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Microbial Co-processing and Beneficiation of Low-rank Coals for Clean Fuel Production: A Review

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Abstract

Coal is a non-renewable fossil fuel combusted to generate power or used to produce liquid or gas fuels with high energy densities. The abundance of coal reserves and the dependence of many countries on coal as a cheap energy resource are the basis for the predicted expansion in coal consumption in the future. However, at the global level, there is a growing concern about the role of coal combustion in environmental pollution, global warming, and climate change. Consequently, it becomes imperative to consider and adopt novel technologies aimed at bridging the gap between demand and supply. These technologies must meet core eligibility criteria and demonstrate substantial benefits, including reduced adverse environmental impacts, improved utilization safety, and decreased gas emissions. This review aims at addressing these issues by means of microbial activities and/or microbial biomass. Firstly, combining low-rank coal and microbial biomass would allow for safe and effective coal utilization through different co-processing technologies to make valuable fuel products. Secondly, microorganisms associated with sulfur removal and dust suppression activities may facilitate "clean" coal production, promoting environmental impact reduction. Furthermore, biogenic coal-to-methane conversion by methanogenic microorganisms is an effective approach to generating high-quality gas. The literature reviewed here demonstrates that these technologies hold great potential for sustainably coping with ever-growing energy demand.

Keywords: Low-rank coal; Coal beneficiation; Microorganisms; Clean fuel; Coal co-processing; Biogenic coalbed methane; Desulfurization; Coal dust suppression.

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1. Introduction

Coal is an abundant and widely distributed energy resource, with relevant reserves in around 70 countries. Due to the increasing global demand for energy, coal continues to be broadly used as a major energy feedstock for electric power generation and other industrial purposes.[1] According to the study by A. Stephen *et al.*, coal provides ~26% of the energy produced worldwide; however, coal consumption presumably emits ~43% of global anthropogenic CO_2.[2] Low-rank coals (LRC), including lignite (brown coal) and some sub-bituminous coals, are primarily used as a fuel source in electricity generation; however, their high moisture content, lower energy density, and risk of spontaneous combustion make them a less preferable fuel worldwide as compared to high-rank coals.[3]

From a historical and economic point of view, coal was a revolutionary energy source. In contrast, from an environmental standpoint, ambient air pollution caused by coal usage represents a severe threat. The major environmental impacts during different stages of coal utilization can be listed as follows.[4-6]

1. Coal mining causes landscape deterioration, soil erosion, water pollution, and local biodiversity disturbance.
2. The formation of post-mining areas and acid mine drainage represent a long-term environmental problem.
3. Coal combustion releases numerous harmful/toxic pollutants, including SO_x and NO_x, particulates, and heavy metals.
4. Coal combustion contributes considerably to global

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greenhouse gas (N_2O and CO_2) emissions.

5. Solid waste from coal combustion (fly ash) contains harmful pollutants that cause significant disposal problems. Moreover, coal mining and exploitation have accumulated billions of tons of coal residues (coal fines, dust, gangue, *etc*.) worldwide, generated as by-products.[7] These components of low quality are considered unusable and uneconomical due to their poor thermal performance and unpredictable chemical composition.

To meet the abovementioned challenges related to the utilization of low-rank and low-quality coals and to reduce environmental pollution in spite of higher energy demand, clean coal technologies could play a key role across the globe. Particularly, *microbiological co-processing, conversion, and cleaning of coals* are paving the way for sustainable and efficient LRC utilization.

Microbial-based clean coal technologies are required to achieve much greener mitigation solutions to meet environmental challenges that arise during different stages of coal utilization, particularly:

1. Coal mining activities often cause environmental losses on an ongoing basis, which can be alleviated by reduced carbon emissions by microalgae cultivation, polluted water treatment by microbial contact, increased soil fertility through carbon sequestration, and coal-to-methane conversion by microbial consortia. Furthermore, coal biodegradation by selected microorganisms is being intensively investigated to develop effective bioremediation and rehabilitation strategies for coal mining areas and waste dumps.[8,9]
2. Persistent and long-term effects of acid mine drainage from coal mining/processing can be mitigated by phycoremediation, bioleaching, and bioremediation, due to their self-renewing capacity, low cost, and sustainable nature.[10]
3. Application of multi-pollutant-resistant microalgae in the biofixation of CO_2 from coal combustion appears to be promising for decreasing CO_2, SO_X, and NO_X emissions.[11,12]
4. Microbial technologies, which are outlined below, exhibit improved coal co-combustion behavior and a low-carbon emission strategy, thus contributing to greenhouse gas reduction.
5. Coal utilization residues and wastes may serve as alternative sources for soil fertility management due to their wide range of macro- and microelement compositions and valuable organic matter content.[13] In addition, bioleaching and biooxidation are potential approaches to removing heavy metals and recovering value-added products from coal waste.[14]

The benefits of microbial biomass over fossil fuels to produce energy are mostly focused on its carbon neutrality and renewable or clean energy features. The use of biomass in thermo-processing has shown encouraging results and is being widely researched for the manufacture of biofuels.[15-17] In this context, microalgae, as a valuable biomass feedstock, offers a range of interesting technological options for their use, especially for *coal-microalgae co-processing*. This process currently gains scientific recognition as a low-risk, near-term, low-cost strategy leading to a significant reduction of net gas emissions. Microalgae, unicellular/multicellular photosynthetic microorganisms, grow naturally in abundance in aquatic environments and even better under controlled conditions. Hannon *et al*. have demonstrated that the simple nutritional requirements of microalgae allow efficient biomass production in different (artificial) ecological niches.[18] In addition, existing abundant activated sludge from biological wastewater treatment units would easily provide considerable microbial biomass for use in innovative coal co-processing systems.[19]

A promising row of LRC-processing technologies is based on the biochemical capability of microbial communities (hydrolytic and fermentative bacteria, and methanogenic archaea) *to convert coal into methane* rather than on the direct combustion of coal. This leads to a reduction in greenhouse gas (GHG) emissions, as pointed out by Burnham *et al*..[20] As key properties for biogenic coal bed methane (CBM) in GHG reduction, methane generation under controlled conditions (either in the existing coal beds or in split process systems), enrichment or de novo design of microbial communities catalyzing methane production, and CO_2 storage in coal seams have attracted significant interest. It is important to note that coals of lower rank are more bioavailable and can produce more methane than coals of higher rank. As coal increases in rank (peat > lignite > sub-bituminous > bituminous > anthracite), heteroatoms such as O, S, and N are lost. At the same time, the aromatic lignin-derived structure of the coal condenses to form polyaromatic compounds and aromatic sheets. The increase in aromaticity is accompanied by the loss of heteroatom moieties amenable to microbial attack.[21,22] This fact, in turn, provides a basis and inspiration for yet another coal beneficiation technology, *microbial coal desulfurization.* This process can be described as microbiological sulfur- and ash removal from various coals and is exploited as either an alternative or a complementary process in clean coal technologies.[23] Furthermore, biomineralization processes are currently receiving extensive attention, as they provide a sustainable and low-environmental-impact approach to coal dust suppression.[24]

In this review paper, we tried to analyze and overview the concepts and state-of-the-art microbial technologies proposed/utilized for coal beneficiation, especially those using co-processing (co-firing, co-pyrolysis, co-gasification, co-combustion, and co-liquefaction) and functional transformation (methane production, biodesulfurization, and coal dust suppression). To our knowledge, this is the first review to address the microbiological aspects of utilizing low-rank coal to produce clean coal fuels.

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2. Coal co-processing

As described above, coal utilization poses numerous severe environmental challenges, including land subsidence, damage to the aquatic environment, mining waste disposal, and air pollution. Therefore, introducing appropriate technology and adopting proper management practices are crucial measures to reduce the levels of coal waste discharged into the environment.

As a possible way to achieve this goal, microbial biomass can be used as a mediator and as an intermediate product in fulfilling energy needs. Compared to non-renewable energy sources such as coal, biomass has a higher moisture content, a lower ash melting temperature, a higher oxygen content, and a lower calorific value. However, combining coal and plant-based biomass is a promising solution to preserve high energy content. This can be realized through various technologies of coal co-processing, such as *co-firing*, *co-pyrolysis*, *co-gasification*, *co-combustion*, and *co-liquefaction* (Fig. 1).

Current international expertise ranks coal co-processing among the best options due to its low operational risks, low costs, high efficacy, and quick beneficial outcome. However, it is also important to notice that LRCs still contain exploitable carbon, *i.e.*, their effective recovery can reduce the environmental burden by generating revenue from coal utilization to yield various fuels and value-added products. Microalgae, as a source of biomass, can be easily employed to treat LRC before or while processing to increase energetic efficiency and reduce environmental impacts. Microalgae possess several advantages over other renewable energy sources, including a fast growth rate, active photosynthesis, shorter rotation, CO_2 fixation efficiency, and no competition with arable land.[25] Estimations made by Li, Y. *et al*. show that microalgae can produce 50 times more biomass than higher plants.[26] Chemically, microalgae biomass is much more complex than coal as it comprises carbohydrates, proteins, and lipids, which are less thermal resistant but have high plasticity properties.[27] Furthermore, combining coal and microalgae can increase energy density and reduce fluctuations in biomass characteristics and supply. The reason why microalgae can improve the reactivity of coal during thermal processes is that biomass contains a significant amount of hydrogen and other species with catalytic activity.

2.1 Coal co-firing

Coal residues, fines, and waste have been considered uneconomic, environmentally hazardous, and difficult to process. However, with proper processing, they could become an essential energy source with improved efficiency. Several studies have recently demonstrated that co-firing different biomasses with coal in power plants has produced high-quality

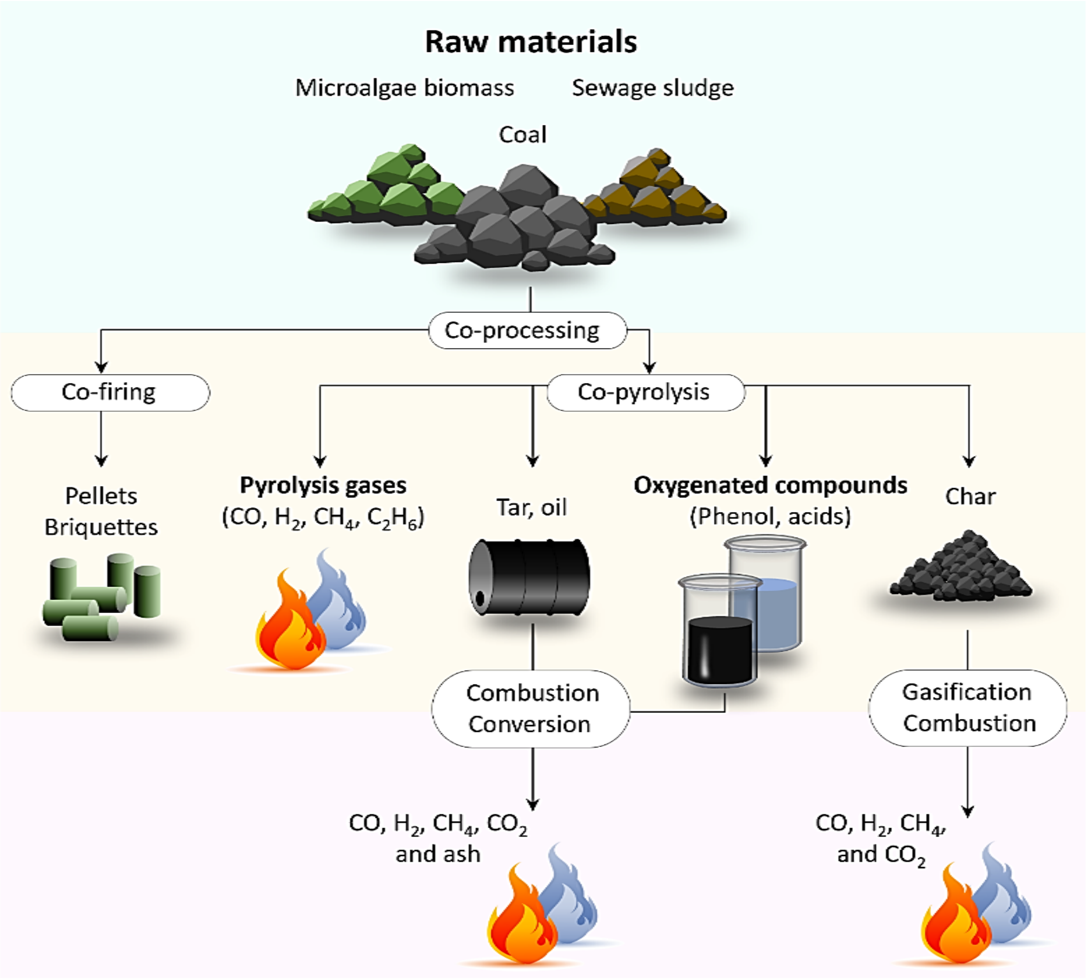

Fig. 1 Co-processing of coal and microbial biomass to produce different fuels.

pellet and briquette fuels.[28-30] Biomass fuels appear in diverse nature, such as organic waste, herbaceous, wood, and microalgae. The ISO 17225 norm establishes a set of specifications for using solid biofuels (various biomasses, along with their combinations or mixtures), broadening the bio-based wastes suitable for commercialization.[31]

Furthermore, Section 6 of this norm promotes the application of non-woody biomass for energy purposes in industries, residential, commercial, and public buildings. Therefore, research on high-quality fuels obtained from microalgae biomass and coal residues is getting more and more advisable.

Currently, especially biodiesel and biogas have become well-studied microalgal-derived fuels, with a wide range of pilot plant facilities in operation to investigate process feasibility and assess novel low-energy and low-cost technologies. Studies by Giostri, A. *et al*., and Magida *et al*. indicated that the combustion behaviors of coal improved when microalgae were used as a supplementary fuel.[11,32] Due to the substantial difference in the combustion characteristics of coal and microalgae biomass, the partial substitution of coal (up to 20%) is reasonable to reduce the degree of performance incompatibility to an acceptable level.[33] This offers several direct benefits, including (1) reduced emissions of NO_x, SO_x, and GHG gases because of the low sulfur, nitrogen, and lean carbon nature of microalgal biomass compared with coal; (2) improved energy conversion efficiency and, therefore, better economics of biomass utilization; (3) straight-forward improvement in the efficiency of power plants with minimal technical risks; and (4) utilization of broad ranges of coals, such as LRC and coal residues.

Microalgae *Chlorella* sp.[34] *Scenedesmus* sp.[11,35], *Tetraselmis* sp.[32], and *Botryococcus* sp.[35] have been successfully employed for co-firing studies. Most strains tested in combustion studies have been reported to have higher volatile matter, a higher gross calorific value, and a lower ash content.[36-38]. Moreover, highly volatile biomass may provide a stable flame, thus further improving combustion, performance, and emission characteristics. In a recent study by Hossain *et al*., the microalgae blended with coal elevated the volatile matter and therefore allowed to reduce the ash content.[39] Kadam K.L. first used power-plant flue gas as a source of CO_2 for microalgae cultivation and then co-fired the obtained biomass with coal for electricity generation.[40] Other researchers have repeatedly highlighted the valuable role of microalgae usage in boosting coal combustion efficiency.[11,12] The main reasons why microalgae biomass beneficially contributes to many life-cycle energy- and cost-saving co-firing technologies can be listed as follows:

1. The co-firing approach readily replaces a portion of the non-renewable fuel – coal – with a renewable fuel – microalgae biomass.
2. Overall production cost savings can be easily achieved by combining coal with inexpensive biomass fuel sources.
3. Domestic coal could be partially substituted with renewable fuel in a flexible way, thus contributing to better energy security.

2.2 Coal co-pyrolysis

Lower ash and higher volatile matter content make LRC more advantageous over many other coals; however, the lower hydrogen-to-carbon ratio typical for LRC hampers its use in thermochemical conversion technologies.[41] Therefore, *co-pyrolysis* of coal blended with various biomasses is seen as a practical solution to allow large-scale utilization of LRC and biomass and mitigate greenhouse emissions.[42,43] Moreover, a higher hydrogen-to-carbon ratio in biomass is essential for volatile end-product formation during the co-pyrolysis process.[44]

In general, the thermochemical conversion of coal and biomass to valuable products can occur via different *co-*options: co-pyrolysis, co-gasification, co-liquefaction, co-combustion, and co-carbonization. Co-pyrolysis is commonly accepted as the starting point of all the technologies mentioned above due to its involvement in all chemical reactions to form solid, liquid, and gaseous products with zero oxygen concentration.[45] Thermochemical conversion targets both gaseous or liquid intermediates and their processing into more valuable forms of energy (such as hydrogen, methane, hydrocarbons, char, *etc*.).

Much research was focused on the co-pyrolysis behavior of coal and terrestrial biomass of various origins, including organic waste, woody, and herbaceous biomass.[46,47] Yet, microalgae biomass possesses here many advantages over the "terrestrial" one. The main organic constituents of microalgae (protein, lipid, and starch) can be readily pyrolyzed under more moderate conditions to produce high-value products.[41,48] Furthermore, microalgae contain less lignin, which makes them more suitable for thermochemical conversion. During co-pyrolysis, microalgae generate many alkyl radicals, and the gain or loss of hydrogen radicals from alkyl radicals yields a normal alkane or α-olefin, respectively. Thus, the pyrolytic oil made of microalgae has a higher hydrocarbon content and a lower oxygen content, promoting tar's yield and quality, as shown by Wu *et al*.[41]

Two such different materials as coal and microalgae biomass, when decomposed simultaneously, give rise to a much more complex chemical scenario where the reactions attributed to the individual materials affect one another. Quan and Gao have proposed three main mechanisms for such interactions that involve synergetic effects: (1) the transferring of active H radicals from biomass to coal, (2) the catalytic action of alkali and alkaline earth metallic species from the biomass, and (3) the mutual heat transfer during co-pyrolysis.[49] Many other research groups studied the synergy characteristics and thermal behavior during coal and microalgae biomass co-pyrolysis (Table 1). These studies provide valuable information for better performance management and process optimization.

Table 1. Reported synergistic effects of the co-pyrolysis of coal and microalgae biomass.

Microalgae	Coal	Analysis	Setup, conditions	Results and observation	Year, Ref.
Blue-green algae mixtures	Bituminous and anthracite	Nitrogen conversions and analytical methods	High-frequency furnace, rapid pyrolysis of the fuel samples at 600–1200 °C	Interactions between biomass and coal decreased char-N yields and increased volatile-N yields	2011[50]
Chlorella vulgaris	Semi-anthracite coal	TGA, differential thermogravimetry	Thermogravimetry analyzer, from ambient to 1000 °C under a nitrogen flow rate of 100 ml min $^{-1}$	Co-pyrolysis presented three stages; interaction between solid phases inhibited the thermal decomposition	2012[51]
Chlorococcum humicola	Victorian brown coal	TGA and modeling	Thermogravimetric analyzer, the final temperature of 1000°C at different heating rates	No chemical interaction between the algae and coal during pyrolysis was indicated	2013[52]
Spirulina	Shenfu coal	Isothermal TGA	Thermogravimetry analyzer, from ambient temperature to 1000 °C	The synergy effect was significant, and the char yield was lower than the theoretical value	2013[53]
Dunaliella tertiolecta	LRC	TGA	Thermogravimetric analyzer, within 200°C-500°C in nitrogen atmosphere	Due to the high content of fixed carbon and ash, the weight loss rate of coal was lower than biomass	2013[54]
Spirulina and model compounds of *Spirulina*	Shenfu bituminous coal	TGA, SEM with energy dispersive spectroscopy	Thermogravimetric analyzer, from 25 °C to 850 °C under a nitrogen flow of 60 ml min^{-1}	Synergistic effects occurred in different forms from co-pyrolysis. The higher volatile yield was found in medium-chain triglyceride and coal mixtures	2017[41]
Nannochloropsis sp.	Colombian bituminous coal	TGA, kinetic models, model-free methods	Thermogravimetric analyzer, non-isothermal conditions at heating rates <80 °C/min up to 900 °C	There were synergistic effects during the co-pyrolysis process, especially for that mixture containing 50 wt% of microalgae	2018[55]
A typical microalga	Shenfu LRC	MS, SEM	A fixed bed reactor, under higher purified argon, from 650 to 850 °C every 50 °C	Biomass promoted the generation of volatiles and liquid products	2018[56]
Nannochloropsis and *Chlorella*	ShenMu LRC	TGA and iso-conversional	Thermogravimetric analyzer, from 250 °C to 850 °C, under nitrogen flow rate of 60 mL min^{-1}	Both microalgae promoted the release of volatiles from coal	2018[57]
Glycine from microalgae	Acid washed LRC	TGA combined with an online MS	Thermogravimetric analyzer, from ambient temperature to 850 °C under 60 ml·min^{-1} of high-purity nitrogen	Both positive and negative synergistic effects from the product distribution were observed. The intensity of H_2 reached a peak value at 700 °C	2018[58]
A typical microalga	Shenfu LRC	Iso-conversional	A fixed-bed furnace, from ambient temperature to 650 °C, 700 °C, 750 °C, 800 °C and 850 °C	Synergistic effects were observed, the biomass decreased the activation energy	2019[59]
Scenedesmus sp.	South African bituminous coal	TGA, thermo gravimetric mass-loss, model-free methods	Thermogravimetric analyzer, the final temperature of 800 °C under inert conditions	Co-pyrolysis had three stages whose kinetics were dominated by the pyrolysis of the individual materials	2020[60]

TGA - thermogravimetric analysis, MS - mass spectrometry, SEM - scanning electron microscopy

As visible from the table, the majority of research so far has focused on kinetic analysis, reaction mechanisms, and product distribution. For the characterization of the thermal behavior of materials, especially popular is the thermogravimetric analysis (TGA).

In good agreement with the Quan and Gao concept, many of the studies listed in the table have evidenced the existence of a synergistic effect in the process of co-pyrolysis of LRC and microalgae biomass. Under certain circumstances, the synergistic effects considerably promote the production of CO_2, CO, CH_4, and H_2.[56,59] The physicochemical characteristics of the co-pyrolysis products, such as char and tar, are essential for a better understanding of the pyrolysis mechanism. The structure of the obtained char, in turn, has a critical influence on its performance: for instance, as fuel for gasification or combustion, or as a functional material with catalytic or adsorbent properties.[49,61]

2.3 Coal co-gasification

Conversion of coal and coal char through gasification and combustion has been practiced for centuries and remains a viable technology. Gasification generates gaseous fuel-rich products (syngas) via the partial oxidation of coal, which is then fired in gas turbines or engines. The principal reactants are coal, steam, O_2, CO_2, and H_2, while the desired products are usually CO, H_2, and CH_4. Compared to combustion, gasification produces fewer greenhouse gases and is considered to have a higher overall energy efficiency (~40%).[62,63] In addition, Xiao, Y., *et al.* pointed out that the moisture content of LRC can facilitate the gasification process.[64] On the other hand, gasification of biomass is a promising conversion technology due to its ability to convert biomass into syngas, which can be used as fuel gas, for power generation, or as feedstock for chemical industries.[65] Recently, the co-gasification of LRC and microbial biomass has been gaining attention for its potential to adjust the feed properties and achieve superior gasification performance.[66] Co-gasification can be divided into two main stages: the initial co-pyrolysis process and the co-gasification reaction of blended char. Co-gasification technology can overcome obstacles that appear when individual components are gasified separately and create many additional advantageous effects, including (1) coal and biomass co-processing causes synergetic interactions leading to better variability in the thermal reactivity of the fuels; (2) coal gasification efficiency can be enhanced through the catalytic effect that comes from biomass; (3) utilization of biomass together with coal allows to overcome the limited availability of biomass due to its seasonal nature; (4) The elevated co-gasification temperature assists in the reduction of tar formation from biomass, enhancing the energy outcome of coal gasification.[67]

Adnan and Hossain used an equilibrium model to investigate the performance (syngas composition, gasification system efficiency, and cold gas efficiency) of an integrated co-gasification of *Nannochloropsis oculata* and Indonesian coal. They found that the reforming and CO_2 absorption enabled the process to provide high-purity syngas in this manner. Furthermore, increasing the coal and microalgae biomass ratio enhanced gasification system efficiency while decreasing cold gas efficiency.[66] Fermoso *et al*. specifically targeted the feasibility of co-processing *Nannochloropsis* sp. with Colombian bituminous coal, both through co-pyrolysis and co-gasification. During the co-pyrolysis tests, they found some synergistic effects at different temperatures, enhancing the devolatilization profile of the mixture components. In contrast, in the co-gasification experiments, both synergistic and inhibiting effects were observed. The synergistic effect at above 800 °C was due to the high Na content of microalgae that promoted coal char gasification.[55]

Shenfu coal slurries prepared with algae were thoroughly studied by Li and Liu, especially in terms of their gasification reactivity, which was compared with that of coal-water slurries. It was revealed that algae could promote coal gasification rate.[68] In a study conducted by Qadi *et al*. using a thermogravimetric analyzer in the temperature range of 800–1000 °C, strong synergetic effects were observed for all ratios of Newlands coal char and *Spirulina* algae char, while the 5:5 blend demonstrated the best co-gasification performance, which may be attributed to the high content of potassium in the algae char, which in turn promotes the catalytic effect.[67] Alghurabie *et al*. studied the fluidized bed gasification of LRC from South Australia with marine microalgae *Tetraselmis* sp. and identified the optimal operating conditions as 1.82 for the air-to-fuel ratio, 0.75 for the steam-to-fuel ratio, and 850 °C for the bed temperature. These conditions resulted in the production of gases with the highest heating value (per mass of fuel fed), the greatest extent of carbon conversion, and the best H_2:CO ratio.[69]

An interesting innovative method to improve the economy of biofuel consists of combining coal-microalgae co-gasification with the chemical looping process. In the framework of this approach, Shen *et al*. studied the co-gasification characteristics of cyanobacteria and Xiaolongtan coal and obtained the highest syngas yield of 1.26 Nm^3/kg in the mixture of 46 wt.% cyanobacteria and 54 wt.% coal under a 0.3 oxygen carrier-to-fuel ratio. Meanwhile, blending cyanobacteria biomass also elevated the coal chemical looping gasification performance measured as the syngas quality, gasification rate, and carbon conversion efficiency.[70]

Due to the variations in microalgae biomass properties, it is still inconclusive whether the synergistic effect certainly leads to better gasification performance, and a high volatile yield. For instance, Zhu *et al.* investigated the co-gasification of Australian brown coal with macroalgae *Derbesia tenuissima* and *Oedogonium* sp. in a fluidized bed reactor and found that more syngas was produced. However, when microalgae *Scenedesmus* sp. was used in the same set-up, the carbon conversion rate decreased, resulting in a lower $CO/CO_2/H_2$ yield than for coal alone. The authors speculated that the high ash content of the microalgae reduced the amount of active sites and lowered the availability of char-to-gas contact.[71]

2.4 Coal co-combustion

Co-combustion of coal and biomass is a novel and progressive technology for power generation. In many cases, "co-firing" and "co-combustion" are used interchangeably; however, co-combustion here refers to combusting coal with biomass using the existing coal-fired boiler systems, while co-firing is related to the combusting of available coal-microalgae pellets/briquettes. Coal co-combustion results in reduced net CO_2, SO_x, and often NO_x emissions and solves the problem of the low efficiency of biomass-firing power plants. Coal co-combustion has a special importance in developing countries as it represents a low-cost, sustainable, and renewable energy option.[72] Due to differences in the burning properties of coal and biomass, their co-combustion characteristics may differ from those obtained for the individual components.[73] Coimbra *et al.* in TGA-experiments showed remarkable differences between the combustion processes of *Chlorella sorokiniana* biomass and León bituminous coal. However, the weight loss and heat release of the used blend were found to be very close to those of coal. The apparent activation energies corresponding to the combustion of the blend and coal showed strong resemblance as well.[74]

Oxy-fuel combustion (combustion in a nitrogen-free atmosphere) is considered an effective technique to facilitate CO_2 capture and storage. In this case, the mixture of O_2 and recycled flue gas is used as an oxidizer. Ye *et al*. compared the co-combustion characteristics of Hengshan bituminous coal, *Nannochloris oculata* microalgae, and their blends under air and oxy-fuel atmospheres. They found that (1) their combustion processes exhibited two stages, while individual coal showed one stage-behavior; (2) with the increasing blending ratio of biomass, flames of volatiles and char became dimmer and smaller, and the average flame temperature decreased, (3) the synthetic effect between coal and microalgae exists, (4) as the N_2 in the combustion atmosphere was replaced by CO_2, the average flame temperature of volatiles and char decreased, and the ignition delay time of fuel increased.[75] Similarly, Tahmasebi *et al*. reported that the weight loss rate curve of *Tetraselmis suecica* microalgae had two peaks while that of coal possessed only one peak, and the increasing microalgae and coal ratio in the blends increased the activation energy of fuel.[76]

2.5 Coal co-liquefaction

Direct coal liquefaction is an ultra-clean coal conversion technology in which LRC is broken down and hydrogenated to produce transportation fuels and aromatic chemicals under high temperatures and hydrogen pressure.[77] However, this process is not considered very competitive due to the high cost of the hydrogen donor solvent and high carbon emissions.[78] More economically attractive options can be developed using by-product hydrogen and hydrogen-rich materials, such as biomass, as hydrogen donors to reduce the high hydrogen cost and cut carbon emissions during direct coal liquefaction.[79] Despite several publications available on the co-liquefaction of LRC and lignocellulose biomasses, only a few studies have been published on coal co-liquefaction with microalgae. Ikenaga *et al*. conducted co-liquefaction using microalga (*Chlorella* and *Spirulina*) and two coal types (Australian Yallourn brown and Illinois No. 6). According to their results, in the reaction with a one-to-one mixture of *Chlorella* and Yallourn coal, 99.8% of conversion and 65.5% of the hexane-soluble fraction were obtained at 400 °C with $Fe(CO)_5$ at excess sulfur to iron (S/Fe = 4). When Littorale and *Spirulina* were used, a similar tendency was observed with the iron catalyst. On the other hand, in the co-liquefaction with Illinois No. 6 coal, which contains catalytically active pyrite, the oil yield was close to the additivity of the respective reaction with $Fe(CO)_5$-S, even at S/Fe = 2.[80]

Bio-liquefaction is another essential aspect of coal liquefaction, in which microscopic fungi convert solid coal to liquid fuels or value-added products at ambient conditions. LRC is an ideal source for bio-liquefaction due to its low aromaticity and high oxygen content compared to high-rank coals. In addition, the condensed polyaromatic structure of LRC is quite similar to lignin, making it more attractive to lignin-degrading fungi.[81] The analytical studies of coal liquefaction products were conducted by Laborda *et al*. using different Spanish coals solubilized by filamentous fungi (*Aspergillus* S10H and *Doratomyces* S3X strains). Their data indicated that the proportion of humic acids extracted from the liquid lignite reached 80%, while for individual lignite, it was only 35%. The remainder 20% were fulvic acids, humin, and alkali-insoluble materials.[82] Kai-yi *et al*. studied the bio-liquefaction of lignite by a fungus called AH from Chinese mine environment. They showed that the main components of bio-liquefied lignite were phenol derivatives, ketones, and aldehydes (UV-VIS). Furthermore, GC-MS analysis revealed 16 high-concentration compounds, of which 11 belonged to aromatic acids or ethers.[83] In contrast, according to Oboirien *et al*., the soluble phenolic compounds were not a major product of the coal bio-liquefaction process when *Trichoderma atroviride* was used.[84] Many other fungal strains with LRC bio-liquefaction ability were found and investigated in other studies. They include *Penicillium* sp.,[85] *Phanerochaete chrysosporium*,[86,87] *Fusarium oxysporum*,[88,89] *Neosartorya fischeri*,[90] *Trichoderma atroviride*,[91,92] *Aspergillus fumigatus* and *Aspergillus oryzae*,[81] *Ganoderma applanatum*, *Pycnoporus cinnabarinus*, *Perenniporia tephropora*, *Pleurotus ostreatus*, *Rigidoporus ulmarius*, and *Xylaria hypoxylon*.[93]

3. Sewage sludge for coal co-processing

The generation of sewage sludge from wastewater treatment plants has been steadily increasing worldwide as a result of rapid urbanization and industrialization. Sewage sludge has a wide variety of microorganisms that may positively impact soil quality if used as fertilizer or energy quality if used as fuel.[94,95] Many factors modulate microbial community structure and composition, which may quickly change from autotrophic to heterotrophic microorganisms depending on sewage origin, treatment conditions, and industrial activity. Overall, *Proteobacteria* is typically the dominant phylum of such habitats, followed by *Bacteroidetes* and *Firmicute*.[96] The existence of different hazards, such as heavy metals, pathogens, and helminth eggs, in sewage sludge poses a great threat to humans and the environment. Thus, adopting energy-efficient and environmentally friendly solutions is imperative to treat and re-utilize sewage sludge in a sustainable way. Unfortunately, recycling in agriculture, dumping into the sea, landfilling, and incineration are still broadly used as sludge disposal methods. However, environmental concerns and high energy consumption limit the future prospects of these technologies.[97] Under adequate control, co-processing sewage sludge with coal may be a secure outlet and generate profits through energy.[98] The dewatering of sewage sludge plays a significant role in minimizing its volume, easing its transportation, and increasing its calorific value. The existing technologies for sludge dewatering prior to coal co-processing include physical (thermal treatment, thawing, sonication, and porous material addition) and chemical (coagulation/flocculation, acid/base treatment) conditioning. However, most of these dewatering technologies are energy-intensive, thereby rendering coal co-processing a potentially efficacious approach for enhancing energy recovery in the form of syngas and tar.

At present, co-pyrolysis, co-gasification, and co-combustion of sewage sludge with LRC are considered the most potent methods for sludge management due to their remarkable reduction in pollutant emissions and energy consumption (Table 2).

4. Biogenic coalbed methane production

Coalbed methane (CBM), captured from underground coal mining activities, is receiving increasing attention, mainly from the perspective of unconventional energy resources and GHG reduction.

Table 2. Proposed schemas for co-processing of coal with sewage and oil sludges.

Co-processing	Coal	Sludge	Analysis, setup	Results and observation	Year, Ref
Co-pyrolysis	Asturian bituminous coal	Sewage sludge	TGA	- The sludge was formed by two organic fractions with different reactivity. - Both fractions were more reactive than coal since they decompose and devolatilize at lower temperatures. - Under oxidizing conditions, the action of oxygen was dependent on the conditioning of sludge.	2005[99]
Co-combustion	Chinese coal	Sewage sludge with straw	TGA, differential thermogravimetry	- Oxygen concentration affects the reaction rate. - The thermogravimetry profiles of each sample were almost the same at different heating rates. - Heating rate could affect the maximum weight loss rate.	2009[100]
Co-combustion	Coal-water slurry	Oil sludge	Fluidized bed incineration system	- The process had good operating characteristics. - The slurry flexibly controlled the dense bed temperatures by adjusting its feeding rate. - All emissions met the local environmental requirements.	2009[101]
Co-combustion	Coal residues	Bio-ferment residue	TGA and kinetic	- Bio-residue favored combustion. - Co-firing mainly consists of four stages. - Reaction mechanisms vary with the combustion stage and blending ratio.	2013[102]

Co-processing	Coal	Sludge	Analysis, setup	Results and observation	Year, Ref
Co-combustion	Different-rank Chinese coals	Sewage-sludge-derived hydrochars	Hydro-thermal conversion	- Hydrochars induced greater heat loss for higher-rank coals, whereas its higher portion further improved the ignition reactivity. - The pre-exponential factor value increased with increasing coal/hydrochars ratio, resulting in more intense synergistic effects in blends.	2014[103]
Co-pyrolysis	Coal residues	Bio-ferment residue	TG – FTIR and kinetic	- Co-pyrolysis occurred in three major stages. - The activation energy was lower than that of individual residues. - Gaseous product yield increased with increasing bio-residue blending ratio.	2014[104]
Co-gasification	Coal residues	Bio-fermenting residue	TGA in an inert gas stream, FTIR, MS	- The main components of evolved volatiles were light gaseous compounds. - Co-gasification had little influence on the syngas composition. - The air emissions from co-gasification basically met the limits.	2014[105]
Co-pyrolysis	Inner Mongolia LRC	Sludge derived fuel	TGA at non-isothermal conditions	- Sludge decomposed at a lower temperature than coal. - There was an inhibitive effect between coal and sludge. - Co-pyrolysis had two or three consecutive first-order reactions, which revealed the possible mechanism of co-pyrolysis behaviors.	2015[106]
Co-pyrolysis	Lignite from Neimeng Province	Anaerobically digested sewage sludge	Fixed bed pyrolysis system, GC-TCD	- Beneficial synergetic effect on gas yield was clearly observed. - Sludge acted as a gasification agent and catalyst provider, promoting CO_2-char and H_2O-char gasification.	2015[107]
Co-gasification	Hard coal	Sewage sludge	Fixed bed Gasifier reactor, TGA	- Sufficient thermal energy was generated in systems with the external heating of the reactor. - The highest hydrogen contents in gas were reported in coal gasification. - The total hydrogen volume decreased with increasing sludge content in a fuel blend.	2016[108]
Co-pyrolysis	Inner Mongolia lignite	Petrochemical wastewater sludge	TGA, packed-bed reactor coupled with FTIR and GC	- TGA showed that the residual weight of blends was higher than the calculated values. - Synergetic effects promoted the release of gas products and left fewer tars/chars. - Pyrolysis temperatures and blending ratios had significant effects on product yield.	2016[109]

Co-processing	Coal	Sludge	Analysis, setup	Results and observation	Year, Ref
Co-gasification	Chinese lignite	Dried sewage sludge	TGA, differential thermogravimetry	- Producer gases that were obtained from the tar-cracking reactor contained high levels of hydrogen (27.7 vol%) and low tar contents. - Upon gasification of the blends, the hydrogen content decreased, and tar content increased with increasing sludge. - Blends with coal/sludge ratios of 70/30 and 50/50 showed a synergetic effect on tar reduction.	2019[110]
Co-pyrolysis	Three representative-rank Chinese coals	Dewatered sewage sludge	TGA coupled with FTIR and a discrete distributed activation energy model	- LRC and moderate-rank coal blended with sludge exhibited the highest synergistic removal of N and S, respectively. - Co-pyrolysis of sludge with high-rank coal favored C retention and N or S removal. - High-rank coal showed the most remarkable synergistic effect on the light hydrocarbon yield.	2020[111]

*TGA - thermogravimetric analysis, FTIR – Fourier transform infrared spectrometry, GC - gas chromatography, GC-TCD – gas chromatography with thermal conductivity detector

CBM is mainly composed of methane (CH_4) with minor amounts of nonhydrocarbon gases (carbon dioxide - CO_2, nitrogen - N_2) and heavier hydrocarbons. Because CBM is rich in methane (80-98%) and its heating value (≈37.4 MJ•m^{-3}) is comparable to that of conventional natural gas, CBM can be used for the same purposes as natural gas.[112] Therefore, over the past few years, a massive impetus for exploring CBM has been given in many countries endowed with ample coal reserves.

Some major coal basins worldwide have proven to contain economic quantities of CBM. Currently, total global CBM resources are estimated to be 113-184 trillion cubic meters (Tm^3), of which a total of 42 Tm^3 is recoverable.[113] The largest CBM resources are located in Russia, the USA, China, Canada, Australia, Indonesia, Poland, Germany, and France. The USA is a leader in CBM production, with over 20 years' experience of successful usage of CBM as an energy fuel. Canada, Australia, China, and India have also continuously stepped up CBM production over the last several years.[112]

Biogenic CBM is of particular interest for natural gas producers because it may represent a renewable type of CBM, and microbial gas resources are located at shallow depths that are relatively inexpensive to extract. In addition, the potential for producing new natural gas from coal deposits previously exploited for CBM is of economic and strategic interest, especially with CBM's existing recovery infrastructure already in place.[114] Therefore, studying the biogenic CBM production process/mechanism in each coal type and understanding the ecological characteristics of geomicrobiological activity in coalbeds, as well as the bioconversion process leading to methane production from coal, are important issues.

Coalbed gas bioengineering, including acclimatization, enrichment, and stimulation of microbial communities, has garnered significant global interest due to its potential to enhance safe CBM production, thereby contributing to the reduction of CO_2 emissions.[115] These technologies have come in place to facilitate the attainment of carbon neutrality via coal methanogenesis, microbial CO_2 sequestration, and low-carbon emission reduction. Furthermore, the combustion of methane results in significantly lower emissions of GHG and toxic gases compared to coal combustion, thereby mitigating the potential for severe environmental issues.[116]

Approximately 85% of dry coal is composed of macerals, *i.e.*, organic materials, which reflect the properties of the precursor biological material. The remainder is typically in the form of aluminosilicate and sand pyrites.[117] Generally, coal's chemical and structural characteristics depend on its rank. The organic fraction of coal consists of a complex mixture of aromatic and aliphatic hydrocarbons as well as N-, S-, and O-containing heterocyclic compounds.[118] Due to the heterogeneity, hydrophobicity, and biochemical recalcitrance of partially aromatic and lignin-derived macromolecules, coal degradation requires microbial communities with a wide range of metabolic strategies. Over the past several years, a number of geomicrobiological and phylogenetic studies of subsurface coal beds and shales have described distinct microbial communities with a high capacity to convert coal to

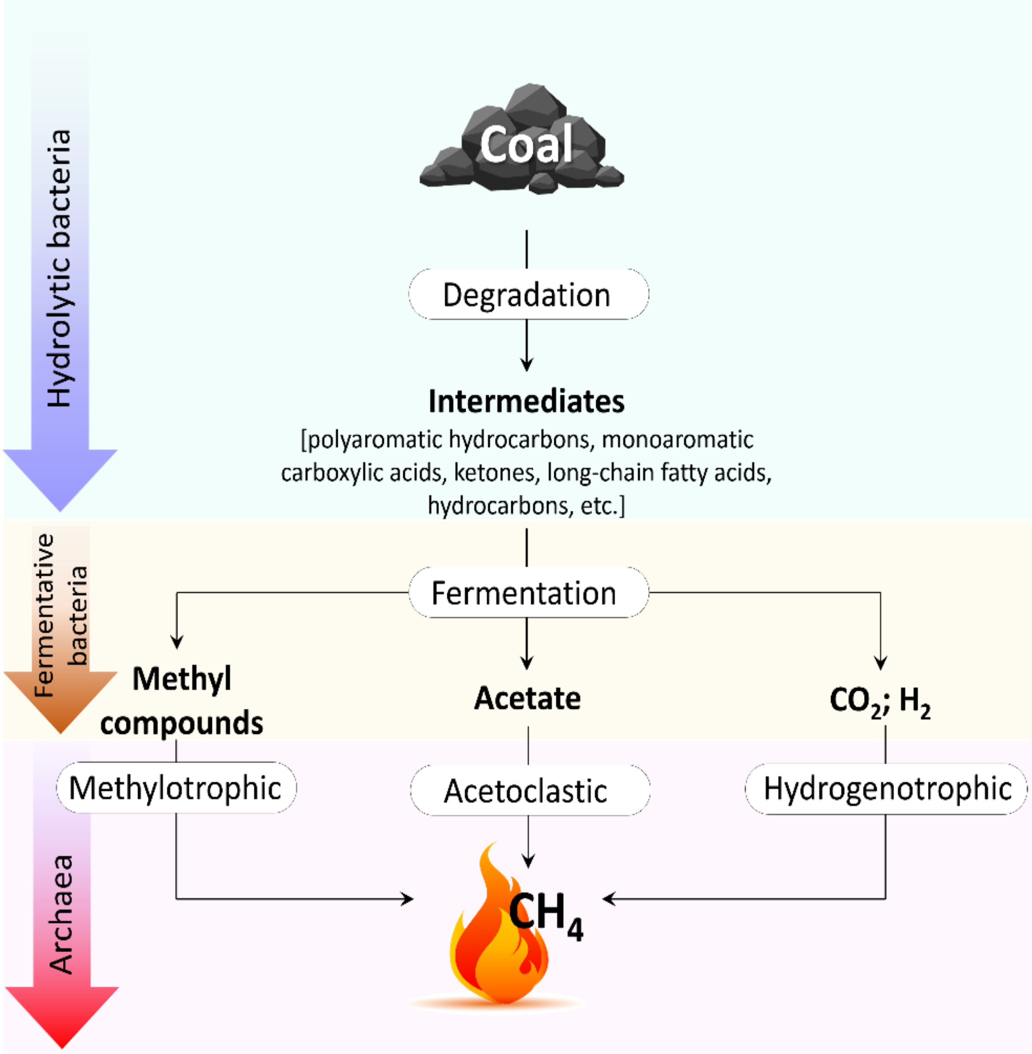

Fig. 2 Proposed pathway for bio-gasifying coal to methane.

methane.[119-123] The process of biogenic coal-to-methane conversion necessarily involves diverse groups of fermentative and acetogenic bacteria syntrophically associated with methanogenic archaea.[22,124,125] (Fig. 2). The bacterial communities, mainly consisting of *Firmicutes*, *Spirochetes*, *Bacteroidetes*, and all subgroups of *Proteobacteria* sequentially break down the complex carbon in coal into intermediate and simple byproducts. Some byproducts of bacterial biodegradation are the substrates required by methanogenic archaea (*Methanobacteriales*, *Methanomicrobiales*, *Methanosarcinales Methanococcales*, and *Methanopyrales*) to produce methane gas. The three primary pathways for archaeal methane production are acetoclastic (Eq. 1), hydrogenotrophic (Eq. 2), and methylotrophic (Eq. 3) reactions:[22]

$$CH_3COOH \rightarrow CH_4 + CO_2 \quad (1)$$

acetoclastic reaction: ΔG = –31 kJ/mol

$$CO_2 + 4H_2 \rightarrow CH_4 + 2H_2O \quad (2)$$

hydrogenotrophic reaction: ΔG = –136 kJ/mol

$$4CH_3OH \rightarrow 3CH_4 + CO2 + 2H_2O \quad (3)$$

methylotrophic reaction: ΔG = –105 kJ/mol

Different coal seams contain various methanogenic communities and therefore display different pathways resulting in methane formation. The requisite methanogenic pathways can vary among coal basins, fields, and wells and, besides that, depend on the physicochemical properties of the given microenvironment.

Unlike thermogenic CBM, which is generated on complex geologic time scales by pressure and heat (mainly at temperatures higher than 100°C), the biogenic CBM process occurs on shorter time scales and under much milder conditions, thus providing many opportunities for novel strategies to increase methane production.[126] Recent studies have indicated that LRC is highly susceptible to microbial transformation due to its organic nature (low aromaticity and low molecular weight).[113,125] In order to enhance methanogenic coal bioconversion, both *in situ* and *ex situ* microbial techniques have been introduced, through *bioaugmentation* of selected microbes and through *biostimulation* of indigenous microbes by adding nutrients.[125,127-131] Coal (pre)treatment to increase its bioavailability for bacterial degradation is yet another proposed strategy for enhancing biogenic CBM production (Table 3).

Table 3. Studies devoted to microbially enhanced CBM generation strategies (within last 10 years).

Strategies	Coal, source	Microbial profile	Maximum CH_4 yield	Results and observation	Year, Ref.
Coal pre-treatment (fungi)	Thar lignites, Pakistan	*Penicillium chrysogenum* isolated from a coal core sample	7 and 11 µmoles per g of coal	Organics released by the fungal treatment could be converted to methane by a mixture of bacteria and methanogens	2013[132]
Coal treatment (potassium permanganate)	Sub-bituminous coal, USA	*Pseudomonas putida* F1-based bioassay that evaluates the coal bioavailability and coal-derived microbial consortia	93.4 µmol CH_4/g coal at day 40	Understanding of the underlying processes involved in the rate-limiting step of coal solubilization and methane generation	2013[133]
Coal pretreatment (dewatering)	Sub-bituminous coal, USA	Coals sampled represented members from aerobic, anaerobic, and facultatively anaerobic	Unsaturated coal: 23-29 µmol methane/g coal, water-saturated: 8-9 µmol methane/g coal	Microbial biomass did not increase in response to dewatering. The dewatered coal had a low residual oxidation potential than water-saturated coals	2013[134]
Biostimulation (coal or acetate/H_2)	CX field, Montana, USA	Methylotrophic and hydrogenotrophic methanogenic archaea and diverse bacterial communities	NA	The *in situ* bacterial communities were more diverse than archaeal. The archaeal populations differed between coal *in situ* and laboratory enrichments	2013[135]
Biostimulation (essential nutrients)	Lignite from Australia, Indonesia, and China	The largest bacterial population was 5.85×10^8 DOI for Indonesian lignite and methanogens of 6.25×10^8 cells/ml	The highest production rate 4.46×10^{-3} mol/(kg·d) (Indonesian lignite)	The Australian lignite also produced methane associated with CO_2 decrease. The Chinese lignite showed a decrease in methanogen counts	2013[21]
Bioaugmentation (mine water)	Jitpur mine coal, India	Methanogens *Methanosarcina mazei* and bacteria *Mircroccus* sp. and *Halomonas* sp.	213.97 µmole per gram of coal, with 92.6% in biogas	The optimum parameters established were pH 7.0-7.5, particle size -60+15, and temperature 35°C	2014[136]
Biostimulation (emulsion nutrition solutions)	Gateway Mine coal, USA	Methanogenic consortia from groundwater	40.32 mL/g day	The supplement could help optimize the microbial balance in the consortia and improve the methane biosynthesis rate	2015[137]
Biostimulation (yeast extract and peptone)	Illinois bituminous coals, USA	The microbial community comprising 185 bacterial and 9 archaea species	828.9 ft^3/ton in 30 days	Decreasing supplement concentrations resulted in decreased methane production	2016[138]
Coal treatment (hydraulic fracturing)	CBM production wells, Australia	The hydraulically fractured well had *Phycisphaerae* class and candidate phylum *Aminicenantes*	NA	Fluids used for hydraulic fracturing affected the native coal bed microbial community structure	2016[139]
Biostimulation (coal loading, coal particle size)	Illinois basin bituminous coal, USA	Formation water microbial community with *Methanocalculus* genus and the *Methanomicrobiales* order	2957.4 ft^3/ton at 32 °C; 201.98 g/L coal loading; <73.99 µm coal particle size	the surface areas increased; substrates were more accessible to microorganisms.	2016[140]

Strategies	Coal, source	Microbial profile	Maximum CH_4 yield	Results and observation	Year, Ref.
Biostimulation (plant-derived carbohydrates)	Sub-bituminous coal, USA	A total of 23,054 sequence reads with 678 OTUs and two archaeal OTUs (*Methanosarcina* and *Methanoregula*)	The highest methane rate was with mannose at 1375 μmol g^{-1} carbon day^{-1}	The microorganisms in the coal anaerobically metabolized monosaccharides to methane to a greater extent than the disaccharide	2017[141]
Bioassay	Three bituminous coals, Pakistan	Mixed methanogenic culture-based bioassay [114]	0.145 – 0.275 μmoles/g of coal	The inhibited susceptibility of higher-rank coal toward biogenic methane generation was observed	2017[142]
Biostimulation (yeast extract and peptone)	Illinois bituminous coal, USA	Microbial community (98.3% Bacteria and 1.7% Archaea species)	24.15 ft^3/ton-day (between days 35 and 55) after treatment	When methane production halts, replacing the used medium and cells could sustain methane release from the coal	2017[143]
Biostimulation (yeast extract and peptone)	Powder River Basin coal seams, USA	In situ microbial community	Treated: 928 μg CH_4/g coal; control: 311 μg CH_4/g coal	Coal-dependent methanogenesis more than doubled when supplements were added	2017[144]
Coal pretreatment (aerobic)	Yima coal Mine lignite, China	Exogenous acclimatized aerobic and anaerobic bacteria	222.50 μmol/g coal	The recalcitrance of coal was mitigated, and the availability of usable substrates increased	2017[145]
Biostimulation (formation water-based recipe)	San Juan coal basin, USA	The microbes in the formation water were composed of 68% of bacteria and 32% of archaea	870.77 ft^3/ton for filtered and 1,041.88 ft^3/ton for unfiltered formation water	Microbial activities toward coal depolymerization stimulated	2017[146]
Biostimulation (algae, cyanobacteria, yeast cells, and granulated yeast)	Powder River Basin coal seams, USA	The bacterial communities for all treatments were *Firmicutes*, *Proteobacteria*, and *Bacteroidetes*.	0.5 g/L amended coal treatments ranged from 1960 to 2185 μg CH_4/g coal	The small amounts of the amendment were not only sufficient but possibly advantageous in faster *in situ* coal-to-methane production	2018[147]
Coal pretreatment (H_2O_2)	Different coal ranks, Australia	Digester sludge with a robust microbial community	223.70 μmol/g for sub-bituminous coal	Methane production increased exponentially with the H_2O_2 concentration but less significantly with pretreatment length	2018[148]
Biostimulation (Fe, methanol, ethanol, and trace minerals)	Illinois bituminous coal, USA	The microbial community comprised 44% bacteria and 56% archaea	1417.35 ft^3/ton yield and 80.7% content	Yeast extract and peptone were found to be indispensable for coal biogasification	2018[149]
Bioaugmentation (methanogenic microbial consortia)	Powder River Basin coals, USA	The feasible co-existence of aerobes, facultative, and strict anaerobes identified in the consortia	14 sft^3/ton (95,700 ppm) after 61 days of incubation	Biodegradation and methanogenesis were stimulated	2018[150]
Biostimulation (ethanol)	Shengli lignite, China	When ethanol was added *Methanobacterium* Increased, while in control *Methanosarcina*	44.86 mL/g at ethanol concentration of 1%	Ethanol changed the structure and composition of coal and facilitated the interaction of microbe with coal	2019[151]

Strategies	Coal, source	Microbial profile	Maximum CH_4 yield	Results and observation	Year, Ref.
Coal pretreatment (H_2O_2)	Powder River Basin sub-bituminous, USA	The obligate hydrogenotrophic *methanobacterium* was the most dominant in the microcosms	552.60 µmol/g at day 184	The enrichment/depletion of the precursor ^{13}C contributed to the shift of the carbon isotopic composition in the subsequence processes	2019[152]
Biostimulation (straw co-degradation)	Qinshui Basin anthracite, China	The methanogenic microflora derived from produced water	592.02 µmol/g coal on the 29th day of cultivation	The enhanced methane production was a result of stimulating the microbial biodegradation of coal	2019[153]
Biostimulation (ethanol)	San Juan and Powder River basin coals, USA	Microbial community with 185 bacteria and 9 archaea species	921.4 ± 1.9 ft3/ton on day 30	Ethanol increased and inhibited methane production from San Juan and Power River coal, respectively	2019[154]
Coal pretreatment (H_2O_2)	Sihe coal mine anthracite, China	The anaerobic microflora enriched from the produced water of active CBM well	254.97 µmol/g at 30% H_2O_2 for 12 hours	Pretreatment increased the oxygen-containing functional groups and decreased the crystal structure of the coal	2020[155]
Coal pretreatment (NaOH)	Qinshui Basin anthracite, China	The microflora was enriched from produced water obtained from the same coal site	240.00 µmol/g coal at 1.5M NaOH for 12 h	The multi-substituted aromatics reduced, the C-O in alcohols and aromatic ethers, and the branching degree of the aliphatic chain increased	2020[156]
Coal pretreatment (electric field)	Qinshui Basin anthracite, China	The bacteria with extracellular electron transfer abilities, such as *Soehngenia*, *Desulfovibrio*, and *Deferrisoma*	63.00 µmol/g coal at 1.2 V	The electric field application changed the microflora structure, promoting extracellular electron transfer and the biodegradation of organic compounds	2020[157]
Microbial acclimation	Erlian lignite basin, China	The diversity of the bacterial and archaeal communities increased	9.26 mL/g after five acclimation passages	The anaerobic digestion efficiency of lignite was significantly improved after acclimatization	2020[158]
Biostimulation ($FeCl_2$ or FeS_2)	Qianqiu Mine coals, China	Microorganisms reacted with Fe to produce several amorphous substances in coal	Methane production of 0.191 mmol/g with FeS_2	The chemical oxygen demand peak value increased after $FeCl_2$ was added	2021[159]
Coal pretreatment (super-critical CO_2)	Anthracite and bituminous, China	Microflora, enriched from the formation water	212.47 µmol/g – anthracite, 202.12 µmol/g – bituminous	More functional groups were formed to increase coal bioavailability after the treatment	2021[160]
Biostimulation (Fe^{2+})	Lignite and bituminous coal, China	The dominant archaea changed from *Methanobacterium* before anaerobic fermentation to *Methanosarcina* after the reaction	For lignite 246 mL and for bituminous coal 197 mL at 10 mg/L Fe^{+2}	Hydrogenase synthesis was promoted, and its activity was enhanced. Soluble sulfides toxicity to bacteria reduced	2021[161]
Biostimulation (microbial electrolysis)	Qianqu mine bituminous coal, China	*Geobacter* and *Methanosarcina* were the most enriched genera in anode biofilms	The cumulative methane production of 6.05 mL/g coal	Significant upregulation in the gene abundances of key enzymes involved in the methanogenic metabolism in hydrolytic bacteria observed	2022[115]

Strategies	Coal, source	Microbial profile	Maximum CH_4 yield	Results and observation	Year, Ref.
Biostimulation (Fe/Cu nanoparticles)	Inner Mongolia bituminous coal, China	3791 OTUs corresponding to bacteria and 110 OTUs corresponding to archaea resulted	6.57 ml/g coal at Fe 1.5 g/L enriched solution	Adding nanoparticles eliminated *Lysinibacillus* in the bacterial community, while *Pseudomonas* prevailed as the dominant bacteria	2022[162]
Biostimulation (corn straw co-fermentation)	Shoushan NO.1 Mine bituminous coal, China	The abundance of *Methanosarcina* and the functional genes observed	26.46 ml/g coal	Fermentation enhanced the hydrogenotrophic and methylotrophic methanogenic pathways	2022[163]
Biostimulation (guar gum co-degradation)	Two bituminous coals, China	Bacteria: *Bacteroidetes* and *Proteobacteria*. Archaea: *Methanosarcina* and *Methanobacterium*	1120.53 and 1167.41 μmol/g	The contents of dissolved organic compounds were enhanced, growth of hydrogenotrophic methanogens were observed	2022[164]

5. Microbial removal of coal impurities and dust suppression

Existing coal processing/combustion technologies emit particulate matter, harmful gases, and GHG into the atmosphere. They also create large amounts of waste/wastewater containing unwanted impurities like ash forming and sulfur compounds.[165] Microbial coal processing methods could remove or reduce most coal impurities and thus would improve both energetic efficiency and environmental effects.[166] In addition, the methods of particulate matter removal and dust suppression using specific microbial strains may be more broadly employed in coal dust prevention and control.[167]

5.1 Removal of sulfur and ash from coal

High sulfur content in coal creates severe challenges to coal processing/application and worsens environmental impact. On average, globally mined coals have a sulfur content varying from 0 to 4 %, but it can also go up to 10 % depending upon the mining geography and depositional history.[3] During coal combustion, sulfur is emitted as SO_2 and SO_3, contributing to air pollution. These compounds also combine with the air's moisture to form H_2SO_4, generating acid rain. Most coal-producing countries have recognized this problem, and have reacted by reducing SO_x emissions and enforcing sanctions.[168] As a result, *coal biodesulfurization* has become a forefront coal processing technology for sulfur removal.

Sulfur is found in fossil coals in various forms and compounds, combining (1) inorganic sulfates (S_s), mainly as ferrous sulfate and gypsum; (2) inorganic sulfides (S_p), generally as ferrous sulfide (pyrite), and (3) as organic sulfur compounds (S_o). Sometimes, a fourth sulfur form is mentioned as well – elemental sulfur (S_{el}), which is detected in weathered coal.[169] The total amount of all types of sulfur content in coal is designated as S_{tot} or S_t. The S_o (mercaptans, thiophenols, thioesthers, dithioesthers, and dibenzothiophenes) may vary from 50 to 70 % of S_t in many coals.[170] The percentage of inorganic sulfur in coal changes from 0.5 to 5 % depending on the rank/type of coal. Some coals are composed of a matrix (condensed aromatic rings), which is highly resistant to microbial action and termed 'hard coal', whereas a mobile phase in coal is composed of nonoxidized compounds or volatile components (phenols, benzoic acids, biphenyls, biphenyl ethers, n-alkenes, n-alkanols, and wax), which can be easily attacked by microorganisms.[3] Most microbial species have been demonstrated to disintegrate the coal structure and utilize the compounds present in coal as a source of carbon and sulfur to meet their nutrient requirements.[171]

Since dibenzothiophene constitutes a major organic portion of coal, microorganisms follow one of the different pathways, including aerobic biodesulfurization: the 4S pathway (oxidative C–S cleavage), the Kodama pathway (oxidative C–C cleavage), and anaerobic biodesulfurization: reductive C–S cleavage.[172] In the 4S pathway, the microorganisms selectively oxidize the sulfur atom in dibenzothiophene without cleaving the C–C bond, thereby maintaining the calorific value. Four different molecular species are formed during sulfur metabolism: dibenzothiophene sulfoxide, -sulfone, -sulfinate, hydroxybiphenyl sulfite. Bacteria *Rhodococcus erythropolis* can desulfurize sulfur from dibenzothiophene by the 4S pathway. Besides, *Gordona* sp., *Paenibacillus* sp., *Xanthomonas* sp., *Mycobacterium* sp. were also reported to remove sulfur from benzothiophene via the 4S pathway.[170] Bacteria, like *Pseudomonas* sp., *Pseudomonas* sp., *Burkholderia* sp., and *Brevibacterium* sp. use dibenzothiophene as their carbon source through a series of enzymatic oxidation steps that lead to the breakage of the C–C bond present in the ring structure. The pathway comprises three major steps: hydroxylation, ring cleavage, and hydrolysis.[173]

Several anaerobes, such as *Desulfovibrio* sp. and *Desulfomicrobium* sp., have shown promising metabolic responses toward dibenzothiophene biodegradation, forming just minor amounts of undesirable compounds such as colored and gum-forming products upon oxidation of hydrocarbons.[174]

S_p comprises a significant portion of inorganic sulfur in coal, with certain chemolithoautotrophic meso-acidophilic bacteria being involved in its degradation. Most studies on biodesulfurization of coal, extensively listed by Osorio *et al.*, account for pyritic sulfur removal using *Acidithiobacillus ferrooxidans*.[175] Regarding the biological oxidation of pyrite, different mechanisms have been proposed, namely, 'direct (contact)' and 'indirect (noncontact)' bioleaching.[176] These mechanisms can be summarized as direct oxidation (Eqs. 4 and 5):

$$2FeS_2 + 2H_2O + 7O_2 \rightarrow 2FeSO_4 + 2H_2SO_4 \quad (4)$$

$$2FeSO_4 + \tfrac{1}{2}O_2 + H_2SO_4 \rightarrow Fe_2(SO_4)_3 + H_2O \quad (5)$$

and indirect oxidation (Eq. 5):

$$FeS_2 + Fe_2(SO_4)_3 \rightarrow 3FeSO_4 + 2S \quad (6)$$

The bacteria oxidize elemental sulfur formed by indirect oxidation to sulfuric acid (Eq. 6):

$$2S + 3O_2 + 2H_2O \rightarrow 2H_2SO_4 \quad (7)$$

Many sulfur-oxidizing microorganisms, including *Leptospirillum ferrooxidans, Beggiatoa, Sulfolobus, Acidithiobacillus thiooxidans*, and *Sulfolobus acidocaldarius* were also reported to degrade sulfur compounds to form elemental sulfur or metal sulfides.[177]

Since coal is a cheap and profuse energy source, the removal of organic and inorganic sulfur from various coals by bacterial strains would have a large-scale promoting effect on environmental safety and energy efficiency. The recently published studies on coal desulfurization are listed in Table 4.

Table 4. Studies on coal desulfurization by different microorganisms (within last 5 years).

Sulfur removal strategy	Coal, source	Microorganism	Maximal sulfur removal	Condition	Results and observation	Ref.
S_t (S_p, S_o)	Jining LRC, China	*Pseudomonas* sp. NP22	~46% (35.8%, 56.2)	pH 5, particle size of −75 + 45 μm, 5% of pulp density, at 35 °C for 8 days	The calorific value of LRC was not affected after desulfurization	2017[178]
S_t (S_o, S_p, S_s)	Tondong-kura coal, Indonesia	*Pseudo-clavibacter sp.* SKC/XLW-1	46.55% (25.43%, 34.74%, 82.82%)	Multi-stage bioprocess with biooxidation (15 or 40 days), bioflotation (30 min) and column flotation	Bacterium's extracellular polymeric substances are responsible in removing S	2017[179]
S_t	NE coal and Rajasthan lignite, India	*Rhodococcus ruber* 9C	29% and 15.87%	MSM with pH 6.5 ± 2, pulp density (w/v) 5%, 150 rpm, 37 °C, 15 days	Treatment increased the coal gross calorific value from 6698 cal/g to 6812 cal/g	2017[180]
S_t (S_p)	Yangquan mine coal, China	*Thiobacillus ferrooxidans*	88.58% (93.89%)	Particle size of –74 μm for 60%, the pulp density was 15% and inoculation amount of 15%, 15 days, pH 1.8, 30 °C, 150 rpm	S_s and S_o were barely removed by *Thiobacillus ferrooxidans*	2017[181]
S_t	Liupanshui coal, China	*Acidithio-bacillus caldus* and *A. thiooxidans*	29.7%	The 9 K medium in 16 days when 1100 mg/L Tween 20 in the shaking test	Tween 20 had a significant effect on coal biodesulfurization	2017[182]
S_t	Tabas Coal Processing Plant, Iran	Mixed culture of Fe- and S-oxidizing microorganisms	31%	pH 1.7-2, 190 days, semi-continuous mode	The coal cleaning efficiency increased from 28% to 36.5%	2017[183]
S_t (S_p)	Wuhai city coal, China	Indigenous and exotic microorganisms	45.3% (77.7%) and 45.8% (87.9%), respectively	pH 1.71-1.73, 10% pulp density, 28 °C, 150 rpm, particle size 0.2 mm, 36 days	Both microbial groups had no effectiveness on S_o removal	2018[184]

Sulfur removal strategy	Coal, source	Microorganism	Maximal sulfur removal	Condition	Results and observation	Ref.
S_t	Çan lignite, Turkey	*Leptospirillum ferriphilum* DSM 14647	56.2%	DSMZ media with 5% (w/v) coal, pH 1.8, 32 °C, at 150 rpm for 21 days	Fe^{2+} in the media improved the bio-desulphurization	2018[185]
S_t	South Karanpura coalfield coals, India	*Pseudomonas mendocina* B6-1	13.79-45.00%	Basal salt medium pH 6.0, 35 °C, pulp density 6.0% (w/v), 7 days, 125 rpm	The treatment caused an increase in the coal's useful heat, gross and net calorific values	2018[186]
S_t	Giral mine lignite, India	*Burkholderia* sp. GR 8-02	50.69%	34 °C, 120 rpm, 30 days, -70 mesh coal size	Treatment increased the calorific value of coal from 5.24% to 20.74%	2018[187]
S_t	Liupanshui coals, China	*Rhodococcus erythropolis* SX-12 and *Acidithiobacillus ferrooxidans* GF	56.83% and 65.23%	Two-step bioleaching for SX-12 (BSM medium) and GF (9K medium), 30°C, pH 7.0, 30 days	Two-step bioleaching overcame the shortcoming of co-culture and had a high rate	2019[188]
S_t	Datong coal mine, China	*Pseudomonas putida*	58.23%	10 days, particle size -125 + 75 µm, pH 6.0, Tween 80 - 0.1%	The coal's energy value was not affected in the biodesulfurization	2019[189]
S_t	Kalimantan coal, Indonesia	*Pseudomonas moraviensis*	17.37%	SKC broth, 180 rpm, at room temperature for 10 days	A reduction in coal ash content after treatment was observed	2019[190]
S_t (S_p)	Huozhou Coal Electricity coals, China	*Acidithiobacillus ferrooxidans*	87.5% - 92.0% (28.2% - 31.6%)	With or without iron-free M9 K medium, 28 °C and 180 r/min for 768 h, pH 2.50	The addition of excessive iron-free M9 K was detrimental to coal biodesulfurization	2020[191]
S_t (S_o, S_p, S_s)	Çan open mine, Turkey	*Acidithiobacillus ferrooxidans* DSM 583	30.84% (20.69%, 32.76%, 30.43%)	A bioreactor with 9 K medium, 12 hours, pH 7.0-7.5, 30-32°C	After treatment, ash content - 33.88% and calorific value - 3982 kcal/kg	2020[192]
S_t (S_p, S_s)	Liejiaqiao coal Mine, China	*Acidithiobacillus ferrooxidans* and *Acidithiobacillus thiooxidans*	From 2.06% to 1.18%. (78.79%, 49.02 %)	The continuous leaching system with 9K medium, flow rate of 10 mL/min for 74 days, initial pH 3.22 and 2.77	The releasement of metal ions in coal, with a leaching concentration of Fe > Mn > Cr	2021[14]
S_t (S_s, S_p, S_o)	Assam coal mine, India	*Acidithiobacillus ferrooxidans*	79.72% (73.64%, 71.42%, 84.75%)	$9K^+$ medium, pH 1.8 ± 0.05, 28 ± 2 °C, 150 rpm, 14 days	Biodesulfurization of the DBT evaluated as well	2022[193]
S_o	Yunnan bumuga high-sulfur coal, China	*Nocardia mangyaensis* and *Pseudomonas putida*	61.58% and 54.19%, respectively	Media with coal of 0.075–0.125 mm particle size, at 30°C and operated at 160 r.p.m for 14 days	The combustion performance of biotreated coal was better than raw coal	2022[194]

Sulfur removal strategy	Coal, source	Microorganism	Maximal sulfur removal	Condition	Results and observation	Ref.
S_t	Coals from different mining areas, China	*Acidithiobacillus ferrooxidans* YQ-N3	40.17%-62.25%	9K liquid medium, 30 °C, 180 r/min, pH 2, 30 days	YQ-N3 can increase the oxidation rate of Fe^{2+} and S^0 and enhance the S^0 hydrophilicity	2023[195]
S_t	Lakhra coal, Pakistan	*Rhodococcus* sp. SL-9	45%	20% (w/v) pulp density, 180 rpm, 20 mesh (850 microns), pH 7	The different process parameters can be used for S removal	2023[196]

Studies also employ clean coal technologies based on fungal strains to upgrade the quality of LRC, removing sulfur. For instance, fungi *Aspergillus* sp. (78%),[197] *Alternaria* sp. (52%),[198] (49.01%),[199] *Trametes versicolor* ATCC 200801 (40%),[200] (29%)[201] demonstrated promising results in sulfur removal from different coals. Şener *et al*. investigated the potential of *Alternaria* sp. through a drum reactor and showed 20% sulfur removal, and a 42% reduction in sulfur emission values.[202]

Most of the LRC have low calorific values, high moisture content, and high ash contents, and their utilization is a priority. While microbiological methods may generally be sufficient for removing sulfur from LRC, supplementary solutions may be required to reduce ash-forming inorganic materials responsible for particulate matter emissions.[168] In their book, Ishfaq *et al*. provide an in-depth review concerning the problem of ash and sulfur removal.[203] A plethora of research has demonstrated the effectiveness of biodesulfurization using bacterial strains, such as *Bacillus subtilis*,[204] *Acidithiobacillus ferrivorans*,[183,188,192,205-209] *Pseudomonas* sp.,[178,186,210] *Sinomonas flava*,[207] *Burkholderia* sp.,[187] and *Rhodococcus erythropolis*[188] for the reduction of high ash contents. Yet some existing studies focus on fungal ash removal by *Aspergillus* sp.[197] and *Alternaria* sp..[198,202]

5.2 Coal dust suppression

The large scale of coal mining, processing, and utilization results in high levels of dust formation in the environment.[211] Coal dust presents a major risk due to its spontaneous combustion,[212] explosion,[213] and pollution,[214] characteristics. In order to overcome the coal dust-related problems, different dust inhibition and control methods have been implemented, such as the usage of dust covering agents, chemical dust suppression, water spray dust removal, and surface curing.[215,216]

A "green" environmental dust suppression technology based on microorganisms would provide here an interesting way for making coal production cleaner and safer. In 2020 Fan *et al*. reported about such biomineralization technology for coal dust suppression.[217] The coal dust-calcium carbonate consolidation layer formed by urease-producing *Aspergillus sydowii* and *Bacillus* DB-6 not only inhibited dust production and explosiveness but also restricted coal contact with oxygen. This allowed to prevent spontaneous coal ignition, which obviously opens broad application prospects for fire prevention and extinguishing in coal. Indeed, microbially induced coal dust-calcium carbonate consolidation has been successfully conducted by many research teams, using urease-producing bacteria *Acinetobacter guillouiae* CIP 63.46 and *Staphylococcus caprae*,[218] *S. succinus* J3,[219] *B. pasteurii*,[220] *B. subtilis*,[221] *Sporosarcina pasteurii*,[222] *B. cereus* CS1 and *S. pasteurii* ATCC11859,[167] *B. pasteurii* and *B. pasteurii*,[223] as well as urease extracts.[224] The adsorption characteristics and interaction mechanism between coal dust and the urease-producing *Bacillus* X4 were studied by Zhang *et al*. in order to further explore the dust suppression mechanism of microbial urease-based dust suppressants.[24] The results of contact angle measurements, spectroscopy analysis, and molecular dynamic simulations demonstrated that *Bacillus* X4 formed a monolayer on coal dust with uneven adsorption, which mainly involved amide groups. The level of dust suppression was positively correlated with the level of bacterial adsorption on coal dust.

Coal dust has high hydrophobicity due to a large number of non-polar groups, including aliphatic hydrocarbons and aromatics.[225] Surfactants, being amphiphilic compounds, can effectively reduce the risk of coal dust by lowering the surface tension of the solution and reducing the hydrophobicity of the coal dust.[226] Biosurfactants are currently receiving extensive research attention due to their low toxicity, high biodegradability, and ecological acceptability.[227] Liu *et al*. screened the biosurfactant-producing *Pseudomonas* PLD01 bacteria from coal and analyzed the ability of their biosurfactants to improve the hydrophilicity of coal dust. The results indicated that biosurfactant (50.0 mg/L) reduced the surface tension of water from 72.8 mN/m to 40.6 mN/m; furthermore, the change in wettability achieved with the biosurfactant was even better than that of the popular synthetic surfactant sodium dodecyl sulfate. Micro-flotation tests conducted by Augustyn *et al*. showed that at acidic pH, the hydrophilicity of coal was significantly enhanced (from 42.9% to 74%) by 15 mg/L surfactin, a biosurfactant of *Bacillus subtilis*.[228] Rhamnolipid-nature biosurfactants produced by *P. stutzeri*[229] and *P. aeruginosa*[230] showed their high potential for coal dust-reduction technologies too.

6. Summary and conclusions: knowledge gaps

Global coal demand is rising at an alarming rate while its reserves are depleting. This tendency creates the prerequisites for more coal production or the introduction of alleviating approaches that make the coal economy more sustainable. Microbial biomasses have huge economic and ecological potential in coal co-processing to produce clean fuels of various forms to be used in different domestic and industrial sectors. However, developing or adopting appropriate technologies for coal beneficiation by removing different impurities without adverse environmental impacts is a significant challenge. The following summary highlights some of the crucial points that should be addressed by future studies in order to deepen our understanding of clean fuel production from LRC:

1. Coal co-processing: despite extensive research, the underlying mechanism of coal and microbial biomass interactions remains unclear. Kinetic analysis and detailed product characterization may reveal coal processing and utilization processes, improving coal usage quality. Moreover, pyrolytic behaviors, kinetics, and gas-releasing characteristics for coal and microalgae/sewage sludge co-processing should be deeply analyzed. More attention needs to be paid to the effect of individual sewage sludge components on co-processing thermal behavior. Ideally, large-scale experiments that deliver better coal co-processing for clean fuel production should be performed.

1.1. Coal co-firing: little or no emphasis has been directed so far toward the co-firing emission profile, thermochemical properties, and kinetic behavior, as well as the environmental and safety qualities of the resulting products. The question should be addressed here: what kinds of operations and processes improve the combustion performance of coal during co-firing? However, for large-scale energy production from coal co-firing, new approaches are being developed. The Coalgae® technology is currently being demonstrated to integrate the various unit operations of the large-scale technology to produce several tons of Coalgae® product.

1.2. Coal co-pyrolysis: although much research has been done on the co-pyrolysis process itself, little kinetic analysis data is available. In addition, further research on the distribution and release characteristics of products from coal and microalgae co-pyrolysis is needed. The questions that remain unaddressed include biomass-fossil fuel ignition and combustion characteristics, fuel reactivity, burnout, and ash deposition behavior. However, large-scale co-pyrolysis operations to produce valuable char products seem to have greater potential; the highest char yields were obtained at ~850 °C under a 50% biomass ratio.

1.3. Coal co-gasification: due to the varieties of coal and microalgae biomass, it is not clear whether co-pyrolysis had a synergistic effect on the volatile yield and its release rate, which is not conducive to guiding efficient gas production. Nevertheless, the available data gives a good insight into the co-gasification behavior under up-draft, down-draft, fluidized bed, and entrained flow reactors. Co-gasification of coal and different microalgae performed using these reactors produced a high syngas yield of <2.0 Nm^3/kg.

1.4. Coal co-combustion: many aspects of coal and microalgae co-combustion must be thoroughly investigated before it can be practically considered a power generation tool in coal-firing boilers. To achieve clarity, research questions should be carefully framed related to the biochemical and physical background of coal and microalgae interactions, their agglomeration, and synergetic effect. Despite shortcomings in some aspects, co-combustion has prospects for practical application. The percentage of biomass co-combusted in distinct power plants with experience ranges from 15% to 100% for grate combustion, 40% to 90% for fluidized bed combustion, and 0% to 100% for pulverized coal combustion.

1.5. Coal co-liquefaction: several areas of co-liquefaction are poorly understood due to scarce studies conducted so far. Improvements in microbial strain performance could be an essential aspect of coal co-liquefaction. Another interesting aspect is whether coal residues of different origins and depositional histories have an adequate co- liquefaction capacity. There is currently no precise prescription for co-liquefaction technological innovation in large-scale practical applications.

2. Biogenic coalbed methane production: understanding the drivers that impact coal gas production may provide a basis for ultimately enhancing biogenic methane yields. Biogenic methane production using various microbial consortia should be examined for economic and biotechnological potential. Important questions remain regarding the environmental impact of unconventional methane production, including formation water quality and toxic metal mobility. The answers to these questions may have implications for both recovery practices and the development of unconventional resources in a sustainable manner. Although some pretreatment, biostimulation, and bioaugmentation methods have been adopted in the laboratory to improve coal bioavailability or biosolubility in order to increase biomethane yield, the majority of these methods are difficult to implement at large-scale operations (*i.e.*, in-situ and/or ex-situ applications). Improving the bioavailability of coal when producing biomethane on a commercial scale urgently necessitates more cost-effective and practical approaches.

3. Microbial sulfur-ash removal and coal dust suppression: isolating and screening novel microbial strains with enhanced desulfurization capabilities may improve their overall performance. The mineralogy of coal with various sulfur and ash levels needs better understanding prior to microbial applications. Coal dust adsorption control and, hence, reliable dust suppression protocols are limited by a lack of in-depth microbial and physicochemical research. Furthermore, screening and analysis of biosurfactant-producing bacteria that improve the hydrophilicity of coal dust are still scant for

developing sustainable coal dust suppression technology. In the coming years, more and more research on this intriguing aspect of coal bio-desulfurization and dust suppression will enhance its distinguishing characteristics and energy value, fostering optimism for the development of an industrial-scale bioengineering system.

Miscellaneous: since coal varies in rank, composition, and occurrence, its in-depth characterization could immensely help to implement microbial resources with maximum performance for developing clean coal technologies. Environmental and health concerns related to coal mining, processing, and utilization increasingly create motivation for the search for better scientific and technological solutions. The development and implementation of novel technologies using microbial biomass are gaining broader recognition. A better understanding and management of microbe-coal interactions would certainly facilitate achieving the goal of engineering versatile and efficient coal-microbial systems.

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Conflict of Interest

There is no conflict of interest.

Supporting Information

Not applicable.

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