

# A review on microbial metabolism to increase coalbed methane generation and coal pretreatment to improve its bioavailability

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**Abstract** Abundant unminable coal in deep strata and abandoned mines are also precious sources of clean gas energy, under which biotransformation is a potential path. In recent years, substantial progress has been made in laboratory research on coal degradation to produce methane by microbial metabolism. This paper systematically reviews the research progress of microbial enhancement and microbial stimulation of coal, physicochemical pretreatments of coal, and environmental factors affecting coal biotransformation. The research idea of coal biotransformation should aim at field production increase and gradually clarify the microbial mechanism of coal degradation and the regional distribution and functional composition of microbial communities on the block scale. The research on coal biotransformation helps improve the development level of coalbed methane and the sustainable development of unconventional natural gas resources.

**Keywords** coal biotransformation, methanogens, microbial mechanism, *in situ* environment, carbon cycle

## 1 Introduction

China's energy consumption structure with a high proportion of coal leads to low energy utilization efficiency and environmental concerns (Fu et al., 2019, 2021; Zhao et al., 2020; Li et al., 2022d). Under the background of carbon peak and carbon neutrality, clean

utilization of coal is particularly important (He et al., 2020; Zhao et al., 2020). microbial means, supplemented by physical, chemical, and engineering technologies, are used to activate microbial metabolism to promote the degradation of coal macromolecules into methane and other gases before mining and utilization to realize the clean and efficient utilization of coal (He et al., 2020; Su et al., 2020). Chinese substantial underground coal reserves provide material sources for coalbed methane exploitation and development opportunities to increase biogenic methane (Davis and Gerlach, 2018).

Shimizu et al. (2007) first reported the microbial community in the coal seam of Hokkaido, Japan. They measured the diversity of bacteria and archaea in the water of the coalbed methane reservoir. Many scholars have found that necessary bacteria and archaea related to coal biodegradation are common in coalbed methane reservoirs worldwide (Bao et al., 2016; Su et al., 2018; Xia et al., 2021). However, under natural conditions, the amount of methane produced by the anaerobic fermentation of coal is tiny (Su et al., 2018). The concept of Microbial Enhanced coalbed methane (MECBM) was proposed, which was initially designed to increase the production of biogenic coalbed methane and improve the permeability of the reservoir (Scott, 1999). In China, Su et al. (2020) proposed combining the metabolism of microorganisms with modern engineering and technical means to accelerate coal degradation into coalbed methane and liquid organic matter.

Coal is a kind of organic material that is difficult to degrade and highly uneven, so it is difficult to characterize its chemical composition fully. Coal degradation requires a series of anaerobic microbial



biotransformation through manual intervention (Davis and Gerlach, 2018; Wang et al., 2019b). At present, the research on increasing coalbed methane production by microbial action mainly focuses on several directions: introducing exogenous high-efficiency microorganisms (microbial enhancement), stimulating microbial metabolism by nutrients (microbial stimulation), pretreating coal to change its physical or chemical properties to improve the bioavailability of coal, and improving environmental conditions to enhance the intensity of microbial metabolism (Chen et al., 2017). This paper systematically combs several essential research directions of microbial stimulation of coalbed methane and puts forward feasible suggestions from laboratory-scale microbial stimulation research to its industrial application.

## 2 Microbial enhancement and microbial stimulation of coal

Microbial enhancement is a method to increase the production of biogenic coalbed methane by adding exogenous microorganisms. The added microorganisms can be a single microbial strain. However, most of them are microbial communities that can degrade coal to produce methane. The added microorganisms are more active than those in the *in situ* reservoir or supplement the lack of necessary microbial communities. The research on high-efficiency functional microorganisms is mainly the domestication or improvement of exogenous microorganisms, carried out only under laboratory conditions and rarely applied to the field (Su et al., 2018). The exogenous microorganisms come from various environments or organisms rich in organic matter and producing methane, including coal with different ranks, lake and river sediments, animal feces, crop soil, and wood-eating termites (Fuertez et al., 2017; Guo et al., 2019b).

In laboratory culture, the microbial communities appear to enrich some strains. The impact of sampling sources on the composition of archaea is limited. The addition of nutrients or culture environment is more important, which affects the species of archaea and the way of methane production. Due to the rich metabolic pathways of bacteria, the composition diversity is affected by the culture stage and the difference in sampling sources (He et al., 2020). Microorganisms can choose their functional potential through their strong environmental adaptability. Once the environment changes in a particular range, highly competitive microorganisms can adapt to the changes of the environment and grow and strengthen specific metabolic functions. Therefore, domesticated microorganisms can maximize their metabolic potential, such as the ability of microbial communities to degrade coal, more targeted, more microbial types to participate in

all stages of methane production, and maintain stable abundance (Zhao et al., 2020).

Under natural conditions, the microbial communities in coalbed methane reservoirs degrade coal slowly, which is affected by limited *in situ* nutrients and the low bioavailability of coal. Microbial stimulation is the addition of nutrients to activate the metabolic rate of microorganisms (Xia et al., 2021). The composition of nutrients in microbial culture medium is different. However, there are standard components: main minerals, organic nitrogen source, vitamins, trace elements, reductant, and redox indicator. The effects of nutrients with different components and concentrations on biogenic methane production are significantly different (Fuertez et al., 2017). In the initial stage of microbial stimulation, the methane production efficiency is high. After repeated addition of nutrients, the methane production efficiency slows down significantly. The continuous and repeated addition of nutrients seems to limit the efficiency of coal biotransformation to some extent. Therefore, a more comprehensive understanding of the microbial mechanism of repeated addition of nutrients and whether there is an inhibition of intermediate product accumulation is needed. The optimal type and concentration of added nutrients should be further studied (Davis and Gerlach, 2018). In addition, a more comprehensive range of fluid environments is likely to affect the cycle and effect of stimulation. Simulating the *in situ* environment on a larger scale is necessary to determine microbial stimulation's actual impact and application prospect (Davis et al., 2019).

Increased biogenic coalbed methane production can be provided by stimulating microbial metabolism with nutrients (Barnhart et al., 2022). The yeast extract stimulated microbial growth in biomethane-free and sulfate-rich sub-bituminous coal seams and achieved good results (Barnhart et al., 2022). By adding nutrients *in situ* coal seam water, the microorganisms of degraded coal were gradually diversified, and the microbial community structure was optimized. Most microbial functions were related to coal metabolism (Li et al., 2022b). The coal gasification medium was optimized by single-factor analysis and discussed the influence of a single component on the microbial community structure and main metabolic pathways. The sediments on the coal surface decreased obviously, and there was no significant difference in the composition and content of liquid organic matter. The composition of microbial community structure and gene function did not weaken with the decrease of nutrients, but a specific and stable microbial community formed (Zhou et al., 2022). Microorganisms in biogenic coalbed methane reservoirs are composed of microbial communities with complementary metabolic activities.

In contrast, a small number of bacteria and methanogens have been detected in some blocks lacking

biogenic coalbed methane, but they cannot provide a suitable growth environment (Su et al., 2018). A long-term evaluation has been made on the stimulation of microorganisms by acetic acid (a critical substrate available to methanogens) for more than 1.5 years. Acetic acid was consumed more than methane. Acetic acid was not only produced by methanogens but also consumed by sulfate-reducing bacteria. However, high sulfate concentration and sulfate reduction could not prevent acetic acid from stimulating the metabolism of methanogens (Beckmann et al., 2019).

Determining the type and concentration of nutrients required by a reservoir has a high cost, so some scholars are committed to finding low-cost universal alternative nutrients. The limited hydrogen content in coal is the limiting factor affecting its biotransformation. Straw is mainly composed of lignin, hemicellulose, and cellulose with high hydrogen, which can be used as an anaerobic hydrogen production material. The structure of straw is simpler than coal, which is more easily degraded by microorganisms. Adding straw promotes the complementary advantages of coal and straw microorganisms in fermentation. Therefore, the co-degradation of straw and coal is a feasible method to improve methane yield (Guo et al., 2018, 2019a, 2020a). In the initial stage of fermentation, the complementation of metabolites and microorganisms in coal and straw enhances the activity of microorganisms and makes the straw fully degraded by microorganisms. The organic matter produced by later degradation can further activate the microbial degradation of coal, which is the main reason for the co-degradation of straw and coal to improve methane yield. The abandoned coal mine filled with straw solves the problems of low filling rates and high investment in filling materials, promotes the regeneration of clean energy, and realizes the recycling of resources. However, the microbial mechanism of the co-degradation of straw and coal is unclear, which is very important for its commercial application (Guo et al., 2020b).

Large-scale biogenic methane production experiments have been conducted on-site, but the results are uneven. The primary way to increase production on-site is to add nutrients to stimulate indigenous microorganisms (Su et al., 2018). Luca Technologies company attempted to produce biogenic methane in areas where biogenic coalbed methane had not previously been discovered. To protect intellectual property rights, the company did not disclose the details of the experiment (Ritter et al., 2015). Luca Technologies has at least 13 patents related to this, mainly through microbial stimulation activities by adding nutrients to coal seams. The goal is to achieve additional coalbed methane production as the coalbed methane production well nears the end of its life. The well can recover to about 50% of the peak productivity with a successful experiment. The coalbed methane production

increased in about five years and then began to decrease, indicating that nutrient solution needs to be re-injected every five years (Ritter et al., 2015). Next Fuel company took microbial stimulation as an essential means to stimulate biogenic methane production. The main target is the lignite coal seam without microbial methane production history in the past, which indicates that there are generally methane-producing microbial communities in the coal seam. The specific nutrients have not been disclosed, but they do not contain carbon (Ritter et al., 2015). It may be an essential research and application direction in the future because the existing infrastructure can be used for biogenic methane yield increase. The cost of yield increase measures is low while the benefits are high (Su et al., 2018).

Because the microbial community is affected by the types of available organic matter, adding a small amount of organic matter can improve the rate of coal biotransformation. In some studies, adding a simple substrate to methanogens as nutrients increased methane production. However, it may be the substrate that is more easily utilized by methanogens, which did not significantly improve the efficiency of coal biotransformation. Therefore, the target microorganisms should be a series of bacterial and archaeal communities in degrading coal macromolecules, not limited to a specific microbial type (Davis and Gerlach, 2018). Microbial methane production in coal seams is limited by microbial metabolic potential and lack of available nutrients and is also affected by the inherent stubbornness of coal itself (Barnhart et al., 2017; Lyles et al., 2017). Therefore, some studies have improved the bioavailability of coal by changing the physical and chemical properties through pretreatment.

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### 3 Influence and response of physicochemical properties of coal on its biotransformation

Coal rank and physicochemical properties are controlled by the sedimentary environment and coalification process, which are internal factors affecting coal biotransformation (Bao et al., 2016). The exploration and development of coalbed methane prove that biogenic coalbed methane can be produced in reservoirs of different coal ranks (Bao et al., 2016; Su et al., 2018; Zhao et al., 2020; Xia et al., 2021). Compared with high-rank coal, low-rank coal has the advantage of biotransformation because of its low degree of molecular aromatization, more oxygen-containing functional groups, and branched chains (Chen et al., 2017). Biogenic coalbed methane has also been found in some areas of medium and high-rank coalbed methane reservoirs, such as the Qinshui Basin. However, biogenic coalbed methane is not enough to form reservoirs due to the

limited available substrate of methanogens or the consumption of biogenic coalbed methane produced by other anaerobic microorganisms (Li et al., 2020a). In addition, there is a corresponding relationship between the abundance and diversity of microorganisms in coal seams and coal rank. Coalbed methane reservoirs with low maturity have high microbial abundance and diversity, but it is unclear whether coal rank is the main factor of coal biotransformation (Li et al., 2019). The microbial metabolism of coal to produce methane is a material transfer process. The surface area of coal in contact with microorganisms will inevitably affect the microbial metabolic efficiency. When the contact surface is large enough, reducing coal particle size will no longer promote improving gas production efficiency. The maceral difference of coal also affects the difficulty of biodegradation. Vitrinite has a high hydrogen carbon atom ratio, low fatty hydrocarbon content, and good pore connectivity. Inertinite has high aromatic hydrocarbon content, a high aromatization degree, and fewer hydrogen and oxygen functional groups. Therefore, vitrinite is easy to biodegrade and has excellent gas production potential compared with inertinite. On the premise of no pretreatment of microorganisms and coal, the biogenic methane production of low-rank coal is generally better than that of high-rank coal. In some studies, the influence of coal rank is weakened due to its well-developed cleat and fracture system (Bao et al., 2016; Chen et al., 2017; Su et al., 2020; Zhao et al., 2020; Xia et al., 2021).

The pore fissure system of the coalbed methane reservoir determines the seepage conditions of fluid. The poor development of reservoir pores and low permeability is also one of the problems restricting coalbed methane exploitation (Li et al., 2020c). Microbial action can significantly improve reservoir pores and fractures and increase the permeability of coal seams. The influence of microbial metabolism on coal seam pores and fractures is manifested in nano and micron scales. It is difficult for microorganisms to enter the nanopores of coal, but their secretion can be transmitted through the nanopores (Su et al., 2020). After enriched and cultured microorganisms with anthracite, bituminous coal, and lignite, the pore volume of large and medium pores of bituminous coal and lignite increases, while the pore volume of micropores and ultra micropores decreases. The surface roughness of anthracite micropores increases, and the complexity of transition pores decreases. The pore changes are caused by the attachment of microorganisms and the retention of metabolic intermediates. Xia et al. (2014) conducted coal biotransformation experiments under laboratory conditions using three different coal rank coals. After microbial action, the macropore volume and proportion of the three coals increased. Furthermore, the micropore volume decreased, reducing their adsorption capacity. Guo et al. (2017b) proved that microbial metabolism promotes the formation of new

pores and the expansion of old pores. Because methanogens are readily adsorbed on the surface of low-rank coal, the effect of microbial action on low-rank coal is particularly prominent. Therefore, coal biotransformation increased the coalbed methane content and was conducive to coal seam permeability enhancement and gas desorption.

Anaerobic fermentation of coal is a highly complex process, which is bound to cause changes in various chemical parameters of coal, such as the decrease of C and O content, the increase of N and H content, the content and occurrence state of trace elements before and after the reaction also change significantly (Wang et al., 2019b). Volatile fatty acids are the intermediate products of coal biotransformation and can be used as essential monitoring indicators (Chen et al., 2017). As the substrate, lignite is used for gas production, divided into two stages. The first stage is the gas production of the humic group, and the second is the gas production of the inert and stable groups. Methanogens produce methane through acetic acid fermentation and carbon dioxide reduction in the two stages. The abundance and activity of methanogens and the content of the humic group determine the gas production (Wang, 2012). Microorganisms preferentially act on saturated hydrocarbons, normal alkanes, and low-carbon alkanes in the degradation process and gradually consume long-chain alkanes later (Guo et al., 2018, 2019b).

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#### 4 Physicochemical pretreatments of coal promote its biotransformation

Pretreatment of coal can significantly improve its bioavailability to accelerate the degradation process of coal. Common pretreatment methods include physical pretreatment and chemical pretreatment. Microorganisms such as degrading bacteria and methanogenic archaea cannot enter the internal molecular structure of coal, and methanogenic activity is limited to the cleavage surface and cracks of coal (Robbins et al., 2016). The microbial communities distributed in the reservoir water differ from those attached to the coal surface. The microbial communities connected to the coal surface play a more significant role in the initial stage of coal degradation. Hydraulic fracturing can provide a method to increase the contact area between microorganisms and the coal matrix. Groundwater flow increases biomass and diversity of microorganisms, reduces salinity, and introduces nutrients and terminal electron receptors, thereby stimulating the growth of microorganisms in coal seams (Raudsepp et al., 2016). The physical pretreatment methods commonly used in the laboratory include heating, photooxidation, ultrasonic treatment, high-energy radiation pretreatment, and swelling treatment.

The hydrophobicity and heterogeneity of coal

significantly limit its bioavailability. The hydrolysis of coal macromolecular is the limiting step of biotransformation (Liu et al., 2019). Chemical pretreatment of coal can substantially reduce the surface hydrophobicity and has higher efficiency in promoting biodegradation (Davis et al., 2019). Chemical pretreatment methods mainly include oxidants, acids, bases, organic solvents, and surfactants (Nie et al., 2019). Oxidants can cleave the coal's molecular structure and increase the soluble organic matter's content. Under laboratory conditions, the oxidant concentration controls the coal's gasification effect. Research on the pretreatment of coal with  $H_2O_2$  is the most common.  $H_2O_2$  pretreatment can significantly improve the bioavailability by reducing the aromaticity and increasing oxygen-containing functional groups of coal (Guo et al., 2017b). The effect of coal pretreatment with  $KMnO_4$  is similar to that of  $H_2O_2$ , which proves that the oxidants can effectively improve the bioavailability of coal.

However, the common problem of chemical pretreatment is that the environment after pretreatment needs to be adjusted to adapt to the growth and metabolism of anaerobic bacteria. After acid pretreatment, coal's chemical bond and ring structure are destroyed. After pretreatment with  $HNO_3$ , the biotransformation rate of lignite by fungi is significantly improved, and the main products are aromatic acids and aliphatic chain hydrocarbons (Guo et al., 2017a, 2019c). Due to its alkali action and strong corrosivity, strong alkali causes a decrease in the crystal nucleus structure of coal, the looseness of the hexagonal aromatic ring structure, and the increase of phenols and alcohols (Jian et al., 2019; Zhang et al., 2019a; Huang et al., 2021). Pretreatment of anthracite in the Qinshui Basin with NaOH solution also reduces the aromaticity of coal and increases the proportion of the C-O bond. Some alkaline substances secreted during microbial growth may be involved in coal degradation. The alkaline metabolites secreted by fungi can ionize the acidic functional groups of low-rank coal and improve the hydrophilicity of coal. Organic solvents can effectively extract organic matter from coal.

$C_2H_5OH$  can also be used as an electron donor, carbon source, and organic solvent (Zhang et al., 2019b). As an organic solvent,  $C_2H_5OH$  improves the solubility of biodegradable substances in coal. It changes microbial structure and methane production mode, which does not lead to the change of coal chemical structure (Yang et al., 2019). Yang et al. (2019) conducted a lignite fermentation and gas production experiment with heterologous bacteria. By adding different concentrations of  $C_2H_5OH$  to the culture medium, it was found that the gas production was the largest after treatment with the 1%  $C_2H_5OH$  solution.  $C_2H_5OH$  can be used as an effective extraction solvent to increase the biotransformation efficiency of coal. It may also inhibit some coal rank coal samples, so various solvents should be developed for

different coal samples (Zhang et al., 2019a). The material transfer between coal and bacteria needs biofilm as the medium. The dynamic and complex biofilm structure is conducive to avoiding the interference of the changeable physical and chemical environment. Surfactants can enhance the surface properties of coal to increase the adsorption capacity of bacteria on its particle surface and promote biofilm formation, which may be the key to further improving the production efficiency of biogenic methane (Guo et al., 2019c).

Microorganisms in the reservoir usually differ from the microbial communities under laboratory culture conditions (Li et al., 2020b; Shi et al., 2021). There are significant differences in the abundance and distribution of microbial communities under different redox conditions, and methanogens are more active in the reducing environment (Shi et al., 2021). The hydrodynamic strength of the No. 3 coal reservoir south of the Qinshui Basin gradually weakens from east to west, forming the transition characteristics from runoff area to stagnation area. The stagnant area has higher reservoir pressure, gas content, and ion concentration than the runoff area. In the stagnant area, the relative abundance of cellulose-degrading microorganisms, methane metabolizing microorganisms, N-cycle related microorganisms, and S-cycle related microorganisms related to C, N, and S cycles increased, indicating that the microbial cycle in the stagnant area is more active (Shi et al., 2021; Li et al., 2022a). Therefore, it is essential to accelerate the study process of *in situ* environments.

#### 4.1 Research progress of $H_2O_2$ pretreatment of coal

A common problem of chemical pretreatment is the impact of chemical residues, pH, and salinity on the environment, so it is necessary to adjust the pretreated environment to make it suitable for microbial survival.  $H_2O_2$  is a more suitable chemical reagent, which has little effect on pH and salinity and will not introduce foreign chemicals (Haq et al., 2018).  $H_2O_2$  can quickly promote low molecular weight organic matter for methane generation in northern Hokkaido, Japan, as a solubilizer into the lignite under high pressure. After pretreatment of bituminous coal with  $H_2O_2$ , dissolved organic matter's type and content increase, including aliphatic carboxylic acids, alcohols, ethers, ketones, and aromatic hydrocarbons (Aramaki et al., 2017). Sub-bituminous coal treated with  $H_2O_2$  increases methane production ten times in 30 days. The methane production increases exponentially with the increase of  $H_2O_2$  concentration and the extension of pretreatment time (Chen et al., 2018). By pretreated sub-bituminous coal from Powder River with  $H_2O_2$ , it is found that the composition and distribution of organic matter after coal degradation under microbial action, unstable compounds such as short-chain carboxylic acids ( $C_1$  to  $C_6$ ) help to shorten the biogas

production cycle and are not easy to be partially degraded by biodegradable components (Wang et al., 2019b).  $H_2O_2$  treatment can also improve anthracite's aromaticity and reduce crystalline carbon content (Zhang et al., 2019b). Guo et al. (2021a) pretreated anthracite with  $H_2O_2$  and found that high concentration and long-term  $H_2O_2$  pretreatment can improve the biogenic methane production of high-rank coal. Aromatic hydrocarbons and aliphatic hydrocarbons depolymerize under the action of  $H_2O_2$  to form oxygen-containing functional groups, which anaerobic bacteria can further degrade. As shown in Fig. 2, according to the coal samples of different coal reservoirs treated with varying concentrations of  $H_2O_2$ , the methane production of low-rank coal treated with high concentrations of  $H_2O_2$  generally increases significantly. Some experiments show that soaking in high concentrations of  $H_2O_2$  can also increase the methane production of high-rank coal. Before field application, the economic feasibility and environmental hazard effects of  $H_2O_2$  *in situ* pretreatment should be comprehensively evaluated (Huang et al., 2021).

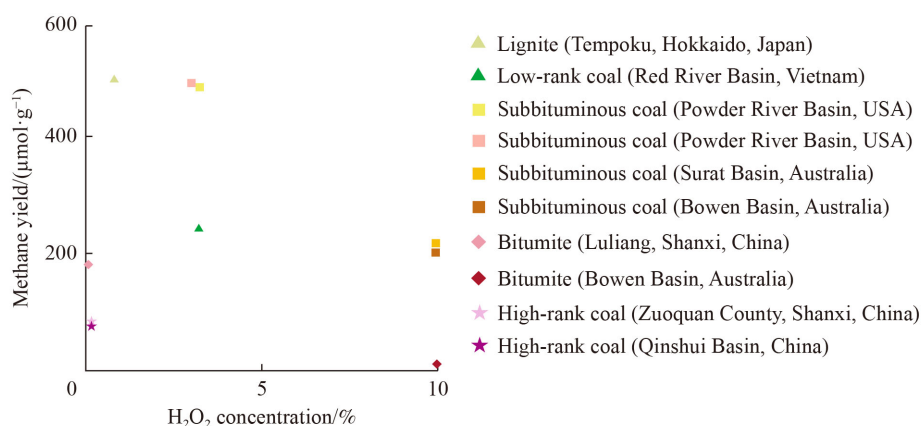
#### 4.2 Research progress of supercritical $CO_2$ pretreatment of coal

The biotransformation of coal has attracted attention due to its uniqueness in coal modification, coalbed methane regeneration, and environmental friendliness (Guo et al., 2021a). However, the effect has not reached the expectation. On the one hand, the structure of coal is complex, and high molecular organic compounds limit microbial degradation. On the other hand, the organic matter available to microorganisms may be trapped in the pore structure of coal, which cannot be fully utilized. Together, they lead to coal's low biotransformation rate (Guo et al., 2019b). Solvent extraction breaks aromatic or aliphatic branched chains and separates functional groups or side chains. The extraction process weakens the irregular three-dimensional cross-linking structure of

coal. The coal structure, with increased pores and cracks, becomes loose. Microorganisms are more likely to enter the internal structure (Gao et al., 2020).

Most biogenic coalbed methane reservoirs discovered so far are produced by  $CO_2$  reduction, and almost all types of methanogens can produce methane by  $CO_2$  reduction. In addition, the  $CO_2$  displacement of coalbed methane has become a conventional method to improve coalbed methane recovery (Guo et al., 2019a, 2019c; Guo et al., 2021b).  $CO_2$  extraction technology is widely used to extract active substances from various mixtures because of its harmlessness and the availability of high-quality soluble organic matter. Some studies have shown that  $CO_2$  extraction can effectively dissolve organics in coal. The destruction of non-covalent bonds involved in the extraction process leads to the separation of small molecular organics from the macromolecular network of coal. Especially under the condition of supercritical  $CO_2$ , the pore connectivity of coal is improved (Guo et al., 2021b). Guo et al. (2021b) improved the biotransformation rate of coal based on the principle of supercritical  $CO_2$  extraction. After supercritical  $CO_2$  extraction pretreatment of anthracite and bituminous coal, the methane yields of anthracite and bituminous coal were increased by 734.85% and 148.15%, respectively. The methane yield of coal treated by subcritical  $CO_2$  extraction has hardly improved. Supercritical  $CO_2$  extraction can form more or even new functional groups, increase specific surface area and total pore volume, and provide more active sites for microorganisms and enzymes.

Geological storage of  $CO_2$  provides a new idea for carbon reduction and has attracted the attention of scholars worldwide. The underground storage of  $CO_2$  has the risk of leakage. Converting  $CO_2$  into clean energy is an effective way to solve this problem. Most underground coal seams are highly reductive environments where methanogens can use bicarbonate as the primary electron acceptor. When the buried depth of the coal seam exceeds



**Fig. 2** Comparison of gas production of different rank coal samples treated with different concentrations of  $H_2O_2$  (Aramaki et al., 2017; Chen et al., 2018; Liu et al., 2019; Wang et al., 2019b; Zhang et al., 2019b; Huang et al., 2021).

a certain depth (more than 800 m), the reservoir's temperature and pressure conditions easily reach the CO<sub>2</sub> supercritical state. Supercritical CO<sub>2</sub> extraction releases soluble organic matter, which microorganisms utilize to produce methane, to realize the dual effects of carbon dioxide emission reduction and coalbed methane production increase. Therefore, when the *in situ* coal seam or artificially transformed underground environment reaches the supercritical CO<sub>2</sub> extraction conditions, it is of great significance for improving the biotransformation of coal and CO<sub>2</sub> emission reduction (Guo et al., 2021b).

## 5 Environmental factors affecting coal biotransformation

External factors such as temperature, pH, salinity, and solid-liquid ratio also impact coal biotransformation (Zhao et al., 2020). Appropriate temperature is conducive to maintaining microbial diversity and metabolic stability (Davis and Gerlach, 2018). Methanogens are sensitive to temperature, and their survival temperature range is extensive (0°C–70°C). However, the temperature in the range of 5°C–45°C is positively correlated with methane production, and the metabolic rate of methanogens is significantly lower than 50°C. The best temperature of various reservoirs is slightly different (Guo et al., 2017b). Although increasing the reservoir temperature *in situ* is not economically feasible, most underground temperatures are more suitable for microbial activities. The physiologic characteristics of microorganisms to adapt to the *in situ* temperature environment can also be improved by domestication (Zhao et al., 2020). The tolerance range of different microbial types to pH is slightly different. Acid-producing bacteria have low sensitivity and can tolerate the pH range of 4.0–8.5, while hydrolytic bacteria are suitable for operating in 5.5–6.5 (Li et al., 2020b). The abundance of methanogens decreases with the increase of salinity. The rise of salinity is not conducive to generating biogenic coalbed methane (Li et al., 2022a). The effect of the solid-liquid ratio on methane generation is similar to that of coal particle size, and excessive coal in a limited space will limit the metabolic efficiency of methanogens (Zhao et al., 2020). Good groundwater conditions affect the transport of nutrients and metabolites of bacteria and archaea. These environmental variables jointly determine coal degradation's fermentation rate and metabolic pathway to methane in coalbed methane reservoirs (Davis and Gerlach, 2018).

The electrochemical battery can reduce CO<sub>2</sub> to CH<sub>4</sub> by applying an external electric field, in which the reduction reaction of the cathode involves electrochemistry and microbial metabolism (Zhao et al., 2022). As shown in Fig. 3, the significant advantage of microbial

electrochemical batteries is that external power supply and cell metabolism replace hydrogen as reducing agents (Giang et al., 2018). The biotransformation of coal depends on the continuous degradation of intermediates. Hydrolysates and degradation intermediates accumulate in the reaction and inhibit methane production. After applying an electric field, the abundance of bacteria with extracellular electron transfer ability increases significantly. The microbial community structure changes significantly (Zhao et al., 2022). Therefore, the applied electric field changes the structure of microbial communities, promotes extracellular electron transfer and biodegradation of organic matter, and even changes the degradation pathway of organic matter in coal (Guo et al., 2020b).

## 6 Feasibility and urgency of the field experiment

Almost all types of methanogens can produce methane through carbon dioxide reduction. The reduction environment of underground coal seams provides conditions for *in situ* microbial anaerobic metabolism (Guo et al., 2019b). The injection of carbon dioxide can accelerate the process of hydrolysis and fermentation, activate methanogens, and even change the microbial community structure and methanogenic pathway. A high concentration of carbon dioxide is conducive to the metabolism of hydrogenotrophic methanogens and inhibits the growth of other kinds of methanogens. The promotion or inhibition of different microorganisms is the root cause of the differences in methane production methods and rates (Guo et al., 2015, 2021a). Laboratory conditions cannot wholly restore the on-site reservoir

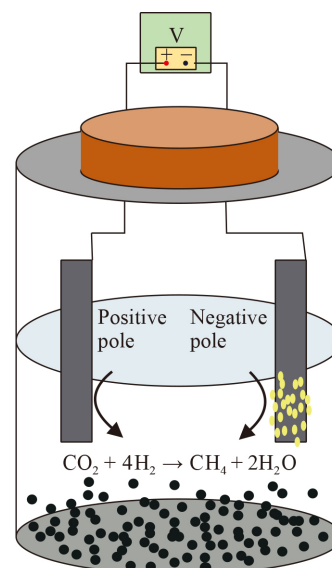
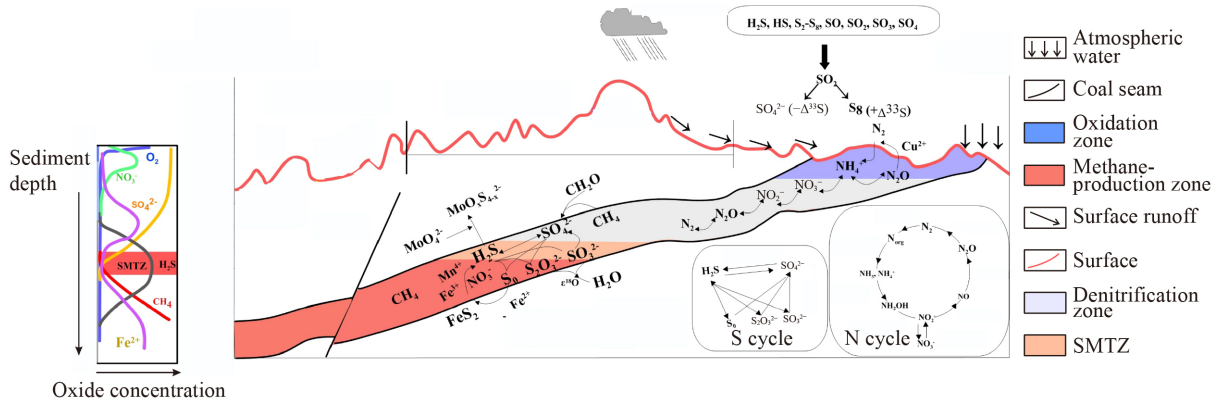


Fig. 3 Schematic diagram of a microbial electrochemical battery reactor.



**Fig. 4** Schematic diagram of biogeochemical cycle mechanism at block scale.

environment, and coalbed methane reservoirs in different places have different underground physical and chemical environments, microbial community types, and metabolic pathways. The field operation environment is relatively complex, but the transformation of the *in situ* reservoir environment can be partially realized (Li et al., 2018). According to the response law of underground physical and chemical environment and microbial community, optimizing injection and production methods and engineering measures can improve the biotransformation efficiency of coal in non-minable coal seams or abandoned mines.

The cultured or domesticated microflora in the laboratory will change significantly, so it cannot represent the type of methanogenic microflora *in situ*. In addition, the synthetic nutrient solution used in the laboratory can control its composition. However, the difference and unknown composition of formation water lead to similar nutrient formulas used in different reservoirs, which may lead to the wrong evaluation of coal biotransformation potential (Davis and Gerlach, 2018; Li et al., 2018, 2020b). Due to the limited availability of substrates under laboratory conditions, the accumulation of metabolic by-products, and inaccurate simulation of under environment and dynamic water flow for microbial communities, the research on industrial production increases *in situ* coal seam biotransformation is limited (Nie et al., 2019). To understand the effectiveness of these strategies in laboratory conditions, it is necessary to improve the laboratory simulation environment or transfer the experiment to the site to promote the biotransformation of coal for industrial production as soon as possible (Giang et al., 2018; Guo et al., 2019c). Taking carbon dioxide as the carbon source of biotransformation provides a new idea for carbon dioxide emission reduction and reuse.

## 7 Conclusions

At present, gasification experiments of coal samples in different reservoirs and coal rank continue to break

through in laboratory conditions. However, the technology of increasing coalbed methane production by microbial action still lacks experimental study and theoretical support for on-site industrialization. Some significant problems need to be solved urgently.

1) The physical and chemical properties of *in situ* reservoirs vary *in situ* environment, and the main factors affecting the coal gasification process may vary. Therefore, it is necessary to strengthen the study on the main controlling factors affecting coal gasification in different reservoirs and the effectiveness and feasibility of various pretreatment methods.

2) Although significant progress has been made in laboratory research, *in situ* reservoir research is still in its infancy. *In situ* reservoir conditions are much more complex than expected, and more control factors remain unknown (including reservoir pressure, coal structure, and fluid flow rate). Therefore, it is necessary to carry out systematic field experiments in time.

3) The general idea of the research is to focus on finding low-cost and high-efficiency engineering and technical means to achieve the long-term stable production increase of the target coal seam. Therefore, the *in situ* microorganisms of the target coal seam are domesticated to improve its environmental adaptability.

4) The systematic study of the microbial structure, metabolic pathways, and synergistic relationship in different blocks is incomplete. It is vital to reveal the functional potential and regional distribution of microbial communities on the block scale. Therefore, it should be gradually clarified that the metabolic mechanism of *in situ* microorganisms and the biogeochemical cycle mechanism on the block scale (Fig. 4).

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