# Study on Influence of Gas Generation Intensity on Gas Content Difference of Different Rank Coal

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**Abstract:** In view of the problems such as the great difference of CBM content in different coal ranks in our country and the unclear understanding of the control mechanism, the gas content test data of surface drilling coal seam coring in dozens of representative areas in China are sorted out. The characteristics of methane content and gas composition under ashless drying conditions of different rank coals are summarized. According to the maximum reflectivity of vitrinite (0.5%, 1.30%, 2.50%, 3.75%), the relationship between methane content and deterioration degree was divided into five stages. The methane content at each stage showed the characteristics of low–rapid increase–slow increase–peak–rapid decrease. The production characteristics of $C_{2+}$ , $H_2O$, $CO_2$, $CH_4$ and $H_2$ at different temperatures were analyzed by means of thermal simulation experiment and pyrolysis infrared spectrum experiment. Based on the characteristics of $CH_4$ generation, the whole process of gas generation was divided into six stages according to the temperature from low to high. It mainly includes the first phase of adsorption and physical precipitation of the absorption state, and the other five stages of chemical reaction formation. Based on the relationship between vitrinite reflectance and experimental temperature during heating, the relationship between the generation intensity of $CH_4$ and the content of $CH_4$ in the coal seam is discussed. It is inferred that the intensity of $CH_4$ generation in different metamorphic stages is the main controlling factor of $CH_4$ content distribution with metamorphic degree.

## 1. Introduction

China is rich in coalbed methane (CBM) resources, with significant variations in geological characteristics. Researchers have conducted studies on the characteristics, differences, and influencing factors of gas content from various perspectives and scales, achieving substantial results [1-9]. Regarding the genesis of gases, researchers generally believe that the types of CBM genesis include biogenic, secondary biogenic, and thermogenic origins. They have established corresponding identification indicators and evaluation methods using techniques such as isotope analysis and thermal simulation experiments [3, 10-15]. In terms of the factors influencing gas content, a significant amount of research has been conducted primarily from the perspectives of reservoir and preservation conditions. This research explores the effects of coal-forming environments, coal accumulation patterns, reservoir formation mechanisms, and preservation conditions [16-22]. As CBM is a self-generating and self-storing gas, gas generation conditions are inherent, while preservation conditions are subsequent. However, there is still no clear understanding of the relative influence or control levels of these two factors on the gas content in coal seams. Based on a summary of the gas-bearing characteristics of typical CBM areas across the country, this paper utilizes gas generation simulation experiments to analyze the relationship between gas generation characteristics and the distribution of gas content, and to explore the gas-bearing characteristics of coals of different metamorphic degrees and their differentiation control

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mechanisms.

## 2. Relationship Characteristics between Coalbed Methane Content and Metamorphic Degree

Gas content test results from coal seam cores obtained by surface drilling in dozens of representative areas in China, including Shanxi, Shaanxi, Anhui, Henan, Xinjiang, Guizhou, Heilongjiang, and Inner Mongolia, were collected and compiled. To eliminate the influence of minerals in coal on the gas content difference, the analysis mainly focused on the dry ash-free $CH_4$ content. As shown in Figure 1, there is a relatively obvious relationship between the dry ash-free $CH_4$ content and the maximum reflectance of vitrinite, which is similar to the statistical results of Ye Jianping et al. [23]. This relationship can be divided into five stages:

In the first stage, with a maximum vitrinite reflectance of less than 0.5%, the $CH_4$ content is very low, less than 1 $m^3/t$.

In the second stage, with a maximum vitrinite reflectance between 0.5% and 1.30%, the $CH_4$ content shows a rapid increase. When the reflectance is between 0.5% and 0.65%, the $CH_4$ content is generally less than 3 $m^3/t$, while in the range of 0.65% to 1.30%, the $CH_4$ content is typically between 5 and 10 $m^3/t$.

In the third stage, with a maximum vitrinite reflectance of approximately 1.3% to 2.5%, the $CH_4$ content shows a slow increase, reaching about 15 $m^3/t$ when the reflectance is around 2.5%.

In the fourth stage, with a maximum vitrinite reflectance between 2.5% and 3.75%, the $CH_4$ content rapidly increases and reaches a peak value range, peaking at about 25 $m^3/t$ around a reflectance of 3.25%.

In the fifth stage, when the reflectance is greater than 3.75%, the $CH_4$ content decreases rapidly, dropping below 1 $m^3/t$ after the reflectance exceeds 4.5%.

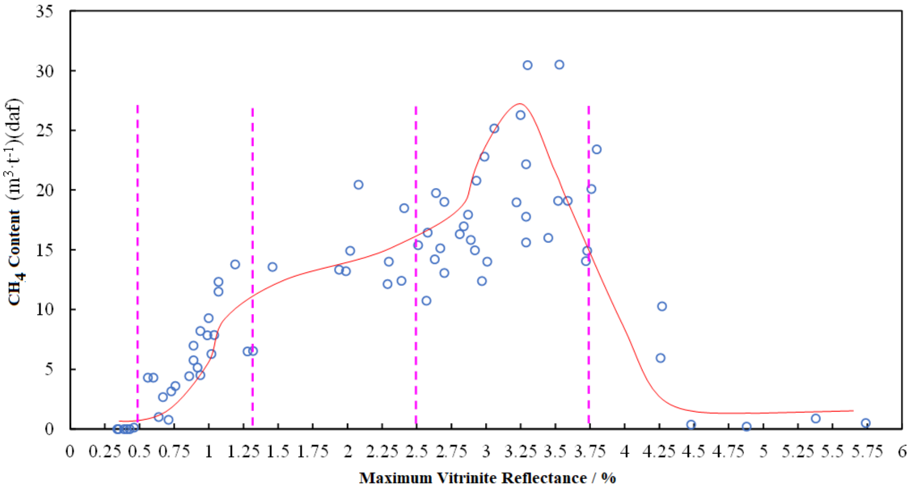



Fig.1 Relationship between the content of dry ashless methane gas in different coals and the maximum reflectance of coal vitrinite

The composition of coalbed methane is mainly $CH_4$, but it also includes gases such as $CO_2$ and $N_2$. According to the statistics by Fu Xuehai et al. [8], the average concentrations of $CH_4$, heavy hydrocarbon gases (collectively referred to as $C_{2+}$), $CO_2$, and $N_2$ in China's coalbed methane are 91.82%, 0.85%, 2.04%, and 5.19%, respectively, with $CH_4$ concentrations of ≥ 90% accounting for 77.44%. This study also compiled gas composition test data from representative areas in China (Table 1) and analyzed the relationship between gas quality and maximum vitrinite reflectance. Generally, coals with a low degree of metamorphism have a lower proportion of $CH_4$, while coals with a medium to high degree of metamorphism have a higher proportion of $CH_4$ and better gas quality. Based on the characteristics of changes in gas composition with maximum vitrinite reflectance, three stages can be roughly identified:

The first stage is low-quality methane, where the maximum vitrinite reflectance is less than about 0.9%, the proportion of $CH_4$ is generally between 60% and 90%, $CO_2$ is generally greater than 2%, $N_2$ is greater than 10%, and there are only small amounts of $C_{2+}$ heavy hydrocarbon components.

The second stage is medium-quality methane, where the reflectance is between 0.9% and 1.9%, the proportion of $CH_4$ is generally between 80% and 95%, $CO_2$ is around 1% to 3%, $N_2$ is generally between 5% and 10%, and the content of $C_{2+}$ heavy hydrocarbon components is generally greater than 1%.

The third stage is high-quality methane, where the reflectance is greater than 1.9%, the proportion of $CH_4$ is generally above 95%, $CO_2$ is generally less than 0.5%, $N_2$ is generally less than 5%, and $C_{2+}$ components are trace amounts.

Table 1 Statistical table of gas composition of coal with different metamorphic degrees

| area | coal rake | Maximum mirror mass reflectivity /% | $CH_4$/ % | $CO_2$/ % | $N_2$/ % | $C_{2+}$/ % |
|---|---|---|---|---|---|---|
| Jimusar, Xinjiang | B1 | 0.45 | 62.89~87.33 | 0.21~0.98 | 11.70~35.72 | 0.37~1.30 |
| JiaoPing shaanxi | 4-2 | 0.60 | 82.96-86.00 | 2.33-2.39 | 11.67-14.65 | 0.07 |
| Urumqi in Xinjiang is uneven | 45 | 0.61~0.70 | 59.6~82.98 | 10.89~18.57 | 3.22~28.7 | Not detected |
| Changji, Xinjiang | B6 | 0.55-0.69 | 86.59-93.31 | 1.26-3.92 | 3.72-9.79 | 0~0.24 |
| Qi dong, Anhui | 7-1 | 0.98 | 89.41~93.35 | 0.42~0.52 | 6.23~10.06 | Not detected |
| Lu Ling, Huaibei, Anhui | 8 | 0.92-1.01 | 78.96~96.37 | 1.12~1.86 | 3.33~19.18 | 0~0.06 |
| Xinjiang worship city | 7 | 1.31-1.37 | 79.17~93.42 | 1.55~2.86 | 5.03~17.97 | Not detected |
| Shouyang, Shanxi | 3 | 1.79 | 92.09 | 6.26 | 1.62 | 0~3.96 |
| Shouyang, Shanxi | 9 | 1.80 | 88.46 | 1.2 | 10.34 | 0~3.32 |
| Shouyang, Shanxi | 3 | 1.91~1.97 | 96.85~98.14 | 0.14~0.38 | 1.40~2.38 | 0~6.74 |
| Shouyang, Shanxi | 9 | 1.98~2.07 | 97.72~99.75 | 0.13~0.37 | 0.09~2.02 | Not detected |
| Shouyang, Shanxi | 15 | 2.02~2.17 | 99.03~99.85 | 0.05~0.26 | 0.06~0.78 | 0.01~0.04 |
| Zhao Zhuang, Jincheng, Shanxi | 3 | 2.36~2.46 | 96.14~99.42 | 0.28~0.44 | 0.21~3.48 | Not detected |
| Gulin Shibao, Sichuan | 17 | 2.61~2.99 | 94.16~98.96 | 0.10~0.37 | 0.94~5.62 | Not detected |
| Guizhou GuanZhai | 2 | 2.63 | 92.69~98.90 | 0.11~0.54 | 0.93~6.71 | Not detected |
| Guizhou GuanZhai | 6 | 2.84 | 91.03~98.12 | 0.17~0.25 | 1.67~8.71 | Not detected |
| Guizhou GuanZhai | 4 | 2.87 | 96.01~98.60 | 0.16~0.33 | 1.15~3.62 | Not detected |
| Guizhou GuanZhai | 11 | 2.89 | 89.65~98.35 | 0.16~0.47 | 1.38~9.85 | Not detected |
| Guizhou GuanZhai | 7 | 3.01 | 94.03~97.11 | 0.18~0.32 | 2.67~5.76 | Not detected |
| Guizhou against Jiangnan | M18 | 3.28 | 96.76~99.35 | 0.23~0.62 | 0.16~2.73 | Not detected |
| Guizhou against Jiangnan | M29 | 3.35 | 98.17~98.74 | 0.34~0.56 | 0.76~1.49 | Not detected |
| Guizhou against Jiangnan | M78 | 3.62 | 92.51~98.80 | 0.28~0.50 | 0.88~7.08 | Not detected |
| Shanxi Qinshui | 3 | 3.56 | 94.36-97.00 | 0.33-0.64 | 2.67-5.00 | Not detected |
| Shanxi Jincheng Temple River | 15 | 3.59 | 98.50~99.69 | 0.21~0.62 | 0.11~0.88 | Not detected |
| Shanxi Jincheng Temple River | 3 | 4.26 | 98.18-99.25 | 0.40-0.44 | 0.24-1.26 | Not detected |

## 3. Gas Generation Thermal Simulation Experiments and Result Analysis

### 3.1. Coal Samples and Experimental Methods

Thermal simulation experiments are an effective means of studying coal structure, reactivity, and gas generation characteristics. In recent years, they have been increasingly applied in research on the genesis of coalbed methane (CBM), gas content, and resource potential. According to the thermal simulation experiment results of peat gas generation at a heating rate of 20℃/h by Tao Mingxin et al. [3, 24-25], it was found that the reflectance of vitrinite increases with the rise in experimental temperature, reaching over 2.5% at about 500℃ and over 4% after 600℃. There is a good linear relationship between vitrinite reflectance and temperature. Considering the complementary effects of temperature and time during coalification, this study adopted a high heating rate thermal simulation experiment method. The heating rate was set at 20℃/min, with a final temperature of 900℃. The experiment was conducted in a nitrogen atmosphere with a flow rate of 45 ml/min using a TGA92 thermal simulation analyzer combined with a QMS422 quadrupole mass spectrometer. Additionally, to more accurately identify the generated gases and to aid in analyzing the gas generation mechanism from a molecular perspective, a thermal simulation infrared spectroscopy experiment was conducted using the CDS-2000-BIO-rad FTS-165 thermal simulation infrared spectrometer. The heating rate for this experiment was set at 5°C/S, with a final temperature of 1000℃, a nitrogen flow rate of 30 ml/min, four scans with a 6-second interval, and a spectral resolution of 8 $cm^{-1}$.

The coal samples used in the experiment were obtained from representative lignite distribution areas in China, and the coal sample information is shown in Table 2. Previous studies have found that minerals such as clays and pyrite in coal can affect the reactivity of the coal molecular structure during heating, thereby affecting the generation intensity of different gases [26]. Therefore, two coal samples underwent demineralization treatment using hydrochloric acid and hydrofluoric acid. After drying the demineralized coal samples in an oven at 50℃ for 5-6 hours, the thermal simulation gas generation experiments were conducted.

Table 2 Coal quality information

| Coal Sample | $M_{ad}$ / % | $A_{ad}$ / % | $V_{daf}$ / % | $R_{max}$ / % |
|---|---|---|---|---|
| sample 1 | 17.38 | 21.12 | 48.42 | 0.33 |
| sample 2 | 8.04 | 19.60 | 41.13 | 0.47 |

**3.2. Main Gas Production Results**

Based on the characteristics of coalbed methane components and the results of thermal simulation experiments, the production characteristics of $CH_4$, $CO_2$, $H_2O$, $H_2$, and $C_{2+}$ gases were primarily analyzed. Since demineralized coal samples significantly reduce or minimize the chemical reactions of minerals themselves and their interactions with the organic structure of coal during heating, thus reducing their impact on the quantity and characteristics of the generated gases, the characteristics of $CH_4$, $CO_2$, $H_2O$, $H_2$, and $C_{2+}$ gases generated from the two coal samples (sample 1 and sample 2) were found to be almost identical (Fig. 2). The production curves of each gas generally showed a unimodal pattern, with the generation of $C_{2+}$ gases being the highest, an order of magnitude higher than other gases. It is speculated that the abnormally high values at the beginning of heating are due to the massive precipitation of aliphatic hydrocarbon radical fragments in the molecular structure of lignite. Among the other four types of gases, $H_2O$ had the highest production and the widest temperature range for gas generation, followed by $CO_2$. $H_2$ had the lowest production and the narrowest temperature range for gas generation, while $CH_4$ was between $CO_2$ and $H_2$.

Based on the characteristics of $CH_4$ generation, the entire gas generation process during heating was divided into six stages (Fig. 2):

First stage: Below approximately 330℃, $CH_4$ is produced in trace amounts, while $CO_2$ and $H_2O$ are produced rapidly. $C_{2+}$ rapidly decreases from an initial high peak to the lowest value, and $H_2$ is not generated.

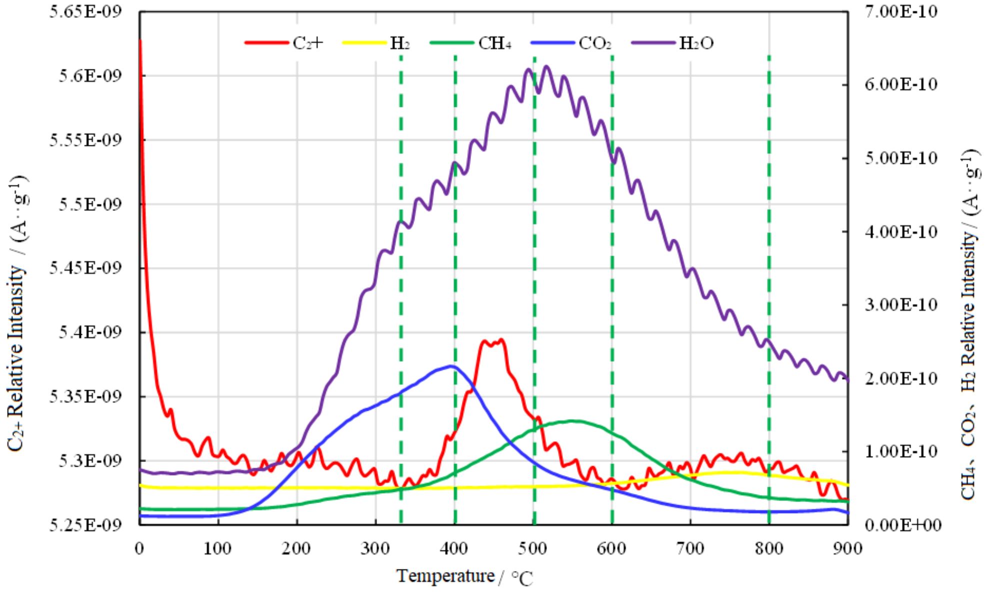
Second stage: Between 330-400℃, $CH_4$ is slowly produced, and $CO_2$ and $H_2O$ are rapidly produced, reaching their peak values. The production rate of $C_{2+}$ sharply increases around 380℃, while $H_2$ is not produced.

Third stage: Between 400-500℃, $CH_4$ production increases rapidly, $CO_2$ and $C_{2+}$ reach their production peaks and then quickly decrease, $H_2O$ tends towards its production peak, and $H_2$ is not produced.

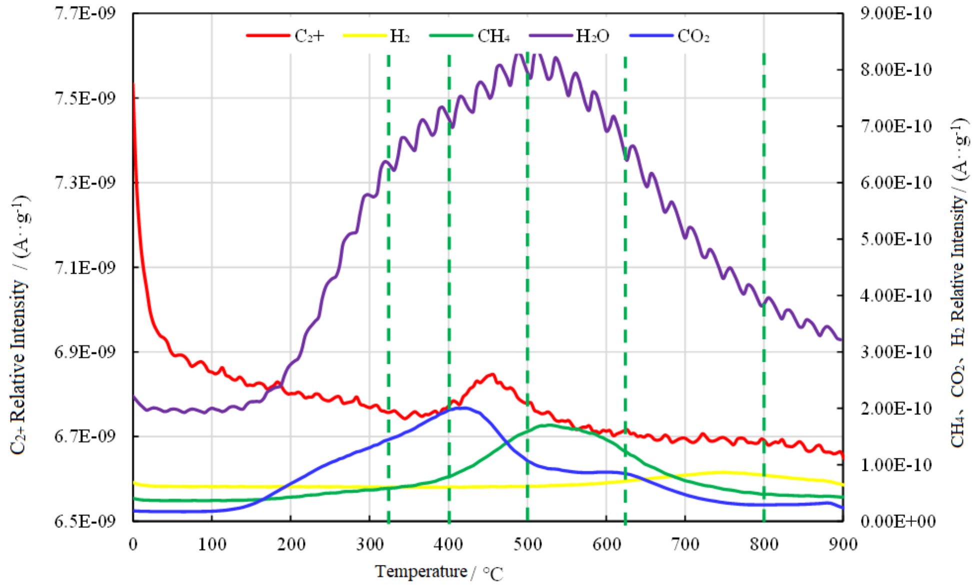
Fourth stage: Between 500-600℃, $CH_4$ production peaks, and $H_2O$ also reaches its maximum value. $CO_2$ has almost decreased to its minimum, and $H_2$ is still not produced.

Fifth stage: Between 600-800℃, $CH_4$ production rapidly decreases to its minimum value. $C_{2+}$ and $CO_2$ are slowly produced to their minimum values, and $H_2O$ decreases rapidly, while $H_2$ begins to be slowly produced and reaches its peak value.

Sixth stage: Above 800℃, the production of $CH_4$, $C_{2+}$, $CO_2$, and $H_2$ remains at their minimum values or ceases, while $H_2O$ trends towards its minimum value.




a) sample 1



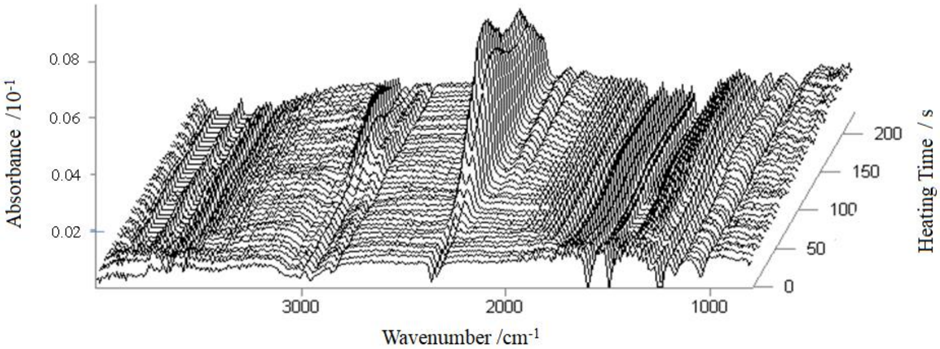

b) sample 2

Fig.2 The formation characteristics of $CH_4$, $CO_2$, $H_2O$, $H_2$, $C_{2+}$ of two coal samples in thermal simulation experiment

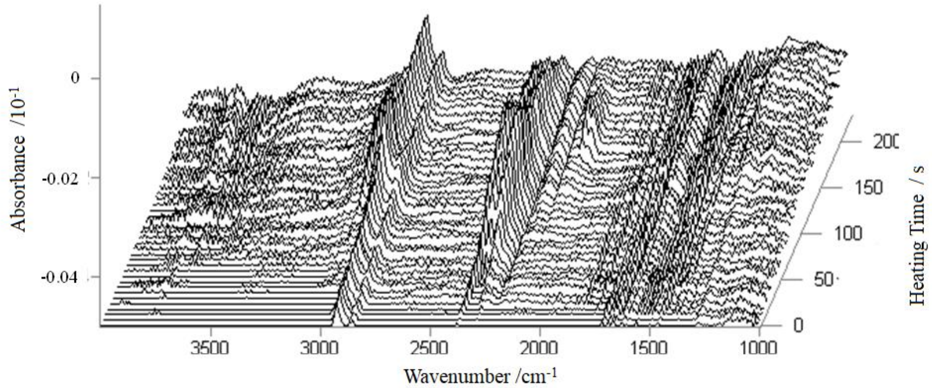
The peak temperatures of different gases in the thermal simulation experiments of samples 1 and 2 are, in order from low to high, $CO_2$, $C_{2+}$, $H_2O$, $CH_4$, and $H_2$ (Table 3), indicating that the generation of $C_{2+}$ and $CO_2$ components in the coal molecular structure during heating is relatively high and the reaction difficulty is relatively low. This is followed by $H_2O$ and $CH_4$, with the generation of $H_2$ being the most difficult.

Table 3 The formation peak temperatures of different gases in two coal samples thermal simulation experiments

| Coal Sample | $CO_2$ | $C_{2+}$ | $H_2O$ | $CH_4$ | $H_2$ |
|---|---|---|---|---|---|
| sample 1 | 395℃ | 446℃ | 515℃ | 550℃ | 765℃ |
| sample 2 | 420℃ | 465℃ | 515℃ | 525℃ | 750℃ |




a) sample 1




b) sample 1

Fig.3 Infrared spectra of two coal samples in thermal simulation experiment

From the results of the thermal simulation infrared experiments of the two coal samples (Fig. 3), it can be seen that the most obvious wavenumbers in the infrared spectra of samples 1 and 2 are mainly distributed at 1100-1400 $cm^{-1}$, 1500-1750 $cm^{-1}$, 2360 $cm^{-1}$, 2800-3100 $cm^{-1}$, and 3500-4000 $cm^{-1}$. Among them, 1100-1400 $cm^{-1}$ and 2800-3100 $cm^{-1}$ are the absorption bands of $CH_4$ molecules.The absorbance of these two bands gradually increases with temperature, reaching a peak and then slowly decreasing. The 1500-1750 $cm^{-1}$ and 3500-4000 $cm^{-1}$ are the absorption bands of $H_2O$ molecules. These bands are widely distributed, and their absorbance remains relatively balanced throughout the entire heating stage, indicating a relatively high source and abundance of $H_2O$ molecules. The 2360 $cm^{-1}$ band is the absorption band of $CO_2$ molecules, with a narrow absorption range and high absorption intensity. The absorbance of $CO_2$ generally shows a pattern of slow increase, peak, shoulder, and slow decrease with the rise in temperature. Overall, the results of the thermal simulation infrared spectroscopy experiments are consistent with those of the gas generation thermal simulation experiments.

## 4. Types and Mechanisms of CH4 Generation

Coal is a mixture composed of a large number of molecules with different molecular weights [27-38]. The macromolecular structure is mainly composed of condensed aromatic nuclei, aliphatic side chains, and oxygen-containing functional groups. Coals with low metamorphic degrees have lower aromatic ring condensation and more aliphatic side chains and oxygen-containing functional groups. In contrast, coals with medium to high metamorphic degrees have higher aromatic ring condensation and fewer aliphatic side chains and oxygen-containing functional groups. In the highly metamorphosed anthracite stage, the aromatic rings are highly condensed [29-33]. Based on this understanding and the results of the gas generation thermal simulation experiments, the types and mechanisms of $CH_4$ generation reactions in the six different stages shown in Fig. 2 are discussed.

As shown in Fig. 4, in the first stage, $CH_4$ is generated physically, mainly through the thermal desorption of adsorbed and absorbed $CH_4$. The anomalous production of $C_{2+}$ hydrocarbons in this stage is likely due to the direct thermal release of small molecular aliphatic hydrocarbons from the coal molecular structure. In the second stage, the generation of $CH_4$ involves chemical reactions such as decarboxylation and decarbonylation, primarily resulting from the detachment of aliphatic side chains containing oxygen functional groups from the aromatic ring structure in coal, producing significant amounts of $CO_2$ and $H_2O$ and a small amount of $CH_4$. In the third stage, the chemical reactions are mainly depolymerization reactions, with decomposition reactions as secondary processes. The depolymerization of long aliphatic chains from aromatic nuclei produces large amounts of $C_{2+}$ hydrocarbons, while some decomposition results in a small amount of $CH_4$.

In the fourth stage, the chemical reactions are dominated by decomposition reactions, with depolymerization reactions being secondary. After depolymerization from the aromatic nuclei, the long aliphatic chains further decompose to generate large amounts of $CH_4$. The fifth stage is primarily characterized by condensation reactions, mainly involving the polymerization of aromatic nuclei and the aromatization of aliphatic hydrocarbons. The C and H atoms produced combine to form $CH_4$. Due to the imbalance in the number of C and H atoms generated, some H atoms combine with each other to form $H_2$, while others combine with hydroxyl groups to form $H_2O$. In the sixth stage, condensation reactions remain predominant. The number of C atoms is insufficient to meet the requirement for combining with H atoms to generate $CH_4$; thus, $CH_4$ production is minimal or negligible, with $H_2O$ and $H_2$ being the main products.

It can be observed that gases such as $H_2O$ and $H_2$ have a competitive effect on $CH_4$ generation by scavenging H atoms and reducing the amount of $CH_4$ produced. Conversely, $C_{2+}$ hydrocarbons, which are generally produced at low temperatures, reduce the quantity of $CH_4$ generated from further decomposition at high temperatures.

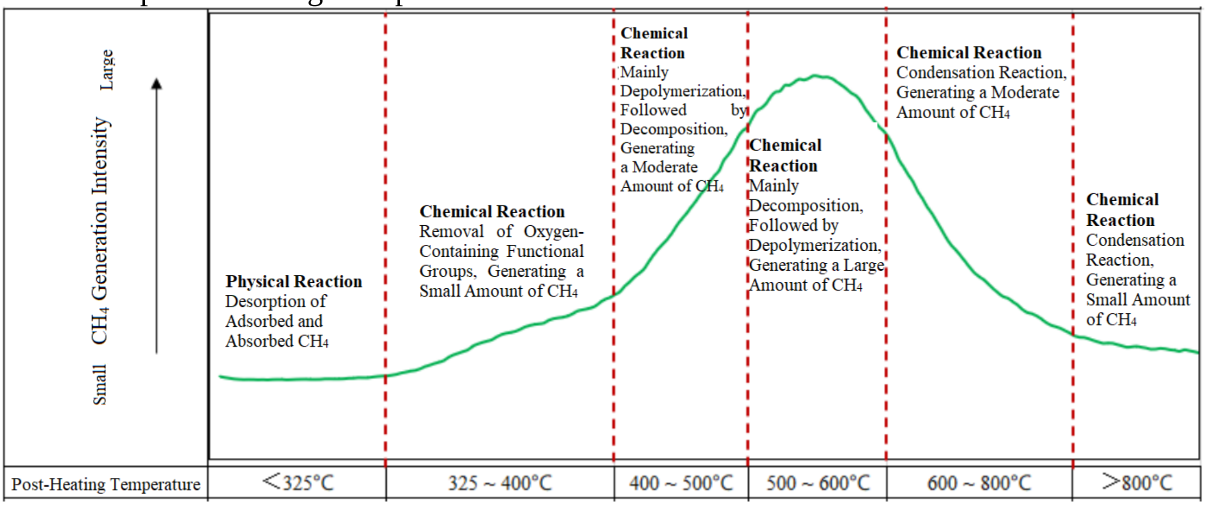



Fig.4 $CH_4$ generation stages and generation mechanism types

## 5. Discussion on the Controlling Mechanism of the Relationship Between Methane Content and Metamorphic Degree

Based on the vitrinite reflectance test data from the thermal simulation experiments conducted by Tao Mingxin et al. [3], a linear relationship between vitrinite reflectance and heating temperature (336–600°C) was established (Fig. 5). Using this relationship, the vitrinite reflectance corresponding to the temperature limits of each stage shown in Fig. 2 was inferred, associating temperature with the coal metamorphic process and $CH_4$ generation intensity with $CH_4$ content. This allowed for an exploration of the influence of gas generation on $CH_4$ content during the coal metamorphic stages (vitrinite reflectance > 0.5%) (Fig. 6).

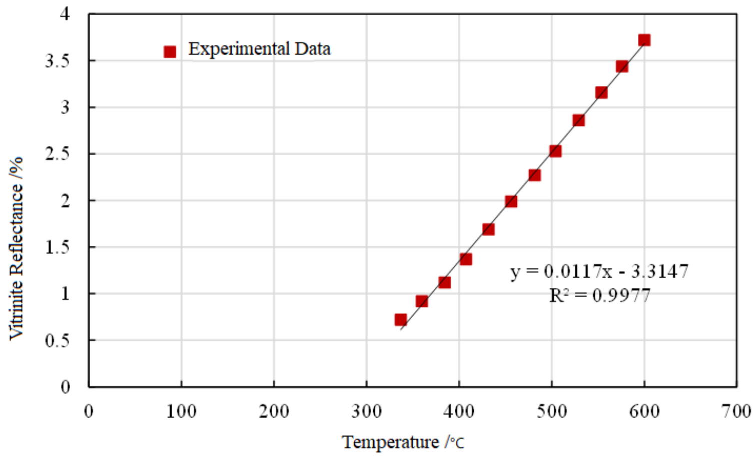



Fig.5 Relationship between vitrinite reflectance and temperature of thermal simulation experiment of coal

As shown in Fig. 6, when the temperature rises from 325℃ to 800℃ and the vitrinite reflectance increases from 0.5% to 6.0%, there is a strong similarity between the $CH_4$ generation curve and the $CH_4$ content distribution curve. At the temperature range of 325–400℃ and vitrinite reflectance range of 0.5%–1.30%, the $CH_4$ generation intensity increases rapidly, and the $CH_4$ content also shows a rapid increase from less than 1 m³/t to around 10 m³/t. At the temperature range of 400–500℃and vitrinite reflectance range of 1.30%–2.50%, the $CH_4$ generation intensity continues to grow rapidly, with $CH_4$ content increasing further from around 10 m³/t to 15 m³/t. In the temperature range of 500–600℃ and vitrinite reflectance range of 2.50%–3.75%, the $CH_4$ generation intensity reaches its peak, and the $CH_4$ content also reaches its highest range, between 15 and 25 m³/t. When the temperature exceeds 600℃ and the vitrinite reflectance is greater than 3.75%,the $CH_4$ generation intensity declines sharply, and the $CH_4$ content also decreases steeply, from less than 15 m³/t to below 1 m³/t.

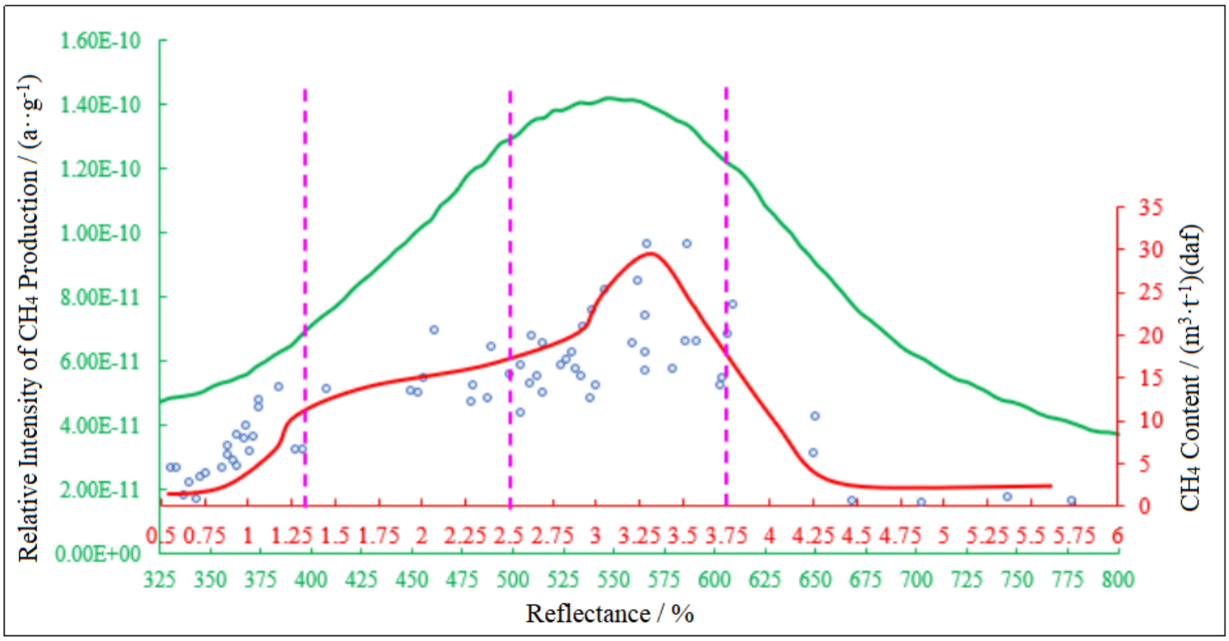



Fig.6 Relationship between $CH_4$ generation during coal metamorphism and CH4 content in different rank coals

In summary, the differences in $CH_4$ generation intensity during coal metamorphism are considered the primary controlling factor causing the current differences in $CH_4$ content in coals of different metamorphic degrees. In the low metamorphic stage, the $CH_4$ generation intensity is low, the gas generation is limited, and the coal seam $CH_4$ content is low. In the medium to high metamorphic stages, the $CH_4$ generation intensity increases, the gas generation increases, and the coal seam $CH_4$ content is high. The abnormally low $CH_4$ content in No. 1 and No. 2 anthracite coal seams is mainly due to the severely insufficient capacity of these metamorphic stage coals to generate $CH_4$. Therefore, it is concluded that the distribution characteristics of coalbed methane content in different regions of China are primarily based on variations in coal metamorphic degrees, while later reservoir and preservation conditions do not exhibit a controlling effect on the differentiation of gas content.

## 6. Conclusions

By compiling gas content test data from coal seam cores obtained by surface drilling in dozens of representative areas in China, it was found that the dry ash-free $CH_4$ content has a relatively obvious relationship with the maximum reflectance of vitrinite. There is also a certain pattern in the proportion of $CH_4$ in the gas composition relative to the maximum reflectance of vitrinite.

Based on the results of the gas generation thermal simulation combined with mass spectrometry experiments and pyrolysis infrared spectroscopy experiments, the characteristics of gases such as $CH_4$, $CO_2$, $H_2O$, $H_2$, and $C_{2+}$ generated from two demineralized lignite samples were analyzed. The production curves of each gas generally exhibited a unimodal pattern. Based on the characteristics of $CH_4$ generation, the entire gas generation process during heating was divided into six stages, and the types and mechanisms of $CH_4$ generation in each stage were studied. These primarily include physical processes at low temperatures and chemical reaction types at medium to high temperatures. A comprehensive discussion was conducted on the relationship between the intensity of $CH_4$ generation and $CH_4$ content in coal. It is concluded that the intensity of $CH_4$ generation at different metamorphic stages is the main controlling factor for the differentiation of $CH_4$ content distribution with metamorphic degree.

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