Origin of Natural Gas in Kekeya Field, Tarim Basin, China *

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ABSTRACT: This paper is mainly concentrated on the geochemical characteristics and origin of gas of Kekeya field in the Tarim basin. NW China. This study shows that Permian mudstone is the main source rock of oil and gas. Based on the carbon isotopes of C1—C4, the carbon isotope of gas in Kekeya field is a little heavier than that in the typical marine-derived gas. The relationship between carbon isotopes of methane and ethane is coincident with Faber equation of gas derived from organic matter I / II. The majority of gas maturity is estimated based on the formula at 1.8 %—2.2 % besides K2 and K18 wells. In addition, the gas derived from 0.9 %—1.2 % R, source rocks may also be mixture. 40Ar/36Ar and 3He/4He ratios from the gas samples also support the mixing process. Moreover, the gas in this region is mainly generated from more mature source rocks although the low mature gas exists.
KEY WORDS: natural gas, origin, Kekeya, source rock.

INTRODUCTION
The Tarim basin is one of the largest basins in the world that has not been well explored. The exploration programs that are special for natural gas systems are becoming increasingly important in the search for new hydrocarbon resources of the region because of higher maturity source rocks. Successful exploration in turn requires a good understanding of all elements of the petroleum system, of which the possible source rock kitchen is a basic element (Chen, 2000). Specially, the southwest depression is an important and difficult region in Tarim basin. The Kekeya field was discovered in 1977. However, the source rocks and origin of gas and oil have not been clarified yet. The complexity of geological and geochemical data, relatively higher maturity oil and gas samples, lower petroleum exploration activity may lead to this situation. The three different source rocks, i.e. Carboniferous-Permian mudstone and limestone, Jurassic coal-bearing strata, Upper Cretaceous-Eogene source rocks have been proposed by many researchers. Li et al. (1999) deduced that the oils and condensates were from the Carboniferous-Permian sections and paraffinic condensates were not formed by thermal cracking of oil by oil and condensate analysis. But their deduction is still not approved because of the lack of source rock data. Some of researchers still believed that the natural gas is mainly derived from coal-bearing strata (Song, Y., et al., 2001. Personal communication). Fortunately, new exploration target in this region provides us with a chance to obtain new oil and source rock samples for the renewal of our current knowledge. The purpose of this study was therefore to determine the origin of natural gas and condensate and to provide new data and evidence for the contradicting gas origin hypotheses of this region.

GEOLOGICAL SETTING
Tarim basin, surrounded by Tianshan
Mountains and Kunlun Mountains, located in the southern part of Xinjiang, is the largest sedimentary basin in China. The southwestern depression of Tarim basin is an important depression and contains four tectonic sub-units: the Maigaiti slope, the Kashi sag, the Yechang-Hetian sag and the Qimuugen salient (Fig. 1). The depression experienced two different sedimentary processes that include Paleozoic platform and Meso-Cenozoic foreland depression. The Eogene depression is located in the foreland basin to the north of suture zone of collision between Eopaleozoic Tarim plate and the middle of Kunlun island arc. Although far from the suture zone in Neogene, the depression is still characterized by the foreland basin in the shape of basin with sedimentary and subside characters and structure. The stratigraphic sequence is relatively complete in this region and the thickness of deposit is about 12 000—20 000 m (Fig. 2). The platform led to the Paleozoic marine deposit that includes strata from Sinian to Carboniferous-Permian. The structure is mainly showing its vertical movement of lithosphere. The shallow marine plateau facies strata of Cambrian, Ordovician, Silurian are steadily and widely distributed in the depression. The Carboniferous-Permian carbonates and clastics are marine transgression strata which was also widely distributed in the depression. The Carboniferous-Permian strata are thinner in Maigaiti slope and Ba-chu lift and thicker in region near Kunlun Mountain because of Caledonian orogeny and Hercynian orogeny. The sedimentary facies changed from shallow marine plateau facies into inland lacustrine facies, And Jurassic deposit is mainly non-marine coal-bearing clastics. Upper Cretaceous-Eocene section is composed of fluvial flood plain to shallow water, lagoon, marine platform facies deposits where the Tethys Sea transgressed along the northern foot of Kunlun Mountains eastward into the area around Hetian. Therefore, five potential source rocks are present in this region which include Cambrian-Ordovician and Carboniferous-Permian marine carbonates and clastics, Jurassic coal-bearing strata and the Upper Cretaceous-Neogene marine sediments. Among these potential source rocks, it is more disputable if coal plays an important role for gas generation.

**ANALYTICAL TECHNIQUES**

The oil, gas and source rock samples have been analyzed in this study. The chemical compositions of gas samples were analyzed with thermal gas chromatography. The composition of gas is given as nitrogen, carbon dioxide, methane, ethane, propane, butane and so on. The stable carbon ratios measured on Finnigan MAT-251 TM isotope ratio mass spectrometer are reported in δ notation in per mil (10^{-3}) deviation, relative to the PeeDee Belemnite (PDB) marine carbonate standard. The rock samples were pulverized and Soxhlet extracted with chloroform. Then asphaltene were precipitated with n-hexane followed by filtration. The asphaltene-free bitumens were further separated with column chromatography into saturated hydrocarbons, aromatic hydrocarbons, resins, using hexane, benzene and alcohol as the solvents, respectively.

**NATURAL GAS COMPOSITION, STABLE ISOTOPIC CHARACTERISTICS AND THEIR ORIGINS**

The natural gases in this region, mostly hydrocarbon gases, include mainly methane, ethane and heavier hydrocarbon (above ethane). The hydrocarbon gases account for above 80 % and are mostly as high as above 90 % (Table 1). The nonhydrocarbon gas compositions are mainly N₂, CO₂, O₂ and some others (Table 1). The nitrogen is mostly below 8 % except in Well K10. Compared with most gas in Tarim basin, the hydrocarbon gas composition has lower methane content that varies from 71.2 % to 88.9 %. The wet gas can reach as high as 11.5 % and is generally related to characteristics of associated gas. But this will not be the real situation as present some other data show that will be dealt with detailed discussion below. A relatively lower C₄/C₃ ratio is found in this region, compared with the gas of typical big gas field in Hetian region in Tarim basin (Fig. 3). The carbon isotope compositions of the hydrocarbon components in gases are mostly not in agree-

![Figure 1. Sketch map showing tectonic unit of Tarim basin and location of Kekeya field.](Image)
mment with the rules that n-alkane gas is of positive carbon isotope series, i.e., $\delta^{13}C_1 < \delta^{13}C_2 < \delta^{13}C_3 < \delta^{13}C_4$ (Table 2). The carbon isotope of propane and butane has its reverse order, i.e., $\delta^{13}C_3 > \delta^{13}C_4$, which may be related to the gas composed of gases in different origins. Generally speaking, the carbon isotopic ratio of methane ($\delta^{13}C_1$), varying from $-43.34 \times 10^{-3}$ to $-36 \times 10^{-3}$, belongs to typical organogenic gases. The carbon isotopic composition of hydrocarbon components in this region is heavier than that of Hetian gas field which is believed to be derived from typical Paleozoic marine source rocks (Fig. 4). The gas in Hetian gas field is dry whose drying coefficient varies from 0.95 to 0.99. Most researchers believed that such a gas belonged to marine high and postmature gas (Zhao and Zhang, 2001), which may give us a hint that the gases in this region were not derived from typical marine source rocks because of its heavier carbon isotopic ratio and more wet gas content.

**GEOCHEMICAL EVIDENCE FOR GAS OF MIXED ORIGIN**

As we know, the mixed gas, commonly found in nature, can be a mixture of abiogenetic and organogenetic gases, or a mixture oil-associated gas and coal-derived gas, or even a mixture of gas generated in different maturation periods of source rock and/or fractionations during dissipation. But how can we identify them? The following data testify our hypothesis that the gases are mixed in different sources and generation stages.

**Carbon Isotopic Composition of Hydrocarbon Gases Heavier than Long Chain n-Alkane**

The carbon isotopic composition of ethane and propane varies from $-23 \times 10^{-3}$ to $-31 \times 10^{-3}$ and...
mostly in the range between $-24 \times 10^{-3}$ and $-26 \times 10^{-3}$ (Table 2). The theoretical study of fractionation is very complex and the simple model shows that the light isotope involves less energy of association than does the heavier one because the breakage of a C$^{12}$ - C$^{13}$ requires more energy than that of a C$^{12}$ - C$^{12}$ (Bordernave, 1993). As a result, the fractionation-based carbon isotopic composition, the $\delta^{13}$ C values of the liquid hydrocarbon, should be heavier than that of the hydrocarbon gases. Actually, the carbon isotopic ratio of long chain $n$-alkane ($> nC_{11}$) ranges normally between about $-28 \times 10^{-3}$ and $-29 \times 10^{-3}$ (Fig. 5). They are lighter than the hydrocarbon gases in carbon isotopic composition, indicating the multi-source for gas generation in this region.

Figure 5. $\delta^{13}$ C values of $n$-alkanes (above nC$^{11}$) in condensate of Kekeya gas field.

Hydrocarbon Gases not in Agreement with Positive Carbon Isotopic Series ($\delta^{13}$C$_{i} < \delta^{13}$C$_{j} < \delta^{13}$C$_{k} < \delta^{13}$C$_{l}$)

In general, the organogenic alkane gas is of positive carbon isotope series, i. e., $\delta^{13}$C$_{1} < \delta^{13}$C$_{2} < \delta^{13}$C$_{3} < \delta^{13}$C$_{4}$. The contents of alkane gas components decrease successively with the increase of carbon numbers in molecules, that is, CH$_{4} >$ C$_{2}$H$_{6} >$ C$_{3}$H$_{8} >$ C$_{4}$H$_{10}$. When gases of different origins are mixed, the original order $\delta^{13}$C$_{1} < \delta^{13}$C$_{2} < \delta^{13}$C$_{3} < \delta^{13}$C$_{4}$ of the mixed gas will be broken, resulting, in part, in reverse or total reverse of order. Figure 4 and Table 2 show $\delta^{13}$ C values of hydrocarbon consist of C$_{1}$ - C$_{4}$. The $\delta^{13}$C value of propane is above the $\delta^{13}$C value of butane in most situations, showing that the gas may be derived from the mixture of different organic matters (OM) or mixture of gases in different maturation periods.

Different Maturities of Gases Testified by Chemical Composition and Carbon Isotopic Composition

In conventional reservoir, such wet gases as ethane, propane and butane were formed primarily in the oil window, whereas methane was formed throughout the entire sedimentary column from diagenesis to metagenesis (Hunt, 1996). Therefore, there should be no more wet gases in high maturity. Based on a data bank, Berner (1989) proposed that the contents of methane, ethane and propane are related to maturity (R$^{o}$, %) for the thermal genetic gas derived from I. II organic matters.

Methane (%) = 9, 1ln R$^{o}$ + 93, 1
Ethane (%) = -6, 3ln R$^{o}$ + 4, 8
Propane (%) = -2, 9ln R$^{o}$ + 1, 9

Although these relations are not accurate, they still give us a hint to estimate roughly the maturity of natural gas based on the composition. R$^{o}$ value can be calculated based on the data of natural gas composition (Table 1). The calculative R$^{o}$ value is in the range of 0.3 % - 1.0 %, and most of R$^{o}$ values are below 0.6 %. This is obviously not real because other geological and geochemical data are not in agreement with this assumption. The carbon isotopes of hydrocarbon gases and biomarker analysis of coexisted light oil all show the relatively high maturity.

Many efforts have been made to interpret plots of $\delta^{13}$ C versus indices of maturity (vitrinite reflectance, R$^{o}$) and to delineate organic sources and identify source rocks.

The maturity of natural gas can be roughly estimated and calculated, using carbon isotope values of methane, ethane and propane etc. Many authors proposed the empirical equation. Dai (1993) proposed the following isotope/maturity relationship based on statistical results

$$\delta^{13}C_{i}(10^{-3}) = 15, 8\log R^{o} - 42, 2$$ (natural gas derived from sapropelic OM)
$$\delta^{13}C_{i}(10^{-3}) = 14, 12\log R^{o} - 34, 39$$ (natural gas derived from humic OM)

Stahl and Koch (1974) established the empirical relationship between carbon isotope and vitrinite reflectance R$^{o}$. Faber (1987) made modifications of the equation for the natural gas derived from type I/II OM

$$\delta^{13}C_{i}(10^{-3}) = 15, 4\log R^{o} - 41, 3$$
$$\delta^{13}C_{i}(10^{-3}) = 22, 6\log R^{o} - 32, 2$$
$$\delta^{13}C_{i}(10^{-3}) = 20, 9\log R^{o} - 29, 7$$

Berner (1989) proposed the isotope/maturity relationship for type III OM as follows

$$\delta^{13}C_{i}(10^{-3}) = 3, 01\log R^{o} - 31, 2$$
$$\delta^{13}C_{i}(10^{-3}) = 3, 32\log R^{o} - 25, 9$$

Therefore, we can calculate and estimate the maturity of gas based on their carbon isotopic compo-
sition. The calculated $R_o$ values can be seen in Table 3 and several deductions can be drawn. (1) Although the equations proposed by Dai (1993) have some differences from the equations by Faber and Berner, the calculated results show the natural gases, in accordance with the relationship of sapropelic OM, are far from real situations by using type II isotope/maturity equations, indicating that the gases are derived from either sapropelic OM or cracking of oil. (2) The chemical compositions show different maturity trends different from isotopic composition. What is the reason for this? The authors believe that the two reasons may be attributed to this. One is related to the mixture of gases from different sources or mixture of gases from different generations. The other reason may be related to the diffusion of fractionation.

Actually we can get more messages based on the relationship between carbon isotopic compositions of methane and ethane (Fig. 6). Overall, the carbon isotopic composition of gases in this region follows the linear equation proposed by Faber (1987). This phenomenon reveals that this empirical formula can be better used to reflect the real maturity of gas. From Fig. 6, it can be seen that the gases in Well K2 and K18 have lower maturity and most gases are generated from source rock at the maturity level of $R_o = 1.8 \%$ because they have similar $\delta^{13}C_2$ values. The carbon isotopic composition of methane varies from $-43 \times 10^{-3}$ to $-36 \times 10^{-3}$, which is related to the mixture of gases of different generations. The gases in Wells K410, K18, K243, K8, K401, K8101, K103 deviate from the straight line of the empirical equation and have lower $\delta^{13}C_1$ values because of the mixture of gases from the lower maturity. Based on statistical data (Whiticar, 1998) the fractionation of carbon isotopic composition will lead to carbon isotopic composition of methane that varies with maturity. The $\delta^{13}C_1$ value is about $-42 \times 10^{-3}$ at $R_o = 0.9 \%$, $-40 \times 10^{-3}$ at $R_o = 1.2 \%$, $-37 \times 10^{-3}$ at $R_o = 2.0 \%$, respectively. Therefore, these gases may be derived from mixture of gases whose $R_o = 0.9 \%- 1.2 \%$ and sometimes that have a higher maturity ($R_o = 1.8 \%$). Another point which we should bear in mind is that the difference of carbon isotopic composition of methane and ethane cannot be used for maturity calculation in the situation of mixing process. In summary, the gas in this region can be estimated by using the carbon isotopic composition of ethane, and the vitrinite reflectance value ($R_o$) of source rock may be in the range of $1.8 \%- 2.2 \%$. There is a gas mixing process in some wells from gases mentioned above with relatively low mature gases of $10.9 \%- 1.2 \%$ ($R_o$).

Similarly, the carbon isotopic composition of ethane and propane can also be used for the identification of the mixing of gases (Fig. 7). In addition, the gases in Wells K2 and K18 have lower maturity with the same source in Fig. 6 and Fig. 7. But it can also be seen that there is a difference for the data point which deviates from the empirical line equation and that is typically a situation of mixing. This is easily understood because the mixing proportions and the composition of hydrocarbons are both different. From Fig. 7, it can be seen that gases in K701, K103, K516 and K428 may be also mixed from different sources. Because the difference in content be-
between methane and ethane varies in different sources, the carbon isotopic compositions of methane and ethane are preferred for us to identify the mixing process of gases. Although it is difficult to know the exact proportion of mixing gas, it can be seen that the main gas is derived from more mature source rocks.

40 Ar/36 Ar and 3 He/4 He Ratios Varying from Gas Samples

The 3 He/4 He and 40 Ar/36 Ar ratios are indirect evidences for the natural gas origin. The 3 He/4 He value, different with the position of mantle and greater than 1.4 × 10⁻⁶, indicates that a considerable amount of helium in given natural gas comes from the

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<td>-25.80</td>
<td>-24.70</td>
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<td>1.92</td>
<td>1.73</td>
<td>0.01</td>
<td>1.07</td>
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<tr>
<td>K243</td>
<td>-41.48</td>
<td>0.31</td>
<td>1.11</td>
<td>-26.15</td>
<td>-24.79</td>
<td>0.97</td>
<td>1.85</td>
<td>1.72</td>
<td>0.00</td>
<td>0.84</td>
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<tr>
<td>K401</td>
<td>-39.16</td>
<td>0.46</td>
<td>1.56</td>
<td>-25.32</td>
<td>-23.87</td>
<td>1.38</td>
<td>2.02</td>
<td>1.90</td>
<td>0.00</td>
<td>1.50</td>
<td></td>
</tr>
<tr>
<td>K410</td>
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<td>0.23</td>
<td>0.85</td>
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<td>-24.58</td>
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<td>1.87</td>
<td>1.76</td>
<td>0.00</td>
<td>0.89</td>
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Note: 1). The calculated Rₑ values based on the equation of δ¹³Cᵢ with Rₑ for type Ⅲ OM by Dai (1993); 2). The calculated Rᵥ values based on the equation of δ¹³Cᵢ with Rᵥ for type Ⅰ / Ⅱ OM by Dai (1993); 3). The calculated Rₑ values based on the equation of δ¹³Cᵢ with Rₑ for type Ⅰ / Ⅱ OM by Faber (1987); 4). The calculated Rᵥ values based on the equation of δ¹³Cᵢ with Rᵥ for type Ⅰ / Ⅱ OM by Faber (1987); 5). The calculated Rₑ values based on the equation of δ¹³Cᵢ with Rₑ for type Ⅰ / Ⅱ OM by Faber (1987); 6). The calculated Rᵥ values based on the equation of δ¹³Cᵢ with Rᵥ for type Ⅲ OM by Berner (1989); 7). The calculated Rₑ values based on the equation of δ¹³Cᵢ with Rₑ for type Ⅲ OM by Berner (1987).

Figure 8. ⁴⁰Ar/³⁶Ar distribution in gas of Kekeya region.

volatile component of mantle. ⁴⁰Ar/³⁶Ar ratio in the mantle is one of the crucial indices in the model of earth degassing. Statistical results show that ⁴⁰Ar/³⁶Ar ratio in the upper mantle, as high as 2 000, can serve as an evidence of abiogenic gas from the deep earth after neglecting the influence of the chronologic accumulation of crust radiogenic ⁴⁰Ar and those of other factors. Besides, different models of mantle degassing will lead to the various ⁴⁰Ar/³⁶Ar ratio in the mantle. In the short-term fast-degassing model, the ⁴⁰Ar/³⁶Ar ratio is 5−6; in the long-term and slowly-degassing model, it is about 2 (Wang et al., 1997).

Furthermore, helium and argon in hydrocarbon source rocks possess a chronologic accumulative effect. The ⁴⁰Ar/³⁶Ar values would increase with the aging of source rocks. Similarly, ³He/⁴He decreases with the aging of source rocks. This difference means that the age of the hydrocarbon source rocks may be estimated by ⁴⁰Ar/³⁶Ar, ³He/⁴He values in natural gas (Xu, 1999).

It can be seen from Table 4 that the natural gas in Shaya region, northern part of Tarim basin is remarkably higher than that in Kekeya region. The ⁴⁰Ar/³⁶Ar value distribution of natural gas in Kekeya region can be divided into two subgroups (Fig. 8). The first subgroup is in the range of 855.4−981.6, and the second one is in the range of 604.6−682.4, indicating various sources for the generation of gas.
Table 4  $^{40}$Ar/$^{36}$Ar and $^3$He/$^4$He ratios for gas in Tarim basin

<table>
<thead>
<tr>
<th>region</th>
<th>No.</th>
<th>age</th>
<th>$^{40}$Ar/$^{36}$Ar</th>
<th>$^3$He/$^4$He</th>
<th>R/Ra</th>
<th>$\delta$D$_{CH_4}$/10$^{-3}$</th>
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<tr>
<td>Ke18</td>
<td>N1</td>
<td></td>
<td>7.9(7.3*)×10$^{-8}$</td>
<td></td>
<td>-170</td>
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<td>Ke2</td>
<td>N1</td>
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<td>0.04</td>
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<td>Ke243</td>
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<td></td>
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<td></td>
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<tr>
<td>Ke351</td>
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<td></td>
<td>718, 8</td>
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<tr>
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<tr>
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<td>N1</td>
<td>604</td>
<td></td>
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<tr>
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<td>N1</td>
<td>768</td>
<td></td>
<td></td>
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<tr>
<td>Ke701</td>
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<tr>
<td>Ke8</td>
<td>N1</td>
<td>982</td>
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<td>0.06</td>
<td>-167</td>
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<tr>
<td>Hetianhe</td>
<td>Ma2</td>
<td>C</td>
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<td>O</td>
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<tr>
<td>Ma4</td>
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<tr>
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<td>Shacan 7</td>
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<td></td>
<td>2 334</td>
<td>2.2</td>
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</table>

CONCLUSIONS

The aim of this investigation was to address the main origin of gas in Kekeya region by utilizing the carbon isotopic composition of light hydrocarbons. The following conclusions have been drawn. (1) The carbon isotope of gas in Kekeya field is a little heavier than that of typical marine-derived gas. The relationship in carbon isotopes between methane and ethane is coincident with that of the gas derived from organic matter I and II. (2) Most maturity of gas is estimated at 1.8 %−2.2 %, besides K2 and K18 wells based on the formula. In addition, the gas derived from 0.9 %−1.2 % Rm source rocks may also be mixed. (3) Many geochemical evidences support that the gas is mixed. However, the gas in this region is mainly generated from more mature source rocks.

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