for all gas samples vary considerably, ranging from -7.7‰ to -3.41‰. In the CO_2 (%) vs. $\delta^{13}\text{C}_{\text{CO}_2}$ plot (Fig. 2), most of the data points fall within the area of inorganic gas (area II), with Bs-4 lying in area IV of mixed gas with inorganic gas being dominant, and H6-2 and H6-4 lying in area III of mixed gas with organic gas being dominant). $\delta^{13}\text{C}_{\text{CH}_4}$ values for all the samples are within the range of -35.00‰– -54.39‰. There is no clear boundary between hydrocarbon and non-hydrocarbon gas pools (Fig. 3).

5 Discussion

Natural gases comprise all kinds of gas. Their chemical composition is an important criterion to distinguish their genesis types and understand their distribution regularities. Natural gases can be classified as hydrocarbon and non-hydrocarbon categories according to their gas components. Hydrocarbon components are mainly CH_4 and C_2H_6 in addition to other heavy hydrocarbons. Non-hydrocarbon components are dominated by CO_2 , with minor N_2 , H_2S , H_2 and some trace gases such as He, Ar and Ne. Because of migration-fractionation and bacterial degradation, different or identical genetic types of natural gases show significant differences in isotopic

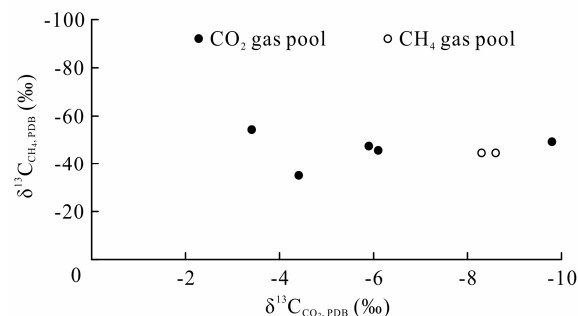


Fig. 3. The correlation between $\delta^{13}\text{C}_{\text{CO}_2}$ and $\delta^{13}\text{C}_{\text{CH}_4}$ in the Gaoqing-Pingnan fault zone.

composition. In terms of the isotope geochemistry data of stable noble gases we can distinguish their origins. Because noble gases are chemically inert, almost no change would occur in their abundance and isotopic composition in the geological processes. So noble gases are regarded as a useful approach to constraining the origin of natural gases.

5.1 Origin of CO_2

In the northern part of the Gaoqing-Pingnan fault zone CO_2 is a predominant component except for Bs-4 well. CO_2 contents are higher than 94%. The formation of such high-purity gas pools requires that they should have originated from carbonate decomposition or be of mantle derivation (Baker et al., 1995; Clayton et al., 1990). Carbonate decomposition is a geological process in which CO_2 is released from carbonates at high temperatures and then accumulated as gas pools under favorable geological settings. Mantle-derived CO_2 was accumulated as CO_2 gas pools during degasification of magma. In the Meso-Cenozoic periods, extensive volcanic activities occurred in the Gaoqing-Pingnan area, as exemplified by the occurrence of diabase in the Es2 member and Mesozoic andesite basalt (Guo Dong et al., 2004). At least, 5 generations of brine inclusions, which are connected genetically with magma filling and CO_2 injecting, were found in well Gq-10 at the depths of 757.24–839.84 m (Li Chenyi et al., 2004). This shows that the multi-phase volcanic activities did occur in the Gaoqing area. In that area the CO_2 gas pools have a close relation with the last phase of volcanism (Fig. 1). The Gaoqing-Pingnan fault is the conduit for mantle magma upwelling and degassing of CO_2 in the condensation process. CO_2 gradually ascended and found its way into thin-layered siltstones of the Guantao Formation, forming gas reservoirs along the fault zone.

Generally speaking, the $\delta^{13}\text{C}_{\text{CO}_2}$ values of carbon dioxide ranging from -4‰ to -7‰ are considered to represent the equivalent values of mantle-derived CO_2

(Thrasher and Fleet, 1995; Koncz, 1983). The carbon isotope data could not provide reasonable interpretations, because the average mantle-derived CO₂ and bulk crustal CO₂ have similar $\delta^{13}\text{C}_{\text{CO}_2}$ values (from -3‰ to -8‰ PDB) (Bredehoeft and Ingebritsen, 1990). In the light of $\delta^{13}\text{C}$ values we can distinguish CO₂ of inorganic genesis. But we cannot ascertain petrochemical genesis or mantle-derived magma genesis. Noble gases can provide further evidence for the origin of CO₂. The results indicate that the $^3\text{He}/^4\text{He}$ ratios of gas samples range from 4.45×10^{-6} – 6.35×10^{-6} in the Gaoqing-Pingnan fault zone, and the R/R_a ratios of air range from 4.45–6.35 (R is the ratio of $^3\text{He}/^4\text{He}$ in the samples, and R_a is the ratio of $^3\text{He}/^4\text{He}$ in air, 1.4×10^{-6}) (Zheng Leping et al., 1997, 2001; Li Chenyi et al., 2004; Che Yan et al., 2001). Usually in mantle-derived CO₂, the helium isotope ratio of R/R_a is higher than 2.5 (O'Nions and Ballentine, 1993; Sherwood-Lollar et al., 1997; Alison et al., 2004). The stable carbon isotope and helium isotope data show that the carbon dioxide resulted from mixing of mantle-derived and crustal-derived end-members. Mantle-derived gas is most abundant in well Gq-3. It is considered that CO₂ in the area is mainly a mixture of gas of magmatic origin and that of petrochemical origin. He Ying et al. (1997) measured the $\delta^{13}\text{C}_{\text{CO}_2}$ values of pyroxene inclusions in the volcanic rocks, which are within the range of -7.2‰– -8.4‰, lower than those of gas samples from gas wells. The most important reason is that in the basement strata of the Gaoqing area exist Ordovician carbonate buried hills (Wang Xingmou et al., 2004). In the process of magma upwelling there was produced a lot of heat, therefore carbonates were baked, leading to the release of massive CO₂. The $\delta^{13}\text{C}_{\text{CO}_2}$ values of CO₂ produced as a result of baking are usually within the range of 3.7‰–3.7‰; those of the mixed gas of mantle-petrochemical origin are higher than those of inclusions in the volcanic rocks. Such contact metamorphism would produce a huge volume of carbon dioxide (Yang Xiaoyong et al., 2003). The migration and accumulation of this kind of CO₂ in the favorable geological settings would promote the formation of gas reservoirs under cap-strata condition. Moreover, in wells Bs-4, H6-2 and H6-4 the $\delta^{13}\text{C}_{\text{CO}_2}$ values of gas samples are low, indicating that a part of oil-type CO₂ was involved in the carbon dioxide of inorganic origin. For this reason, the $\delta^{13}\text{C}_{\text{CO}_2}$ values are decreased.

5.2 Origin of CH₄

The shallow oil-type gases are believed to have originated from hydrocarbon source rocks dominated by lacustrine shale mudstones of the Shahejie Formation in the Gaoqing-Pingnan fault zone (Cheng

Youyi et al., 1995; Dai Jinxing et al., 2001a). Gas-source correlation is an important approach to understanding natural gas systems in hydrocarbon exploration. The isotopic composition of methane in natural gases is dependent on the type and maturity of source rocks. This has been established from natural samples and pyrolysis experiments (Chung and William, 1979). According to Dai Jinxing et al. (1992), the relation between the ^{13}C concentrations in methane and the vitrinite reflectance (R_o) is described as follows:

$$\text{Oil-type gases: } \delta^{13}\text{C}_1 = 15.80 \lg R_o - 42.20 \quad (1)$$

Vitrinite reflectance (R_o) ranges from 0.16% to 0.69%, as revealed by the above equation and some data of Table 2. The source rocks are generally at the low-mature to mature stage of evolution. Vitrinite reflectance (R_o) (2.86%) for well Gq3 is much higher than that for other wells. This is attributed to multi-episode magmatic eruption in the Gaoqing area. The geothermal temperature of sedimentary basin would rise under the action of deep thermal fluids which have strong solubility and diffusion capacity. The high temperature would accelerate the thermal evolution of hydrocarbon source rocks, making local organic matter become abnormally over-mature. The stable carbon isotopic composition of methane offers useful information about the origin of methane. Bacterial methane has been demonstrated to be rich in ^{12}C and the $\delta^{13}\text{C}_1$ values are less than -55‰. The $\delta^{13}\text{C}_1$ values higher than 50‰ are considered as an indicator of thermogenic gas (Jenden et al., 1993; Wang Wanchun et al., 2003, 2005; Zheng Jianjing et al., 2006). From Fig. 3 we can see that the $\delta^{13}\text{C}_1$ values range from -35.00‰– -49.44‰. There is no obvious difference between CO₂ reservoir and CH₄ gas pool. Hydrocarbon gases could be, for the most part, designated to thermogenic gases. In the sedimentary basin hydrothermal systems would supply methane with intermediate $\delta^{13}\text{C}_1$ values, ranging from -30‰ to -50‰. Recent experimental results have shown that in the process of diffusion methane is always significantly lighter in carbon isotopic composition than that in the source rocks during migration from depth to shallow level (Pernaton et al., 1996; Prinzhofer and Pernaton, 1997). The $\delta^{13}\text{C}_1$ values ranging from -30‰ to -50‰ implied that the methane may have come from degassing of the upper mantle or the interaction of CO₂ with water in the redox buffer of FeO. In the light of the carbon isotopic composition of this methane it is difficult to distinguish it from thermogenic methane. Recently, Prinzhofer et al. (2000) have developed a completely independent way of distinguishing these two types of gases differing in origin in terms of the ratios of the two radiogenic

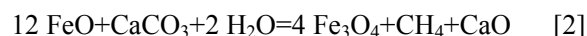
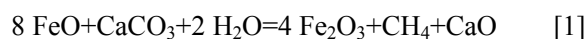
noble gases, ^4He and $^{40}\text{Ar}^*$ (referring to radiogenic portion of the total ^{40}Ar), as they have not been affected by secondary chemical processes. Additionally, physical processes should not lead to fractionation of helium and argon over the range of observed variations. If some helium is of mantle deviation, it should diffuse more easily than argon. $^4\text{He}/^{40}\text{Ar}^*$ ratios are expected to be high for either methane resultant from upper mantle degassing or that from interaction of CO_2 with water in the redox buffer of FeO . On the other hand, ^4He and $^{40}\text{Ar}^*$ molecules would promote their effective transfer at high forming temperatures of thermogenic gas, and $^4\text{He}/^{40}\text{Ar}^*$ ratios in these gases are relatively low, always close to the average crustal values. Owing to the existence of hydrocarbon source rocks in the sedimentary basin, magma upwelling, could produce huge heat that could lead to the production of alkane gas in large amounts. Methane, when migrating into the sedimentary basin, may be considered as alkane gas of organic origin. According to the geological and geochemical conditions of formation of natural gases (Dai Jinxing, 2006), there exists methane derived from deep rift basins in eastern China.

Zhang Xuehua and Zheng Yongfei (1997) considered that the $\delta^{13}\text{C}_1$ values within the range of -30‰ – -50‰ may be indicative of inorganic origin. Inorganic alkane gases typically showed a negative stable carbon isotope sequence of $\delta^{13}\text{C}_1 > \delta^{13}\text{C}_2 > \delta^{13}\text{C}_3 > \delta^{13}\text{C}_4$. Computation (Li Chunyuan et al., 1999) showed that a completely reverse carbon isotopic distribution pattern for C_1 to C_4 hydrocarbons can be attributed to mixing of two kinds of biogenic gas with a normal carbon isotopic distribution pattern. Meanwhile, a normal carbon isotopic distribution pattern in natural gases can also be produced by two reverse end members. In the first case, the two end members should be different in source, formation mechanism, or evolutionary stage. We considered that in this area hydrocarbon gas is a mixture of organic and inorganic gases, according to multi-episode magma eruption and the geochemical features of fault structures.

There are two main sources of abiogenic methane. One is methane volatile escaping from the magma. Xie Hongsen (1997) suggested that the magma contains a considerable amount of volatile carbon species, primarily CO_2 and CH_4 . At the time of magma upwelling along the fault, the pressure and temperature would be reduced, and methane would escape from the magma (Zhu Yongfeng, 1997). There were found calcite veins of mantle deviation. The $\delta^{13}\text{C}$ values of the calcite range from -7.3‰ – -7.9‰ . Raman spectral analysis of fluid inclusions in the calcite showed that the gas phase CH_4 accounts for 9.0% – 14.2% (mol) and the liquid phase CH_4 accounts

for 0.08% (mol). This has proven that developed in this area is mantle-source methane (Jin Zhijun et al., 2002). The mantle-source methane with high $\delta^{13}\text{C}_1$ values, when mixed with organic methane, will inevitably lead to light $\delta^{13}\text{C}_1$ values.

Other abiogenic hydrocarbons are generally formed from the reduction of carbon dioxide. A process is thought to occur during magma cooling and more commonly in the hydrothermal systems of water-rock interactions. Water heated by magma reacted with minerals such as olivine, which contain high levels of the catalyst iron. During serpentinization, hydrogen liberated from water would react with carbon from carbon dioxide to form methane. We analyzed the volcanic rock samples for their Fe_2O_3 and FeO contents (He Ying and Liao Yongsheng, 2001). Significant differences are noticed in Fe_2O_3 and FeO contents of the volcanic rocks between CO_2 gas-fields and CH_4 gas-fields. The contents of FeO in the CO_2 gas-fields are much higher than in the CH_4 gas-fields, however, the contents of Fe_2O_3 in the CH_4 gas-fields are higher than in the CO_2 gas-fields. This shows that FeO was oxidized into Fe_2O_3 in the CO_2 gas-fields, and the corresponding mantle-derived CO_2 has been partially converted to CH_4 . The experimental results (Scott et al., 2004) showed that hydrocarbons were produced as a result of carbonate reduction at upper-mantle pressures and temperatures. Methane was derived from interaction of FeO , CaCO_3 (e.g. calcite) with water at pressures between 5 and 11 GPa and temperatures ranging from 500 to 1500 . The experimental results demonstrated the existence of pathways for the formation of abiogenic hydrocarbons in the Earth's interior. The possible reactions are presented as follows:



It is further evidenced that hydrocarbon gases have mixed with some hydrocarbons of abiogenesis in the region studied.

5.3 Differences for gas reservoirs on both sides of the fault zone

Gas pools show significant differences on both sides of the Gaoqing-Pingnan fault zone. CO_2 gas pools occur on the northern side while hydrocarbon gas reservoirs occur on the southern side. Such differences may be attributed to extensive magmatic activities.

Magmatic activities during the Triassic are very extensive in the Huiming depression and Dongying

depression, covering an area of 1000 km² and a thickness of one kilometer. In the early time magmatic activities led to the release of CO₂ gas but it is impossible to form any effective gas trap because of the influence of open faults. Figure 1 indicates that the distribution of CO₂ is related with igneous rocks of the Cenozoic alkali basalt series. Volcanic activities tend to weaken following the Neogene period, and the intense phase occurred mainly in northern Gaoqing-Pingnan fault zone. Alkali basalt is the dominant product of volcanic effusion. Abundant CO₂ was produced in the late period of magmatic activity.

Figure 3 indicates that the $\delta^{13}\text{C}_1$ values of gas samples taken from both sides of the fault zone show no difference. This implicates that the hydrocarbon reservoirs were formed in the same period. In early magmatic activities, the fault became the channelway of magma upwelling. Massive magmatic materials occurred as fillings on both sides of the fault zone. Magma eruption produced huge heat energy which accelerated the transformation of organic matter into hydrocarbons. So gases in the area studied are dominated by thermogenic gas, and mixed with part of CH₄ volatile component and mantle-source CH₄. The accumulation of gaseous hydrocarbons would give rise to the formation of gas reservoirs under favorable geological settings. When crustal stress was released, faulting would stop and the faults would close. Faults on the southern side provided no pathway for gas source, and the gas pools have not been destroyed by overlying igneous rocks. A phase of weak magma activity occurred on the northern side of the fault zone. Following magmatic invasion and eruption, heat energy was released and the temperature gradually decreased, an igneous rock column was formed in the process of condensation. Carbon dioxide found its way into the Guantao Formation thin-layered siltstones to form gas reservoirs.

Under the same cap rock conditions, CH₄ is different in molecule radius from CO₂. CH₄ molecules diffused very rapidly because of its small molecular structure and strong molecular activity. So, CH₄ is relatively of easy leakage. The gas pools are composed mainly of CO₂ with minor CH₄ in the Gaoqing area.

6 Conclusions

(1) In the Gaoqing-Pingnan fault zone CO₂ gas pools are composed of mixed gases of mantle-derivation & petrochemical origin; and gaseous hydrocarbon pools are primarily composed of mixed thermogenic and abiotic gases.

(2) A comparison of the Fe₂O₃ and FeO contents of volcanic rocks indicates that in the gas reservoirs there exists alkane gas resultant from reduction of

mantle-derived CO₂ in the gas pools.

(3) The primary reason for the different distributions of gas pools on both sides of the fault zone is magmatic activity.

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References

- Alison M.S., David R.H., Colin, G.M., and John M.S. (2004) The CO₂-He-Ar-H₂O systematics of the Manus back-arc basin: Resolving source composition from degassing and contamination effects [J]. *Geochimica et Cosmochimica Acta*. **68**, 1837–1856.
- Baker J.C., Bai G.P., Hamilton P.J., Golding S.D., and Keene J.B. (1995) Continental-scale magmatic carbon dioxide seepage recorded by dawsonite in the Bowen-Gunnedah-Sydney Basin system, eastern Australia [J]. *Journal of Sedimentary Research*. **A65**, 522–530.
- Bredehoeft J.D. and Ingebritsen S.E. (1990) Degassing of carbon dioxide as a possible source of high pore pressures in the crust. In *The Role of Fluids in Crustal Processes* (eds. J.D. Bredehoeft and D.L. Norton) [M]. pp.158–164. National Academic Press, Washington, D.C.
- Che Yan, Jiang Huichao, and Mu Xing (2001) Gas reservoir type and reservoir-forming rule of Huagou gas field [J]. *Petroleum Geology and Recovery Efficiency*. **8**, 32–34 (in Chinese with English abstract).
- Cheng Youyi, Song Lailiang, Guo Jin et al. (1995) Favorable conditions for shallow (Neogene) gas accumulation in Jiyang Depression [J]. *Petroleum Exploration and Development*. **22**, 16–19 (in Chinese with English abstract).
- Chung H. Moses and William M. Sackett (1979) Experimental confirmation of the lack of carbon isotope exchange between methane and carbon oxides at high temperatures [J]. *Geochimica et Cosmochimica Acta*. **43**, 273–276.
- Clayton J.L., Spencer C.W., Koncz I., and Szalay A. (1990) Origin and migration of hydrocarbon gases and carbon dioxide, Beks Basin, southeastern Hungary [J]. *Organic Geochemistry*. **15**, 233–247.
- Dai Jinxing (2001a) *Geology and Geochemistry of Natural Gases* [M]. pp.225–226. Petroleum Industry Press, Beijing (in Chinese).
- Dai Jinxing (2006) Characteristic of abiogenic gas resource and resource perspective [J]. *Natural Gas Geoscience*. **17**, 1–6 (in Chinese with English abstract).
- Dai Jinxing, Pei Xigu, and Qi Huofa (1992) *Chinese Geology of Natural Gases* [M]. pp.55–63. Petroleum Industry Press, Beijing (in Chinese).
- Dai Jinxing, Shi Xi, and Wei Yanzhao (2001b) Summary of the abiogenic origin theory and the abiogenic gas pools (fields) [J]. *Acta Petrolei. Sinica*. **22**, 5–11 (in Chinese with English abstract).
- Dai Jinxing, Song Yan, Dai Chunsen, Chen Anfu, Sun Mingliang, and Liao Yongsheng (1995) *Inorganic Gases and the Formative Conditions of the Inorganic Gas Pools in Eastern China* [M]. pp.131–150. Science Press, Beijing (in Chinese).

- Guo Dong Qiu Longwei, and Jiang Zaixing (2004) Development features of igneous rocks in Jiyang Depression and its relationship with CO₂ reservoir forming [J]. *Petroleum Geology and Recovery Efficiency*. **11**, 21–24 (in Chinese with English abstract).
- Guo Dong, Xia Bin, Wang Xingmou et al. (2006) Relationship between faulting and reservoiring of CO₂ in Jiyang Depression [J]. *Natural Gas Industry*. **26**, 40–42 (in Chinese with English abstract).
- He Ying and Liao Yongsheng (2001) Auriferous CO₂ rich fluids in Shengli Oilfield, Shandong Province and their origin [J]. *Geological Review*. **47**, 454–464 (in Chinese with English abstract).
- He Ying, Wang Dingyi, Liu Hongying et al. (1997) Origin of carbon dioxide gas reservoir Shengli Oilfields [J]. *Oil & Gas Geology*. **18**, 82–82 (in Chinese with English abstract).
- Hou Guiting, Qian Xianglin, Song Xinmin, and Fan Liangxing (1996) The origin of carbon dioxide gas fields in Jiyang Basin [J]. *Acta Scientiarum Naturalium Universitatis Pekinensis*. **32**, 712–718 (in Chinese with English abstract).
- Huang Gaojian, Chen Jianyu, Zhang Dongmei et al. (2002) Origin and accumulation of CO₂ gas in the Huagou-Gaoqing gas field, Jiyang depression [J]. *China Offshore Oil and Gas (Geology)*. **16**, 295–301 (in Chinese with English abstract).
- Jenden P.D., Hilton D.R., and Kaplan I.R. (1993) A biogenic hydrocarbons and mantle helium in oil and gas fields. In *The Future of Energy Gases* (ed. D.G. Howell) [M]. pp.31–56. United States Government Printing Office, Washington.
- Jin Zhijun, Zhang Liuping, and Zeng Jianhui (2002) Compound genesis alkenes relate to mantle source CO₂ fluid [J]. *Science in China (Series D)*. **47**, 1276–1280 (in Chinese with English abstract).
- Koncz I. (1983) The stable carbon isotope composition of the hydrocarbon and carbon dioxide components of Hungarian natural gases [J]. *Acta Mineralogica-Petrographica, Szeged*. **XXVI**, 33–49.
- Li Chenyi, Fan Tailiang, and Zheng Herong (2004) Formation mode of carbon dioxide gas reservoirs in Yangxin-Huagou area [J]. *Acta Petrolei. Sinica*. **25**, 35–39 (in Chinese with English abstract).
- Li Chunyuan, Wang Xianbin, and Xia Xinyu (1999) The possibility and explanation of complete reverse isotopic distribution of light hydrocarbons in natural gas [J]. *Acta Sedimentologica Sinica*. **17**, 306–311 (in Chinese with English abstract).
- Li Pilong, Wang Xingmou, Guo Dong et al. (2004) *Comprehensive Study of CO₂ Gas Exploration Shandong Area Shengli Oilfield* [M]. pp.70–72. China Petrochemical Ltd., Shengli Oilfield.
- Liao Yongsheng, Li Juyuan, Li Xiangchen, and Xu Shougen (2001) A discussion of CO₂ genesis in Jiyang Depression by using C, He, Ar isotopes [J]. *Bulletin of Mineralogy, Petrology and Geochemistry*. **20**, 351–353 (in Chinese with English abstract).
- O'Nions R.K. and Ballentine C.J. (1993) Rare gas studies of basin scale fluid movement [J]. *Philosophical Transactions of the Royal Society of London*. **344**, 141–156.
- Pernaton E., Prinzhofer A., and Schneider F. (1996) Reconsideration of methane signature as a criterion for the genesis of natural gas: Influence of migration on isotopic signature [J]. *Revue de l'IFP*. **51**, 635–651.
- Prinzhofer A. and Pernaton E. (1997) Isotopically light methane in natural gases: Bacterial imprint or segregative migration? [J]. *Chemical Geology*. **142**, 193–200.
- Prinzhofer A., Mello M.R., Da Silva Freitas L.C., and Takaki T. (2000) *A New Geochemical Characterization of Natural Gas and Its Use in Oil and Gas Evaluation* [C]. Hedberg Conference, Rio de Janeiro, November 1997. Special publication of the AAPG.
- Shen Weizhou, Xu Shijin, Wang Rucheng, and Lu Jianjun (1998) The study on the isotopic characters and the origin of the CO₂-rich gas deposits of Jiyang Depression [J]. *Journal of Nanjing University (Natural Sciences)*. **134**, 308–313 (in Chinese with English abstract).
- Sherwood-Lollar B., Ballentine C.J., and O'Nions R.K. (1997) The fate of mantle-derived carbon in a continental sedimentary basin: Integration of C/He relationships and stable isotopic signatures [J]. *Geochimica et Cosmochimica Acta*. **61**, 2295–2307.
- Scott H.P., Hemley R.J., Mao H.K., Mao H.J., Herschbach D.R., Fried L.E., Howard W.M., and Bastea S. (2004) Generation of methane in the Earth's mantle: In-situ high pressure-temperature measurements of carbonate reduction [J]. *PNAS*. **101**, 14023–14026.
- Thrasher J. and Fleet A.J. (1995) Predicting the risk of carbon dioxide 'pollution' in petroleum reservoirs. In *Organic Geochemistry: Developments and Applications to Energy, Climate, Environment and Human History* (eds. J.O. Grimalt and C. Dorronsoro) [C]. pp.1086–1088. Proceedings 17th International Meeting on Organic Geochemistry, San Sebastian, Spain.
- Wang Wanchun, Liu Wenhui, and Gao Bo (2003) Genetic identification of natural gas in shallow combination reservoirs in China [J]. *Natural Gas Industry*. **23**, 20–23 (in Chinese with English abstract).
- Wang Wanchun, Liu Wenhui, Xu Yongchang, and Shen Ping (2005) Genetic identification of natural gases from shallow reservoirs in some oil- and gas-bearing basins of China [J]. *Chinese Journal of Geochemistry*. **24**, 90–95.
- Wang Xingmou, Qiu Longwei, and Jiang Zaixing (2004) The relativity of igneous activity and CO₂ gas reservoir in Jiyang Depression [J]. *Natural Gas Geoscience*. **15**, 423–426 (in Chinese with English abstract).
- Xie Hongsen (1997) *An Introduction to Material Science in the Earth's Interior* [M]. pp.215–216. Science Press, Beijing (in Chinese).
- Yang Xiaoyong, Liu Deliang, and Chen Yongjian (2003) Reservoir formation model for inorganic natural gas under contact metamorphism: Exemplified as Shuangshan area in the south of Tanlu fault belt [J]. *Acta Petrolei. Sinica*. **24**, 19–22 (in Chinese with English abstract).
- Zhang Xuehua and Zheng Yongfei (1997) A theoretical model for the effect of out gassing on the carbon isotope composition of anorganic natural gas [J]. *Bulletin of Mineralogy, Petrology and Geochemistry*. **16**, 81–85 (in Chinese with English abstract).
- Zheng Jianjing, Hu Huifang, and Sun Guoqiang (2006) Carbon isotopic characteristics of hydrocarbon gases from coal-measure source rocks—A thermal simulation experiment [J]. *Chinese Journal of Geochemistry*. **25**, 167–172.
- Zheng Leping, Feng Zujun, Liao Yongsheng, and Xu Shougen (1997) Genesis of the non-hydrocarbon gas reservoir (CO₂, He) in Jiyang Depression [J]. *Journal of Nanjing University (Natural Sciences)*. **33**, 76–81 (in Chinese with English abstract).

Zheng Leping, Feng Zujun, Xu Shougen, and Liao Yongsheng (1995)

CO₂ gas pools originated from the earth interior in Jiyang Depression [J]. *Chinese Sci. Bull.* **40**, 2264–2266 (in Chinese).

Zheng Leping, Wang Shujun, Liao Yongsheng, and Feng Zujun (2001)

CO₂ gas pools in Jiyang Sag, China [J]. *Applied Geochemistry*. **16**,

1033–1039.

Zhu Yongfeng (1997) Mantle fluid and earth degassing [J]. *Earth Science Frontiers* (China University of Geoscience, Beijing). **5** (suppl.), 71–75 (in Chinese with English abstract).