Geochemical Characteristics of Natural Gases in the Sichuan Basin

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Abstract

The Sichuan basin is one of the largest gas-oil-bearing basins in China. Ool and gas pools occur in Mesozoic, Paleozoic and Proterozoic strata in this basin, with natural gases being dominant.

A good wealth of data from 2000 drill wells on the distribution of natural gases (hydrocarbons: CH_4 $C_2H_6^+$; non-hydrocarbons: H_2S, CO_2, N_2; noble gases: He, Ar) show that natural gases in the basin are predóminated by oil-thermocracked and coal-series gases.

Geological-factor analysis of the geochemical characteristics of natural gases provide evidence suggesting that the occurrence of natural gases, especially dry gases, is attributed to the high maturity of organic matter, and the multi-productive formation has a great bearing on the multi-source rocks; the anomalies of some components (e. g. H_2S) are related not only to the type of primary organic matter, but also to the lithological characters of reservoir beds.

Also discussed in this paper are some geochemical characteristics of coalseries and non-coal-series gases at the same degree of maturat ion, demonstrating that the former is characterized by high proportions of CH_4 and gaseous Hg, high C_1/C_2 ratio, high $\delta^{13}C$, low C_2^+, and high iC_4/nC_4.

The Sichuan basin is one of the important oil and gas pools in China. Oil and gas occur extensively in Mesozoic——Proterozoic strata with the latter being dominant. However, little research has been made on the tempo-spatial distribution and geochemical characteristics of the gas occurrences. This paper is intended to discuss these problems and the affecting factors in an attempt to provide clues for oil and gas exploration.

Distribution and Development of Natural Gases in Time and Space

It is generally accepted that the Sichuan basin was formed during the third episode of the Yanshanian movement on the basis of extensive sedimentation. Recent evidence, however, has suggested that it was formed later during the Himalayan movement[1]. There is a thick sequence of sedimentary rocks in the basin, ranging in age from Sinian to Cretaceous. The sedimentary rocks are well developed except for Devonian and Carboniferous rocks which are absent in some locations. Tertiary sediments are also observed in local places. The total thickness of the sediments reaches more than 10^4m in the western and northern parts. The Middle-Triassic series is underlain by the sediments composed mainly of carbonate rocks of the marine or marine-continental facies. In going upwards from the Upper Triassic they gradually give way to the continental facies dominated by detrital rocks.

Geomorphically, the Sichuan basin is rhombohedron-like with an area of about 2.3×10^5 km². Based on the nature of base rocks, the development of sedimentation and the

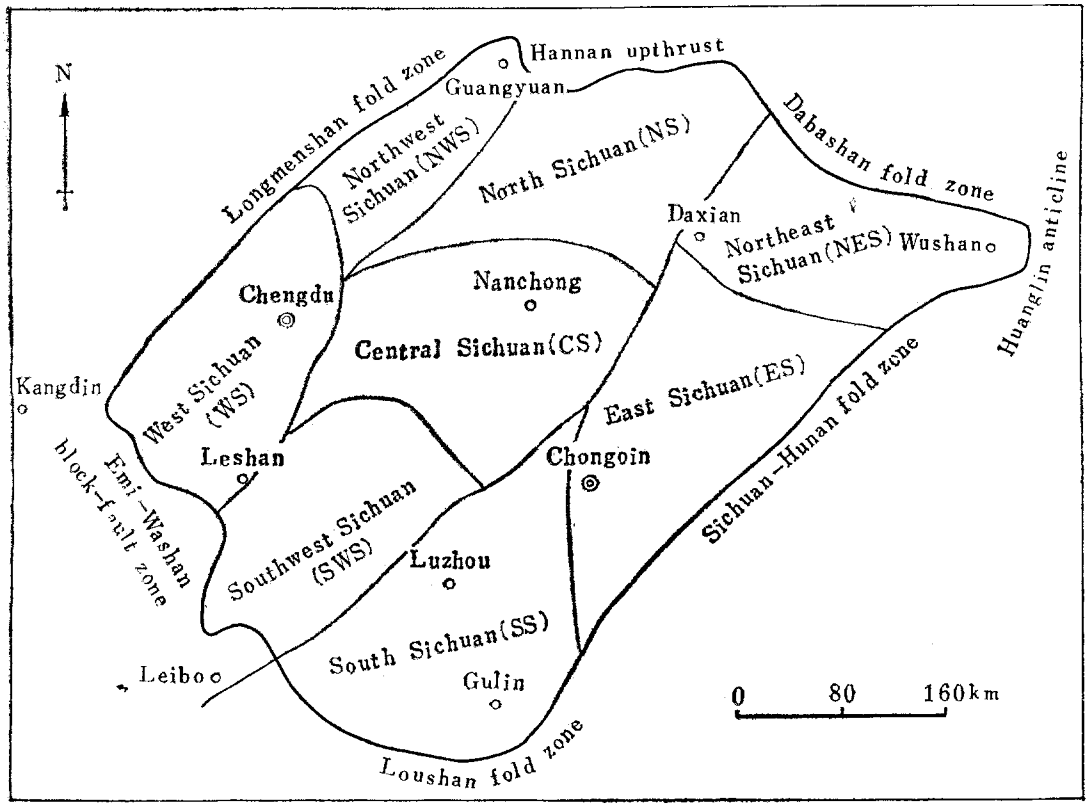

Fig. 1. Sketch map showing the structural division of the Sichuan basin.

structural features, the basin can be divided into three large areas or eight small ones (Fig. 1).

According to the data available from more than 2000 deep wells, gas occurrences can be seen in almost every series, but those of commercial values are concentrated mainly in the Sinian, Permian-Carboniferous and Triassic series. Oil is thie dominant product in the Jurassic series. The deepest gas indication found up till now is at about 7175 m where the oldest strata are pre-Sinian basement rocks (granite). Strata below the Triassic in the basin contain mainly gases, but at the fringe of the same series or in local places oil seepage, oil-bearing soft bitumen, crude oil and condensate can still be observed. As shown in Fig. 2, Lower-Paleozoic oil seepage and soft bitumen occur in the southeastern part of the Longmenshan fault-fold zone in northwestern Sichuan and in the junction of the Dabashan fold zone with the northeastern part of Sichuan. Oil seepage is also observed in the north wing of the Sinian Hannan uplift in the north. In the Upper-Paleozoic strata, in addition to the above regions, oil seepage is also recognized both in the Huanglin geoanticline and on the northwestern side of the Jiangnan paleouplift. The above areas with oil-gas indication are all included in a large sedimentary world.

Condensate, light crude oil and oil seepage are also observed in the core of the Luzhou paleouplift in southern Sichuan and locally in the east of the basin. Oil occurrences are frequently observed in Mesozoic strata in the basin and even oil flows, including

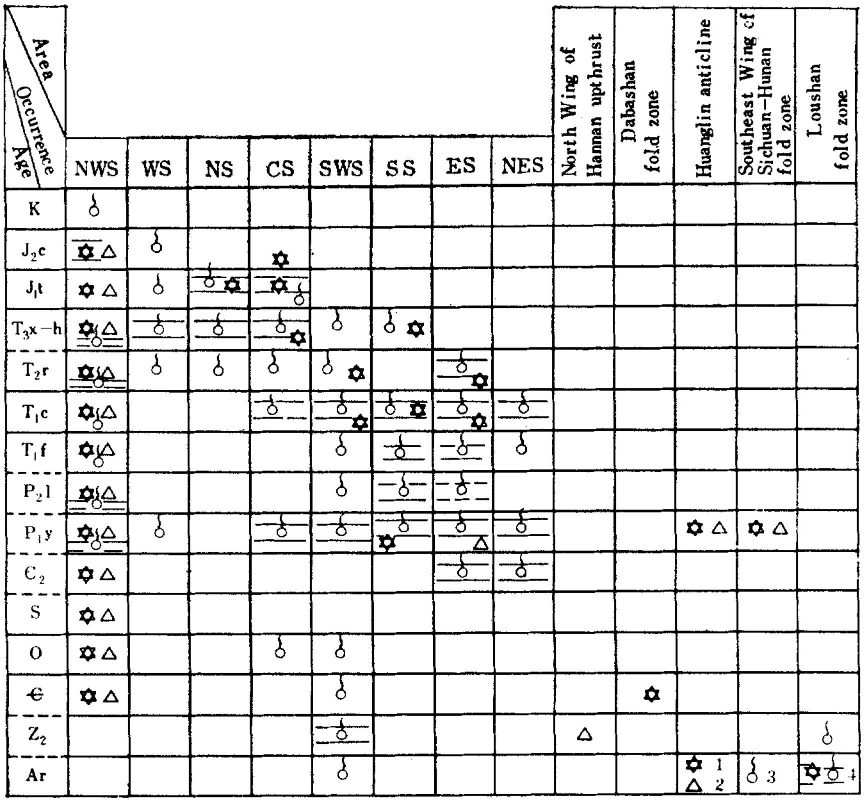

Fig. 2. Oil and gas occurrences in the Sichuan basin.
1. Crude oil, condensate and oil seepage; 2. soft bitumen;
3. gas occurrence; 4. oil-gas pool.

condensate of commercial importance, have been extracted from some regions.

Geochemical Characteristics of Natural Gases

In general, because of the differences in boiling point, critical temperature and critical presure of natural gases with different compositions, CH_4 occurs invariably in the gaseous state under stratigraphical conditions, and so does C_2H_6, but C_3H_8, C_4H_{10}, C_5H_{12} are usually in the liquid state, or exist indenpendently or dissolve in the media. So gaseous components and their contents in oil-gas pools show a tendency of variation in different stages of exploitation. Therefore, the analytical data should be used with cautions. The data chosen by the author are mainly from oil-gas wells under early test or in normal production. The use of analytical data obtained after acidizing operation should be avoided. For acidic gases, because they are water-soluable, used were those analyzed in drilling spots. In the discussion of geochemical characteristics, the general tendency of "group-point" distribution is taken into consideration.

Natural gases from the Sichuan basin consist mainly of CH_4, C_2H_6 and C_3H_8 with minor amounts of C_4H_{10} and C_5H_{12}. The total amount of hydrocarbons in natural gases normally exceeds 95% except in the Sinian strata where it is slightly less than 90%. So they are all combustible gases. Non-hydrocarbon (inorganic) gases are composed mainly of CO_2 H_2S, N_2, He and Ar with a certain amount of gaseous Hg. The principal characteristics of natural gases in different stratigraphical systems and regions are depicted in the following.

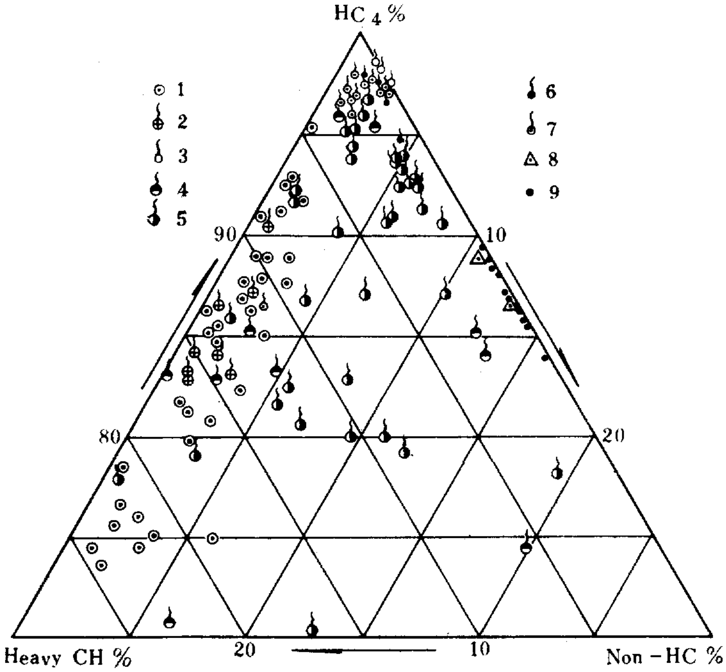

Fig. 3. Triangular diagram of CH_4, heavy HC and non-HC gases.
1. J_{1t}; 2. T_{3x-h}; 3. P_{2l}; 4. T_{2r}; 5. T_{1c}; 6. C_2; 7. P_{1y}; 8. Є; 9. Z_2.

Compositional characteristics of gaseous hydrocarbons

1. Variation of CH_4 and heavy hudrocarbon contents As the strata become old, CH_4 increases remarkably, whereas heavy hydrocarbon content decreases sharply (Fig. 3). Mesozoic samples contain 80—90% CH_4, far away from the highvalue line of non-hydrocarbon gases (except for T_{1c} and T_{2r} in local places). The CH_4 content in Upper Paleozoic samples is generally>95%, but locally <90%, as in Luzhou. Although the CH_4 content in Lower Paleozoic-Proterozoic samples is also <90%, it keeps distant from the high-value line of heavy hydrocarbons (C_2^+) and features a markedly increased non-hydrocarbon content (mainly N_2, CO_2).

If the effect of non-hydrocarbons is not taken into account, the vertical variation as described above will become more pronounced. For example, the proportion of heavy hydrocarbons in total HC is variable in different regions and stratigraphic systems, especially in the same structure or region, or in the same well (Fig. 4). This implies that the content of heavy hydrocarbons tends to decrease with the aging of the strata. High heavy hydrocarbon contents are reported from Mesozoic strata, such as J_{1t} in central

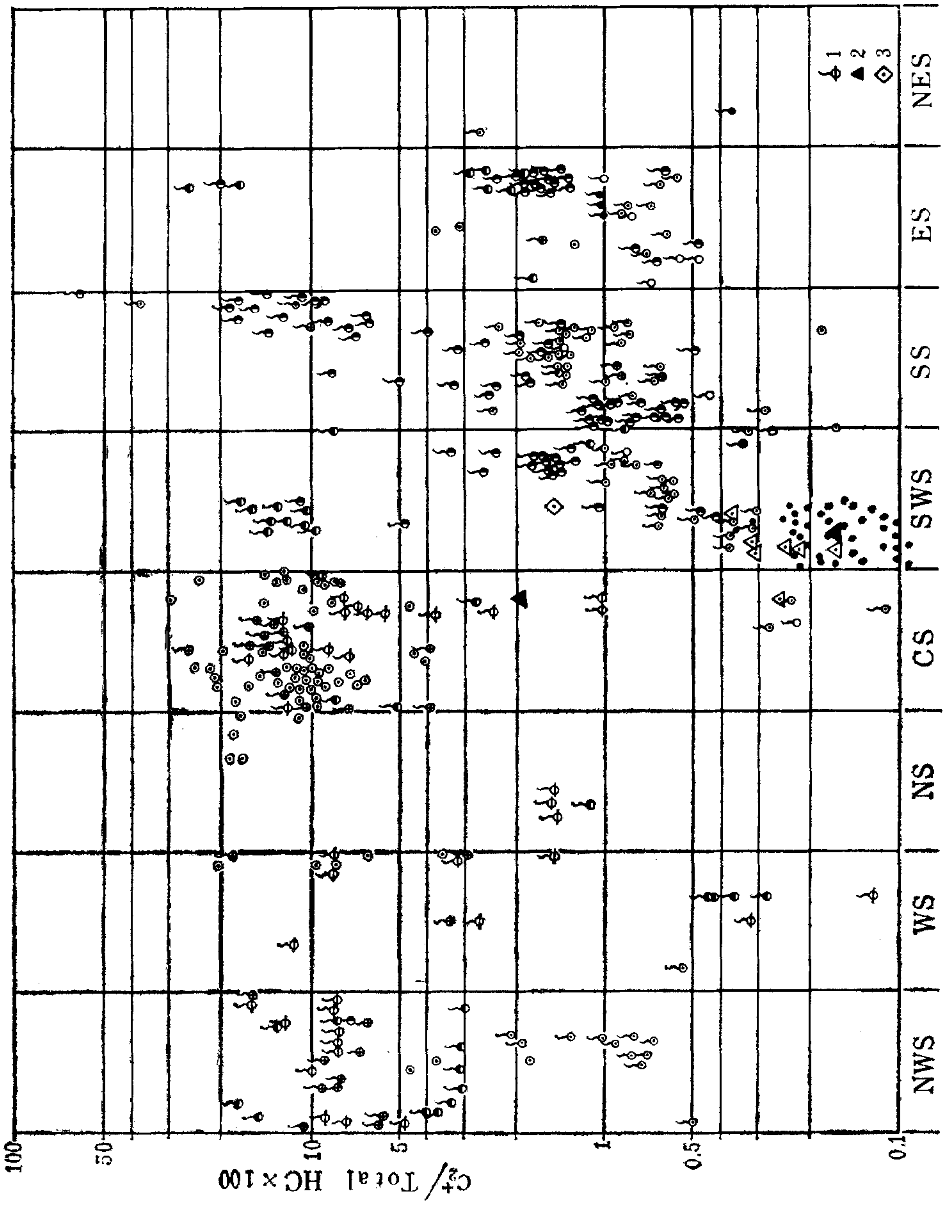

Fig. 4. Percentage of heavy HC in total HC. 1. T_{3r}^{1};2. S-O;3. Ar.
(for the rest explanations, see Fig. 3)

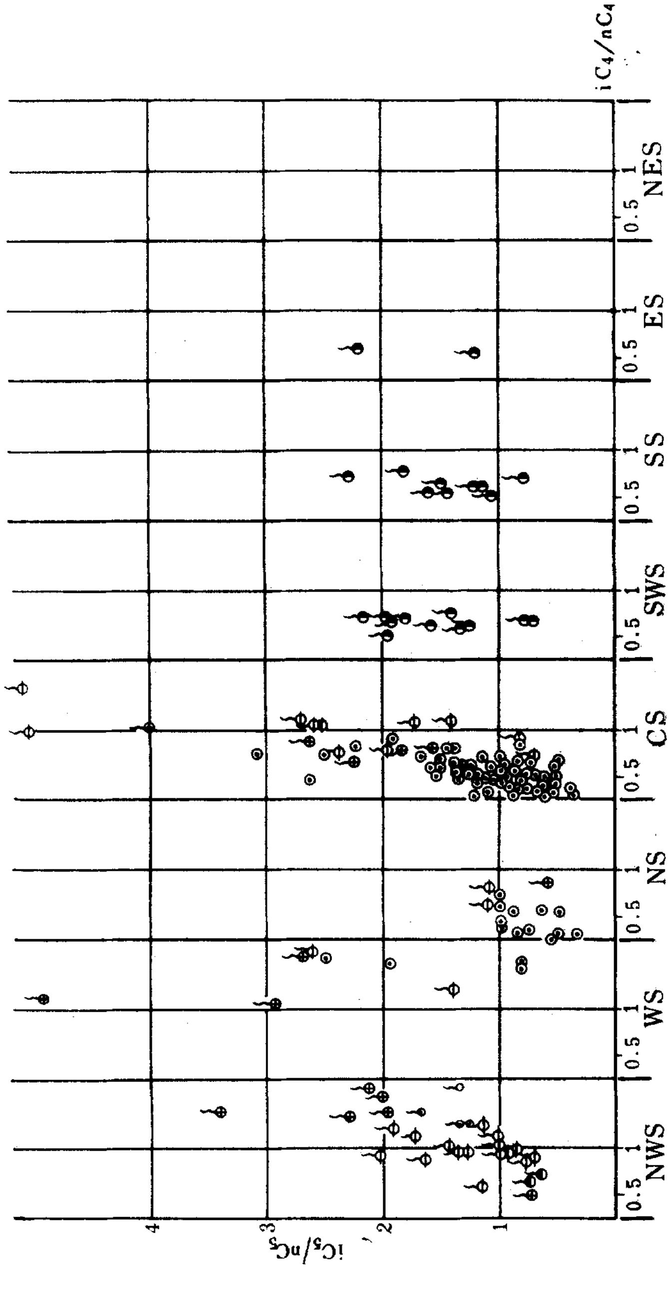

Fig. 5. Ratios of isomer to normal in C_4H_{10} and C_5H_{12}.

Sichuan, T_{2r} and T_{1c} in the Weixi district of southwest Sichuan, Luzhou of south Sichuan and Wolonghe district of east Sichuan, generally higher than 10%. In general, the heavy hydrocarbon content in Upper Paleozoic strata in southeastern Sichuan is about 1—2%, but in the Deshen syncline in the Luzhou district it reaches more than 10%, whereas it is higher in Mesozoic strata than in Paleozoic strata of the same region. In Sinian strata of Proterozoic the heavy hydrocarbon content is usually less than 0.2%, while in Cambrian strata of Lower Paleozoic it is, in most cases, slightly higher than 0.3%. But there are some exceptions. For example, in the T_{3x-h} and P_{21} series the heavy hydrocarbon content is lower than that in the older series of the same region.

2. Variation of normal and isomer in C_4H_{10} and C_5H_{12} As can be seen from the J_{1t}, T_{3x-h}, T_{2r} and T_{1c} series containing more heavy hydrocarbons, the isomer content of alkane is usually higher than normal. The same holds true for most other regions and series where the ratio iC_5/nC_5 is frequenty greater than 1 (mostly >2 in T_{3x-h}). Exceptions were only seen in T_{3h} 2 of central Sichuan, T_{3x} of northwest Sichuan, J_{1t} and T_{3x-h} of west Sichuan where the iC_4/nC_4 ratio is frequently greater than 1 (Fig. 5).

3. Variation in $\delta^{13}C_{PDB}$ of CH_4 As can be seen from the relationship between C_1/C_{2+3} and $\delta^{13}C_1$ (Fig. 6), the degree of maturation of organic matter increases with the age of the strata, and so does the $\delta^{13}C_1$ value. This agrees with the ranges for crude oil, condensate——wet gases and dry gases divided by Shaokaili[2]. $\delta^{13}C_1$ values of the gases associated with the J_{1t} oil-pool associated gases in central Sichuan are within the range of —42——58‰. In going northward to Bajiaochang the maturity increases and $\delta^{13}C_1$ values fall within —36——42‰, corresponding to the range for condensate-wet gases. $\delta^{13}C_1$ values for Paleozoic strata are between —20——36‰, corresponding to the range for dry gases. Those for Triassic strata also

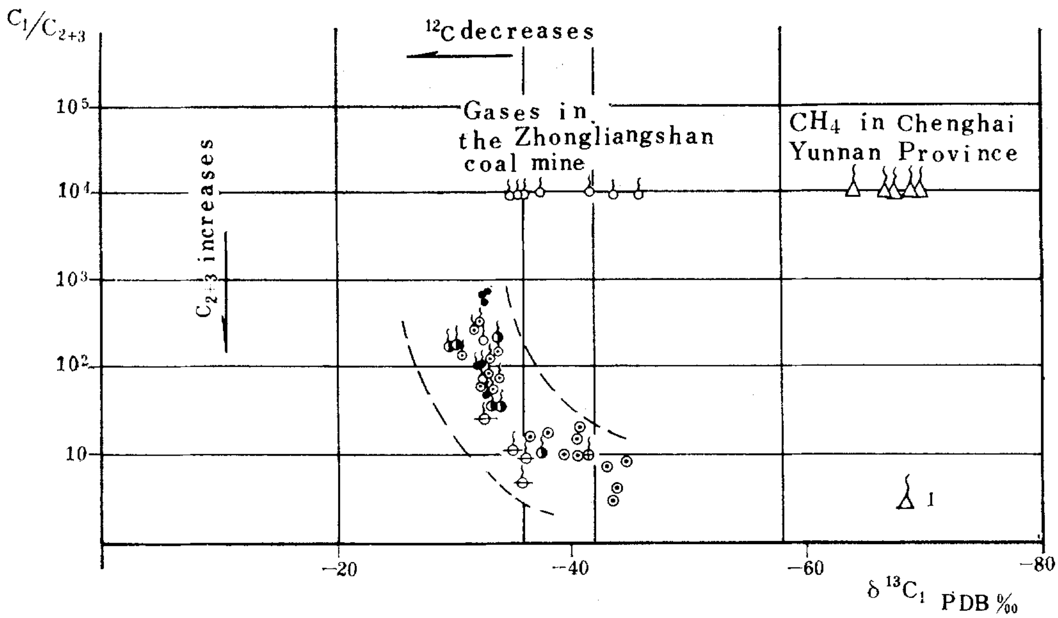

Fig. 6. Relationship between C_1/C_{2+3} and $\delta^{13}C_{1PDB}$‰.
1. Q. (for the rest explanations, see Fig. 4)

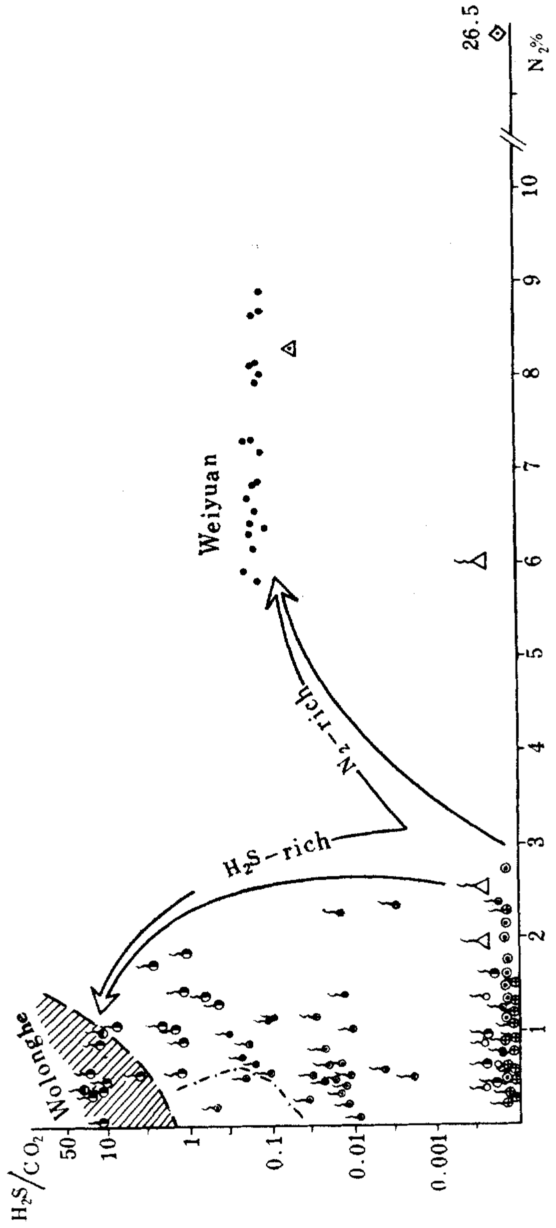

Fig. 7. Relationship between H_2S/CO_2 and N_2 (for the explanations, see Fig. 4).

fall within -36——$-42‰$. $\delta^{13}C_1$ values of the gases in coal bed P_{21} in Zhongliangshan near Chongqing are around $-38‰$, which is distinguishable according to its extraordinarily lowC_1/C_{2+3} value ($\sim10^4$) (Fig. 6). $\delta^{13}C_1$ values for Sinian strata are also high (rich in ^{13}C), which can be distinguished from those for Upper Paleozoic strata by higher C_1/C_{2+3} ratios (>600). This rule is consistant with the mechanism of isotopic fractionation, i.e., in general under thermal action the older the strata, the higher the temperature or the longer the heating process. Thereby, the free-energy of ^{13}C is low relative to ^{12}C, so the former is more stable. The order of stability can be expressed as ^{13}C-^{13}C $>$ ^{13}C-^{12}C $>$ ^{12}C-^{12}C.

Compositional characteristics of non-hydrocarbon gases

1. Variation of acidic components The acidic components are predominated by CO_2 and H_2S/CO_2 is usually less than unity. But the natural gases in T_{1c} and T_{2r} of Middle and Lower Triassic are enriched in H_2S, especially in the gas pools of the Wolonghe region in east Sichuan where the H_2S/CO_2 ratio reaches about 10, and one cubic meter of natural gas contains about 100 grams of H_2S. In contrast, the natural gases in Triassic and Jurassic strata contain no or little H_2S (Fig. 7).

2. Variation of N_2 The N_2 content in Sinian strata is within the range of 6—9%, but mostly less than 2% in other strata. N_2 contents listed in Fig. 7 are all based on the relation $a=\frac{Ar\ 100}{N_2\ 1.18}>1$. Because the ratio of N_2 over Ar in the air is 78.03:0.932, i.e., Ar is 1.18% of N_2 in concentration, a ratio of less than 1 indicates that N_2 is of non-air origin.

Compositional characteristics of noble gases He and Ar

1. Gaseous He According to the statistics made by B. P. Tissot and D. H. Welte[8], the He content in Paleozoic strata (609 gasfields) is >1000 ppm, that in Mesozoic strata (1983 gasfields) $100\sim1000$ ppm, and that in Cenozoic (178 gasfields) <100 ppm. The distribution of He in the Sichuan basin (Fig. 8) agrees with the above statistical rule, i.e., it is greater than 1000 ppm in the Sinian and Cambrian series and 100—1000 ppm in the Triassic and Jurassic series. But in the Carboniferous and Permian series it is lower than statistical value, generally within the range of 200—800 ppm. T_{3x-h} and J_1 in some local regiens have a He content range characteristic of Cenozoic or Mesozoic strata. In addition, as can also be seen from the figure, the He content in Permian and Carboniferous strata of Upper Paleozoic is similar to that in Middle-Lower Triassic strata. In view of the fact that He originates from the decay of radioactive elements (U, Th, K) and is accumulated with the aging of strata and occurs with the enrichment of soluble gases and free gases, the older the strata, the higher the He content (known as the "stratum-accumulation effect"). Disagreement of He variation in gas pools with this rule may probably be accounted for by the poor closure of the strata which led to the diffusion and thus decrease of He, or by the migration of deep gas sources which caused the increase of He in the new-hosting strata.

2. Gaseous Ar In natural gases Ar takes two forms —— one is of air origin and the other of radiogenic origin, so it is usually a mixture. According to the data obtain-

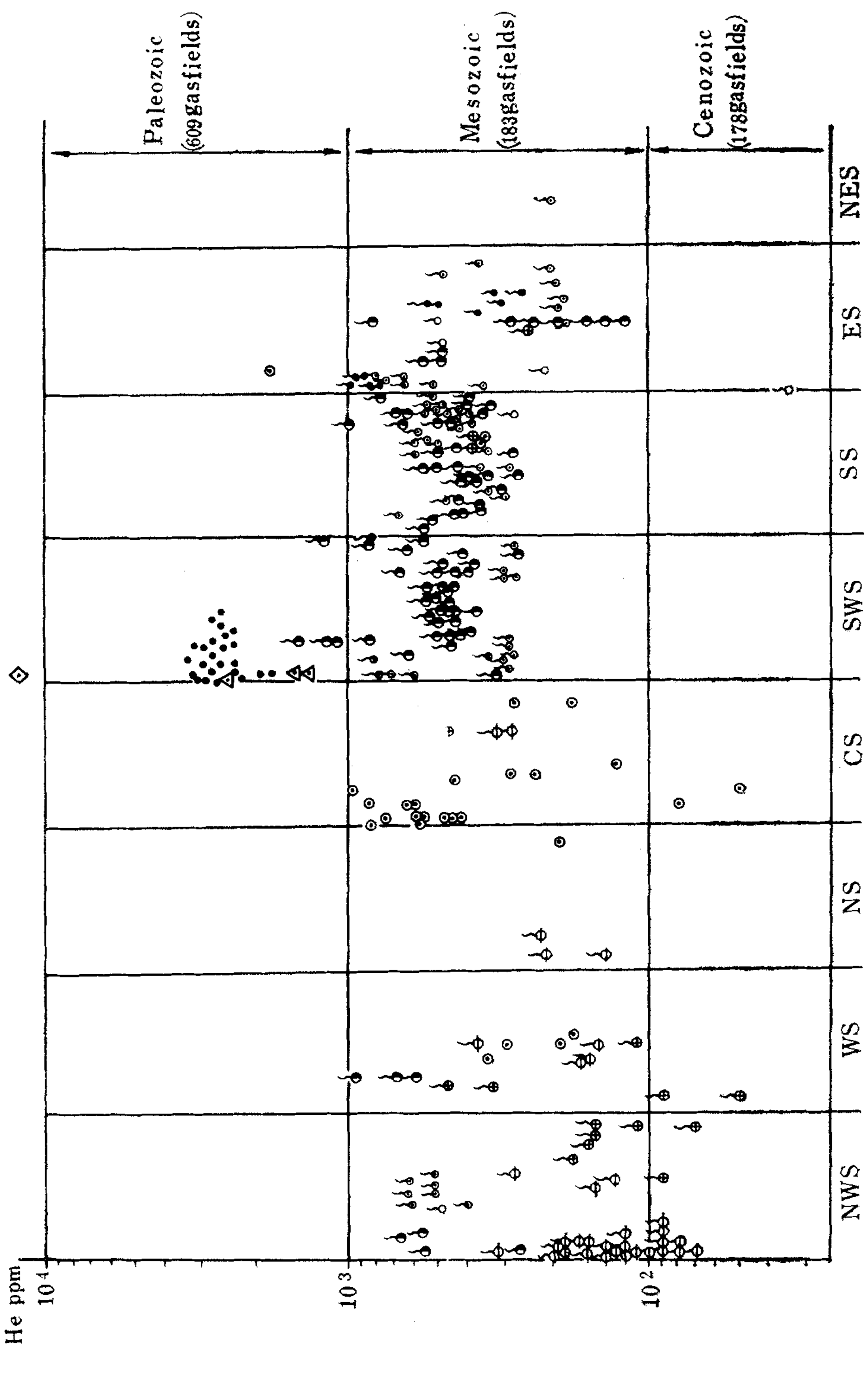

Fig. 8. Distribution characteristics of He in different series (according to the statistical values for 970 gasfields through out the world, after B. P. Tissot and D. H. Welte; for the explanations,see Fig. 4).

ed from 60 wells in the Sichuan basin, Ar_{air} and He/Ar increase, but Ar_{radio} and $^{40}Ar/^{36}Ar$ decrease with the aging of the strata.

Table 1.

Variation of Ar in Samples of different ages and from different areas**

Age \ Area / Item	Southwest Sichuan (SWS)				South Sichuan (SS)*				East Sichuan (ES)			
	Ar_{radio}	$^{40}Ar/^{36}Ar$	Ar_{air}	He/Ar_{radio}	Ar_{radio}	$^{40}Ar/^{36}Ar$	Ar_{air}	He/Ar_{radio}	Ar_{radio}	$^{40}Ar/^{36}Ar$	Ar_{air}	He/Ar_{radio}
J_1									96	686	74	18.23
T_3	36	445	80	9.74								
$T_{1,2}$	32~70	435~1057	17~181	6.11~14.57	32~67	758~1348	15~27	7.73~15.63	25~63	442~1371	15~92	8.57~20.80
P_1	36~45	694~1069	17~21	5.59~12.40	37~55	268~1148	14~37	9.64~12.93	19~58	568~1422	13~57	5.22~23.18
Z_2	363~468	4440~9255	12~34	5.39~6.14								

* Data from T_{1c} only.

** Revised from the data of Lanzhou Institute of Geology, Academia Sinica (1978) (in ppm).

Characteristics of gaseous Hg abundance

From Fig. 9 plotted according to the data of the Ministry of Geology and the Lanzhou Institute of Geology, Academia Sinica, it is evident that the general tendency of variation in Hg content of natural gases in the Sichuan basin is: Hg is higher in terrestrial facies than in marine facies, in detrital rock formations than in carbonate formations, and in coal series than in non-coal series. Natural gases in the Upper Triassic coal-series of terrestrial facies have the highest Hg, mostly amounting to more than several thousand ng/m^3 (of 28 wells, 19 contain more than 1000 ng/m^3 of gaseous Hg) with the maximum being 39000 ng/m^3 (according to the Lanzhou Institute of Geology, the maximum is 50000 ng/m^3). In Triassic, Permian and Carboniferous marine-facies carbonate gas pools the Hg content generally reaches several hundred ng/m^3 (of 41 wells, 29 contain more than 100 ng/m^3 of Hg) with the maximum being 3600 ng/m^3 (according to the Lanzhou Institute of Geology, the maximum is 1250 ng/m^3). In Sinian dolomite gas-pools the Hg content is usually low (in 50% out of 10 wells the Hg content is less than 100 ng/m^3 with the maximum being 8085 ng/m^3). Natural gases in oil pools occurring in the Jurassic non-coal series have a high Hg content. All the three wells tested contain more than 1000 (1440, 2133, 2680) ng/m^3 of gaseous Hg. As shown above, the Hg content in natural gases is much higher than its average in the air (3.6 ng/m^3 determined by Geological Brigade 101, Ministry of Geology), indicating that all the Hg determined falls within the Hg range of natural gases (Fig. 9).

Analysis of Geological Factors

As stated above, the characteristics of natural gases in the Sichuan basin are described in the following:

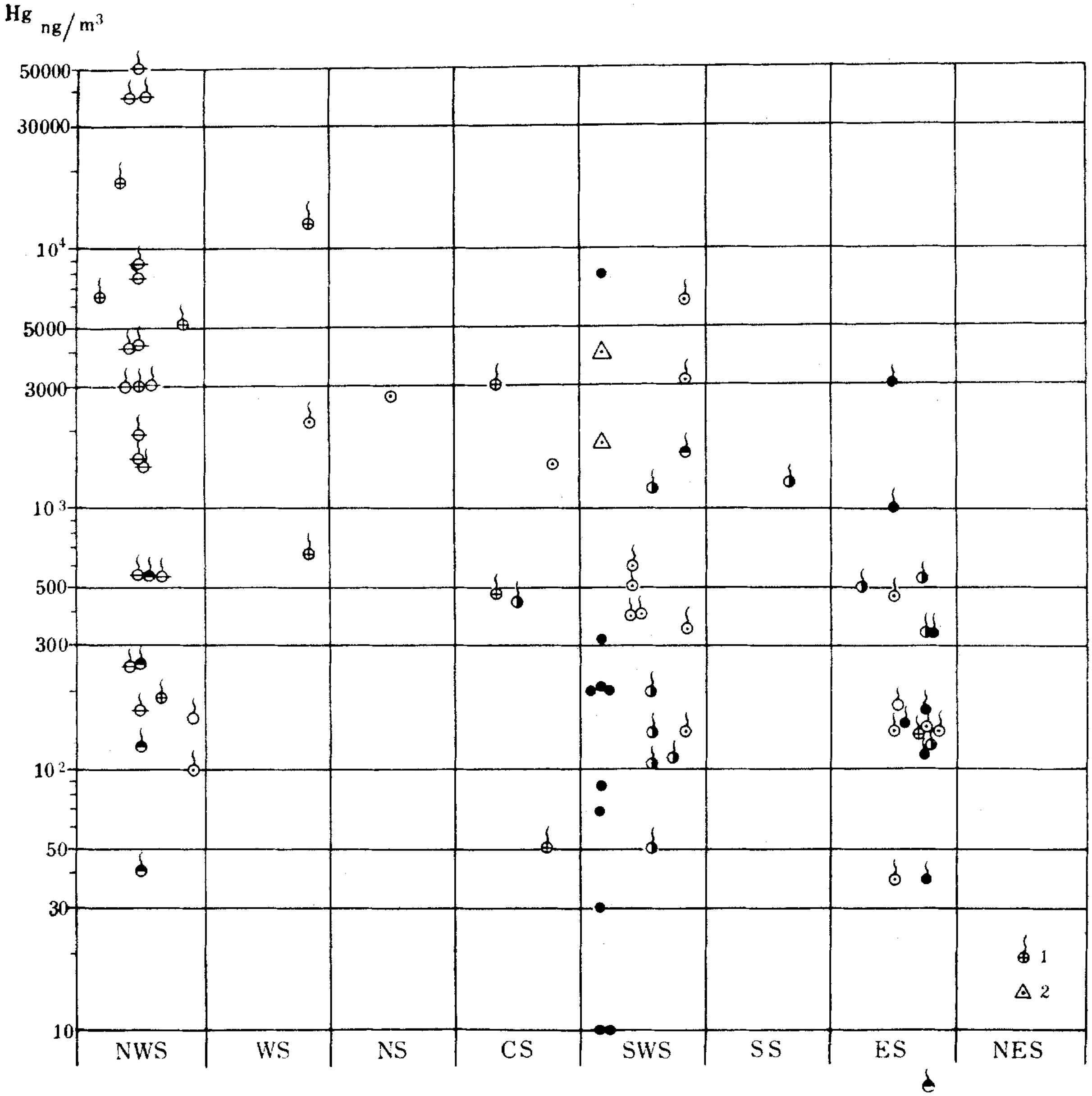

Fig. 9. Gaseous Hg content in natural gases from different series (for the rest explanations, see Fig. 4). 1. $T_{3x}^{4}\ \frac{T_{3h}^{4}}{T_{3h}^{2}}$; 2. $Є_{2+3}$.

1. The maturity of organic matter is a main factor affecting the compositional variation of natural gases. Temperature and time are the essential causes which determine the maturity of organic matter. Oil and gas were derived from the decomposition of organic matter (kerogen) under the action of temperature and time, and further evolved with increasing maturity in the sequence of oil→condensate→wet gases→dry gases. The characteristics of Sichuan natural gases in both longitudinal and lateral distributions have reflected the generality of the mechanism described above. In the northwestern

part of the basin the maturity of organic matter is lowest ($R^o<1.35\%$)[4], so oil seepage, soft bitumen and bitumen veins occur either in Paleozoic or in Mesozoic strata there. Again, in the core of the Luzhou paleouplift the Upper Paleozoic strata still remain at the early stage of high maturity ($R^o=1.35-2.00\%$), and, as a consequence, light crude oil and condensate were observed in Permian limestone beds at Yangxin, where the content of heavy hydrocarbons in natural gases is increased remarkably. But in the other regions of southeast Sichuan remaining at the late stage of high maturity ($R^c=2.00\sim 2.50\%$) no condensate was observed and the content of heavy hydrocarbons in natural gases is, in most cases, around 1%. In going northward to Laoguanmiao, the H_2S content in natural gases also tends to increase relative to the regions mentioned above. Judging from $R^o=3.199\%$ determined from $P_{21}1$, the strata should designated to the early stage of overmaturity ($R^o=2.50\sim 4.00\%$). Up to Mesozoic, the vast area of central and southeast Sichuan remained at the mature stage. so crude oil or oil occurrences were extensively discovered in the Jurassic series. In north Sichuan crude oil has generally evolved into condensate —— wet gases because of deep burial and high maturity. Crude oil or condensate also occurs in the Triassic series in southeast Sichuan where the content of heavy hydrocarbons in natural gases is generally higher than that in the productive beds of the underlying Paleozoic strata. The major commercially productive region in the Lower Paleozoic and Sinian strata is the Weiyuan gasfield where the R^o value of carbonaceous bitumen is greater than 4.60%, indicating a late stage of overmaturity ($R^o=4.00-6.00\%$). Thus, the gas occurrences in the field are predominated by dry gases with a small amount of CH_4. Meanwhile, the composition of carbon isotopes in CH_4 is typified by a high $\delta^{13}C_1$ value which is indicative of the increase of maturity. Therefore, it should be assigned to the product of the late stage of thermal evolution (pyrolysis).

In short, the distribution and development of oil and gas and the geochemical characteristics of natural gases are all closely related to the maturity stages. This may well explain the obvious longitudinal zonation of natural gases (Table 2).

2. The type of source matter directly affects the compositianal characteristics of natural gases The geochemical characteristics of natural gases vary with the degree of maturation, which is reflected mainly in oil and gas derived from the source matter (type-I and -II kerogen). If the source matter responsible for natural gases is composed mainly of dispersed organic matter or humic coal of type III the geochemical characteristics of natural gases also vary accordingly, but don't agree with the maturity stages of type -I and -II source matter. In other words, crude oil facies does not necessarily appear at the mature stage, instead, condensate-wet gases or dry gases will be the major products. For example, organic matter in the Upper Triassic series is a mixed type composed mainly of type-III source matter, and, as a consequence, although this series in central or northwest Sichuan is at the mature stage ($R^o<1.35\%$), condensate-wet gas assemblage is still observed and CH_4 has heavy values (generally around—36‰) as such is the case with the J_{1t} mature stage in central Sichuan, but $\delta^{13}C_1$ values for oil-pool associated gases derived from type-II source matter are generally lower (—40——40‰). For another example, in the coal formations of the Upper Paleozoic Longtan Group which are extensively developed in central and south Sichuan, organic matter is of humic type and the gas pools in the overlying Chanxing limestones in the Luzhou dis-

Table

Relationship between the maturity (stage) of organic matter

Age	Maturity		Natural gas type		$C_2H_6^+$/ Total HC%	
	Low maturity area	High maturity area	Low maturity area	High maturity area	Low maturity area	High maturity area
J_{2c}	Immature ~ NWS	Mature ~ CS		Associated gas		20±
J_{1t}	Early mature stage ~ CS	Late mature stage ~ NS	Associated gas	Wet gas	10~20±	15±
T_{3x-l}	Late mature stage ~ NWS	Early stage of high maturity ~ NS	Wet gas	Dry gas	10±	<5
T_{2r}~T_{1c}	Early stage of high maturity ~ SS	High maturity stage ~ SES	Wet gas	Dry gas	10~15±	<5
P_{21}^{2}	Early stage of high maturity ~ SS	Late stage of high maturity ~ SES	Dry gas with high CH_4	Dry gas with high CH_4	1.5±	<1±
P_{1y}	Early stage of high maturity ~ SS	Late stage of high maturity ~ SES	Wet gas	Dry gas with high CH_4	10±(2±)	1±
C_2	Early stage of high maturity ~ ES	Late stage of high maturity ~ NES and ES	Dry gas with high CH_4	Dry gas with high CH_4	1±	<1
Є$_{2+3}$	Early stage of over-maturity ~ Weiyuan	Early stage of over-maturity ~ CS	Dry gas	Dry gas	0.3±	0.25±
Z_2	Late stage of over-maturity ~ Weiyuan	Late stage of over-maturity ~ CS	Dry gas with relatively low CH_4 (C_1<90%)	Dry gas with relativeyl low CH_4 (C_1<90%)	<0.3	<0.2

* South Sichuan is referred mainly to the Luzhou region; all the data cited are those with a

2.

and the geochemical characteristics of natural gases

Dryness moduls C_1/C_2^+		$\delta^{13}C_1$‰		Note	
Low maturity area	High maturity area	Low maturity area	High maturity area		
	<5				Maturity R^0%
5±	10±	−44±	−40±	Oil-pool associated gas (thermocataly tic gas)	<0.50 (immature stage)
10±	20±	−35±	−33±	Gases from Type III and Type II-III kerogen	0.50 ≀ 1.00 (early and late stages of maturity) ≀ 1.35 ≀ 2.00 ≀ (early and late stages of high maturity) 2.50 ≀ 4.00 ≀ (early and late stages of over-maturity) 6.00
<10 (>10±)	50±	>−36 (−34±)	−32±	In low maturity area only wet gases occasionally occurring	
100±	>200	P_{21}^{2} −33± −38 P_{21}^{1} ∼ −39	−32±	Gases from Type-III kerogen dominant	
10± (50±)	200±	(−34±)	−32±	In low maturity area only wet gases occasionally occurring	
100±	200±	−33±	−32±	Low maturity area referred to Xiangguoshi	
300±	400±	−32±		Gas-occurrence in CS	
>500	>1000	−32±		Gas-occurrence in CS (more than 99.5% CH_4 if nonHC gases are excluded)	

frequency of>50%.

Table 3.

Comparison between oil-thermocracked and coal-series gases

Age	Area	Maturity stage	Heavy HC/Total HC%	Dryness module C_1/C_2^+	Isomer/Normal C_4 >1	Isomer/Normal C_4 <1	Isomer/Normal C_5 >1	Isomer/Normal C_5 <1	Carbon isotopic value $\delta^{13}C_1$‰	Carbon isotopic value δC_2‰[1]	Gaseous (Hg(-ng/m^3)) Measured by Ministry of Geology	Gaseous (Hg(-ng/m^3)) Measured by Lanzhou Institute of Geology, Academia Sinica
$J_{1t}{}^{5}_{4}$	CS	Mature stage	10~15±	5±	1.4%/1	100%/2 98.6%/70	50%/1 56.3%/36	50%/1 43.7%/28	(-43.6~-44.8) -44±/4 (-40.2~-43.0) -40±/3	(33.4~-34.0) -34±/4 (-26.7~-30.3) -29±/3		1440/1
$T_{3x}{}^4T_{3h}{}^4T_{3h}{}^2$	NWS	Mature stage	5±	>10	100%/5		100%/5		(34.5~-38.6) -36±/4	(-25.1~-27.4) -27±/3 (CS)	(5067~18150) >5000/3	188/1
$T_{3x}{}^2$	Zhongba, NWS	Late mature stage	10±	10±	39.0%/9	61.0%/14	43.5%/10	56.5%/13	(-34.8~-36.2) -36±/7	(-25.0~-26.7) -26±/7	(170~39200) >2000±/11	(250~50000) ≥3000/4
$T_{2r}{}^3$ $T_{2r}{}^1$	Zhongba, NWS	Early stage of high maturity	5± 15±	20± 5±		100%/2	50%/1	50%/1	-34.5/1		(41~550) <500±/4	
C_2	Wolonghe' ES	Late stage of high maturity	1±	200±	no	C_4~C_5			(-32.2~-32.3 -32±/2	-35.6/1	(4~330) <200/3	38/1
Z_2	Weiyuan, SWS	Late stage of overmaturity	<0.3	>500	no	C_3~C_5	C_3-0~tr.		(-31.6~-32.8) -32±/7	(30.8~-35.3) -32±/4	(10~8050) <300/9 <100	88/1

trict of south Sichuan belong to the early stage of high maturity, but neither condensate nor wet gases are found, and the heavy hydrocarbon content is extremely low (usually ~1%). That is precisely because the hydrocarbons derived from humic matter consist mainly of gaseous hydrocarbons, and even though some liquid hydrocarbons and heavy hydrocarbons could be formed during coalification, they could be hardly released owing to being adsorbed on coal beds or carbon-high beds. Therefore, light crude oil and condensate are frequently observed in the P_1^3 limestone-gas pool in the same region, and the content of heavy hydrocarbons in natural gases also tends to increase. So it is of no doubt that these findings are all related to the type of source matter.

3. Some geochemical characteristics of natural gases in the coal series

Take natural gases in the Upper Triassic coal series in northwest Sichuan for example, for more data are available. Stratigraphically, the series is composed of two gas-generating assemblages (upper and lower). In the series, $T_{3x}^{1,3,5}$ consists of dark-colored argillaceous rocks and contains abundant coal beds or coal lines, which constitutes the parent rocks and also serves as a cover and $T_{3x}^{2,4}$ consists of grey sandstones and serves as a reservoir; T_{3x}^{1-3} is the lower assemblage and T_{3x}^{3-5} the upper assemblage. The series itself is a complete system for gas formation, preservation and capping. Natural gases in the upper assemblage seem unlikely to have come from external sources. This makes it possible to discuss the geochemical characteristics of coal-series gases. Industrial gas flows have been discovered one after another in the structures of Zhongba, Yuquan, Xiaoquan, Laoguanmiao, Wenjiachang and Tobachang. They have now become the major gas pools in the region. In the following we'll compare coal-series gases with oil-pool associated gases in the adjacent Jurassic series as well as with oil-thermocracked gases in the Carboniferous series in the Wolonghe region, east Sichuan and those in the Sinian series in Weiyuan, southwestern Sichuan. As can be seen, they feature high dryness module (high CH_4), high content of gaseous Hg, high superiority of isomer, high carbon isotopic value and low contant of heavy hydrocarbons (Table 3.)

1) High dryness module: Comparisons made in the same region, especially in the same structure of between adjacent regions at similar maturity stages indicate that T_{3x} has a module of 10± or >10, T_{2r}^3 20±, and T_{2r}^1 and J_{1t} in central Sichuan 5±, indicating that the coal series strata are rich in CH_4.

2) High gaseous Hg: The enrichment of gaseous Hg is generally thought to be characteristics of the gases formed in coal series strata, especially during coalification. For the time being, its formation mechanism is interpreted by such a process that a considerable amount of Hg was adsorbed on organic matter during coalification, and then the adsorbed Hg was gasified as the temperature rised and took part in the migration and accumulation of natural gases. As can be seen from Table 3, the Hg content in T_{3x-h} gases is mostly greater than 2000 or 5000 ng/m^3 with the maximum being 5000—39200 ng/m^3, which is the highest value so far reported.

3) High isomer superiority: Statistics on isomer superiority, which is so called when iC_4/nC_4 and iC_5/nC_5 are greater than 1, indicates that the ratios in gases from 5 wells in the upper gas assemblage T_{3x-h} are all greater than 1, from 23 wells in the lower gas assemblage are, for about 40%, greater than 1, and for J_{1t} and T_{2r}^3

about 50% of the sites tested have a iC_5/nC_5 ratio of greater than 1; the rest all exhibit normal superiority.

4) High carbon isotopic value: The $\delta^{13}C$ value of CH_4 in T_{3x} is generally —36‰, greater than that in the overlying J_{1t} (—40, —44‰). The $\delta^{13}C$ value of C_2H_6 is quite different from that of oil-thermocracked gases. It usually falls within the range of —26～—27‰. So the variation of ^{13}C value seems to have an important bearing on the type of gas-generating source matter.

5) Relatively low heavy hydrocarbon content: In natural gases in the upper assemblage heavy hydrocarbons comprise 5% of the total HC, while in the overlying J_{1t} and underlying T_{2r} it amounts to 10—15%, which runs counter to the interpretation on the basis of maturity stages, i.e., it is not only lower than that at the same maturity stage, but also than that at higher maturity stages. The geochemical characteristics of coal series gases mentioned above reflect the basic characteristics of the coal series in Upper Triassic strata. Their source matter is believed to be composed mainly of type-III kerogen or type-II-III kerogen with type III dominant. Their precursors are some higher plants, such as coniferous and broadleaf trees and pteridophyta which are typified by the enrichment of xylon and cellulose (usually >40—50%), the depletion of H, low H/C ratio, high O, N and S, dominating aromatic HC structure, high carbon isotopic value (close to the average of woods—25‰)[5], low saturated HC in soluble organic matter and S/A ratio <1.

4. Great influence of wall-rock assemblages on the acidic composition of natural gases

The characteristics of wall-rock affect remarkably the composition of natural gases, especially the content of gaseous H_2S (Fig. 6). Natural gases rich in H_2S occur in the Middle-Lower Triassic series. The profile associated with gas pools consists of gypsum and carbonate interbeds, the former serving as a good cover for the gas pools and also creates an environment favourable for the reduction of sulphates in the presence of hydrocarbons. This feature is consistent with the general tendency observed elsewhere in the wold——H_2S in natural gases tends to increase in the strata where the carbonate-sulphate assemblage appears.

5. Influence of the age of gas pools on the composition of noble gases in natural gases

The noble gases He and Ar in gas pools stem mainly from the decay of radioactive elements in the strata[6]. They were accumulated at the time of formation of gas sources, and gathered, migrated and renewed along with soluble and free gases, thereby the gas sources could be traced. As evidenced by the data from the Sichuan basin, the abundances of noble gases in Paleozoic, especially in Lower Paleozoic strata coincide with the statistical levels given by B. P. Tissot[3], i.e., He content is higher than 1000 ppm and, in Sinian strate, it falls mostly around 2500 ppm; 9r content is as high as 363—468 ppm with the $^{40}Ar/^{36}Ar$ ratio standing between 4440 and 9255, which makes the Sinian strata distinguished from the Paleozoic ones. There is almost no difference in He and Ar contents between Upper Paleozoic strata and Middle-Lower Triassic strata. With a few exceptions (e.g. J_{1t} in Shapingba, east Sichuan) the He and Ar contents in Mesozoic strata also agree with the statistical abundances[3], i.e., He falls within the range of 100—1000 ppm. The above distribution characteristics of the noble gases are in good agreement with the development and distribution of oil-gas source strata in the Sichuan basin. The oil source rocks are mainly Lower Cambrian and Lower Silurian dark claystones, Lower Permian clay and micritic limestones, and Lower Jurassic dark claystones. The gas source rocks are principally the Upper Permian and Upper Triassic coal series. It may be concluded that the differences in formation time for different gas sources may reflect the above-described differences in noble gas composition for natural gases.

Conclusions

1. Oil and gas in the Sichuan basin are of extensive distribution in time and space from Sinian to Jurassic with natural gases being dominant.

2. Natural gases in the Sichuan basin consist mainly of oil-pool associated and coal-series gases. Their formation is closely related to maturity stages. Longitudinally, they are mainly related to the high-maturity and over-maturity stages except for Jurassic gases which are related to the mature stage. They also feature a clear zonation.

3. Significant differences are noticed in geochemical characteristics between the coal-series gases and the oil-pool associated gases. For instance, coal-series gases in the Upper Triassic strata of northwest Sichuan are typified by high dryness module, high gaseous Hg content, high isomer superiority and high carbon isotopic value, and low heavy hydrocarbon content.

4. The type of wall-rock assemblage has an important bearing on the composition of non-hydrocarbon gases. For example, the profile of carbonate-sulphate rocks is favorable to the enrichment of H_2S.

5. The distribution characteristics of noble gases are more or less consistent with the ages of oil-gas sources. In other words, the compositional distribution of noble gases is related to the age of a gas source.

References

[1] Te Tianhua, Natural Gas Industry (in Chinese), 2(1982), 2—12.
[2] Zhang Zishu, Geology-geochemistry (in Chinese), 3(1980), 51—57.
[3] B. P. Tissot and D. H. Welte, Petroleum Formation and Occurrence, New York, 1978, 189—190.
[4] Huang Jizhong and Zhang Zishu, Petroleum Exploration and Development (in Chinese), 1(1982), 12—25.
[5] Organic Geochemistry, Ed. Laboratory of Organic Geochemistry, Inst. of Geochem., Academia Sinica, Science Press, Beijing (in Chinese), 1982, 189—195.
[6] Natural Gases (in Japanese), translated by Japanese Association of Natural Gas Mining, 11(1981), 29—35.