Genetic Classification of Natural Gases in the Oil-Gas Zone and Its Application in the Sichuan Basin

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Abstract

On the basis of the carbon isotopic compositions of methane (CH_4) and its homologues and the differences in isotopic values for CH_4 and ethane (C_2H_6) and the correlation and compositional characteristics of hydrocarbon gases, the author has proposed a genetic classification of natural gases in the oil-gas zone. They are classified as biogenetic and abiogenetic gases in terms of the types of hydrocarbon-generating precursors (or parent materials) and their thermal evolution stages. Biogenetic gases can also be further divided into two series: biochemical and thermochemical gases, with the latter formed at different evolution stages. Gases generated from type-I and -II_1 organic matter are called oil-series gases, those from type-III, coal-series gases, and those from type-II_2, mixture-type gases. Gases generated from two or more than two types of precursors are called mixture-source gases.

According to those mentioned above, natural gases from the major oil-gas pools in the Sichuan Basin have been discriminantly analyzed, and the results are concordant with the distribution and development of hydrocarbon-source rocks as well as with their characteristics, indicating a prospective application.

Genetic Classification of Natural Gases

Since the 1930's many researchers have devoted their efforts to the study of the compositions, environments and sources of natural gases and proposed a number of schemes of classification, which can roughly be generallized as follows: (1) according to their generating environments, natural gases can be classified as atmospheric gas, surface gas, sedimentary-rock gas, oceanic gas, metamorphic-rock gas, magmatic-rock gas, mantle gas and cosmic gas (Sokolov, V.A., 1966; 1971); (2) according to the precursor types, natural gases can be classified as organic and inorganic gases, with the former being further divided into biochemical gas, thermally catalytic gas, pyrolytic gas, etc. (Tissot, B. P. and Welte, D. H., 1978); (3) according to their hydrocarbon compositions and C and H isotopic compositions, natural gases can be classified as biogenetic gas (terrestrial facies, marine facies), thermogenetic gas (petroleum-associated gas, condensate-associated gas, sapropel-type dry gas), mixture-type gas (shallow and deep levels), deep-source gas, etc. (Stahl, W., Schoell, M., 1981; 1983); and (4) in terms of phase changes natural gases can be classified as free gas, dissolved gas, adsorbed gas, solid gas, etc.

In recent years a rapidly increasing demond for natural gases, oil/gas exploration has been greatly expanded and researches concerned have also been deepened. Therefore, a number of classification schemes have been put forward, dividing natural gases into oil-type and coal-generating gases (Dai Jinxing et al., 1982); oil-type and coal-type gases (Xu Yongxun et al., 1985); oil-generating and coal-generating gases (Wang Hanyun and Fu Jiamo, 1987); oil-series and coal-series gases (Huang Jizhong, 1983; 1984), etc. Nevertheless, non of these classification schemes has been well accepted. The possible reasons may be: (1) As compared with oils, natural gases are diverse in origin (organic→ inorganic), involve a variety of gas-generating processes (Macdomald, G. J., 1983, who put forward six models for gas generation; Xiong Shousheng et al., 1984, presenting eight gas-generating processes), and are characterized by a long duration of gas generation (they can be formed at ≤75 ℃, 75 – 200 – 300 ℃, even > 300 ℃) and easy flow (As gas molecules are small in diameter, they are easy to migrate and diffuse). So natural gases obtained under natural conditions are always characteristic of multiple origin, which is not only related with hydrocarbon-generating precursors, but also with hydrocarbon-generating environments (temperature, pressure, wallrock, medium, etc.), the path and distance of migration and diffusion and trap conditions. (2) Because of the simple composition of natural gases, the detecting methods available cannot completely present the direct indicators to distinguish natural gases of different origins. Although much work has been done in this aspect and many discriminant indicators have been established, they are still inapplicable to the actual geological conditions encounterd in different rock series as well as in different regions. Strictly speaking, it is difficult to find end-member indicators to distinguish natural gases of different origins. In most cases natural gases are of multi-origin (derived from different sources at different evolution stages).

Figures and Tables for Genetic Classification of Natural Gases

Considering the figures and tables for the genetic classification of natural

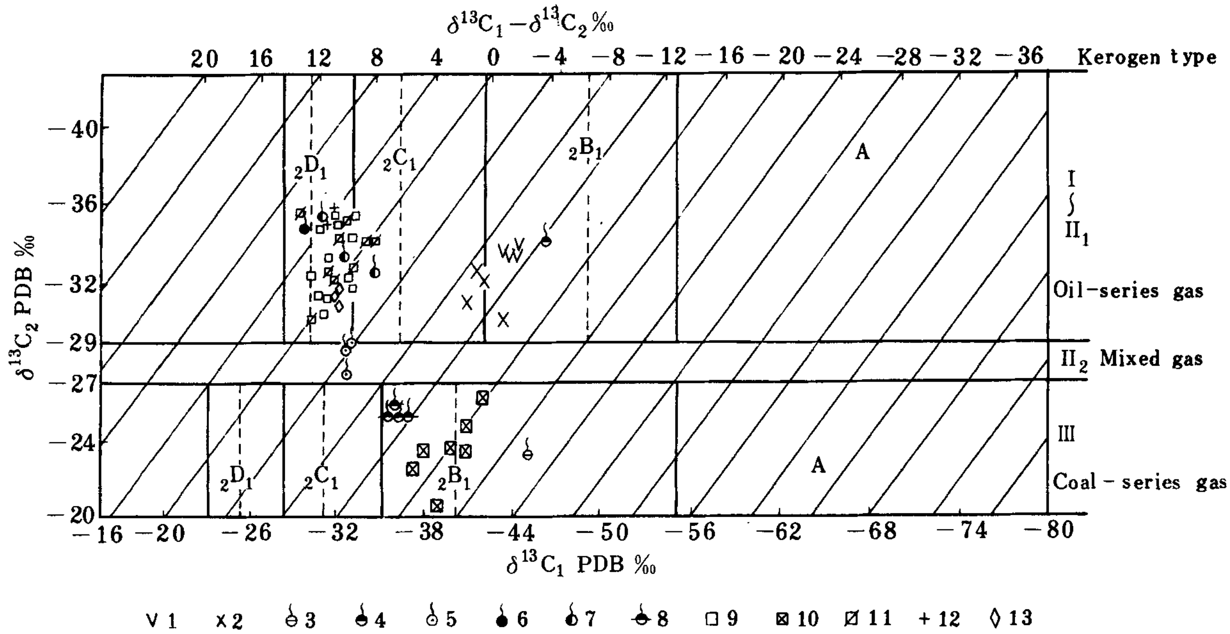

Fig.1. Genetic classification of natural gases in the oil-gas zone.

1. J_1t^5; 2. J_1t^4; 3. T_3x; 4. T_2r, 5. T_1c^5; 6. T_1c^3; 7. T_1c^1; 8. T_1f; 9. P_2l^2; 10. P_2l^1; 11. P_1y^3; 12. C; 13. Z_b.

gases, one should pay attention to the compositional changes of hydrocarbon gases and the carbon and hydrogen isotopic values of CH_4 and its homologues. So the genetic classification has been done in accordance with hydrocarbon-generating precursors and their geological settings (evolution stage). Different hydrocarbon series have resulted from different precursors. For example, oil-series precursors will generate petroleum, condensate, wet gas, dry gas, low methane gas, etc., while coal-series precursors will produce condensate, wet gas, dry gas, low methane gas, etc., instead of liquid hydrocarbons in large amounts. At different evolution (degradation, pyrolysis) stages there will be different products with different geochemical signatures. Therefore, the genetic classification in terms of precursor types and evolutionary environments (Table 1, Fig. 1) is conducive to gas exploration.

Table 1. Genetic classification of natural gases in the oil-gas zone

Evolution stage (R^0_{max}%) \ Type	Biogenetic (organic) $\delta_{13}C_2$ ‰: Oil-series (type-I, -II$_1$) (<-29)		Mixture-type (type-II$_2$) (-27 ~ -29)	Coal-series (type-III) (≥-27)		Abiogenetic (inorganic): Hydrocarbon gas $\delta_{13}C^-$ ‰	Non-hydrocarbon gas $^3He/^4He$
Immaturity I	$\delta^{13}C_1$ ‰	C_2^+/C_1^+ %	Biochemical gas	$\delta^{13}C_1$ ‰	C_2^+/C_4^+ %	$\delta^{13}C_1 > -20$ ‰; $\delta^{13}C_1 > \delta^{13}C_2 > \delta^{13}C_3 > \delta^{13}C_4$	≥8RA; PA=1.4 ×10^{-6}
0.5	-55 ~ -60	<1		-55 ~ -60	<0.1		
Maturity stage II: (1) Early stage			Thermochemical gas				
1	-49	>10		-40	>5		
(2) Late stage							
1.35	-42	10		-35	5		
High-maturity III: (1) Early stage							
2	-36	5		-31	3		
(2) Late stage							
2.5	-33	3		-28	1		
Over-maturity IV: (1) Early stage							
4	-30	1		-25	<1		
(2) Late stage							
6	-28	<1		-23	<0.1		
Metamorphism V							

*$\delta^{13}C$ relative to PDB.

As can be seen in Table 1 and Fig. 1, the stable isotopic values at molecule level cannot only reflect isotopic differences, but also rule out the influence of mixed hydrocarbons Carbon isotopic values for CH_4 and C_2H_6 are more impor - tant, as CH_4 and C_2H_4 are the components most widely distributed in oil and gas fields, and they are relatively stable under the same conditions. Our studies show that $\delta^{13}C_1$ values, which appear more important in the geological sense, are related out only with the precursor type, but also with the thermal evolution stage, $\delta^{13}C_2$ values bear information about the parent sources and the differences between them can reflect isotopic fractionation and source. The more negative the differences, the more possible it will be that biochemical gas is involved. On the contrary the more positive the differences, the more possible the deep mixture-source will be. Significant differences are noticed for hydrocarbon gases, especially the proportion of heavy hydrocarbon (C_2^+) due to different precursor sources. Differences would also be recognized at different thermal evolution stages, although the same precursor source is considered. In consideration of both inorganic and organic categories, changes in $^3He/^4He$ are also taken into account in addition to $\delta^{13}C_1$ values. Although the latter is a rare gas, it is very useful.

Natural gases are roughly classified as biogenetic (organic) and abiogenetic (inorganic) gases

1. Biogenetic gases can also be further divided into two series: biochemical and thermochemical. According to their thermal evolution stages, thermochemical gases are classified as mature gas, highly mature gas and over-mature gas. As viewed from hydrocarbon-generating precursor sources, type-I and $-II_1$ precursors will produce oil-series gases, type-III will produce coal-series gases, and transitional type-II_2 will generate mixture- type gases.

Table 1 and Fig. 1 are not restricted to the current classification scheme, i.e., natural gases are generally classified as biochemical, pyrolitic and deep-source gases. Natural gases of thermal origin should be classified as detailed as possible because the maturity of organic matter is normally high in sedimentary rocks of Sichuan or even of the whole southern China. Organic matter mostly entered into the high-maturity stage, even into the over-maturity stage during the Paleozoic- Proterozoic. In some regions where organic matter is deeply buried the thermometamorphic stage is expected. According to the maturity of organic matter natural gases can be classified as immature, mature, highly mature and over-mature gases. This scheme of classification is not so ideal that the author tries to classify them as thermochemical gases generated at different evolution stages (early-stage and late-stage gases), followed by their nomenclature. In this way the geochemical implications and indicators are well defined for different evolution stages. In addition the best gas-generating period (or stage) is indicated, i.e., from the high-maturity stage to the over-maturity early stage. The over-maturity late stage is a period of low gas production and thus infavourable to the preservation of hydrocarbon gas (Huang Jizhong, 1986).

2. Coal-series gases (humic-type precursors consisting of two subtypes: dispersed and concentrated) are very important in energy exploration, so this type of gas is listed independently. But it still belongs to the thermochemical gas series

because it was produced as a result of thermodynamic action. Coal-series gases are substantially different from oil-series gases due to their different hydrocarbon-generating precursors.

3. Gases generated at the immaturity stage were derived from the biochemical action of anaerobic bacteria under low temperature, oxygen-deficiency and low sulphate conditions. This kind of gas is produced either for oil series or for coal series, and is also assigned to immature gases.

4. Pyrolitic gases are defined as thermochemical gases, which cannot only correspond to biochemical gases, but also are different in nature. The weak bonds of macromolecules are broken and degradate to form liquid hydrocarbons and gaseous hydrocarbons under the action of heat energy contributed by organic matter. With increasing temperature the evolution will continue and then lead to the formation of hydrocarbons low in carbon number and molecular weight. This process is actually a very complicated process, and it can also be considered a process of redistribution of hydrogen.

5. In the study of the classification of natural gases the transitional type — mixture-type gases are extremely difficult to deal with. They comprise not only gases generated from type-II_2 precursors, but also those from different types of precursors, including those mixing with biochemical gas and with the gases generated at different evolution stages; oil-series gases mixing with coal-series gases, organic gases mixing with inorganic gases and so on. For the purpose of distinguishing, we name those generated from type-II_2 precursors the mixture-type gases. The other types are named mixture-source gases. The gases generated from more than two kinds of precursors or at different evolution stages are called composite-type natural gases.

Parameters and indicators for classifying natural gases

1. To distinguish biogenetic gases from abiogenetic gases (derived from deep levels of the crust and the upper mantle) relies on the carbon isotopic values of CH_4. In the case of $\delta^{13}C_1 \geq -20$ ‰ PDB, the gases are classified as inorganic gases as the accepted $\delta^{13}C_1$ values range from -15 to -17.6 ‰ for inorganic gases from the mid-rise in the East Pacific Ocean. According to the carbon isotopic values of CH_4 and its homologues, the gases characterized by $\delta^{13}C_1 < \delta^{13}C_2 < \delta^{13}C_3 < \delta^{13}C_4$ are classified as biogenetic gases, and inversely as abiogenetic gases. As is known, biogenetic gases show regularities concerning isotopic fractionation, but those of inorganic origin often show a random trend. As viewed from the difference in isotope series between CO_2 and CH_4 (Guchalo, 1981), the $\delta^{13}C_1$ values of inorganic gases are with the range of $-7 \sim -41$ ‰, while the $\delta^{13}C_{CO_2}$ values, $-27 \sim -70$ ‰. In addition $^3He/^4He$ ratios for rare gases can also be used to distinguish biogenetic from abiogenetic gases. For example, $^3He/^4He = n \times \geq 10^{-5}$ (Yakucheni, 1976) would indicate an inorganic origin. In this paper $n \times \geq 8RA$, $RA = 1.4 \times 10^{-6}$ is an indication of inorganic origin. Up to now, such high values have not yet been reported in China. Nevertheless, high values, if any, should be dealt with on the objective basis because rare gases would by no means be considered the same as hydrocarbon gases. Both of them may not come from the same source, and they are probably of mixture source in origin.

2. Differences between oil-series gases and coal-series gases are determined mainly by $\delta^{13}C$ values of ethane. $\delta^{13}C_2 \geq -27‰$ is indicative of coal-series gases, $\delta^{13}C_2 < -29‰$ of oil-series gases, and $-27‰ < \delta^{13}C_2 \geq -29‰$ of transitional type or mixture source. Why do we use the $\delta^{13}C_2$ values of C_2H_6? That is because $\delta_{13}C_2$ values show little variation with the thermal evolution of C_2H_6. When R° increases from 0.78 to 3.13%, $\delta^{13}C_2$ values only show an increase of 1 — 2 ‰ (Tang Xiuyi et al., 1987). Moreover, as can be seen from the statistical data on the $\delta^{13}C_2$ values for coal-series gases from China, 78.6% of the 84 samples have $\delta^{13}C_2$ values $\geq -27‰$ and 94.5 ‰ of them have $\delta^{13}C_2$ values as high as up to $\geq -28‰$. From the statistical data on coal-series gas samples from Australia one can see that of the 115 samples those with $\delta_{13}C_2 \geq -27‰$ account for 80%, and those with $\delta^{13}C_2 \geq -28‰$ for 92.17%, i.e., the former is dominant. Similarly, of the 62 oil-series gas samples collected in China 95.16% have $\delta^{13}C_2 < -29‰$ (Shao Jianjun, 1987). As compared with high-molecule alkanes, C_2H_6 would be less affected by biological degradation under the same conditions. So we consider that the $\delta^{13}C_2$ values may substantially reflect the types of hydrocarbon-generating precursors.

3. Further classification of thermochemical gases can reveal the gases generated at various evolution stages.

(1) On account of the maturity of organic matter natural gases can be classified as maturity early-stage gas, maturity late-stage gas, high-maturity early-stage gas, high-maturity late-stage gas, over-maturity early-stage gas, over-maturity late-stage gas, immaturity-stage gas and metamorphic-stage gas, with the corresponding R^0_{max} values being $\leq 0.5\%$, 0.5 ~ 1 ~ 1.35%, 1.35 ~ 2 ~ 2.5%, 2.5 ~ 4 ~ 6%, and >6%.

(2) The classification is based on the compositions of hydrocarbons $-C_1/C_2+C_3$, $C_1/C_1^+\%$ and $C_2^+/C_1^+\%$.

(3) Based on the carbon isotopic values of CH_4. With reference to the boundary values for crude oil, condensate-wet gas, and dry gas (Stahl, W. et al., 1981), maturity early-late-stage oil-series gases have $\delta^{13}C_1 = -60 \sim -55‰ — 49 \sim -42‰$, high-maturity early-late-stage $\delta^{13}C_1 = -42 \sim -36 \sim -33‰$, and over-maturity early-late-stage $\delta^{13}C_1 = -33 \sim -30 \sim -28‰$. The corresponding values of coal-series gases are: $-60 \sim -55 \sim -40 \sim -35‰$, $-35 \sim -31 \sim -28‰$, and $-28 \sim -25 \sim -23‰$. $J_1t^4(II_1)$ type $< T_3x(III)$ type — $-4 \sim -8‰$ +, showing a $\delta^{13}C_1$ decrease by $-5 \sim -7‰$. The $\delta^{13}C_1$ values of mixed gases are intermediate between those of the two types.

(4) Based on the correlation between the composition of hydrocarbon gas and the carbon isotopic composition of CH_4.

(5) Based on differences in carbon isotopic values between CH_4 and C_2H_6 (Figs.2, 3 and 4).

4. The problem of transition-type and mixture-source gases is a hardnut to crack in the classification of natural gases. On the hand, they may have been formed from mixture-source parent materials (type-II_2), and on the other they may have been derived from mixing of oil-series and coal-series gases, or of biochemical and oil-series gases, or of biochemical and coal-series gases, or of low-maturity gases at shallow levels and high-maturity gases at deep levels, or of oil-series or inorganic hydrocarbon gases. In other words, natural gases

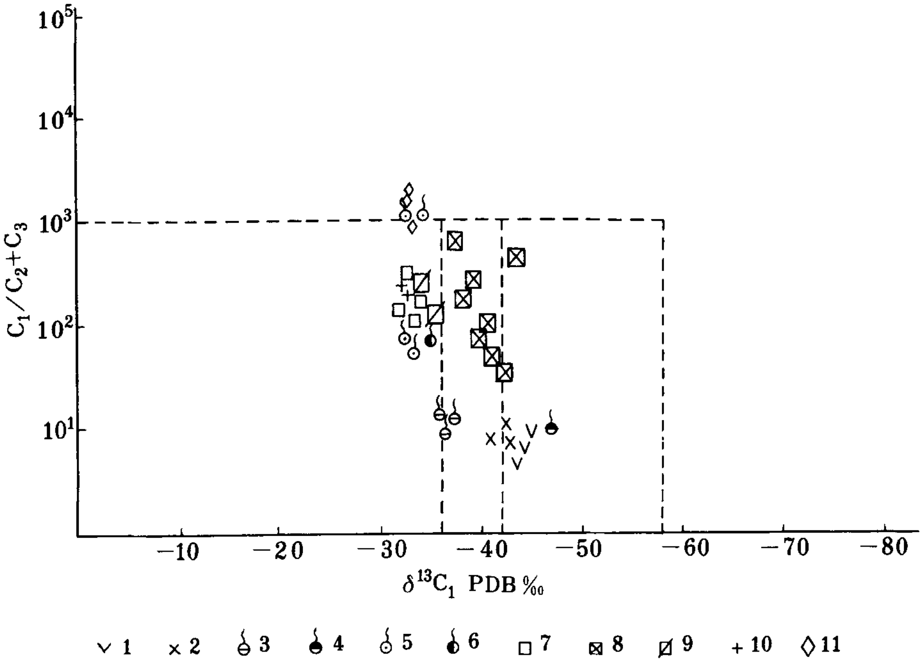

Fig.2. Plot of C_1/C_2+C_3 vs. $\delta^{13}C_1$ ‰ .

1. J_1t_5 ; 2. J_1t^4 ; 3. T_3x ; 4. T_2r ; 5. T_1c^5 ; 6. T_1c^1 ; 7. P_2l^2 ; 8. P_2l^2 ; 9. P_1y^3 ; 10. C; 11. Z_b .

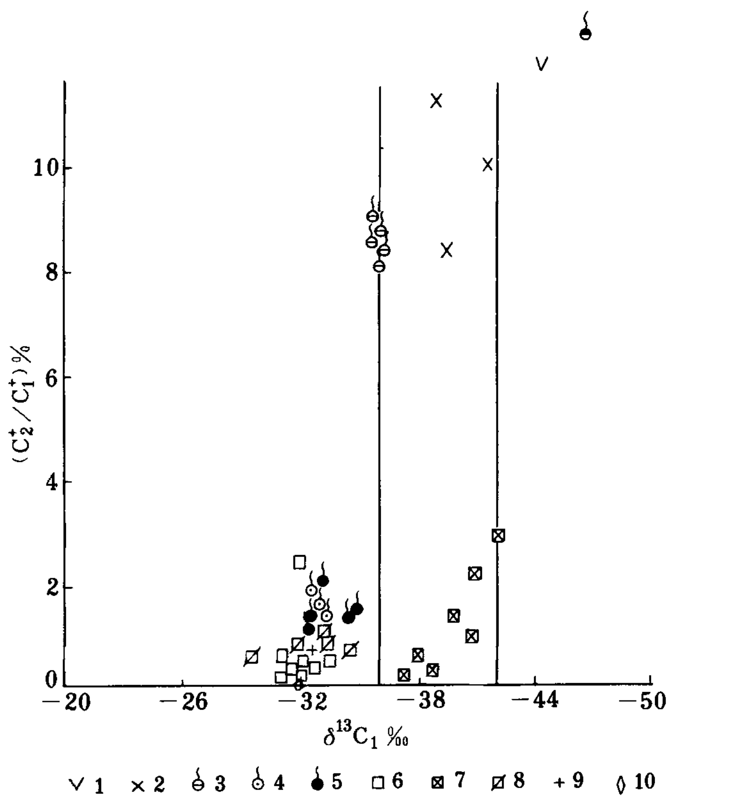

Fig.3. Plot of $C_2^+./C_1^+$ ‰ vs. $\delta^{13}C_1$ ‰ .

1. J_1t^5; 2. J_1t^4; 3. T_3x ; 4. T_1c^5; 5. T_1c^3; 6. P_2l^2 ; 7. P_2l^1 ; 8. P_1y^3 ; 9. C ; 10. Z_b .

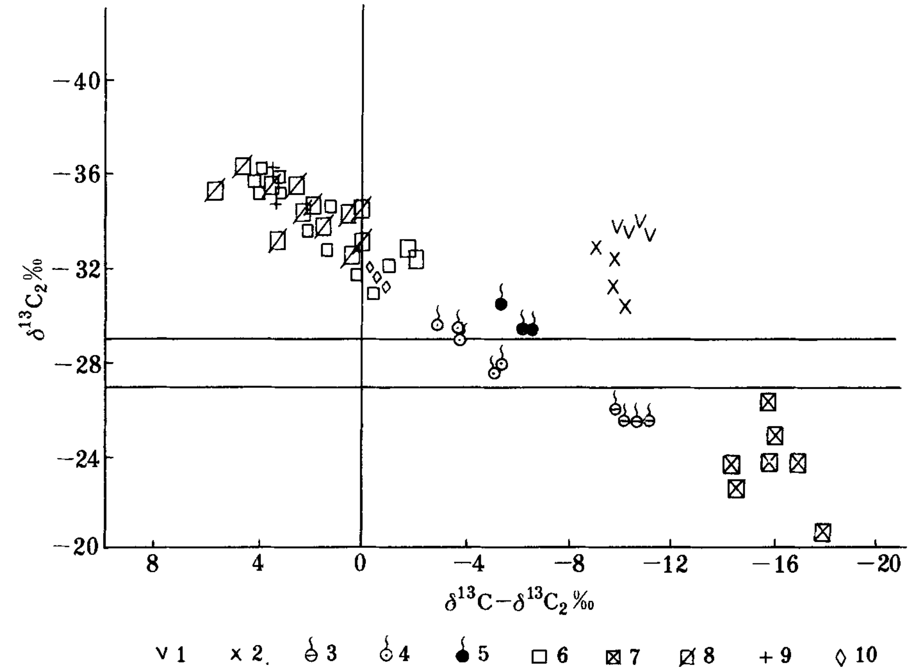

Fig.4. Plot of $\delta^{13}C_2$‰ vs. $\delta^{13}C_1-\delta^{13}C_2$‰ .
1. J_1t^5; 2. J_1t^4; 3. T_3x; 4. T_1c^5; 5. T_1c^3; 6. P_2l^2; 7. P_2l^1; 8. P_1y^3; 9. C; 10. Z_b.

gases only bear limited information. In this paper it is evidenced that in the case of $\delta^{13}C_2 < -27 \sim -29$‰ natural gases should be classified as type$-II_2$. i. e., mixture-type gases, and the others are called mixture-source gases.

5. In the practical application of the above indicators local geological conditions should be taken into consideration at the same time. For example, in case natural gases have $\delta^{13}C_2$ values ranging from $-27 \sim -29$‰, kerogen type and the maturity stage of the reservoir should be comprehensively analyzed so as to gain relatively true results.

6. Natural gases formed from more than two different sources or at different evolution stages are called composite-type gases (the mixing of gases generated at different stages is commonly seen in the thermochemical gas series).

Application of Genetic Classification of Natural Gases in the Sichuan Basin

Natural gases are distributed widely in the Sichuan Basin from Jurassic to Sinian. Industrial crude oils are produced in the Jurassic system and condensate in the Middle and Upper Triassic series. The other strata produce mainly natural gases. The major industrial production aquifers are Triassic, Permian, Carboniferous and Sinian. Natural gases tend to increase in the Lower Jurassic series in the northern part of central Sichuan. The Middle and Lower Triassic (T_2, T_1), Permian (P), Middle Carboniferous (C_2) and Sinian (Z_2b) are distributed in southeastern Sichuan, covering an area accounting for about half of the basin. The Middle and Upper Triassic are distributed in northwestern, western and northern Sichuan. Natural gases are produced mainly in the Paleozoic in local places. Recently, industrial scale gas deposits have been discovered in the Middle

Triassic-Upper Permian (P_3) in the transitional zone between central and southern Sichuan. It can be seen clearly that natural gases are not only abundant, but also are of extensive occurrence in the Sichuan Basin.

Description of hydrocarbon-source rocks in the Sichuan Basin

Studies have shown that the main hydrocarbon-source rocks are Lower Cambrian ($€_1$) and Lower Silurian (S_1) dark-colored mudstones in the Sichuan Basin. The strength of organic matter there is high ($10^4 t/km^2$). It is generally > 100 in the Lower Cambrian mudstones, i. e., 100 ~ 300 in northwestern Sichuan, 100 ~ 200 in southwestern and southern Sichuan, 100 ~ 600 in northeastern Sichuan, and < 100 in the other locations. And the value of S_1 is about 100± in southwestern and southern Sichuan, and 50± in eastern Sichuan. Organic matter is designated to type-I and $-II_1$, indicative of oil-source rock series. Organic matter is high in maturity and over-maturity early-stage organic matter is dominant in the basin (R_{max} = 2.5 ~ 4%), except for maturity-stage organic matter at the edges of northwestern and northeastern Sichuan. Petroleum produced has been transformed into natural gases through thermal Evolution (degradation, pyrolysis). In the Lower Permian muddy material and micritic carbonate rocks organic matter is higher in strength than in the Lower Paleozoic equivalents, with $Q_c > 200 \times 10^4 t/km^2$ in the Maokou Formation P_1y^3 except in western Sichuan, and > 500(Q_c) in Hunan, Hubei, Guizhou and the southwestern part of southern Sichuan. Type-II_1 is the major type of organic matter, and type-II_2 comes next, indicative of oil-source rock series (Huang Jizhong and Jiang Huaicheng, 1986). Organic matter is of high maturity (R^0_{max} = 1.35 − 2.5%) except at the margins of northwestern Sichuan where organic matter is at the maturity stage and in the Nanchuan area of eastern Sichuan where it is at the late stage of maturation. However, deeply buried organic matter (western and northern Sichuan) has come into the over-maturity early stage (R^0_{max} = 2.5 — 4%). Oils generated from hydrocarbon-source rocks have also been transformed into natural gases (Huang Jizhong, 1984). Coming next are the Lower Jurassic dark-colored mudstones where organic matter is high in strength (Q_c = 100±) in central, northern and eastern Sichuan. Type − II_1 organic matter is dominant, with type − II_2 recognized in local places, which is generally at the late stage of maturation (R^0_{max} = 1 ~ 1.35%), indicative of oil-source rock series with oil as its main product. The Upper Permian and Upper Triassic dark-colored mudstones and coal seams are gas-source rock series, in which organic matter is so high in strength as to be 1000 ~ 3000 in the Upper Permian at the juncture of central, southern and eastern Sichuan, but 300 ~ 500 ~ 1000 in other locations of the basin; 1000± in the Upper Triassic (except in southern Sichuan). Type-III organic matter is dominant (only in T_3x^1 of western Sichuan is recognized type-II_2 organic matter). Natural gases are derived mainly from dispersed- and concentrated-type organic matter. Upper Permian organic matter is generally at the high-maturity stage (mostly at the high-maturity early stage), deeply buried organic matter (western, northwestern and northern Sichuan) is at the high-maturity late stage, and at the over-maturity early stage in local places. Upper Triassic organic matter is generally at the maturity late stage, but locally at the high-maturity stage.

From the above it may be concluded that natural gas sources of the major gas pools in the Sichuan Basin are the Lower Permian, Upper Permian, Lower Jurassic and Upper Triassic series which are of authigenous-source type. However, the Sinian, Middle-Upper Triassic and Carboniferous are of allothigenous-source type.

Analysis of the genetic types of natural gases in the Sichuan Basin

It is known from Fig.1, which was computer-charted on the basis of the data for natural gases in different production aquifers in the Sichuan Basin, that:

1. In the Sichuan Basin natural gases in different production aquifers are all organic in orgin as indicated by the carbon isotopic compositions of CH_4 and C_2H_6 in hydrocarbon gases, as well as by the $^3He/^4He$ ratios $= n \times 10^{-8}$ for natural gases from the Sinian of Weiyuan, Sichuan. As a result, the possibility has been ruled out that high $^{40}Ar/^{36}Ar$ and He/Ar ratios would be indicative of inorganic origin.

2. There is no biochemical gas, i.e., immaturity-stage gas of commercial importance in the Sichuan Basin because of $\delta^{13}C_1 \geqslant -55 \sim -60‰$.

3. Natural gases in the Sichuan Basin all belong to thermochemical gases generated at different evolution stages, and hence can be classified as oil-series and coal-series gases in terms of the $\delta^{13}C$ values of C_2H_6.

(1) The $\delta^{13}C_1$ values of natural gases from the T_3x^2 gas pool in the Upper Triassic series are close to those of natural gases from Upper Permian (P_2l^1) and Upper Triassic (T_3x) coal seams ($> -27‰$). There is no doubt that both of them are coal-series gases, i.e., authigenous-source coal-series maturity late-stage gases. This is in concordance with the fact that organic matter in the $T_3x^{1,3}$ argillaceous hydrocarbon-source rocks is of type III and $R^0_{max} = 1 \sim 1.35\%$ at the late stage of organic maturation.

(2) Natural gases from gas pools in the Lower Jurassic (J_1t), Lower Permian (P_1y), Middle Carboniferous (C_2) and Sinian (Z_2b) are all assigned to oil-series gases, with $\delta^{13}C_2 < -29‰$. According to $\delta^{13}C_1$ values and their correlations with C_1/C_2+C_3, C_2^+/C_1^+ and natural gases with $\delta^{13}C_1 - \delta^{13}C_2$ values, they belong to naturity late-stage gases, high-maturity stage gases and over-maturity early-stage gases (Fig.2, 3 and 4).

a. There are two groups of oil/gas pools in the Lower Jurassic Lianggaoshan Formation (J_1t^5), and Da'anzhai Formation (J_1t^4), the former's $\delta^{13}C_1$ values ranging from -43.6 to $-44.8‰$, and the latter's from -41.028 to $43.62‰$; the former's $\delta^{13}C_1 - \delta^{13}C_2$ values from -9.9 to $-11.2‰$ and the latter's from -9.828 to $-12.7‰$. In some regions such as Mingyuechang, Jinhua Township in northwestern Suining, $\delta^{13}C_1 > -40‰$ and $\delta^{13}C_2 > -29‰$ for J_1t^4 characteristic of type II_2.

It is concluded that natural gases from J_1t belong to authigenous-source oil-series gases generated at the late stage of maturation.

b. Gas pools in the Maokou Formation (P_1y^3) and the Xixia Formation (P_1y^2) of the Lower Permian have $\delta^{13}C_1$ values from -32.25 to $-34‰$ in southern Sichuan and from -30.51 to $-34.7‰$ in eastern Sichuan. $\delta^{13}C_2$ values range from -33.07 to $-35.378‰$ and -30.499 to $-35.796‰$; $\delta^{13}C_1 - \delta^{13}C_2$

values from -2.069 to $2.2‰$ and from -0.3 to $5.627‰$, respectively. It may be concluded that natural gases from P_1y mostly belong to high-maturity late-stage gases of authigenous-source oil series.

c. The Carboniferous (C_2) gas pools have $\delta^{13}C_1$ values ranging from -32.17 to $-32.25‰$ in the Wolonghe gas field, and -33.6 to $-34.32‰$ in the Xiangguosi gas field. $\delta^{13}C_1-\delta^{13}C_2$ values are positive, that is, $\delta^{13}C_1>\delta^{13}C_2$. in the range $3.13\sim3.86‰$ and $2.01\sim3.04‰$.

It is suggested that Carboniferous natural gases belong to over-maturity early-stage gases of allogenous-source oil-series. Moreover, signs of the presence of mixture-source gases are noticed as $\delta^{13}C_1-\delta^{13}C_2$ values are apparently positive.

d. Gas pools in the Dengying Formation (Z_2b) of the Upper Sinian have $\delta^{13}C_1$ values ranging from -32.33 to $-32.5‰$, and $\delta^{13}C_2$ values from -31.193 to $-31.907‰$, with $\delta^{13}C_1-\delta^{13}C_2<0‰$, within the range of -0.393 to $-0.998‰$.

It is evidenced that natural gases from Z_2b belong to over-maturity early-stage gases of allogenous-source oil-series.

(3) Although natural gases from gas pools in the Changxing Formation of the Upper Permian series are distributed in the range of oil-series gases, their $\delta^{13}C_1$ values are variable from -31.52 to $-34.97‰$ (-32.08 to $-32.82‰$) in southern Sichuan, and from -30.643 to $-33.3‰$ ($-31.143\sim-32.24‰$) in eastern Sichuan. $\delta^{13}C_2$ values are in the range $-31.93\sim-36.03‰$ ($-34.17\sim-35.87‰$) and $-30.731\sim-35.51‰$ (-31.50 to $-35.51‰$), with $\delta^{13}C_2<\delta^{13}C_1$. The former $\delta^{13}C_2$ values are in the range $-1.22\sim4.4‰$ ($1.25\sim4.14‰$), while $\delta^{13}C_1$ values, $-0.655\sim-3.84‰$ ($-0.61\sim3.17‰$). $\delta^{13}C_1$ values are apparently discrepant with the maturity of organic matter in gas pool-hosted limestones. Studies have shown that in the P_2l^2 high-maturity early-stage natural gases ($R^0_{max}=1.35\sim2\%$) are dominant (Jiang Huaicheng and Huang Jizhong, 1986). However, natural gases are found distributed in the range from high-maturity late stage to over-maturity early stage. This is obviously discrepant with geological facts, but in consistence with the evidence developed from $\delta^{13}C_1$ values for coal-series gases.

Natural gases from P_2l^2 belong to mixture-source gases resulting from mixing of upward-migrating coal-series gases with P_2l^2 oil-series gases (diluted by CH_4 and coal-series gases).

(4) Although natural gases from T_1 are in the range of oil-series gases, evidence suggests the upward migration of high-maturity natural gases as $\delta^{13}C_1$ values are in the range -29.91 to $-34.718‰$ (-31.32 to $-34.58‰$). There is a significant difference in maturity from organic matter in gas reservoir strata. The latter's organic matter is at the high-maturity early stage ($R^0_{max}=1.35\sim2\%$), i.e., at the condensate-wet gas stage. Condensate was found in the process of drilling, but its $\delta^{13}C_1$ values $>-36‰$, and $\delta^{13}C_2$ values $>-30‰$ in the Lower Triassic ($T_1c_1^5$ and T_1c^3) in the Wolonghe gas field, generally within the range of $-27.5\sim-29.918‰$ ($-28.87\sim-29.61‰$). These values are not only significantly different from Carboniferous equivalents ($-35\sim-36‰$), but also from those of $P_2l^2-P_1y^3$ gas pools, which range from $-30.731\sim-35.796‰$ ($-31.636\sim-35.6‰$). It is demonstrated that the T_1c itself has a limited gas supply source, but mostly from the underlying strata. Similarly, $\delta^{13}C_1$ values are

in the range $-31.32 \sim -35.22$ ‰, and $\delta^{13}C_2$ values in the range $-32.97 \sim -35.73$ ‰ for T^1c^1 gas deposits in the Naxi gas field, southern Sichuan; $-32.09 \sim -32.951$ ‰ ($-32.25 \sim -32.91$ ‰) and $-35.14 \sim -35.44$ ‰ ($-35.17 \sim -35.378$ ‰) for P_1y^3 gas deposits. Similar gas supply source is also expected. That is to say, most of the CH_4 is derived from upward-migrating gases in the Lower Paleozoic.

Conclusions

1. According to the figures and tables for the genetic classification of natural gases in terms of $\delta^{13}C$ values of CH_4 and C_2H_6, genetic types of natural gases can be distinguished: organic and inorganic gases. $\delta^{13}C_2$ values $\geqslant -27$‰ are indicative of coal-series gases; < -29 ‰, oil-series gases; $-27 \sim -29$ ‰, mixture-type gases. As viewed from the correlations between $\delta^{13}C_1$ values and CH_4 and their homologues, natural gases are classified as biochemical and thermochemical gases. The latter is also classified as gases generated at different evolution stages (degradation, pyrolysis).

2. Distinguished in accordance with the classification figures, natural gases from the major oil/gas pools in the Sichuan Basin can be divided into $J_1t^{4,5}$ maturity late-stage gases of authigenous-source oil series, T_3x maturity late-stage gases of authigenous-source coal series, P_2l^2 high-maturity maixture-source gases of authigenous-source coal-oil series, P_1y high-maturity late-stage – over-maturity early-stage gases of authigenous-source oil series, C_2 over-maturity early-stage gases of allogenous-source oil series, and Z_2b over-maturity early-stage gases of allogenous-source oil series. All of them are concordant with the distribution and characteristics of hydrocarbon-source rocks so far known in the Sichuan Basin. Therefore, this classification scheme is of great potential application in gas exploration.

3. Although natural gases from gas pools in the Upper Permian Changxing Formation are distributed in the range of oil-series gases, their $\delta^{13}C_1 - \delta^{13}C_2$ values are positive, i.e., $\delta^{13}C_1 > \delta^{13}C_2$ ‰. So they are considered to be mixture-source gases. Considering the development of P_2l^1 coal-series in eastern and southern Sichuan and P_1l^2 is situated in the platform area in favour of the development of hydrocarbon-source rocks, P_2l^2 oil-series gases are the result of the extensive dilution by P_2l^1 coal-series gases. As coal-series gases are characterized by high contents of CH_4 and high $\delta^{13}C_1$ values, P_2l^2 oil-series gases are characterized by light isotopic values, although present in small amounts. Once they are both mixed, the coal-series and oil-series gases would be noted for heavy CH_4 isotopic values and light C_2H_6 isotopic values. We distinguished them as high-maturity-stage gases in the light of CH_4 carbon isotopic values of coal series, in consistence with the maturity of organic matter in the Upper Permian series. The explanation has proved itself to be reasonable.

4. Natural gases in the Middle-Lower Triassic series possess characteristics of mixture-source gases. That is to say, they are derived mainly from mixing of oil-series and coal-source gases of high maturity at lower levels.

5. Studies have shown that the favourable hydrocarbon-generating stage for oil-source gases is the high-maturity to over-maturity early stage ($R^0_{max} = 1.35 \sim 2$

~ 2.5 ~ 4%) and that for coal-source gases is the high-maturity late stage – over-maturity early stage. The over-maturity late stage is not only a stage at which hydrocarbon production is minimized, but also a stage which is not favourable to the preservation of preexisting hydrocarbons. Therefore, in the exploration practice attention should be paid not only to the distribution of hydrocarbon-source rocks, but also to the present thermal maturity stage so as to gain better results in gas exploration.

References

Dai Jinxing et al., 1982, The conception and classification of coal-generating gases: Geological Review, v. 28, p.370 — 372 (in Chinese).

Fu Jiamo, 1987, Origin and evaluation of natural gases: Natural Gas Industry, v.7, p.7 — 11 (in Chinese).

Guchalo, L.K., 1981, A Collection of Papers Compiled by the Academy of Sciences of USSR: v.2, 259p. (in Russian).

Huang Jizhong, 1986, Thermally evolved gases and their prospecting in the Lower Archean-Proterozoic in the Sichuan Basin: Natural Gas Industry, v.6, p.8 — 13 (in Chinese).

Huang Jizhong, 1985, Geochemical characteristics of coal-series gases in the Upper Triassic in the western and northern parts of the Sichuan Basin: Natural Gas Exploration and Development, n.4, p.73 — 83 (in Chinese).

Huang Jizhong, 1984, On the occurrence of oil and gas in the Yangxin limestone series in the Sichuan Basin: Journal of Petroleum, v. 5, p. 9 — 18 (in Chinese).

Huang Jizhong, 1985, Geochemical characteristics of natural gases in the Sichuan Basin: Chinese Journal of Geochemistry, v.4, p.343 — 361.

Huang Jizhong, 1991, Geochemical characteristics of natural gases in the Sichuan Basin —— A further discussion: Chinese Journal of Geochemistry, v.10, p.45 — 57.

Huang Jizhong and Jiang Huaicheng, 1986, On the origin of Ordovician natural gases in the Sichuan Basin: Journal of Petroleum, v. 7, p.11 — 23 (in Chinese).

Jiang Huaicheng and Huang Jizhong, 1986, The color of Upper Permian conodonts in relation to the maturity of organic matter in the Sichuan Basin: Acta Sedimentologica Sinica, v.4, p.127 — 137 (in Chinese).

Madconal, G.J., 1983, The origin of natural gas: Journal of Petroleum Geology, v.5, p.341 — 362.

Stahl, W. et al., 1981, Near-surface evidence of migration of natural gas from deep reservoirs and source rocks: AAPG; v. 65, p.1543 — 1549.

Schoell, M., 1983, Genetic characterization of natural gases: AAPG, v.67, p.2225 — 2238.

Shao Jianjun, 1987, Carbon isotopic characteristics of humic-type and sapropel-type natural gases from China: Journal of Petroleum, p.249 — 258 (in Chinese).

Sokolov, V.A., 1965, Formation and Migration of Petroleum and Natural Gas: Moscow, "Nieder", p. 10 — 53 (in Russian).

Tang Xiuyi et al., 1987, Some problems of the composition of coal-generating gas and carbon isotopic characteristics of methane: Annual Report of Lanzhou Institute of Geology, Academia Sinica, p.240 — 252 (in Chinese).

Tissot, B. P. and D.H. Welte, 1978, Petroleum Formation and Occurrence: New York, Springer-Verlag, p. 67 — 224.

Xu Yongchang et al., 1985, Geochemical characteristics of coal-type gases in oil-gas zones in central and northern China: Acta Sedimentologica Sinica, v.3, p.11 — 26 (in Chinese).

Xiong Shouseng et al., 1984, A contribution to the model of multi-action and multi-origin for natural gases in China: Experimental Pectroleum Geology, v.6, p.213 — 228 (in Chinese).

Yakucheni, V. P., 1976, Natural Gases in Sedimentary Rocks: Leningrad, "Nieder" (in Russian).