

Evolution of the CBM reservoir-forming dynamic system with mixed secondary biogenic and thermogenic gases in the Huainan Coalfield, China

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Abstract The secondary biogenic gas is an important original type of the coalbed methane (CBM) in China. Based on the analyses of sedimentary and burial history of the Permian coal-bearing strata, combined with thermal history and gas generation process of coals, the CBM reservoir-forming dynamic system with mixed secondary biogenic and thermogenic gases in the Huainan Coalfield is subdivided into four evolutionary stages as follows: (i) shallowly-buried peat and early biogenic gas stage; (ii) deeply buried coal seams and thermogenic gas stage; (iii) exhumation of coal-bearing strata and adsorbed gas lost stage; and (iv) re-buried coal-bearing strata and secondary biogenic gas supplement stage. The Huainan CBM reservoir-forming model has the features of the basin-centered gas accumulation. The evolution of the reservoir-forming dynamic system proves that the thermogenic gas is not the main gas source for the Huainan CBM reservoir. Only the secondary biogenic gases as an additional source replenish into the coal bed after basin-uplift, erosional unroofing and subsequent scattering of thermogenic gases. Then this kind of mixed CBM reservoirs can be formed under suitable conditions.

Keywords: coalbed methane (CBM), secondary biogenic gas, thermogenic gas, evolution, reservoir-forming dynamic system, the Huainan Coalfield.

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1 Introduction

The Huainan Coalfield has been recognized as an important contributed base for the Eastern China energy supply. In recent years, many scholars^[1-5] have conducted the demonstrations on the Permian coalbed methane (CBM) resource potential and prospect in some details, and the CBM exploration and development have been given rise to great attention by some related departments.

Unfortunately, the selection of the optimum target area or "sweet spot" for exploration and development is not so well advanced because of the limited knowledge for the CBM accumulation mechanism.

Coal is both an excellent hydrocarbon source rock and an unusual reservoir. The coal bed is different from the conventional reservoir in that usually all but a negligible amount of the gas is sorbed on to the coal matrix rather than occurring as free gas or dissolved in formation water. Most of the stored gas in coals is adsorbed, i.e. densely packed as a single molecule layer on the pore walls in coal matrix. It behaves like a liquid rather than following the ideal gas law like the conventional free gas. The study on the reservoir-forming dynamic system of conventional natural gas has obtained some important advances^[6-9]. However, there have been few papers published on the CBM reservoir-forming mechanism^[10-13]. The purpose of this paper is to discuss the evolutionary problems of the CBM reservoir-forming dynamic system, and to provide a basic understanding of some parameters involved in assessing a target area for CBM exploration and development in the Huainan Coalfield.

2 CBM genesis and buried history of coals

2.1 Geological setting

The Huainan Coalfield located in the southeastern corner of the North China Platform is a typical Permian coalfield. Its east is the Tanlu (Tancheng-Lujiang) Fault Belt, and in the south it is not far from the Qinling-Dabieshan Orogenic Belt. Structurally, it is a synclinorium plunged to the east. As a small portion of the thrust nappe tectonic belt in the southern North China Platform, the Huainan Synclinorium is boundaried by the Fufeng (Fuyang-Fengtai) Nappe Structure^[14] onto the south margin of the coalfield.

The thickness of the Permian coal-bearing strata, including the Taiyuan, Shanxi, Lower Shihezi and Upper Shihezi Formations in an ascending order, varies considerably from 610 to 920 m in the Huainan Coalfield. Lithologically, it is mainly composed of limestones, terrigenous clastic sediments and coal beds. The sequence of the Permian rocks is deposited in the sedimentary cycles from the clastic shoreline or/and carbonate platform, to delta or/and fluvial systems. About 38 seams from the sequence in the coalfield mainly occur in the Shanxi, Lower Shihezi and Upper Shihezi Formations. Among them, Nos. 1, 8, 11-2 and 13-1 seams are the main targets for CBM exploration. The late Mesozoic (Yashanian) aplite and lamprophyre dikes are only well exposed in the coalmines in vicinity of Panji and Dingji^[11].

2.2 Geochemical evidence for coalbed gas origin

Gases can be generated from coals in three main phases: (i) early or primary biogenic gas formed by the bacterial

activity during the conversion from peat to coal ($R_o < 0.3\%$); (ii) thermogenic gas formed by thermal processes during the main coalification; and (iii) secondary or late biogenic gas formed by the bacterial activity after the coal has reached thermal maturity^[15–17]. The CBM origin is excellently determined by many researchers using the gas componential, carbon isotopic and gas-chromatographic evidences, and combined with the analytical results on the coal rank and basin hydrogeology. Among them, the methane carbon isotopic composition is the main indication for identifying the CBM origin. It is often considered that the methane $\delta^{13}\text{C}$ value being equal to -55% is the critical one for distinguishing the biogenic and thermogenic gases^[15,17,18]. Traditionally, the gas with methane $\delta^{13}\text{C}$ values less than -55% is regarded as the biogenic origin; conversely, it is assumed to belong to the thermogenic origin^[18–20].

The methane carbon isotope values of coalbed gas measured in the Huainan Coalfield vary from -49.22% to -72.27% (Table 1). It is narrower than the distribution range of methane $\delta^{13}\text{C}$ values (-24.97% – -73.70%) from the measured gas-samples in all-China^[21]. They are universally lighter than the methane $\delta^{13}\text{C}$ values from the Qinshui Basin in Shanxi Province, Jingyuan Coalfield in Gansu Province and Nantong Coalfield in Sichuan Province and so on, but very close to those from the Xuzhou Coalfield in Jiangsu Province, Tangshan Coalfield in Hebei Province, Fengcheng Coalfield in Jiangxi Province and San Juan Basin in the United States^[21–27].

The methane $\delta^{13}\text{C}$ values of the gas samples collected from the Huainan Coalfield are mostly less than -55% (Table 1), and totally they show the feature of biogenic gas. From the measurement of vitrinite reflectance ($R_o = 0.8\%$ – 1.0%), the coal rank has exceeded the stage of early biogenic gas, and is at the onset stage of the intense thermogenic gas generation. Therefore, it probably belongs to the secondary biogenic origin rather than the early biogenic gas. It has to be noticed that in about a quarter of the gas samples from the Huainan coals, the methane $\delta^{13}\text{C}$ values are ranging between -55% and -49.22% , and it seems to be the carbon isotopic features of thermogenic gas. However, based on the conversion

result of thermal maturity of coals, the methane $\delta^{13}\text{C}$ values of thermogenic gases should be ranging between -36.90% and -34.50% , and it is differentiated largely from the measured methane $\delta^{13}\text{C}$ values. On the other hand, the ethane and propane $\delta^{13}\text{C}$ values measured are ranging from -15.9% to -26.2% and from -7.7% to -28.4% respectively (Table 1), and they show the thermogenic origin.

Besides, the CBM origin can be characterized by the gas dryness index, which is the ratio of methane to heavier hydrocarbons (C_1/C_{1-5} value). The heavier hydrocarbon content of coalbed gas is very low (0.03% – 0.37%) in the Huainan Coalfield, and the values of gas dryness index vary between 0.993 and 1.0. This shows that it is the very dry gas composed of methane. After Scott et al.^[27], only the biogenic and pyrolysis gases show the features of very dry gas. However, the thermal maturity of the Huainan coals has not attained the over-mature stage generating cracking methane yet, and it is the secondary biogenic origin. As mentioned above, it goes without saying that the CBM in the Huainan Coalfield is the mixed secondary biogenic and thermogenic gas concurrently^[28,29].

2.3 Burial history of coal-bearing strata and tectono-thermal events

According to the Global stratigraphic chart published by IUGS and Permian chronostratigraphic correlation in China^[30–32], the deposition of the Permian coal-bearing strata in the Huainan Coalfield commenced at about 295 MaBP in the initial Asselian and ceased at about 253 MaBP in Wuchiapingian.

The evidence furnished with the apatite fission track analysis (AFTA) from the relevant horizons^[33–35] indicates that, after the Permian coal measure was deposited, three tectono-thermal events in the Huainan Coalfield had been encountered during 240–220 MaBP (from Middle Triassic Anisian to Late Triassic Carnian), 160–120 MaBP (from late Middle Jurassic Callovian to Early Cretaceous Valanginian) and 80–60 MaBP (from Late Cretaceous Campanian to Paleocene Danian) respectively. Among them, the first tectono-thermal event is equivalent to the early Indosinian movement, and the two later events are

Table 1 The carbon isotopic data of coalbed gas from main coal seams in the Huainan Coalfield (PDB)

Formation	No. Seam	$\delta^{13}\text{C}_1$ (‰)	$\delta^{13}\text{C}_2$ (‰)	$\delta^{13}\text{C}_3$ (‰)	$\delta^{13}\text{C}_{\text{CO}}$ (‰)
Upper Shihezi Fm	13-1	-66.80–-49.22	-22.90–-17.40	-28.40–-10.80	-17.80–-16.20
	11-2	-72.27–-57.00			
	9	-56.90			
	8	-68.00–-50.70			
Lower Shihezi Fm	7	-56.20–-55.40	-24.10–-23.30	-21.90	-39.00–-25.00
	6	-54.70–-49.20	-21.00		-19.80
	5	-51.10	-20.40	-24.20	-29.20
Shanxi Fm	1	-58.70–-55.60	-26.20–-19.60	-25.30	-19.10–-6.00

relatively close to the middle and late Yanshanian tectonic movements. In the east half of the coalfield, the two later events are probably magma intrusion thermal events confirmed by the Yanshanian aplite, aplitic diorite and lamprophyre dikes in vicinity of Panji mine area^[1].

The subsidence analysis completed using the stratigraphical section and the AFTA results from relevant horizons have demonstrated that, after the coal-bearing strata were formed, the basin has been settled quickly. The main coal-bearing member attained its maximum buried depth during 240–220 MaBP (Fig. 1). Because rock records were lacked, the Middle-Late Triassic, Jurassic and Cretaceous subsidence and sedimentary history could not be recovered directly, but the apatite fission track ages were centralized in 90–60 MaBP^[33,34], it reflected that the main tectonic uplift events in the Huainan Coalfield occurred from the Late Cretaceous to middle Paleocene in age. Besides, a part of strata in the apatite fission track-annealing zone in the study area has been eroded away, and the overlying rocks in the non-annealing zone have been denuded wholly. Based on the data of the earth surface temperature and palaeo-geothermal gradient, it is inferred that the thickness of the non-annealing zone varies from 1570 to 2100 m. Additionally, the Cenozoic rocks have a thickness between 50 and 900 m. It follows that the thickness of the eroded rocks during the main tectonic uplift in the Huainan Coalfield should be more than 2000 m.

3 Evolution stages of the CBM reservoir-forming dynamic system

Based on the burial history of the Permian coals, and combined with the tectono-thermal events, features of palaeo-geothermal field, coalification degree and CBM origin, four evolution stages of the CBM reservoir-forming dynamic system in the Huainan Coalfield can be subdivided (Fig. 1) as follows.

3.1 Shallowly buried peat and early biogenic gas stage (253–250 MaBP)

The evidence from subsidence-buried and thermal evolution analyses indicated that, during 253–250 MaBP after the coal-bearing strata were formed, the buried depth of coal seams was less than 1400 m. The palaeotemperatures on the top surface of the Taiyuan Formation are from 50°C to 60°C (Fig. 1(a)), and the Permian top surface have kept the palaeotemperatures between 35°C and 40°C. It follows that the palaeotemperatures of the main coal-bearing member including the Shanxi, Lower and Upper Shihezi Formations are roughly ranging from 40°C to 55°C. Therefore, the main seams have not entered into the threshold of the thermogenic methane generation as yet, and only the early biogenic gases are generated.

The early or primary biogenic methane generated from peat at relatively low temperatures (<56°C) and shallowly

burial depth or low coal rank ($R_o < 0.3\%$) is closely similar to the biogenic gases formed shortly after burial in the marine sediment^[36]. Practically, at the peatification stage after the coal-forming plants were accumulated, the early biogenic methane could be generated from peat swamp. Although peat has a high surface area, the high moisture content of peat suggests that water molecules occupy many of the potential adsorption sites for the biogenic gases. Therefore, the significant early biogenic gas probably does not occur because of the low pressure and limited adsorption sites. Most of the early biogenic methane and carbon dioxide are probably dissolved in formation water and subsequently removed from the coal reservoir system during compaction and coalification^[18]. Therefore, it has no substantive significance for the CBM reservoir in the Huainan Coalfield.

3.2 Coals deeply buried and thermogenic gas stage (250–120 MaBP)

The Permian coals in the Huainan Coalfield were deeply buried during 220 MaBP, with the maximum burial depth of about 3200 m. Thereafter, the basin was uplifted slowly, and up to 140 MaBP, the main seams all were buried in the depth of 2000–3000 m. The analysis results of tectonic history and AFTA have proved that, in the period from 250 to 120 MaBP, the coal seams had undergone through two tectono-thermal events in 240–220 and 160–140 MaBP respectively^[33,34].

During 240–220 MaBP of Middle and Late Triassic, the top surface of the Taiyuan Formation had temperatures between 90°C and 130°C, and the main seams, with reservoir temperatures from 80°C to 120°C (Fig. 1(b)), should enter into the thermogenic gas generation stage. Thereafter, the temperature was slightly declined. During 160–120 MaBP of Middle and Late Jurassic, the geo-temperature was risen again, and the temperatures of 90°C–130°C on the top surface of the Taiyuan Formation were steadily maintained (Fig. 1(c)). It is clear that the coalification step of seams in the Shanxi Formation and lower member of the Lower Shihezi Formation in the vicinity of Panji mining area was increased from gas coal rank to fat coal rank because of the magmatic activity represented by the thermal event of 160–120 MaBP, and entered into the stage of intense thermogenic methane generation.

After Scott^[19], early thermogenic gases are generated from coal at the high-volatile bituminous rank (R_o between 0.5% and 0.8%). Moreover, the coalbed gases produced at the stage of intense thermogenic methane generation are generated by coals that have attained vitrinite reflectance values between 0.8% and 1.0%. The thermogenic gas generated from the Permian coals in the Huainan Coalfield was mainly conducted by the plutonic (or regional) metamorphism pattern during Middle and Late Triassic

As mentioned above, the Permian coals in the Huainan Coalfield are commonly characterized with the dominance of gas coal, and less importance of fat and coking coal ranks. Tang et al.^[37] consider that the economic quantities of methane are generated from coals that have attained vitrinite reflectance values between 0.7% and 1.0%. That is to say, the threshold of significant thermogenic methane generation occurs when coal attains the high-volatile bituminous rank.

In the light of thermal simulation results of coal, the gas yield from brown coal to gas coal is roughly ranging from 20 to 100 m³/t^[1,25,38]. Using the data from the isothermal adsorption test of the Huainan coals, the authors calculate the maximum capacity of gas adsorbed by coals varying from 12 to 18 m³/t in the burial depth from 2000 to 3000 m. That is to say, under the ideal status, only a small amount of thermogenic gas can be stored in the Huainan coals. Even so, it still is enough to lead the coal reservoir in the over-saturated and/or saturated status, and the surplus thermogenic gases will be migrated from coals to the adjacent sandstone reservoir, or in seam it is laterally migrated to the part with the lower coal rank^[12].

3.3 Exhumation of coal-bearing strata and adsorbed gas lost stage (120–23 MaBP)

The main exhumation of the Permian coal-bearing strata in the Huainan Coalfield was probably commenced at about 120 MaBP in the mid-Early Cretaceous and ceased at 23 MaBP in the early Miocene. In this process, although the tectono-thermal event “remembered” universally by the apatite fission track occurred during 80–60 MaBP in Late Cretaceous, it mainly resulted in the regional thermal uplift^[33–35], and almost had not any impact on the superimposed metamorphism of coals. In this situation, the top surface of the Taiyuan Formation always had temperatures between 40°C–56°C (Fig. 1(d)), and the thermogenic gases were no longer generated from the Permian coals. With respect to the CBM storage, the important fact of this evolution stage is that with the regional uplift and the exhumation of the coal-bearing strata, a part of adsorption gas in coals is desorbed and diffused, and it

is scattered largely through the filtration.

In order to illustrate the content variation of methane adsorbed by coals during the uplift process, using the Langmuir parameters at different temperatures (20°C, 30°C, 40°C and 50°C) of the main seams in the Huainan Coalfield, and combined with the data of the pressure and paleo-geothermal gradients^[33–35], the authors calculate the comprehensive influence of temperature and pressure (depth) on CBM storage capacity (Fig. 2). The results indicate that when the paleo-geothermal gradient is 35°C/km, the paleo-buried depth in which the seam attains the maximum adsorption capacity (13.5 m³/t) is 1100 m. Assuming that the coal seam is uplifted by 2000 m from a maximum buried depth of 2780 m after gas generation, adsorbed gas content will not change significantly, and is decreased only by 8.3% (Fig. 2, ΔI_1). Significant degassing occurs only at the burial depth less than 700 m, and as the seam is uplifted to earth surface from that depth, it will lose the whole adsorbed gas by 90% (Fig. 2, ΔI_2).

Based on the calculation, the basin uplift rate of the Huainan Coalfield at this stage is approximately 0.21 mm/a. The coal sorption capacity increases with increasing pressure and decreasing temperature. Because the reservoir pressure and temperature gradually decline during the basin uplift, the degassing of adsorbed gas resulting from exhumation of coal-bearing strata is also very slow. Under this situation, although the adsorbed gas is decreased continuously, the coal reservoirs will still remain saturated with respect to methane.

The given data of gas-bearing parameters indicate that, the main seams in the Huainan Coalfield have the gas saturation values between 33.85% and 71.60%, and all undersaturated with respect to methane. Fig. 3 shows a possible interpretation of the formative mechanism of the unsaturated coal reservoirs. When the seam is uplifted, the adsorbed gas in coals is continuously desorbed and scattered (Fig. 3-a). Thereafter, the basin is settled, and when the seam is re-buried to the present depth, the coal reservoirs become unsaturated with respect to methane (Fig. 3-b) if there has no supplement from the additional source of gas. However, the causes resulting in the unsaturated

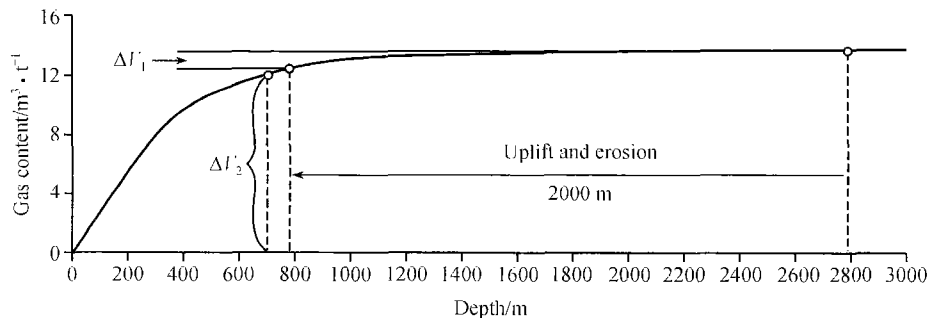


Fig. 2. The gas content variation in Permian coals during basin uplift in the Huainan Coalfield.

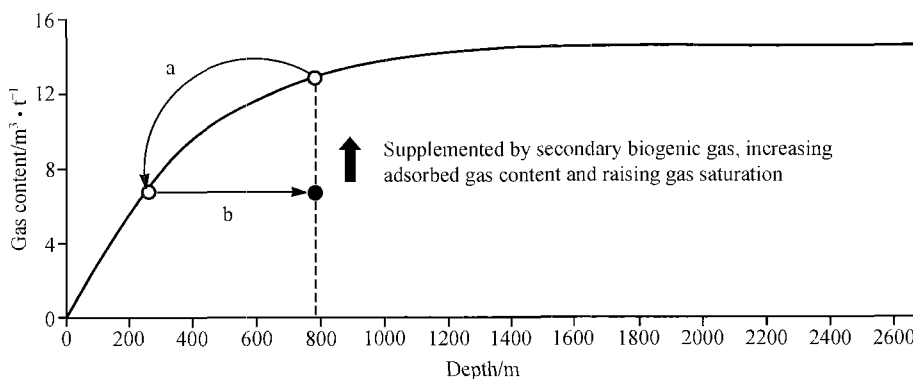


Fig. 3. The gas saturation variation in the elevation process of coal reservoir in Huainan Coalfield and the supplement of the secondary biogenic gas on the gas-bearing property of seam. a, The uplift of seam. It is the pressure and temperature reduction process, and is saturated; b, the re-settling and re-buried of seam. It is the pressure and temperature increase process, and is unsaturated.

coal reservoir are more complicated, and the interpretation only by the re-settling and re-buried of seam is incomplete.

The variations of the basin geothermal gradient during different evolution stages could be one cause having the influence on the adsorption capacity in the coal seam. For example, in the Huainan Coalfield, the geothermal gradient in the geological time was 35°C/km, and the maximum adsorption capacity of the seam under the comprehensive influence of the temperature-pressure (depth) was 13.5 m³/t (Fig. 2); the current geothermal gradient is 31.0°C/km, and the maximum adsorption capacity of the seam is 14.5 m³/t (Fig. 3), the difference between them is 1 m³/t. Therefore, under the situation that it has no supplement from additional sources of gas, the primary saturated coal reservoir could be currently unsaturated with respect to methane.

The change of hydrological conditions also has the influences on the gas content and saturation in coal reservoir^[12]. As is well known, the gas diffusing in coal seams is largely controlled by diffusion gradient rather than reservoir pressure incompletely. In a stagnant groundwater system, the gas adsorbed by coal matrix could be balanced with the dissolved gas in coal cleats. However, once the stagnant groundwater system is replaced by an active groundwater system, the gas adsorbed by coal-matrix must be diffused and entered into the cleat system. Then, it was carried off by the actively alternative groundwater flow. It follows that the reduction of the adsorbed gas content and gas saturation within coal reservoir occurs. This is the so-called sweeping action of groundwater on the coalbed methane^[12,39]. From an examination of the deposystems and unconformity interfaces at different evolution stages after the coals buried, it can be determined that the different hydrodynamic regimes during different geological time intervals in the Huainan Coalfield have occurred. Therefore, it is natural that the decrease in gas content within the coal reservoir results from

the alternative swashing and filtering by meteoric water recharge and groundwater.

The generation, desorption and scattering of CBM are completed in a dynamically balanced complex system. When the primary balanced system is broken, and the new balance yet has not established, the coal reservoir is unsaturated with respect to methane. In the Huainan Coalfield, in the uplift process of about 100 Ma during Cretaceous and Paleogene, a considerable part of thermogenic adsorbed gas in coals had been desorbed and scattered due to the impacts of the exhumation and unloading of the coal-bearing strata, and the coal reservoirs are universally in the unsaturated status. This process is very important for correctly understanding the coal reservoir features and the CBM reservoir-forming mechanism.

3.4 Re-buried coal measure and secondary biogenic gas supplement stage (23 MaBP to recent)

Since 23 MaBP, the Neogene and Quaternary sediments with total thickness of about 50–900 m had been deposited in the Huainan Coalfield, and then, the Permian coal seams were re-buried. The present geothermal gradient is slightly lower than the palaeogeothermal gradient. The top surface of the Taiyuan Formation has a temperature between 40°C and 56°C (Fig. 1(e)), and the main seams above it also are roughly in the same geothermal regime. Therefore, the superimposed thermogenic gas cannot be generated from the coal reservoirs during this stage.

The CBM in the Huainan Coalfield is the mixed origin in which it is mainly the secondary biogenic gas, as well as the co-existed residual thermogenic gas^[28,29]. The secondary biogenic gas generated from coals is closely related to the formation water, and the relatively low geothermal field provides a necessary environment for its generation. According to Scott et al.^[27], the secondary biogenic gases are generated by the metabolism of the microorganism entering into seam after the coal-bearing strata are deeply buried and then uplifted and erosively

unroofed. The generation of the secondary biogenic methane requires at least the following conditions: (i) coalification or coal rank from brown coal to low-volatile bituminous coal (R_o 0.5%–1.5%); (ii) geotemperature less than 56°C; (iii) adequate coalbed permeability; (iv) sufficient recharge along basin margins; and (v) bacteria being effectively transported basinward by groundwater. That is to say, most biogenic gases in coal beds are probably associated with groundwater flow and generated by secondary biogenic processes. Secondary biogenic gases are generated through the metabolic activity of bacteria, introduced by meteoric waters moving through permeable coal beds or other organic-rich rocks. The bacteria or microorganism carried by groundwater metabolize wet gas components, heavy hydrocarbons and other organic components in coals at relatively low temperatures to generate methane and carbon dioxide.

K and Na ions are the primary elements for growth of all organisms including the bacteria; their concentration has a direct influence on growth and flourishing of methane-forming bacteria. Generally, the methane-forming bacteria can well live in the water medium with the salinity not exceeding 4000×10^{-6} [40]. When K+Na ions concentration in methane bacteria bodies exceeds 20000×10^{-6} , their metabolism will be restrained strongly. The results of the chemical analysis of groundwater samples in the Huainan Coalfield have proved that the salinity is less than 1800×10^{-6} . Therefore, the features of the coal seam and the Cenozoic geological and hydrogeological environments in the study area fully possess the generation conditions for the secondary biogenic methane.

From an examination of the Cenozoic settling features and the formative and evolutionary history of the basin, it can distinguish that the secondary biogenic gases in the Huainan Coalfield probably have been generated since 23 MaBP. Just due to this process, it is supplemented by the secondary biogenic gas as the additional gas source, and the gas content and gas saturation are increased (Fig. 3)

after the thermogenic adsorbed gas in the coal reservoir is scattered largely.

4 The CBM reservoir-forming geological model

The CBM reservoir formation is controlled by various geological and hydrological factors. For the CBM generation, and desorption, scattering and storage of adsorbed gas, except the influences from the physicochemical conditions of coal seam itself, the tectonic action is one of the guiding factors undoubtedly. The tectonic force is the most active geological stress within the earth crust or/and lithosphere, and it is also the main motive power for promoting CBM enrichment and accumulation in reservoirs. Therefore, the tectonic style must controls on the basic features of the CBM reservoir. The synclinorium in the Huainan Coalfield was fallen into the pattern during Late Jurassic and Cretaceous^[14]. If the small anticlines and synclines distributed along the northern margin of the coalfield are not considered, the most important structure forms are the Panji anticline and Xieqiao syncline, as well as the Fufeng nappe structure, which is thrust on the southern flank of the synclinorium (Fig. 4).

With respect to the Late Paleozoic coalfield strongly reformed by many episodes of tectonic movements, the settling and deeply buried of coal-bearing strata, the capacity and dynamic process of hydrocarbon generated from coal seams, and coal maturity are only the precursory factors for the CBM reservoir. The main factors controlling on CBM reservoir formation are the basin uplift in later period, the scattering and storage of adsorbed gas, as well as the *in situ* stress, recent tectonic stress field and geothermal field. The presence of appreciable secondary biogenic gas indicates an active dynamic flow system with sufficient permeability. The secondary biogenic gas is an additional source of gas beyond that stored on the coal matrix during thermogenic gas generation. Understanding the tectonic evolution and hydrogeology of the coal basin, the timing of secondary biogenic gas generation and trap development are important for delineating area for CBM

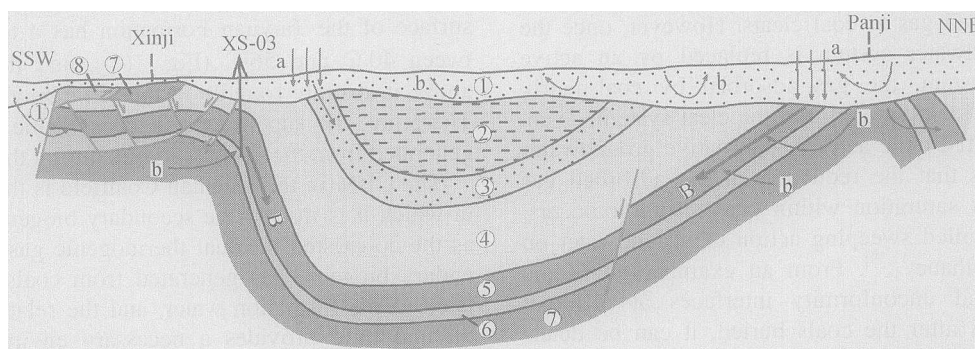


Fig. 4. The CBM reservoir-forming geological model in the Huainan Coalfield. ①Cenozoic sediments; ② red mudstones of the Shiqianfeng Formation; ③ sandstones at the bottom of the Shiqianfeng Formation; ④ granophyric mudstones in the upper part of the Upper Shihezi Formation; ⑤ main coal-bearing member (Nos. 1–13 seams); ⑥ Taiyuan Formation; ⑦ Lower Paleozoic limestones; ⑧ Precambrian metamorphic rocks. a, Centripetal flow of meteoric water permeating downward; b, discharge by cross-formational flow and evaporation; B, secondary biogenic gases carried by advancing front of centripetal flow are accumulated at tectonic hingeline.

exploration.

Because CBM generally occurs in an adsorbed form rather than in a free state, a conventional-type trapping mechanism is not entirely required. Many scholars^[12,17,27] recognize that the basin-centered gas accumulation or/and synform enrichment is the most common CBM reservoir-forming model. It implies that its shape is like a bowl rather than having the domed configuration of most conventional traps, and the gas is below the hydrostatic head rather than above it.

The recent hydrodynamic system in the Huainan Coalfield has been established since the Neogene. It includes generally the local hydrodynamic units as follows: (i) the centripetal flow of meteoric water permeating downwards; (ii) discharge by cross-formational flow or/and cross-formational flow-evaporation, and (iii) the water-stagnant area. The centripetal flow of meteoric water recharge is induced by gravity potential effect of meteoric water due to influence from the topographical height difference at the basin edge (Fig. 4-a). The discharge by the cross-formational flow is to be flow of groundwater from high potential area to low potential area, and totally, it appears as the flow from depth to shallow and/or earth surface (Fig. 4-b). It generally includes both cross-formational and evaporation discharges. The evaporation discharge is reduced gradually with depth increasing; it follows that the discharge by the cross-formational flow from the Lower Palaeozoic karst limestones to the coal-bearing rocks becomes a main pattern. During the evolution of the hydrodynamic system, the groundwater permeating downward is obstructed in the basin depth, and the water-stagnant will occur.

After the thermogenic gas is generated from coal seams in the Huainan Coalfield, it is followed by the basin uplift and erosion with large scale, as well as coal-measure reburied, so that the coal reservoirs are mostly in unsaturated status. However, when the meteoric water permeating downwards is passing through the seams, the bacteria carried by it metabolize wet gas, heavy hydrocarbon and other organic matters produced during coalification, and generate the secondary biogenic methane and carbon dioxide (Fig. 4-b). As the groundwater flows basinward, the dissolved biogenic gases carried by advancing front are pushed to the vicinity of the impermeable barrier or hinge line (e.g. fault or syncline inflection end) and are stagnated. Ultimately, those gases are resorbed by coal matrix, or are filled in coal cleats, and then supplement the deficient coal reservoirs. It results in that the gas content and saturation in coal reservoirs increased (Fig. 3 and Fig. 4-b). Of course, the water-bearing capacity and permeability of the coal reservoir in local units are very poor, and the *in situ* secondary biogenic gases generated only can supplement the loss of thermogenic gas in some degrees.

The dissolution of methane in formation water is realized mainly through two ways as follows: (i) the methane

and water molecules form the methane-hydrated molecule; (ii) the methane molecules are filled in water molecule gap^[41]. The dissolution capacity of formation water is depended on the temperature, pressure and salinity of water^[42–47]. Because quantity of methane dissolved in formation waters is $0.2 \text{ m}^3/\text{m}^3$ commonly^[7], it seems that its contribution to the CBM enrichment is insignificant.

Based on the measured results on isotope deuterium (1.7–8.1 TU) in the water in coal-bearing strata (samples collected in Nos. –341 and –326 machinery roadways in Xinji Mine) and the drained water from some CBM boreholes (Fig. 4) (water samples after drainage by 1 a), it is calculated that the age of the groundwater is 35–65 a. It shows a close relationship between the groundwater in coal-bearing strata and meteoric water. Of course, all measured water samples are collected in the vicinity of the groundwater alternative zone, and their ages are younger, while the age of stagnant water in depth can be counted by ten thousand years or even million years. Therefore, although the methane-dissolving capacity of groundwater is not higher, the accumulated quantity of water-dissolved methane transported to the impermeable or low-permeable boundary of coal reservoirs cannot be underestimated, due to the long-term accumulation.

5 Conclusions

(1) The Permian coals in the Huainan Coalfield have vitrinite reflectance values from 0.80% to 1.00%, the methane $\delta^{13}\text{C}$ isotopic values (PDB) of gas samples are ranging from -49.22‰ to -72.27‰ , and the gas dryness indices range from 0.993 to 1.00. Coalbed gases in the Huainan Coalfield represent a mixture of secondary biogenic and thermogenic gases. The former is produced by the bacterial activity after the coal has reached thermal maturity, and the latter is generated by coal when its constituents volatilize as it increases in rank due to increasing temperatures encountered with greater depth of burial or proximity to igneous activity.

(2) Based on the comprehensive analyses on burial and tectono-thermal history of the Permian coal-bearing strata, and CBM generation history, four evolutionary stages of the CBM reservoir-forming dynamic system of the Huainan Coalfield can be subdivided as follows: (i) shallowly-buried peat and early biogenic gas stage (253–250 MaBP); (ii) deeply buried coal seams and thermogenic gas stage (250–120 MaBP); (iii) exhumation of coal-bearing strata and adsorbed gas lost stage (120–23 MaBP); and (iv) reburied coal-bearing strata and secondary biogenic gas supplement stage (23 MaBP to recent).

(3) The secondary biogenic gas is an additional source of gas beyond that sorbed on the coal surface during the thermogenic gas generation. The evolution of the CBM reservoir-forming dynamic system with mixed secondary biogenic and thermogenic gases has proved that the ther-

mogenic gases are not the main gas source to form this CBM reservoir. Only the secondary biogenic gases as an additional source replenish into coal beds after basin-uplift and erosional unroofing, and subsequent scattering of thermogenic gases. This CBM reservoir in the Huainan Coalfield can be formed under the suitable conditions.

(4) The geological model for the Huainan CBM reservoir is a basin-centered (or synformal) gas accumulation. This model emphasizes that after the Permian coals reaches their thermal maturity, it has suffered the basin uplifting and cooling, unload and re-buried of coals. It results in that coal reservoirs are unsaturated with respect to methane, owing to the fact that thermogenic gases will most likely be lost to leakage. When meteoric water transporting bacteria from the recharge area permeates downward to coal seams, the carried bacteria metabolize wet gases and heavy hydrocarbon in coals to produce secondary biogenic gases. During the centripetal flow process of groundwater, the dissolved biogenic gases carried by the advancing front are pushed to the vicinity of permeable barrier or fault and syncline hingeline and are stagnated. Those gases are resorbed by coal matrix, or filled in coal cleat system. Ultimately it results in that the secondary biogenic gases replenish the unsaturated reservoirs to form the saturation CBM reservoirs. Therefore, the enriched CBM reservoir with mixed secondary biogenic and thermogenic gases is mainly controlled by the coupling of evolution of the CBM reservoir-forming dynamic system, tectonic styles and hydrodynamic fields.

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