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**Several Basic Scientific Problems in Geochemical Studies On Deep Natural Gases**

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The traditional theory on the hydrocarbon generation by the late thermal decomposition of kerogen reveals the formation, evolution and distribution rules of conventional hydrocarbons, describes various stages of hydrocarbon generation and destruction, and consequently produces a concept of the hydrocarbon “deadline”. When thermal maturity exceeds the “deadline”, oils and natural gases will not be able to form a commercial hydrocarbon reservoir, from which an “economical deadline” in commercial exploration and development is inferred.

Deep oils and natural gases refer mainly to hydrocarbon resources that are buried below 4000m~4500m with geo-temperature over 135°C~150°C and a high thermal maturity ( $R_o > 1.35\%$ ), especially for the deep natural gas resource. Deep oils and natural gases in different basins have varying burial depths due to different geothermal gradients and thermal evolution history, therefore, the “deep oil and gas” concept should be defined as hydrocarbon resources that have high thermal maturity ( $R_o > 1.35\%$ ). The key to deep hydrocarbon resources is that the formation and evolution of deep oils and natural gases occur in a relatively close system, thus, the constraint mechanism of physicochemical conditions in a thermodynamic equilibrium system should be fully taken into account as the hydrocarbon generation and accumulation system is investigated.

Temperature still remains as an important influence factor that plays a dominant role in the evolution of organic matter in a deeply buried environment. However, the evolution of organic matter under physicochemical conditions in a relatively stable chemical thermodynamic equilibrium system at deep depth is also constrained by various factors, including the following aspects:

1. geochemical characteristics of deeply-buried source rocks and hydrocarbons;
2. the effect of temperature on the evolution of organic matter;
3. constraints of abnormal pressure on the evolution of organic matter;
4. the effect of allogenetic hydrogen on the formation of hydrocarbons;
5. catalysis of transition metal elements (V, Co, Fe, Ni, etc.);
6. thermodynamic effect in the deep gas system (studied by means of simulation experiments and calculations);

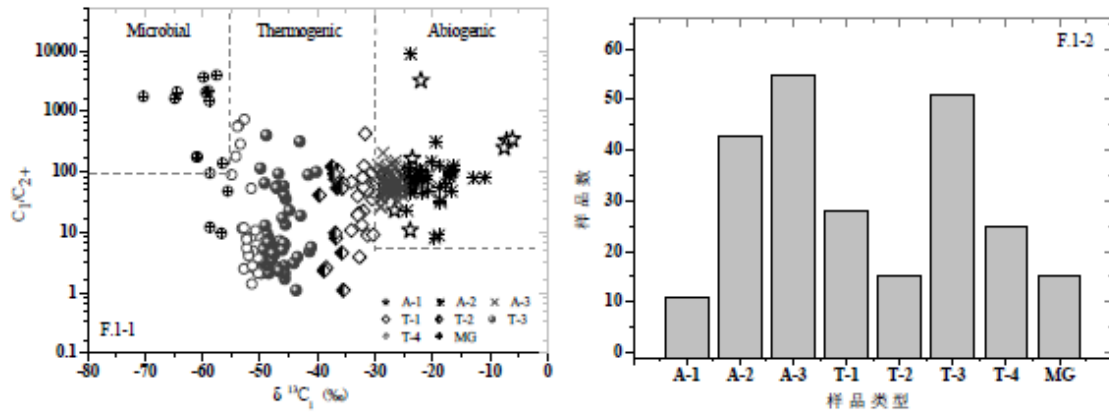
Theoretical studies on deep oils and natural gases have indicated that oils and natural gases in deeply buried environments below their traditional “economical deadline” still have a considerably high thermodynamic stability. Deep high pressure conditions constrain the thermal maturity and thermal destruction of organic matter and favor the preservation of hydrocarbons. Deep media conditions, input of allogenetic hydrogen and catalysis of transition metal elements would accelerate the generation of hydrocarbons and promote the production rate of hydrocarbons. With the increase of burial depths and the gas-to-oil ratio, a gas reservoir

dominated by methane would be finally formed. Under deep conditions, the organic matter system is accelerated toward to an equilibrium stable state and finally reaches a thermodynamic equilibrium. The evolution of organic matter (source rocks, oils and gases) and characteristics of molecular chemical compositions in the system are constrained by temperature, pressure and chemical (mineral) compositions of the system and have not correlated with burial time. Ages of source rocks and reservoirs can be ignored in thermodynamic calculations.

In addition, another key scientific problem worthy of investigation encountered in geochemical studies of deep natural gases is how to distinguish them from conventional thermogenic natural gases and abiogenic gases. Therefore, the formation mechanism of natural gases and the corresponding isotopic fractionation of carbon and hydrogen in alkanes demand to be investigated in detail. Meanwhile, it is necessary to integrate comprehensive geochemical studies of natural gas samples (including isotopic compositions of alkanes, non-hydrocarbons and noble gases) with geological tectonic backgrounds of studied basins, aiming at establishing an effective geochemical criterion system for the identification of deep natural gases.

Figure 1 shows distribution of  $\delta^{13}C_1$  versus  $C_1/C_{2+}$  and frequency distribution of  $\delta^{13}C_1$  values for natural gases from the Songliao Basin and data of 11 samples in literature, in which a-1 represents  $\delta^{13}C_1$  values of gases from the serpentinization of ultramafic rocks [1], crystalline rocks [2] and the mid-ocean ridges [3]. These  $\delta^{13}C_1$  values of abiogenic methane ( $-7.1\text{‰} \sim -29.0\text{‰}$ ) provide significant evidence for the identification of abiogenic hydrocarbon gases. Based on the typical  $\delta^{13}C$  values of abiogenic, thermogenic and microbial methane, natural gases from the Songliao Basin can be in principle classified into three types, abiogenic as (a-2,  $-10.9\text{‰} \sim -24.9\text{‰}$ ; a-3,  $-25.2\text{‰} \sim -29.8\text{‰}$ ), thermogenic gas (t-1,  $-30.2\text{‰} \sim -34.1\text{‰}$ ; t-2,  $-35.5\text{‰} \sim -38.9\text{‰}$ ; t-3,  $-41.1\text{‰} \sim -49.9\text{‰}$ ; t-4,  $-50.3\text{‰} \sim -54.9\text{‰}$ ) and microbial gas (mg,  $-55.6\text{‰} \sim -70.3\text{‰}$ ). It is evidently hard to interpret the distribution of  $\delta^{13}C_1$  data for a-3 by the abiogenic gas theory and for t-1 by the conventional thermogenic gas theory. Given the geological settings of high heat current, high geothermo-gradient and frequent magma activities in the Songliao Basin, it may be more reasonable to interpret the  $\delta^{13}C_1$  distribution for both a-3 and t-1 by the deep natural gas theory. In other words, the data for a-3 and t-1 can be incorporated to be classified as a novel type of natural gases-deep natural gas.

Figure 2 are characterized by a normal distribution(A), indicating these natural gases are products of sedimentary organic matter via the degradation of hydrocarbon formation, and a partially reversed distribution of  $\delta^{13}C$  values(B). These samples may be related to deep natural gas generated by sedimentary organic matter under thermodynamical condition deep down in the Songliao Basin.



Type	Sample number	$\delta^{13}C_1$ (‰) PDB	$C_1/C_{2+}$	Source
A-1	11	-7.1~-29.0	10.81~342.5	Others*
A-2	43	-10.9~-24.9	8.1~8907.3	This paper
A-3	55	-25.2~-29.8	26.2~205.3	This paper
T-1	28	-30.2~-34.1	3.9~429.1	This paper
T-2	15	-35.5~-38.9	1.1~105.8	This paper
T-3	51	-41.1~-49.9	1.1~404.3	This paper
T-4	25	-50.3~-54.9	1.4~588.6	This paper
MG	15	-55.6~-70.3	9.6~3671.3	This paper

\* Abrajano et al.,1990; Sherwood et al.,1993; Charlou et al.,1996.

Figure 1: The distribution of  $\delta^{13}C_1$  values for natural gases from the Songliao Basin.

F.1-1  $\delta^{13}C_1$  versus  $C_1/C_{2+}$  distribution of natural gases;

F.1-2 Type-frequency distribution of natural gases.

The distribution of  $\delta^{13}C_1$  values for natural gases from the Songliao Basin by classifying genetic four types: abiogenic gas (A-2), thermogenic gas (T-2,T-3 and T-4), deep gas(A-3 and T-1) and microbial gas (MG).

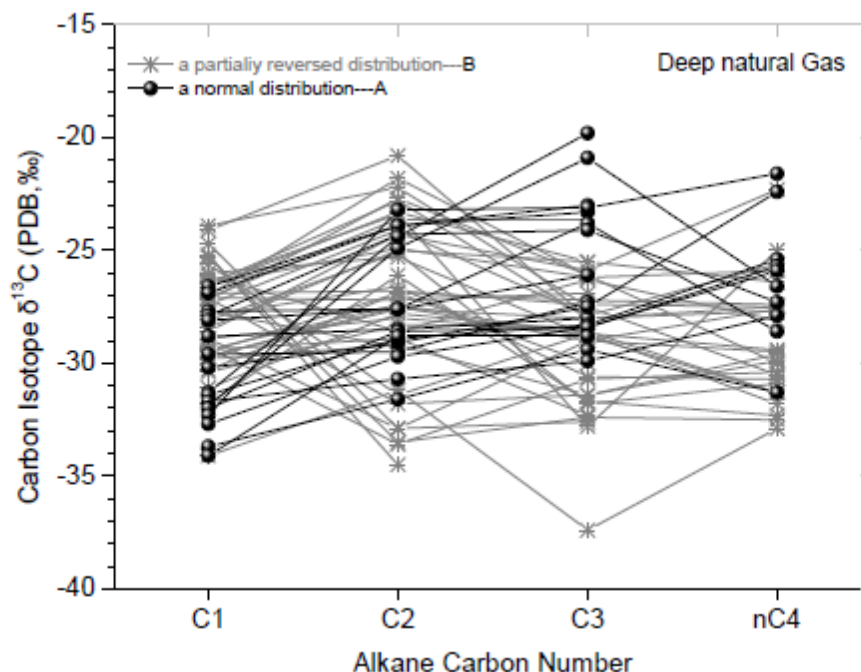


Figure 2: Distribution of alkane carbon numbers versus the  $\delta^{13}\text{C}$  value of the deep natural gases from the Songliao Basin.  
 A– normal distribution ; B–a partially reversed distribution

Researches on deep oils and natural gases should focus on breaking through the traditional “thermal maturity deadline” to search for new oil-gas resources, especially deep natural gas resources. Deep horizons of ultra-deep hydrocarbon-bearing basins will be significant targets and provinces in future natural gas exploration.

### References

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