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RESEARCH ARTICLE

Deep-seated gas generation and preservation condition in China

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Abstract Deep-seated gas is defined in this paper as natural gas generated under the combined action of high temperature, high pressure, and environment media. As to organic matter vertical evolution, deep-seated gas is natural gas, which is generated and deposited under the position of an oil generation window. Deep-seated gas exploration is an important potential field for oil-gas exploration. Also, it is an inexorable trend to the further development of oil and gas provinces. In this paper, the authors will examine and distinguish the concept of deep-seated gas, and discuss the distribution and gas source of deep-seated gas. It is pointed out that kerogen, assemble dissoluble organic matter and disperse dissoluble organic matter all have contributed to deep-seated gas generation, especially disperse dissoluble organic matter in composite and superimposed sedimentary basin. In the end, according to the structural evolution and hydrocarbon source rock depositional distribution, the distribution of deep-seated gas in China is predicted.

Keywords deep-seated gas, oil generation window, gas source, disperse dissoluble organic matter

1 The conception of deep-seated gas

The definition of deep-seated gas in exploration should be in agreement with the idea of that in research. In exploration work, deep-seated gas can be regarded as a technical qualification, but in oil and gas geology research, it is difficult to give a simple definition. In a given sedimentary basin, deep-seated gas should have an unambiguous definition. The existence of inorganic origin methane is widely recognized, but the resources for exploitation are mostly CO_2 and N_2. By far, there is not enough evidence for unmixed inorganic origin methane gas reservoir that can meet the requirement of an industrial gas reservoir. Thus, the deep-seated gas studied in this paper is mainly organic methane.

It is necessary to define the concept of deep-seated gas before deep-seated gas generation and preservation condition are discussed. Since the concept of deep-seated gas was defined by Sugisaki (1981), the research of natural gas in deep sedimentary formations has attracted many oil and gas geologists at home and abroad. Shi et al. (2003) summarized in detail the study of deep-seated gas and had pointed out its importance. Deep-seated gas, just as its name implies, is the gas generated and accumulated in deep sedimentary formations. According to the present viewpoints, the upper limit of the depth of the buried formation is from 2 800 m to 5 000 m. Each viewpoint has its own geologic background and practical significance. Different sedimentary basins have different main depth circumscriptions of oil and gas generation, so deep-buried formation of deep-seated gas just contains a relatively wide sense of depth. While the buried formation is being discussed, another question is that deep-seated gas not only includes “deep generated and deep accumulated” gas but “deep generated and up accumulated” gas. It is said that the gas can still be called deep-seated gas, if its production formation is shallow but its source is in a deep formation. Through the above analysis, deep-seated gas can be definitely defined as natural gas generated under the combined action of high temperature, high pressure, and environment media, especially the natural gas generated and deposited under the position of an oil generation window.

The theory of oil generation window based on the zonation of oil generation states that the main oil generating zone, which depends on sedimentation rate and earth temperature conditions, is determined by the zone of earth temperature 70–120°C. The end of the main oil generating zone corresponds to the stages II–III of catagenesis (*R*o, 1.10%–1.15%). The main oil generating zone becomes the main condensate oil generating zone, and with the increase of depth becomes

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the dry gas generating zone (Mielienievskij, 1999). In studies, the geochemistry marker and vitrinite reflectivity (*R*o) are commonly used to represent the degree of evolution. *R*o usually varies between 0.4% and 4.0% and *R*o = 1.34% is believed to be the critical value of the existence of oil. If the geothermal gradient is 2.5–3.0°C/100 m, the generation of mass liquid hydrocarbon comes to the end and gaseous hydrocarbon begins to generate when the depth is about 4 000–5 000 m. Tissot and Welte (1984) and Xu (1994) respectively established the typical pattern of oil and gas generation according to the organic evolution. With the development of hydrocarbon prospecting and geology theory, the markers about the threshold of liquid hydrocarbon and gaseous hydrocarbon generation, especially about the preservation conditions of deep-seated liquid hydrocarbons, show their limitations.

2 The generation of deep-seated gas

2.1 The source of deep-seated gas

Samvielav (1995) believed that there are two kinds of deep-seated gas: one is from deep-seated liquid hydrocarbon degradation and the other is from dispersive kerogen thermal evolution at the catagenetic stage. Exploration practice and simulation experiments indicate that liquid hydrocarbon is able to degrade into gaseous hydrocarbon at high temperature and pressure conditions. The degradation of early generated liquid hydrocarbon in an oil reservoir is an important source of deep-seated gas. While reaching oil generation peak, as a whole, the total amount of oil generated continuously decreases and that of gas generated continuously increases. As will be readily seen, there is not only kerogen degradation but oil degradation at the stage of condensate oil and wet gas and the main production is oil degradation and wet gas. But at the dry gas stage, there is primary hydrocarbon degradation and the main product is methane.

Deep-seated dispersed organic matter consists of soluble organic matter (dispersed oil), insoluble organic matter (kerogen) and deep-seated dispersed insoluble organic matter which mainly refers to kerogen. Kerogen at the stage of high evolution degree is a key factor for deep-seated gas resource. Its chemical constitution determines the composition of its main hydrocarbon products. Zhou et al. (1997) experimented with the hydrocarbon generation capacity of different kinds of kerogen. The results show that when the experimental temperature is 350°C (up to *R*o = 1.2%), the III kerogen will release all the alkyl carbon and hydrogen, 3.5% alkyl carbon remains in the II kerogen and 9.0% alkyl carbon still remains in the I kerogen; when the temperature is 500°C (up to *R*o = 21.2%), less alkyl carbon and more alkyl hydrogen remains in the I kerogen with a certain extent of hydrocarbon generation potential. The simulation experiments indicate that the main product is methane if *R*o > 2.0%. McNeil (1996) believed that the main product of the remaining kerogen is almost pure methane after it releases oil. In their kerogen model, the alkyl joining aromatic ring can easily creak between the first carbon atom and the second carbon atom to produce methyl, and the alkyl rejoins the aromatic ring and then leads to gas-enriched methane in the form of broken-off methyl. This model can explain why the abundance of methane always increases at the stage of any temperature not just the stage of bitumen generation peak.

The different kinetic models of petroleum formation (Horefield et al., 1992; Pepper and Corvi, 1995; Pepper and Dodd, 1995; Behar et al., 1997; Knauss et al., 1997; Tsuzuki et al., 1997) show that the low earth temperature conditions of sedimentary basins are good for deep-seated gas generation. The III kerogen always generates more gas than I kerogen in any other earth temperature conditions. The oil remaining in source rocks generate gas as much as III or II kerogen at low temperature (1°C/Ma) conditions which can be a source of deep-seated gas, but it generates less gas than any kind of kerogen if the earth temperature is higher than 10°C/Ma. Hydrous and nonhydrous simulations indicate that gas generated by accumulation of oil is four to seven times as much as by kerogen if the earth temperature is 1°C/Ma, and it is six to twenty times if the earth temperature is 10°C/Ma, which is consistent with the thermodynamics stability of oil in these conditions (Fig. 1). Simulations indicate that water can increase the stability of oil, and water widely exists in geological conditions, thus it is good for the formation of deep-seated gas. Though oil can generate much more gas than equiponderant kerogen, the amount of kerogen in deep-seated source rocks is a few orders of magnitude as much as that of crude oil. Thus, the kerogen at high degree of evolution is still the main gas source.

Dispersed oil refers to the soluble organic matter preserved in a geologic body, which is "hand-in-glove" with the continuity of gaseous hydrocarbon generation. There are two kinds of occurrence of dispersed soluble organic matter. One which coexists with insoluble organic matter and never departs from the geologic body (source rocks) preserving the insoluble organic matter (kerogen) consists of the soluble organic matter originally sedimented, and the soluble organic matter formed during the process of evolution but never departed from the ancestral body. Soluble organic matter originally in sediment is the primary source of immature-low mature oil (Huang, 1996; Xia and Zhang, 2001). The soluble organic matter formed during the process of evolution is the primary source of kerogen cracked gas. This kind of soluble organic matter contributes to the generation of deep-seated gas. The other kind of soluble organic matter is derived from insoluble organic matter (including soluble organic matter originally sedimented) with the increase of evolution degree, and part of it accumulated as soluble organic matter oil reservoir but most of it dispersed in the migrating channel or dissipated through many ways. Through simulation, Li (2004) concluded that oil generation accumulation ratio is generally 5%–10% and the gas generation accumulation ratio is generally 0.5%–1.0%, that is, over 90% of hydrocarbon dissipates during the process of accumulation. The migration, accumulation and dissipation are continuous processes. Part of the dissipated hydrocarbon as soluble organic matter remains in the migrating channel and geologic body and part of it has been dissipated to the atmosphere or has been destroyed by faulting, denudation or biodegradation, which appears as the show of oil and gas, seep oil or rock with bitumen. The accumulated hydrocarbon can be divided into industrial hydrocarbon reservoir and non-industrial hydrocarbon reservoir. The industrial hydrocarbon reservoir refers to the reservoir with commercial exploitation value but most of the hydrocarbon exists as non-industrial dispersed form and in this condition the hydrocarbon appears as oil spots or smears. Accumulated soluble organic matter (hydrocarbon reservoir) is also able to appear as dispersed soluble organic matter after catagenesis. If the traps were destroyed during the geological process, partial or entire hydrocarbons can be dissipated by biodegradation or water wash, which leads to oil in reservoir becoming bitumen and reservoir bitumen, and sandstone with bitumen is the product of this process. Solid bitumen is dispersed in rocks as amorphous phase and all these dispersed soluble organic matter, except tar sand, cannot be used as resource. But, this kind of dispersed soluble organic matter dispersed in a geological body can be the source of gas in certain conditions. With the simulation experiments about cretaceous bitumen from the Karamay oil field in Xinjiang, Feng and Wu (1995) found that one third of the bitumen turned into light oil while the temperature was 400°C and the primary product is gas if the temperature is higher and the conversion can reach one third of total bitumen. This means that bitumen can be an excellent source of gas and generates hydrocarbon as a secondary function.

2.2 Influence of organic-inorganic interaction on deep-seated gas

During the generation of deep-seated gas, the influence of organic-inorganic interaction on the two primary kinds of gas sources—dispersed soluble organic matter and accumulated soluble organic matter—is hydrogenation and catalysis. At the beginning, liquid hydrocarbon is being generated; the hydrogenation is not a key factor, but with the increase of thermal evolution degree and product, the carbon chain becomes short, the ratio of H/C increases and now it needs more hydrogen. Hydrogenation is the result of organic-inorganic interaction and the joining of external hydrogen will accelerate macromolecular compound, for example, the kerogen turns into micromolecule hydrocarbon for less hydrogen in organic matter than in hydrocarbon. The chemical process is simply a process of destroying original organic matter by being hydrogenated. The key factor of gas generation in big superimposed basins with high evolution degree is the source of hydrogen. Hydrogenation is important for gas generation. There are two sources (organic and inorganic) of hydrogen. Hydrogen from organic sources is formed in the thermal evolution of organic matter, especially in the biochemical process but it usually appears as a compound for its active chemical property. The hydrogen from inorganic source is the primary source of hydrogenation and its source is water. It is believed that hydrogen from water is related to the thermal evolution of sediment organic matter that challenges the traditional viewpoint that organic original hydrogen in source rocks is the key factor of hydrogenation for hydrocarbon generation. The pyrolysis of water, decompounds of water in radiate condition, catalytic reaction of water with ferrous oxide at temperatures of 300–500°C, and deep-seated fluids are all origins of hydrogen. All these kinds of hydrogen migrate to sediment organic formation with tectonic movement and magma movement can be the important source of organic matter hydrogenation for generating hydrocarbons. The Fe^{2+} in sedimentary rock has notable acceleration for hydrogenation, so abundant Fe^{2+} is an excellent catalyst for hydrogenation. However, clay mineral is the important catalyst at low temperatures. Therefore, hydrocarbon generation is the result of the organic-inorganic interaction. The carbon of hydrocarbon is from organic matter but its hydrogen is partly from syn-sedimentary water decompound, deep-seated supply, rock chemical reaction, and so on. The essence of hydrocarbon generation with organic-inorganic interaction is that the carbon is abundant. However, hydrogen is deficient at the stage of high evolution degree of organic matter generating hydrocarbon and it offers part of the hydrogen. For example, it has been found that there is hydrogenation of kerogen by fluid with abundant hydrogen (Yang and Jin, 2001; Jin et al., 2002). The catalysis of clay mineral and transition metals, such as Ni, Co, Fe, Cu and some metal oxides, can accelerate the combination of hydrogen from an inorganic source with organic matter to form hydrocarbon (Frank and Joe, 1997; Frank, 2001).

It is generally recognized that clay mineral is a kind of effective catalyst. Its main characteristic is that it has obvious absorptive capacity and allows clay to become an organic complex. The mineral catalyst can accelerate kerogen pyrogeneration and its adsorption cannot be ignored either. The absorbability of the mineral makes chemical reaction with sediment organic matter easily and forms organic-inorganic complex. The adsorption and flocculation are important for quick sedimentation and preservation of organic matter. The formation of organic-inorganic complex is an important geochemical factor for catalytic cracking of organic matter to generate hydrocarbons (Li et al., 2002).

3 Preservation condition of deep-seated gas

3.1 Trap condition of deep-seated gas

Before the preservation condition is discussed, the temperature and pressure zone in which gas can steadily exist must be ascertained. Baskin (1997) believed that methane can exist at the temperature of 750°C, for gas is steadier than oil in

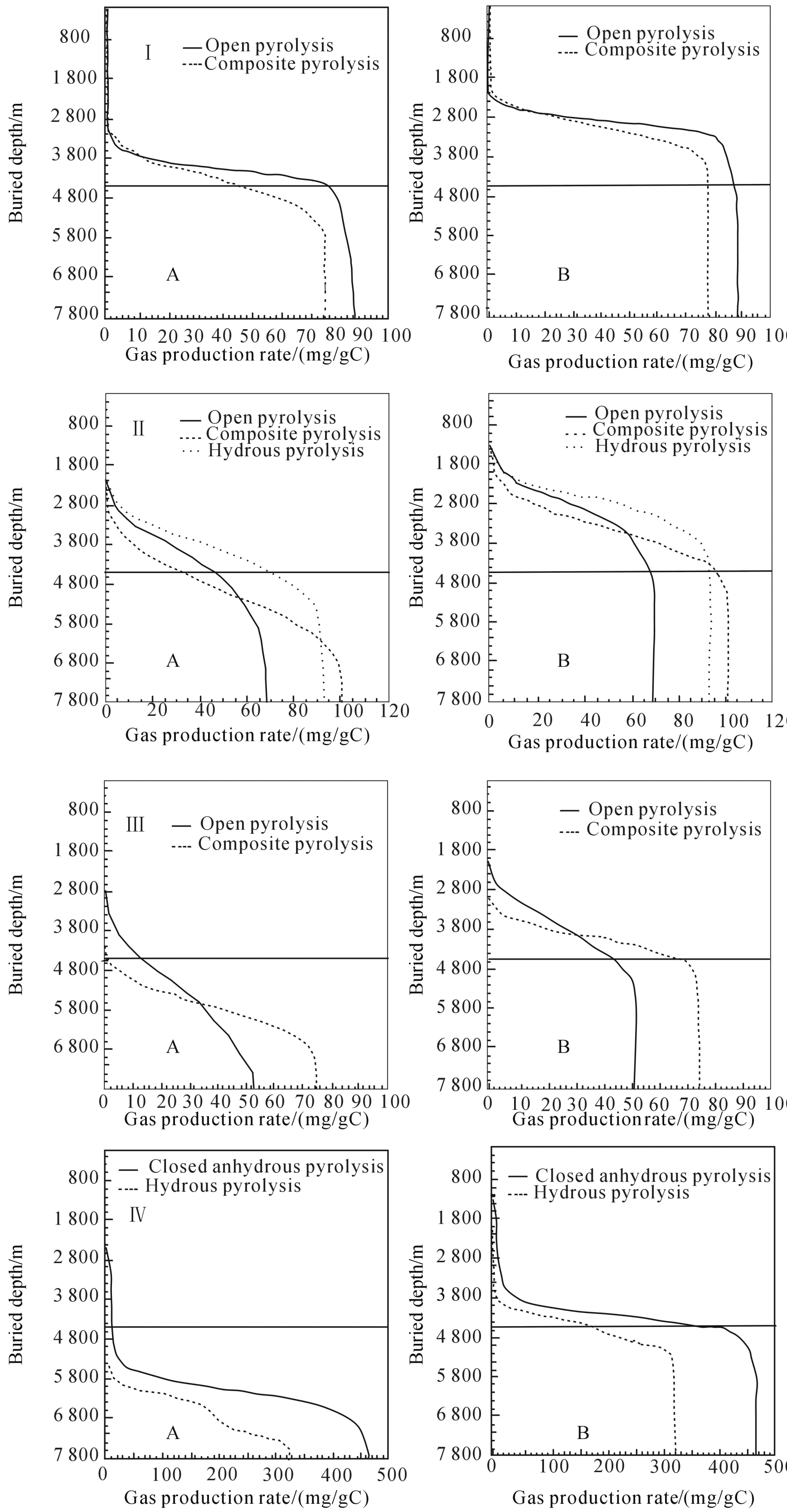

Fig. 1 Gas production rate (C_1–C_5) of different types of kerogen and oil in different simulation experiment conditions with buried depth changing.

I: I type kerogen; II: II type kerogen; III: III type kerogen; IV: oil; *A*: 1°C /Ma; B: 10°C /Ma (according to Horsfield et al., 1992; Pepper and Corvi, 1995; Behar et al., 1997; Knauss et al., 1997; Tsuzuki et al., 1997, and perfected ratherish) deep-seated condition. Oil has its own disappearing threshold, so it cannot be preserved deeply. However, gas can be preserved even at the bottom and there is evidence for it. Since the factor of deep-seated gas generation is related to the depth of its generation and degree of evolution and the gas generation can be accumulated in both deep and upper reservoirs, the preservation condition of deep-seated gas must first meet the condition of common gas reservoirs. It is still determined by trap condition whether gas has accumulated to the reservoirs or not. The primary traps of deep-seated gas are structural traps (Qiu et al., 1997). The different basins have different force mechanisms and evolution histories, so the traps for deep-seated gas in these basins are different and multifarious. For example, the Bohai Bay Basin is the tectonic basin with extensional stress on the base of paleoplatform, so the traps are mainly formation-burial hill traps; the basins in the central and west of China are under compressive stress, so the deep-seated gas trap is always connected with complicated anticlines. Deep-seated fluid is more or less reform reservoir strata. Thermal fluid after magma, volcano thermal fluid, metamorphic thermal fluid and underground water thermal fluid can fill up the pores to result in compactly cemented strata which can be local cover strata. The stratum heterogeneity and mineral differential diagenesis results in diachronous sealed formation and abnormally pressured fluid compartment, which is a favorable condition for deep-seated gas traps.

3.2 Evaluation on reservoir and seal condition of deep-seated gas

Under compaction and diagenesis and with the increase of depth, primary pore decreases gradually and the primary pore hardly exists. However, the induced pore becomes the primary reservoir space at the stage of late diagenesis if the depth is more than 4 000 m. Qiu et al. (1997) has summarized that there are four kinds of mechanisms of induced pore: dissolution of organic acid and carbon dioxide acidic water, supergene leaching under a plane of unconformity, dissolution of organic acid formed from thermochemical sulphate reduction, and dissolution for underground water heat cycle and convection. All these can change the physical property of a reservoir for the better. In some deep basins and sags where the depth is 4 000–5 000 m, such as the Yinggehai Basin and Qikou Sag, the porosity of the reservoir sand bed is as high as 18%–20% (Tian and Li, 1997). According to the experience from deep well drilling, there is no evidence to show that the rock seal capability weakens irreversibly, including the disappearance of rock seal capability as a result of irreversible de-aquation, compacting action or the formation of cracks in the transition from mudstone to slate. The representative characteristic of deep structure is that there is no connection between the lower structural layer and upper structural layer. For example, the upper and lower gas combination in the Sichuan Basin and the upper and lower structural style in Kuqa depression are inconsistent. Because of high temperature, abnormal pressure, high fluid viscosity and micromolecule hydrocarbon, the deep hydrocarbon reservoir needs better seal conditions than the middle and upper hydrocarbon reservoirs. The regional seal can compact the hydrocarbon into different systems of beds to generate serial reservoirs with different depths and at different strata. For example, the middle Triassic gypsum rock as the regional seal has vertically sealed all the gas (there is only dry gas in Triassic and lower strata) in the Paleozoic group. As another example, the Upper Permian mudstone as the effective regional seal has vertically sealed gas in the Paleozoic group in Shaanxi-Gansu-Ningxia Basin (Schimmelmann et al., 1999, 2001).

4 Distribution of deep-seated gas in China

Because of its particularity, deep-seated gas is mainly distributed in inherited superimposed basin, reformed superimposed basin, intermediate massif basin, and foreland basin. Deep-seated gas reservoirs discovered in China are distributed in different strata from Precambrian to Neogene and the reservoir rocks are mainly carbonate rocks.

4.1 Distribution of high evolution deep-seated gas

In China, there are four large gas-bearing domains: the Lower Paleozoic—Precambrian basins mainly formed with carbonate rocks; the Upper Paleozoic—Triassic gas-bearing domains mainly formed with carbonate rocks and coal measure strata; the Neocene—Quaternary gas-bearing domains mainly formed with bio-thermocatalytic transitional zone gases; and secondary hydrocarbon reservoirs. Two of the four domains are formed with source rocks under Paleozoic. Though Dai et al. (1997) have indicated that the two domains do not necessarily belong to the category of deep-seated gas, the gas in continuously subsided basins is attributed to deep-seated gas indubitably.

There are mainly marine carbonate rocks in Paleozoic in China but there are also non-marine clastic rocks from Mesozoic to Cenozoic, such as in the Sichuan Basin, Chuxiong Basin, Ordos Basin, Tarim Basin, and Bohai Bay Basin. Most of these basins successively came into being from Mesozoic to Cenozoic, which leads to the better preservation of source rocks from Proterozoic to Paleozoic and continuous hydrocarbon thermal evolution. Most of the hydrocarbon is highly matured or over matured. This makes it an important source of deep-seated gas in China. But the hydrocarbon reservoirs in Proterozoic and Paleozoic in basins where the tectonic movement is strong from Mesozoic to Cenozoic have been reformed repeatedly. As a result, the paleo-reservoirs have been greatly reformed and the hydrocarbon is complexly distributed after having migrated and accumulated many times.

The Proterozoic and Paleozoic source rocks are mainly carbonate rocks and dark mudstones. They are mainly distributed in the central and western China (Sichuan Basin, Ordos

Basin, and Tarim Basin) and North China in the east of China. In the Tarim Basin, limestone with dark mudstones from Cambrian to Lower Ordovician are mainly distributed in the Manjar region and the organic matter is mainly type I and over matured at present; the carbolic marine-terrigenous facies are mostly distributed in the southwest of the Tarim Basin and the organic matter is mainly type III at the stage of turning from highly matured to over matured. The Middle and Upper Ordovician limestone with dark mudstones are mainly distributed in the central and northern Tarim Basin. The organic matter is mainly type I and exists in the oil generation window which is believed as the main source of marine oil by some scientists. In the Sichuan Basin, all the source rocks that include the Lower Cambrian black mudstones and carbonaceous shale, the organic-matter-enriched graptolite shale, and the Lower Permian plumbeous biolithites are at the stage of being over matured. In the North China craton the Lower Ordovician lithites, Benxi Formation and Taiyuan Formation in Upper Carboniferous, coal formations in Lower Permian Shanxi Formation developed. The lithologic characteristics are various and there are dark mudstones, coal formations and carbonate rocks. In the Ordos Basin, the two series of source rocks are at the stage of turning from highly matured to over matured and mainly generate dry gas. In North China, the maturity is different in different regions for different fluctuating extent and there are likely second generation and accumulation. The above series of source rocks can be the source of deep-seated gas and control the distribution of deep-seated gas.

4.2 Distribution of deep-seated gas in sediments rapidly buried foreland basins

Foreland basin refers to the sedimentary basin between orogeny anterior border and craton. The foreland basins are advantageous regions for hydrocarbon generation, but the geologic condition of the foreland basins in China is very complex for most of them are intra-continental foreland basins. The terminal collision always controls huge sediments which are favorable for generating deep-seated gas. There are huge sediments from Mesozoic to Cenozoic in the Kuqa depression in the Tarim Basin. The complex thrust structures formed at the Himalayan stage control the hydrocarbon distribution and there is an abundant gas resource in the Kuqa depression where much deep-seated gas has been discovered. The molasse in the foreland basin before the Kunlun Mountains is 4 000–7 000 m and the southwest Tarim foreland thrust zones control the hydrocarbon distribution from the Mesozoic to Cenozoic in the southwest Tarim Basin. Some oil and gas fields, such as the Kekeya oil and gas field, have been found. Other foreland basins are advantageous regions for deep-seated gas, such as the foreland basin in the southern border of the Junggar Basin, the foreland basin in the western margin of Ordos Basin and the Longmen Mountain foreland basin.

4.3 Distribution of deep-seated gas in some basins in China

According to the above analyses, it can be concluded that coal formation, III kerogen and low geothermal gradient are advantageous conditions for deep-seated gas, so coal formations with low ground temperature in the middle and west of China are the main source of deep-seated gas. There are also some mainly preserved in Paleozoic and other age-old formations. The buried depth of deep-seated fault zones is over 10 000 m in the Songliao Basin. It is near the heat source of the earth's mantle, which results in high geologic temperature. The broad distribution, huge thickness and high thermal conductivity lead to shallow depth of the gas reservoir, but there are still some deep-seated gas generation conditions (Li, 2004). The discovery of the Changde deep-seated gas field in 1998 and the present breakthrough in Xujiaweizi prove the large potential of deep-seated gas in the Songliao Basin. There are 30 sags with advantageous targets for deep-seated gas exploration. There are 13 deep-seated gas reservoirs in the Jizhong depression and seven of them are oil reservoirs, two of them are gas cap reservoirs, three of them are condensate oil reservoirs and the depth of the deepest reservoir is about 5 200 m (Tuo et al., 1999; Tuo, 2002). Some explorations have been done and some traps and reservoirs have been discovered in some other depressions in the Bohai Bay Basin.

The Sichuan Basin has oil and gas generating formations, and most of the depth of the major gas reservoir is within the range of that of deep-seated gas. The Paleozoic east of the Sichuan Basin and the complex traps in the west margin of Huaying Mountain in the middle of Sichuan Basin and the largest Le Mountain-Longnv Temple uplift is the most likely deep-seated exploration target for the future and some industrial hydrocarbons have been found in some regions. In the south and southwest of the Sichuan Basin, the Lower Paleozoic is the new long-term exploration region for its large resource potential and low exploration degree.

The Paleozoic formation located north of the Ordos Basin is a major exploration target for deep-seated gas and the west border of the Ordos Basin is also a likely region. The Tarim Basin has the largest amount of gas in China. According to exploration instances in the past ten years, there are lots of targets in the Tarim Basin. Most of the gas resource is in the deep formation with buried depth of over 4 000 m. Kuqa depression, Tabei uplift, Tahong uplift and the northern slope are all likely directions of deep-seated gas fields for their good hydrocarbon sources, large resource potential and advantageous preservation conditions. Bachu Uplift and the southwest of Tarim Basin are also important regions for deep-seated gas. In the Junggar Basin, the deep-seated gas is mainly distributed in the south border and the second and the third rows of the structures near Tianshan Mountain which are mostly targets. The inner region is a possible long-term region.

5 Conclusions

In the Phanerozoic, the mainland of China underwent separation-coalition evolution of the continental plate and oceanic plate in the Paleozoic and the basin-mountain coupling evolution in the Cenozoic. This has resulted in the formation of polycyclic (superposed basin) basins where the Mesozoic—Cenozoic continental formations superposed the Paleozoic marine and paralic formations. There are large sets of source rocks for deep-seated gas and deep-seated gas resource is abundant. The China marine source rocks are characterized by high evolution degree. The main characteristic of deep-seated gas generation is that the paleo-petroleum reservoir continuously adjusts and evolves into adjusted accumulations in the paleo-uplift regions and the dispersed organic matter can be the gas source. The organic matter dispersed in minerals should be attached with great importance because of better catalysis conditions. According to the deep-seated gas generation characteristic, the coal-bearing basin with low geothermal gradient in the west of China should be the major region for deep-seated gas exploration.

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