



Original research paper

Further discussion about the origin of natural gas in the Sinian of central Sichuan paleo-uplift, Sichuan Basin, China[☆]

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Abstract

The origin of Sinian natural gas in the paleo-uplift in the central Sichuan is controversial. The major geo-chemical evidence to prove the gas origin was studied again, and it indicates that: the relationship of the Cambrian and Sinian ethane carbon isotope and ethane content complies with the Rayleigh distillation model, hence, the differences in the $\delta^{13}\text{C}_2$ was not due to the different parent materials. As for the Sinian methane, the δD_1 abnormally decreases with the increase of the $\delta^{13}\text{C}_1$. This can be caused by the isotopic exchange with the formation water at an extremely high maturation stage rather than the different water salinities of depositional environment of their source rocks. Meanwhile, for the Sinian bitumen, the alkyldibenzothiophene molecular ratios 4-/1-MDBT is higher in the southwest and lower in the northeast, showing the hydrocarbon laterally migrated from the aulacogen Cambrian Qiongzhusi shale. The C_7 light hydrocarbon was dominated by methylcyclohexane and a lack of toluene, demonstrating that the gas is from oil cracking instead of carbonate source rock. Therefore, the point that there is significant contribution from the Sinian source rock lacks evidence at present, and the major contribution of the Sinian gas is from the aulacogen of the Cambrian shale.

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Keywords: Gas origin; Carbon isotope; Hydrogen isotope; Sinian; Cambrian; Central Sichuan paleo-high

1. Introduction

Ever since the Weiyuan Gas Field was discovered, the origin of natural gas in the Sinian of central Sichuan paleo-uplift has been controversial. In the past few years, natural gas exploration in the Sinian has gradually made significant breakthroughs such as verifying two super-giant integrated gas fields (Gaoshiti and Moxi). The controlled gas-bearing area in the Dengying Formation is 7500 km², with an expected 3P reserves of more than 1×10^{12} m³. The origin of natural gas in

the Sinian and formation model of gas fields have a close relationship with next exploration directions, thus, they must be studied in detail.

After the discovery of the two super-giant gas fields, namely, Gaoshiti and Moxi, as there are some differences between the natural gas in the Sinian Dengying Formation and Cambrian Longwangmiao Formation, some scholars consider that the natural gas in the Dengying Formation is chiefly generated and stored inside the Sinian system [1,2], or the Sinian source rocks have an important contribution to the Dengying Formation gas source [3,4]. The major evidence include: (1) the carbon isotope values of ethane ($\delta^{13}\text{C}_2$) in the Dengying Formation and Longwangmiao Formation are apparently different, which mainly reflects different parent materials; (2) the hydrogen isotope values of methane ($\delta^2\text{H}_{\text{CH}_4}$) are much different, which chiefly reflects different water salinity values in parent materials; (3) the alkyldibenzothiophene molecular ratios 4-/1-MDBT of bitumen in the

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Dengying reservoir are between that of the Dengying Formation and the Sinian source rocks, being the mixture of the Cambrian bitumen with lower maturity and the Sinian bitumen with higher maturity. We disagree with the overhead presented evidence, and consider that the evidence for gas generation in the Sinian source rocks is controversial.

2. The origin of carbon and hydrogen isotopes in natural gas

The carbon isotope values of methane ($\delta^{13}C_1$) are similar in the Sinian and Cambrian systems in the paleo-uplift of the central Sichuan Basin (the average $\delta^{13}C_1$ value in the Sinian system is only 0.3‰ higher than that of the Cambrian system), but there are apparent differences between the $\delta^{13}C_2$ and $\delta^2H_{CH_4}$ (Table 1), which is regarded as the most essential evidence for gas generation in the Sinian source rocks. However, the overall evaluation regularity of carbon and hydrogen isotopes in the over-mature gas in the Sichuan Basin is still debatable. Firstly, for the $\delta^{13}C_2$ values in the Gaoshiti and Moxi regions, there's more than -29.5‰ in the Sinian system (average -28.1‰), while there's less than -32.3‰ (average -33.4‰) in the Cambrian system. The carbon isotope of ethane is regarded as having a good parent material inheritance [5], thus, it is the commonly used index for identifying gas sources. The apparent difference between them is easily regarded as the difference of the parent materials. However, the recent explorations have results of shale gas being at the high and over-mature stages, show that there may be some problems in applying carbon isotope of ethane to identify gas sources in over-mature stage [6].

As the natural gas maturity increases, the natural gas wetness ($Wetness = \Sigma C_2 - C_5 / \Sigma C_1 - C_5$) decreases gradually. For oil-type gas, the $\delta^{13}C_2$ values can undergo three processes:

normal increase during initial cracking stage of kerogen ($R_o < 1.3\%$ or 1.5%), inverted decrease during secondary cracking stage (R_o between 1.3% and 2.2%) [6,7], and a quick increase in the Rayleigh distillation during the over-mature stage ($R_o > 2.2\%$). Heavy hydrocarbons are almost cracked. Ethane is only cracked, without newly generated ethane [8,9] (Fig. 1). Laser Raman R_o values of the residual bitumen in the Qiongzhusi Formation in the Gaoshiti well field are larger than 3.0% , which apparently indicates that it has entered the Rayleigh distillation stage. Due to the activation energy differences, ^{12}C is favorably cracked, and the carbon isotope values of the remaining ethane increase gradually alongside the decrease of ethane content.

To solely consider ethane cracking, the carbon isotope values of ethane satisfy the following equation [10]:

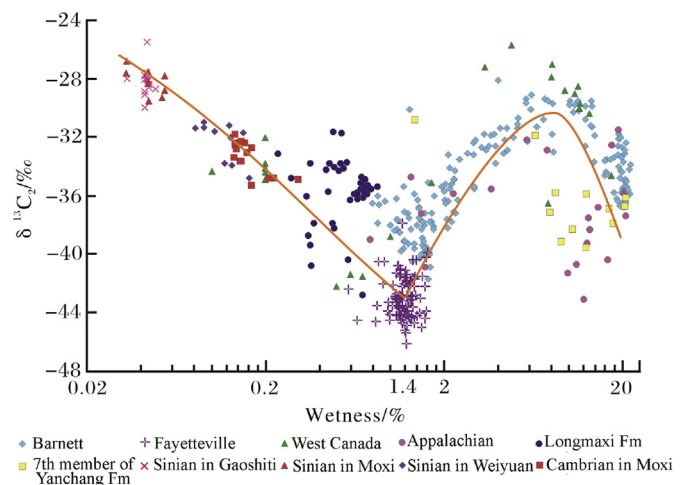


Fig. 1. The relationship between the $\delta^{13}C_2$ and wetness in gasses with different maturities [1,3,6,8,9,11–16].

Table 1
Natural gas components and isotope in the paleo-uplift of the central Sichuan Basin [1–4,6].

Well name	Strata	Depth/m	Major components of natural gas					$\delta^{13}C/\text{‰}$ (VPDB)		$\delta^2H_{CH_4}/\text{‰}$ (VSMOW)
			CH ₄	C ₂ H ₆	CO ₂	H ₂ S	N ₂	CH ₄	C ₂ H ₆	
Gaoshi 1	Upper Deng 4	4956–5195	91.22	0.04	6.35	1	1.36	-32.3	-28.1	-137
Gaoshi 2	Deng 2	5300–5390	82.65	0.04	14.19	0.85	2.12	-32.3	-28.7	-137
	Upper Deng 4	5018–5020	92.61	0.04	6.41	0.83	0.03	-33.74	-28.02	-137
Gaoshi 3	Deng 2	5799–5810	89.39	0.03	8.2	1.57	0.76	-32.6	-28	-149
Gaoshi 6	Deng 4	4958–5210	94.72	0.04	3.99	0.2	1.02	-33.78	-28.06	-140
Moxi 8	Deng 4	5107–5172	91.4	0.04	5.87	0.96	1.66	-32.8	-28.3	-147
Moxi 9	Deng 2	5421–5459	91.42	0.04	6.01		2.46	-32.3	-27.5	-147
	Deng 4	5423–5459	91.82	0.05	4.24	2.75	0.96	-33.5	-28.8	-141
Moxi 10	Deng 2	5759–5165	92.03	0.05	4.64	2.4	0.86	-33.9	-27.8	-139
Moxi 11	Deng 4	5148–5208	90.49	0.03	7.05	0.91	1.45	-33.9	-27.6	-138
Wei 39	Deng 2	5455–5486	89.87	0.03	7.32		2.32	-32	-26.8	-150
	Deng 4	2833–2986	84.94	0.11	4.73	1.04	8.86	-32.42	-33.91	-142
Wei 100	Deng 2	2959–3041	86.9	0.13	5.07	1.18	6.47	-32.38	-31.82	-136
Zi 1	Deng 2	3944–4044	93.59	0.12	4.31	0.75	1.22	-37.1		
Wei 201-H3	Qiongzhusi	3642–3655	96.52	0.35	1.24		1.75	-35.4	-40.8	-145
Moxi 8	Longwangmiao	4697–4713	96.85	0.14	1.78		0.6	-33.1	-33.6	-133
Moxi 9	Longwangmiao	4575–4607	95.16	0.13	2.35		2.35	-32.8	-32.8	-134
Moxi 10	Longwangmiao	4646–4697	97.64	0.14	2.1		0.1	-32.1	-33.6	-134
Moxi 11	Longwangmiao	4674–4681	97.09	0.13	2.04		0.67	-32.5	-32.4	-132

$$R_{s,t} = R_{s,0} f^{(\alpha-1)} \quad (1)$$

Wherein the $R_{s,t}$ is the absolute carbon isotope value of ethane after cracking; $R_{s,0}$ is the initial absolute carbon isotope value of ethane; f is the percentage of reaction residues (remaining ethane content/initial ethane content), $f = C_{2,t}/C_{2,0}$, α is isotope fractionation factor.

To change Equation (1) to be in the $\delta^{13}C_2$ form, which is more commonly used, take reference in the following:

$$10^{-3}(\delta^{13}C_{2,t} + 1)R_{\text{standard}} = 10^{-3}(\delta^{13}C_{2,0} + 1)R_{\text{standard}} f^{(\alpha-1)} \quad (2)$$

To eliminate R_{standard} in the Equation (2), calculate the natural logarithm of its two sides to get the following equation:

$$(\alpha - 1)\text{Ln}f = \text{Ln}[(10^{-3}\delta^{13}C_{2,t} + 1)/(10^{-3}\delta^{13}C_{2,0} + 1)] \quad (3)$$

If x is smaller, $\text{Ln}(1 + x) \approx x$, the Equation (3) can be approximately simplified to the following equation:

$$1000(\alpha - 1)\text{Ln}(C_{2,t}/C_{2,0}) = \delta^{13}C_{2,t} - \delta^{13}C_{2,0} \quad (4)$$

To change Equation (4) to the following equation:

$$\delta^{13}C_{2,t} = \delta^{13}C_{2,0} - 1000(\alpha - 1)\text{Ln}C_{2,0} + 1000(\alpha - 1)\text{Ln}C_{2,t} \quad (5)$$

Equation (5) means that during the Rayleigh distillation stage, there is a linear relationship between the $\delta^{13}C_{2,t}$ and the natural logarithm of the remaining ethane content ($\text{Ln}C_{2,t}$). We analyzed the published data of the Sinian and Cambrian systems in the central Sichuan Basin [1–3]. We drew the relationship graph between the $\delta^{13}C_2$ and $\text{Ln}(\text{mol}\%C_2H_6)$ (Fig. 2). We found out that the relationship between the carbon isotope and the content of ethane in the Sinian Dengying Formation and Cambrian Longwangmiao Formation in the central Sichuan paleo-uplift is as follows:

$$\delta^{13}C_2 = -40.63 - 3.822 \text{Ln}(\text{mol}\%C_2H_6), R^2 = 0.876 \quad (6)$$

Equation (6) complies with the idealized mathematical model of the Rayleigh distillation (Equation (5)). Assuming

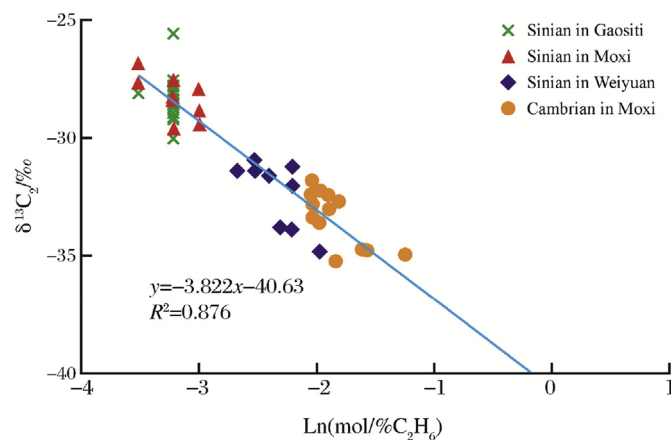


Fig. 2. The relationship between the $\delta^{13}C_2$ and $\text{Ln}(\text{mol}\%C_2H_6)$ of gas in the Sinian-Cambrian of the central Sichuan paleo-uplift [1–4].

Rayleigh distillation started when the ethane content in the Sinian and Cambrian systems was 1.4%, it can be inversely calculated that the $\delta^{13}C_{2,0}$ was -41.92‰ during the initial cracking, which is already very close to the average breakpoint in shale gas observation (Fig. 1). The Rayleigh distillation equation of a deep basin gas is related to the Utica shale in the Appalachian Basin, and it can be approximately expressed as follows [9]:

$$\delta^{13}C_2 = -45.26 - 15\text{Ln}(\text{mol}\%C_2H_6) \quad (7)$$

We consider that the Rayleigh distillation of natural gas that is related to Utica started when ethane content was 1.2% and the $\delta^{13}C_{2,0}$ was -48‰ [9], which means that the $\delta^{13}C_{2,0}$ values are different in various shales.

As the mathematical relationship between carbon isotope and content of ethane in the Cambrian and Sinian systems of the study area accord with the Rayleigh fractional principle, statistics show that the correlation coefficient is high. The deduced formula and the current results of shale gas exploration can also be verified mutually. We have more reasons to believe that the difference in the $\delta^{13}C_2$ values of natural gas in the Sinian and Cambrian systems is caused by ethane cracking. During the initial cracking stage, the $\delta^{13}C_{2,0}$ values in the Sinian and Cambrian systems are almost the same, thus, it can be concluded that they came from the same gas source; hence, the distinct difference is that the Sinian ethane was cracked more efficiently, thus, the current ethane content is much less, and the difference of parent materials is not the major reason.

Secondly, let's discuss the reason of hydrogen isotopic difference of methane. Some researchers [1–3] think that the hydrogen isotopic difference of methane mainly reflects the difference in water salinities of the corresponding source rock depositional environment. The $\delta^2H_{CH_4}$ values of natural gas in the Longwangmiao Formation is more than -138‰ (average -134‰), and that in the Dengying Formation is less than -135‰ (average -141‰), which means that there was apparently a different depositional water environment of source rocks, and they came from different gas sources. This explanation seemed plausible, but it neglects one occurrence: the $\delta^2H_{CH_4}$ values inside the Sinian system decrease gradually with the increase of the $\delta^{13}C_1$ values. In the Cambrian system, within the Moxi region, the $\delta^2H_{CH_4}$ values already have a decreasing trend with the increase of maturity; to the Sinian system (especially the 2nd member of the Dengying Formation), this trend becomes more seemingly. The overall trend of the $\delta^2H_{CH_4}$ values is summarized as follows: the 2nd member of the Dengying Formation < the 4th member of the Dengying Formation < the Longwangmiao Formation (Fig. 3), which is opposite to the trend of thermal evolution degree. The difference of water salinities could not lead to the event that the $\delta^2H_{CH_4}$ values inside the Sinian system decrease with the increase of $\delta^{13}C_1$ values.

The abnormal phenomenon, the $\delta^2H_{CH_4}$ values decrease with the increase of $\delta^{13}C_1$ values, disaccords with the normal principle of isotopic thermodynamic distillation. Such

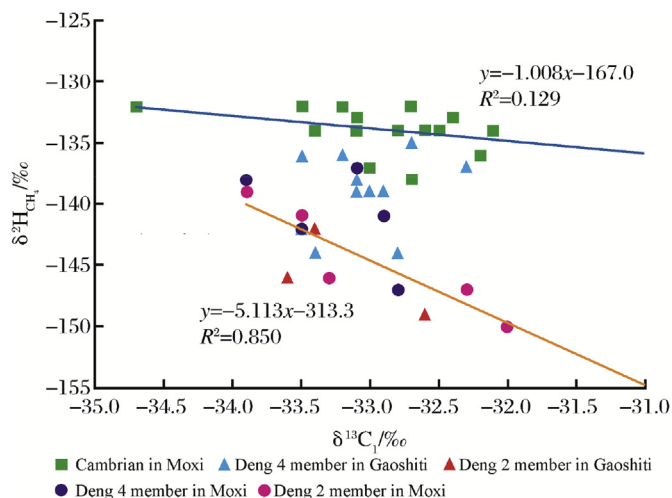
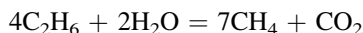


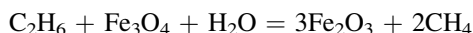
Fig. 3. The relationship between the $\delta^{13}\text{C}_1$ and $\delta^2\text{H}_{\text{CH}_4}$ of gas in the Sinian-Cambrian of the central Sichuan paleo-uplift.

phenomenon can be observed in the natural gas that has an abnormally high thermal evolution degree such as the shale gas in part the Longmaxi Formation of the Sichuan Basin ($R_o > 2.5\%$) [6,8] and deep basin gas in the Appalachian Basin [9]. This is considered to be the result of a special reaction that involved water during the later period.

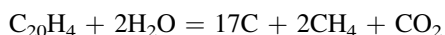
The ethane cracking that has been discussed above can occur in the following reaction:



If there is transition metal catalysis, the following reaction is more likely to occur:



There is a great deal of anthracolite in the central Sichuan paleo-uplift. Such hydrogen deficient organic matter at the anthracite rank can react with water during the later period:



All the methane generated in the three mentioned reactions [9] is from the “hydrogen source” in the formation water. The higher the thermal evolution degree, the more special methane there is. Hence, the $\delta^2\text{H}_{\text{CH}_4}$ values can decrease in such an “abnormal” style.

There are two vital differences between carbon and hydrogen in natural gas in the Cambrian and Sinian systems in the paleo-uplift of central Sichuan that are related to the cracking process that involves water. Experiments in laboratories also prove that water can promote hydrocarbon cracking [17]. The thermal evolution degree of the Sinian system in this study area is higher than that of the Cambrian system; moreover, water saturation in the Sinian reservoir in this area is higher than that in the Longwangmiao Formation in the chief part of the Moxi region. The Connate water was exclusively found in the Longwangmiao Formation in some areas of the Moxi 203 and Moxi 204 well fields. On the other hand, water was only found in the low areas of the gas reservoir periphery

in the 4th member of the Dengying Formation (Well Moxi 22, 5243 m). Meanwhile, water is widely contained in the lower part of the gas reservoir in the 2nd member of the Dengying Formation (gas/water contact is 5167.5 m in the Moxi region and 5170 m in the Gaoshiti region; both are higher than the trap lines). Gas-water contact in the 2nd member of the Dengying Formation is the most adequate, thus, its cracking during the later period has a higher evolution degree, which can cause less ethane content in the Sinian system, heavier carbon isotopic composition of ethane, and an abnormal decrease of hydrogen isotope values of methane.

3. Gas source correlation

There are two major proofs that the source rock in the Dengying Formation contributes greatly to gas source specifically for the component correlation of carbon isotopes and aromatic parameters of bitumen.

The carbon isotope values of bitumen in the Sinian reservoir in the central Sichuan Basin are between -34.5% and -37.0% (average -35.7%), and the bitumen is rich with ^{12}C . Through the comparison of this feature with the carbon isotope values of the kerogen in dolomite in the Sinian Dengying Formation (between -31.5% and -28.8% , average -30.7%), mudstone in the third member of the Dengying Formation (between -33.5% and -29.0% , average -31.9%), and shale in the Cambrian Qiongzhusi Formation (between -36.8% and -29.9% , average -32.8%) some former researchers inferred that they mainly come from the Cambrian Qiongzhusi Formation [18,19]. However, as the $\delta^{13}\text{C}_2$ values of the Sinian gas grows larger than the $\delta^{13}\text{C}_{\text{kerogen}}$ values in the Qiongzhusi Formation, according to ordinary isotope distillation principle, such natural gas is regarded to be from the dolomite kerogen being rich in ^{13}C in the Sinian system [3]. Just as discussed above, due to the super high evolution degree, ethane only enters the Rayleigh distillation stage once cracked, without newly generated ethane. Hence, the $\delta^{13}\text{C}_2$ values at that time were not controlled by the carbon isotope values of kerogen or bitumen but mainly controlled by the secondary distillation degree of the cracked isotope itself. Consequently, the relationship between the $\delta^{13}\text{C}_2$ and $\delta^{13}\text{C}_{\text{kerogen}}$ values cannot be used as the index for gas source correlation. During super high thermal evolution stage, even the $\delta^{13}\text{C}_1$ values may be greater than the $\delta^{13}\text{C}_{\text{kerogen}}$ values [6], the correlation of carbon isotope values between gas and kerogen is not the most effective evidence, and its assurance level is lower than the correlation of carbon isotope values between bitumen and kerogen.

The DBTs compounds can act as the parameters of the bitumen maturity. The 4-MDBT/1-MDBT and 4,6-DMDBT/1,4-DMDBT are commonly used as indexes [20]. Taking 4-MDBT/1-MDBT ratio as the example, the ratios in the study area are overall featured with: the Longwangmiao Formation < the Qiongzhusi Formation < 4th and 2nd members of the Dengying Formation < 3rd member of the Dengying Formation (Table 2). Wei et al. [3] thought that this feature is caused by the mixture of the hydrocarbon source in

Table 2
The DBTs' bitumen and source rock parameters in the central Sichuan paleo-uplift [3].

Well name	Strata	Depth/m	Lithology	4-MDBT/1-MDBT	4,6-DMDBT/1,4-DMDBT
Gaoshi 6	Longwangmiao Fm	4553.3	Bituminous dolarenite	3.24	1.52
Moxi 11	Longwangmiao Fm	4885.4	Bituminous dolomite	2.94	1.19
Gaoshi 1	Deng 4 member	4975.7	Bituminous dolomite	4.40	1.40
Gaoshi 6	Deng 4 member	5034.62	Bituminous dolomite	4.42	1.37
Gaoke 1	Deng 4 member	5026.55	Bituminous dolomite	4.57	1.73
Moxi 8	Deng 4 member	5109.55	Bituminous dolomite	3.94	1.59
Moxi 8	Deng 4 member	5113.32	Bituminous dolomite	4.43	1.62
Gaoshi 2	Deng 2 member	5396.17	Bituminous dolomite	4.33	1.53
Moxi 9	Deng 2 member	5461.2	Bituminous dolomite	3.82	1.42
Moxi 10	Deng 2 member	5470.7	Bituminous dolomite	3	1.68
Moxi 10	Deng 2 member	5459.2	Bituminous dolomite	3.93	1.43
Moxi 11	Deng 2 member	5482	Bituminous dolomite	3.32	1.43
Moxi 9	Qiongzhusi Fm	4965.6	Mudstone	3.87	1.39
Zi 9	Qiongzhusi Fm	4229.4	Mudstone	3.57	1.12
Gaoke 1	Deng 3 member	5343	Mudstone	3.73	1.38
Gaoke 1	Deng 3 member	5352.5	Mudstone	5.65	1.43
Hanshen 1	Deng 3 member	5129.8	Mudstone	4.77	/

the Qiongzhusi Formation with lower maturity and that in the 3rd member of the Dengying Formation with higher maturity. Even though all the bitumen in the 4th member of the Dengying Formation comes from the Qiongzhusi Formation, its maturity values should be between that of the 3rd member of the Dengying Formation and the Qiongzhusi Formation. Meanwhile, considering burial depth, it should not be considered to be from the bitumen in the 3rd member of the Dengying Formation with higher maturity just because its ratio is between the aforementioned. Moreover, the 4-MDBT/1-MDBT ratios of the 2nd member of the Dengying Formation in the Well Moxi 9 and Well Moxi 10 are between 3.00 and 3.93, respectively, and they are generally less than that in the 3rd member of the Dengying Formation (3.73–5.65). This can only prove that such amount of bitumen mainly comes from the Qiongzhusi Formation with lower maturity (3.57–3.87) rather than that in the 3rd member of the Dengying Formation; it also cannot be proven that it was generated and stored in the Dengying Formation.

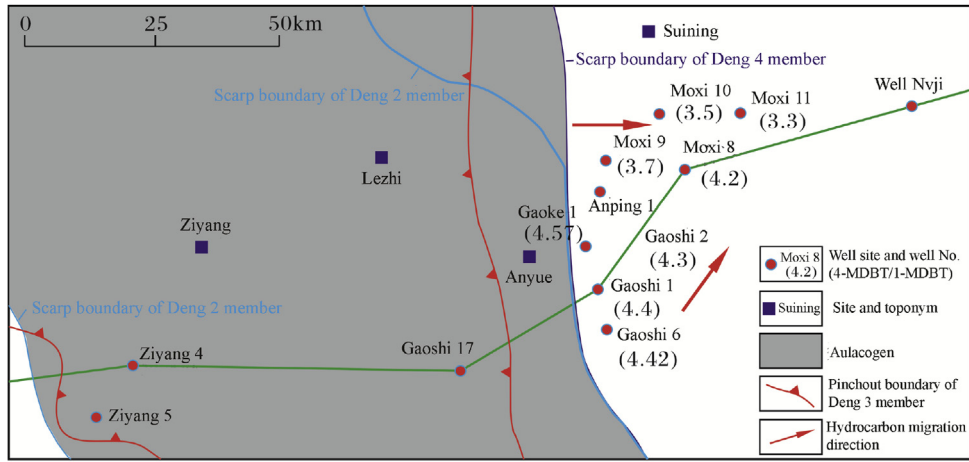
DBTs can be used to represent the maturity of organic matter and trace the charging direction of the oil reservoirs. During the period in which paleo-oil reservoirs form, the crude oil maturity decreases, which can subsequently be used to trace the pathways of the oil filling. In addition, with the increasing distance of migration, the alkylidibenzothiophene isomers can shield the migration fractionation effect of the DBTs' hydrogen bond, and it can lead to the decrease of 4-MDBT/1-MDBT ratios. Based on the two factors (thermal stability and hydrogen bond), the crude oil with higher 4-MDBTs/1-MDBT ratios is relatively closer to the sources [20]. According to planar distribution (Fig. 4a), 4-MDBT/1-MDBT ratios in the Gaoshiti-Moxi regions decrease from the southwest to the northeast; generally being proximal in the southwest and distal in the northeast. Aromatic parameters in the reservoir bitumen also reflect that the Sinian bitumen predominantly came from the Cambrian system. As large aulacogen [21] (also called erosion valley [22]) developed in

the Deyang-Anyue areas, high-quality gas source rocks in thick Qiongzhusi Formation (and Maidiping Formation) also developed in the aulacogen region. They laterally come in contact with the reservoir beds in the Dengying Formation (Fig. 4b). If the early crude oil migrated from the aulacogen laterally, the lateral and vertical distribution laws of the current 4-MDBT/1-MDBT ratios can be interpreted reasonably.

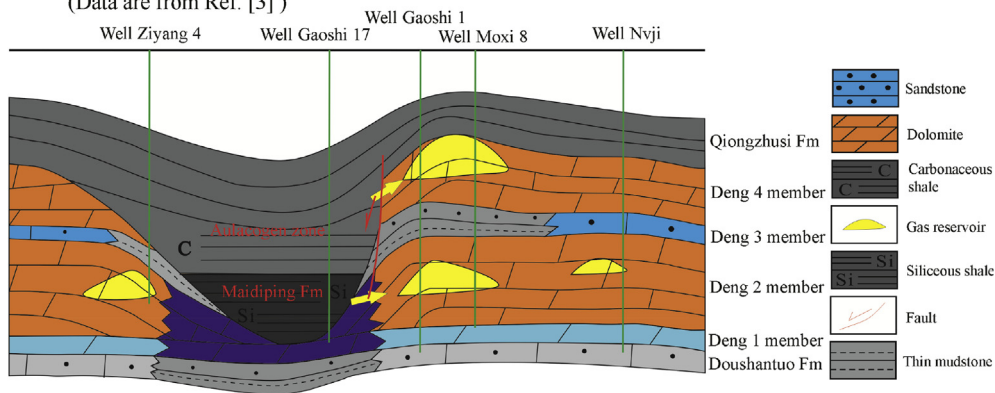
Apart from the aromatic parameters of bitumen, Cheng et al. [23] used acetic acid and hydrogen peroxide, instead of the sterane and terpane parameters of the early absorbed hydrocarbons and entrapped hydrocarbons, to conduct mild oxidation within the Sinian bitumen in the Well Gaoshi 1 and Well Anping 1. Xu et al. [24] also studied the biomarker parameters in the Well Anping 1 and Well Gaoke 1. The research results indicated that the Sinian bitumen in this area mainly comes from the Cambrian Qiongzhusi Formation.

4. Light hydrocarbons

There is no seeming difference in the C₇ light hydrocarbon in the Sinian and Cambrian systems in the paleo-uplift of central Sichuan Basin. In the triangular diagram of toluene, methyl cyclohexane, and normal heptane, most of the Sinian gas has methyl cyclohexane predominance [5]. Either the oil cracking gas [25] or the dispersal of liquid hydrocarbon cracking gas [26] have methyl cyclohexane predominance. Similar phenomena can also be found in the shale gas of the Qiongzhusi and Longmaxi Formations [8]. Therefore, it is ascertainable that the Sinian gas chiefly came from oil cracking gas [3,19]. In addition, the aromatic hydrocarbon contents in the light hydrocarbons that were generated in carbonate rocks are higher [15]. The predominant content of toluene was also observed in light hydrocarbons of the thermal simulation gas at 500 °C in carbonate rocks in the 4th member of the Dengying Formation in the Well Gaoshi 1. However, in Fig. 5, the toluene contents in wellhead gas in the Sinian system are generally less than 30%, which also means that the



(a) Planar distribution of 4-/1-MDBT ratios in Dengying Formation and hydrocarbon migration direction (Data are from Ref. [3])



(b) Well-tie profile during deposit stage of Canglangpu Formation (Its location is shown in Fig.4a)

Fig. 4. The hydrocarbon accumulation model in the central Sichuan paleo-uplift.

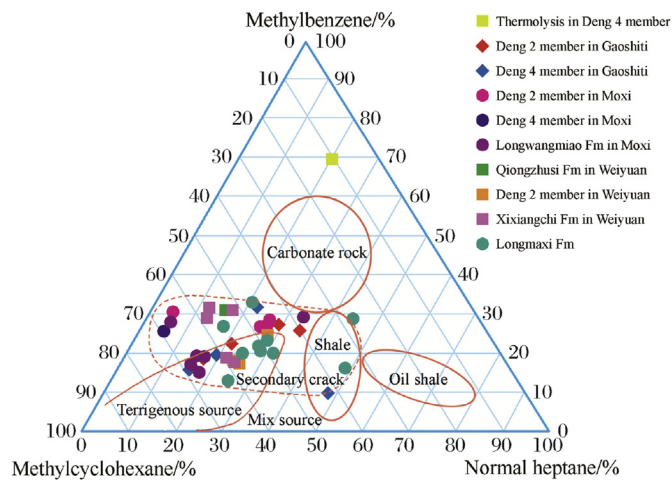


Fig. 5. The diagram of the C₇ light hydrocarbon for the gas in the Sinian-Cambrian of the central Sichuan paleo-uplift (modified from Ref. [15]).

Sinian carbonate rocks have less contribution. Mudstone only develops in the 3rd member of the Dengying Formation in the Sinian system, but its thickness and lithology are unstable. It was completely eroded in the aulacogen zone. The organic-rich shale (TOC > 0.5%) develops along the Gaoshi 1-Moxi

9 well field, with a thickness of about 20 m, and it gradually changes to muddy dolomite and sandstone eastwards (to the east of the Well Moxi 8). The gas generation scale is much less than that of the Qiongzhusi Formation. The formations that are exposed by well (Gaoshi 1, Nvji, Wei 117, Wei 28, etc.) drillings within the Doushantuo Formation are sandstones in coastal facies, or sandy and gypsum dolomite in diamicitic continental shelf facies, without further discovery of source rock, and there is an apparent change in facies within the basin periphery. Hence, light hydrocarbons are capable of proving that the Sinian gas is chiefly sourced in the Cambrian system.

5. Conclusions

The $\delta^{13}C_2$ and $\delta^2H_{CH_4}$ values of natural gas in the Sinian and Cambrian systems in the paleo-uplift of the central Sichuan Basin have evident differences. According to a long-established understanding, there are two types of natural gas coming from different sources. Whilst considering the intrinsic relationship between $\delta^{13}C_2$ values and ethane contents, the difference between the $\delta^{13}C_1$ and $\delta^2H_{CH_4}$ values in this area are more likely to be caused by various thermal evolution maturities of the same gas source. Relevant evidence from the Sinian bitumen and light hydrocarbons also prove that natural gas mainly

comes from the Cambrian Qiongzhusi Formation. It is still controversial whether the Sinian source rock has an apparent contribution to the Sinian gas source. It is necessary to further study the gas generation capacity of the Sinian source rock and its contribution to the current gas reservoirs. According to the successful cases in the Gaoshiti and Moxi regions, we believe that the Sinian oil and gas mainly comes from high-quality source rocks in the aulacogen via lateral migration. This respective hydrocarbon accumulation pattern is the priority issue in future exploration in the Sichuan Basin. However, there's no sufficient geochemical evidence for hydrocarbon accumulation pattern proving that hydrocarbons were directly expelled downward from the overlying source rock, or being self-generation and self-storage in the Sinian system.

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Conflict of interest

The authors declare no conflict of interest.

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