



Opportunities, challenges and modification methods of coal gangue as a sustainable soil conditioner—a review

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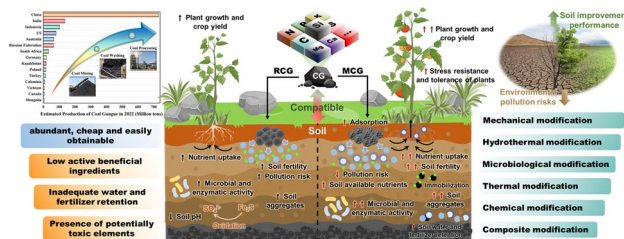
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Abstract

The persistent reliance on coal has resulted in the accumulation of substantial coal gangue, a globally recognized problematic solid waste with environmental risks. Given the coal gangue properties and global land degradation severity, the resourceful utilization of coal gangue as soil conditioners is believed to be a universally applicable, cost-effective, high-demand and environment-friendly model with broad application prospect. The direct application of raw coal gangue faces challenges of low active beneficial ingredients, inadequate water and fertilizer retention, presence of potentially toxic elements, resulting in limited efficacy and environmental contamination. This paper provided a comprehensive review of various modification methods (including mechanical, chemical, microbiological, thermal, hydrothermal and composite modifications) employed to enhance the soil improvement performance and reduce the environmental pollution of coal gangue. Furthermore, an analysis was conducted on the potential application of modified coal gangue as a multi-function soil conditioner based on its altered properties. The modified coal gangue is anticipated to effectively enhance soil quality, exhibiting significant potential in mitigating carbon emissions and facilitating soil carbon sequestration. This paper provided innovative ideas for future research on the comprehensive treatment of coal gangue and restoration of degraded soil in order to achieve the dual goals of zero-coal gangue waste and sustainable agriculture.

Graphical Abstract



Keywords Coal gangue · Soil conditioner · Modification · Resourceful utilization · Soil remediation · Sustainable

Introduction

Coal is the largest source of global energy, accounting for over 26% of the world's primary energy with a total consumption of 161.47 EJ in 2022 (IEA 2023). Despite renewable energy sources (e.g., wind, sun, biomass) gain the increasing global focus and are generally proposed as potential substitutes for fossil fuels in recent years (Brockway et al. 2019; Devlin et al. 2023; Smith et al. 2022), fossil fuels particularly coal, still dominate the world energy

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consumption as illustrated in Fig. 1a. The global coal consumption witnessed a growth of over 7% in 2022, with China and India accounting for more than 70% (Debiagi et al. 2022). Consequently, the dominant role of coal in global energy supply is anticipated to remain unchanged for the foreseeable future.

However, a substantial amount of coal gangue was generated and discharged during the coal mining, washing and processing processes. Coal gangue, a black-gray rock with lower carbon content and heat value compared to coal, accounts for about 15%-20% of coal production (Li et al. 2006). The production of coal gangue was conservatively estimated based on the published coal production data from the Energy Institute (Energy Institute 2023), as shown in Fig. 1b. As the world's largest coal producer, China produced nearly half of coal gangue. The amount of accumulated coal gangue in China has exceeded 7 billion tons as of 2022 (Wang et al. 2023), and is still rising at an alarming rate of more than 300 million tons per year (Li and Wang 2019). The utilization rate of coal gangue in China only reached 73.1% as of 2023 with a consistent flat trend as depicted in Fig. 1c (Li and Wang 2019; Wu et al. 2023a). Therefore, the utilization amount of coal gangue still falls short in comparison to its vast production and reserves, making it one of the largest industrial solid wastes (Wu et al. 2017).

The accumulation of enormous coal gangue not only occupies a significant amount of land resource, but also causes severe safety and environmental issues (Fig. 1c), including soil deterioration, spontaneous combustion, the release of toxic gases, heavy metal contamination and geological hazards (Ma et al. 2019a; Sun et al. 2021; Wu et al. 2023b). Hence, numerous countries have been actively exploring various utilization ways to eliminate the threats triggered by coal gangue accumulation. At present, considerable research has been conducted on the resourceful utilization of coal gangue as value-added products. The global comprehensive utilization of coal gangue encompasses various sectors, including construction materials (e.g., cement and concrete) (Qin et al. 2021), energy generation (e.g., power generation) (Peng and Li 2018), filling applications (e.g., underground backfill) (Li et al. 2020c) and emerging industries (e.g. chemical products) (Cao et al. 2021). The applications mentioned above are extensively employed in economically developed and densely populated areas, yielding remarkable economic and social benefits. Conversely, in underdeveloped areas with a substantial stock of coal gangue, they fail to achieve the anticipated outcomes due to the low local demand and exorbitant export costs.

Moreover, the excessive cultivation and prolonged use of chemical fertilizer have resulted in the gradual depletion

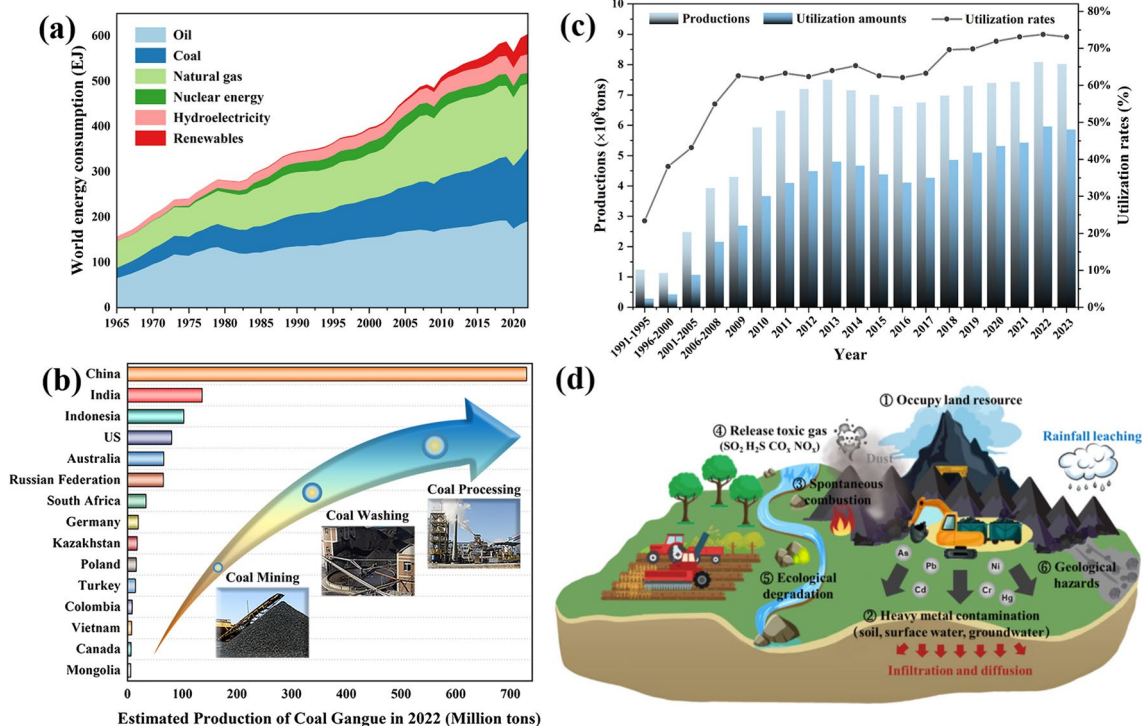


Fig. 1 **a** The global energy consumption by source in exajoules (EJ) from 1965 to 2022; **b** The coal gangue generation links and estimated production of coal gangue in various countries in 2022; **c** Produc-

tion, comprehensive utilization, and utilization rate of coal gangue in China from 1991 to 2023; **d** The harm caused by the accumulation of enormous coal gangue

of organic matter and humus in soil, leading to increasingly serious soil degradation (Babla et al. 2022). Severe soil degradation has occurred in one-third of the world as a result of soil erosion, desertification, salinization, compaction, and pollution (Hou et al. 2020), thereby diminishing soil productivity and often necessitating soil conditioners to replenish the soil (for the lost nutrients). However, the utilization of commercial soil conditioners incurs substantial costs, constituting 10 to 90% of the overall expenses associated with farm treatment (Yunusa et al. 2012). Previous studies have indicated that scientific and reasonable addition of coal gangue holds the potential to improve soil quality, which will effectively overcome the negative effects caused by traditional fertilizers on soils (Tang et al. 2014). Hence, the utilization of coal gangue as soil conditioners is presumed to be a universally applicable, affordable, high consumption and efficient sustainable approach, which can tackle the issues of coal gangue management and soil degradation simultaneously.

Currently, an increasing work has been reported on the resourceful utilization of coal gangue as soil conditioners, however there is a lack of comprehensive summary in this field. To fill this vacancy, the properties of coal gangue with a specific focus on the analysis of its overlooked agricultural value were summarized in this paper. Then, this review identified the opportunities and challenges related to using raw coal gangue as soil conditioners from previous studies. Furthermore, the mechanisms, effects, and limitations of several modification methods for coal gangue were comprehensively integrated and evaluated in order to enhance soil improvement performance and ensure environmental safety. The potential application and environmental implication of modified coal gangue were also discussed. This review aims to provide valuable insights and references for utilizing coal gangue in soil practices and sustainable agriculture.

Properties of coal gangue

Coal gangue is a complex industrial solid waste mixed with organic compounds, inorganic compounds and minerals (Fabiańska et al. 2013). Its major chemical compositions are SiO_2 , Al_2O_3 and Fe_2O_3 , with traces of CaO , P_2O_5 , MgO and MnO_2 . The chemical composition of coal gangue from various sources and its comparison with soil reference values were tabulated in Table 1. It can be concluded that the chemical composition of coal gangue and soil exhibits similarities, suggesting compatibility between them, which may be attributed to the fact that they are both formed in strata (Wang et al. 2016b). It is worth noting that the majority of coal gangue samples present a high loss on ignition (LOI), indicating a higher proportion of organic carbon (Zhao et al. 2022b), which is conducive to the soil utilization of coal

gangue. Besides, coal gangue predominantly consists of quartz and clay minerals (kaolinite, illite, etc.) (Cao et al. 2016). The abundance of clay minerals provides convenient for the enhancement of soil structure and fertility (Grim 1962).

There is a growing stockpiles of the industrial byproducts including coal gangue, fly ash, red mud and flue gas desulfurization gypsum which may potentially give rise to environmental concerns (Koshy et al. 2019). The comparison with other industrial solid wastes (Table 2) reveals that coal gangue exhibits higher levels of nitrogen (N), phosphorus (P), potassium (K) and sulfur (S), contributing to improve soil fertility. Although sulfur is considered detrimental in certain coal processing and utilization projects, it can effectively stimulate crop growth and productivity in moderate quantities (Mu et al. 2021; Yuan et al. 2021). Coal gangue possesses developed pores and specific surface area, thereby aiding soil aeration and preventing compaction (Zhang et al. 2022a). The organic carbon content in coal gangue is typically 2 to 10 times higher than that found in soils, making it possible to produce multi-functional soil conditioners using coal gangue as a raw material.

In addition, coal gangue contains various trace elements such as Mn, Mo, B, Zn, Cu, Cr, Cd, Pb, Ni, As, Hg, Se, and Co (Table S1), both as essential micronutrients and environmental pollutants (He et al. 2005). Compared with other industrial solid wastes, the potential hazardous trace elements of coal gangue are generally within acceptable limits, as displayed in Fig. 2. This means that coal gangue contains adequate and safe levels of trace elements to ameliorate the soil and support plant growth more securely and effectively. Currently, the research on trace elements in coal gangue primarily focuses on their adverse aspects, including the distribution, leaching, diffusion and environmental risk assessment of heavy metals such as zinc (Zn), copper (Cu), chromium (Cr), cadmium (Cd), nickel (Ni), arsenic (As), mercury (Hg), among others (Ashfaq et al. 2020; Gao et al. 2021a), the migration and release of potentially deleterious trace elements during combustion (George et al. 2020; Wang et al. 2021b). However, there is a lack of attention to the nutritional value and benefits associated with trace elements like molybdenum (Mo), boron (B), selenium (Se) contained within coal gangue.

Utilization of coal gangue as soil conditioners

Owing to excessive reclamation and intensive use of fertilizers, the organic matter and humus in soil are gradually depleted, soil degradation has become a global issue. Because of the remarkable effect of soil conditioner in improving soil quality, it has received extensive attention in

Table 1 Chemical composition of coal gangues (wt. %) sourced from regions and cities

Sources	SiO ₂	Al ₂ O ₃	CaO	Fe ₂ O ₃	Na ₂ O	MgO	Ti ₂ O	K ₂ O	SO ₃	P ₂ O ₅	MnO ₂	LOI	Refs
Beijing, China	49.90	24.41	0.82	6.42	1.46	1.59	0.88	2.06	0.12	-	-	11.76	(Li et al. 2006)
Beijing, China	56.11	16.78	3.11	7.02	1.84	1.68	-	6.98	-	-	-	6.38	(Li et al. 2010)
Xuzhou, China	55.50	18.15	3.38	5.42	0.64	1.23	-	1.67	0.64	-	-	13.20	(Li et al. 2006)
Shanxi, China	56.56	36.78	0.62	1.95	0.42	0.22	2.10	-	-	-	-	-	(Ma et al. 2019b)
Shanxi, China	55.65	29.00	0.53	5.01	0.85	0.49	1.13	1.81	5.01	-	-	-	(Li et al. 2022a)
Shanxi, China	54.63	24.63	0.15	2.17	0.349	0.815	0.682	2.52	-	0.074	0.006	13.45	(Li et al. 2023)
Inner Mongolia, China	45.90	16.00	0.74	4.71	0.99	1.37	0.78	3.36	-	-	-	8.03	(Guan et al. 2021)
Inner Mongolia, China	59.33	30.23	0.41	4.88	0.07	0.75	1.12	2.35	0.26	0.11	0.09	-	(Li et al. 2020a)
Hebei, China	57.74	30.58	0.20	4.31	1.10	1.00	-	2.76	0.16	-	-	-	(Guo et al. 2022)
Heilongjiang, China	58.82	27.87	0.78	8.31	-	-	-	1.74	0.51	-	-	1.97	(Qin and Gao 2019)
Anhui, China	58.20	32.00	1.01	3.96	0.34	-	1.23	1.74	-	-	-	1.52	(Jin et al. 2022)
Anhui, China	53.24	22.09	1.43	5.97	0.32	0.39	-	0.95	0.08	-	-	13.36	(Wang et al. 2022a)
Anhui, China	51.13	25.51	0.44	3.10	0.30	1.16	0.88	1.10	-	0.32	0.06	-	(Fu et al. 2018)
Wuhan, China	69.30	20.80	0.24	3.81	0.36	0.43	1.34	2.07	1.25	-	0.05	11.5	(Liu et al. 2021c)
Henan, China	51.66	17.77	3.82	3.67	0.53	1.01	0.71	1.21	-	0.05	0.04	19.53	(Xu et al. 2017)
Spain	43.70	21.35	0.89	5.57	0.11	0.77	1.05	0.16	1.02	0.16	-	25.18	(Frias et al. 2012)
	57.20	18.69	1.86	6.25	0.46	1.42	0.95	0.14	0.08	0.14	-	9.28	
	46.86	17.15	7.60	7.67	0.15	0.99	0.78	0.16	0.34	0.16	-	15.80	
Spain	49.79	21.77	3.84	4.07	0.13	0.64	1.07	2.74	0.27	0.13	0.08	15.18	(Canales-Martínez et al. 2019)
Brazil	55.91	24.81	0.28	8.73	-	-	1.17	2.02	2.11	-	0.01	4.50	(Frasson et al. 2019)
Poland	58.95	20.50	0.35	6.63	0.54	1.93	1.05	3.19	0.17	0.06	0.05	5.50	(Jabłońska et al. 2017)
Poland	45.80	24.00	0.60	4.30	0.40	0.90	1.30	2.30	1.90	0.50	-	-	(Záleská et al. 2023)
India	52.70	22.60	2.45	6.37	0.65	1.22	0.98	2.48	0.43	-	-	12.65	(Ashfaq et al. 2020)
India	59.20	18.00	2.10	7.29	-	-	2.67	2.71	5.23	-	0.096	-	(Sahu et al. 2019)
Iran	58.80	23.10	1.59	5.30	-	1.68	2.69	3.23	0.95	-	-	2.28	(Mohammadi et al. 2019)
Morocco	52.40	21.90	0.81	4.55	1.53	1.26	-	2.24	3.54	0.135	-	10.70	(Moussadik et al. 2022)
Australia	59.24	23.41	2.16	9.53	-	1.16	0.98	0.67	0.45	-	-	-	(Tang et al. 2020)
Columbia, USA	33.99	13.54	0.34	3.49	0.25	0.51	0.53	1.19	4.97	0.14	0.02	40.96	(Rodríguez et al. 2021)
Soil reference value	37.10–77.70	6.86–32.38	0.50–5.19	2.18–11.33	0.48–4.90	0.41–4.90	0.23–1.83	1.34–5.69	-	-	-	-	(Wang et al. 2018)

L.O.I. represents the loss on ignition

Table 2 Comparison of physicochemical properties in several typical industrial solid wastes

Physicochemical properties	Coal gangue ^a	Coal fly ash ^b	Red mud ^c	Flue gas desulfurization gypsum ^d
pH (1:5)	6.90–8.93	3.1–12.8	9.0–13.1	6.5–8.46
EC (ms cm ⁻¹)	0.26–2.2	3.2–7.6	10.0–60.8	0.84–2.6
Specific surface areas (m ² /g)	0.52–7.24	2.5–14.8	5.31–64.09	0.38–6.68
Organic carbon (%)	3.5–21.2	0.45–1.7	0.60–1.28	0.16–2.4
Total nitrogen (%)	0.19–0.87	0.01–0.12	<0.02	<0.01
Total phosphorus (%)	0.23–0.47	0.01–1.08	0.01–0.08	0.01–0.42
Total potassium (%)	0.86–2.71	0.01–0.28	0.10–1.82	0.05–1.35
Total sulfur (%)	0.55–4.90	0.01–0.58	0.10–0.60	8.2–20.9

a (Ashfaq et al. 2020; Du et al. 2020; Jabłońska et al. 2017; Li et al. 2020a; Li et al. 2022a; Tong et al. 2008; Zhou et al. 2015; Zhou et al. 2012)

b (Bhattacharya et al. 2012; Chen et al. 2010; He et al. 2017; Jankowski et al. 2006; Sarbak et al. 2004; Singh et al. 2010; Yunusa et al. 2012)

c (Anam et al. 2019; Berta et al. 2021; Garau et al. 2011; Gräfe and Klauber 2011; Hua et al. 2017; Liu et al. 2014; Liu et al. 2021b)

d (Aakriti et al. 2023; DeSutter et al. 2014; Li et al. 2022b; Liu et al. 2021a; Sun et al. 2014; Wang et al. 2017)

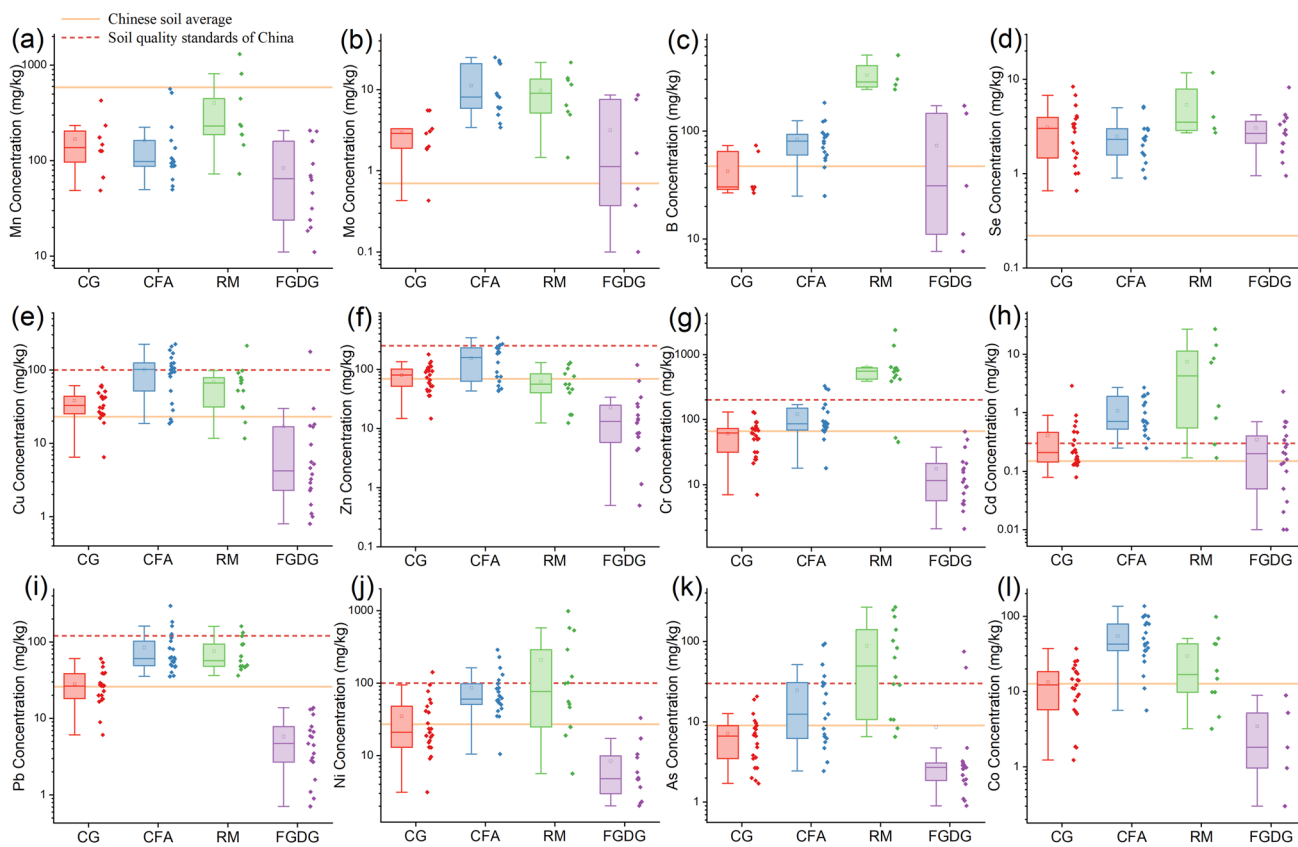


Fig. 2 The summary and comparison of trace element contents in coal gangue (CG), fly ash (CFA), red mud (RM), and flue gas desulfurization gypsum (FGDG) based on the existing literature

recent years (Amoah-Antwi et al. 2020; Rakshit et al. 2019). Current research mainly focuses on artificial synthetic

polymer soil conditioners, natural soil conditioners and microbial soil conditioners (Saha et al. 2020; Song et al.

2018; Zhou et al. 2019). Coal gangue, the main object of this review, is a kind of natural soil conditioner. As a kind of industrial solid waste, it is abundant, cheap and easily obtainable, and its composition closely resembles that of known soil (Tong et al. 2008). Consequently, the coal gangue-derived soil conditioner exhibits superior compatibility with soil compared to artificial synthetic and microbial soil conditioner (Han et al. 2021), enabling its extensive and prolonged utilization while achieving mass-scale production at low cost. If the substantial accumulated coal gangue can be utilized to ameliorate and remediate extensive areas of potentially available degraded soil worldwide, it will give a promising solution for addressing both coal gangue management and soil degradation problems simultaneously. It represents an effective and feasible way for local resourceful utilization of coal gangue as soil conditioners.

Soil improvement application of raw coal gangue

As mentioned above, coal gangue is primarily composed of clay minerals, quartz and carbon. Its organic matter content ranges from 15 to 25%, and it contains nutrients such as nitrogen, phosphorus, potassium, calcium, magnesium, silicon and trace elements required for plant growth (Tong et al. 2008). The coal gangue-derived soil conditioner has the combined effects of natural organic and mineral conditioners, which can be used not only for light soils with poor organic matter and nutrients (NPK), but also as a fertilizer (supply mainly Si, Ca, Mg and trace elements) or to restore degraded soil (Garbowski et al. 2023; Long et al. 2019). Previous studies have documented the excellent efficacy of coal gangue in optimizing soil structure, enhancing soil water retention, augmenting soil fertility and stimulating plant growth (Babla et al. 2022), as detailed in Table 3.

Furthermore, numerous scholars have explored the impact of the amount and particle size of raw coal gangue on soil properties, nutrient content and plant growth characteristics. The research findings of Zhou et al. (2010) demonstrated that as the addition of coal gangue increases, both the infiltration rate and saturated water conductivity decrease while the water retention performance of sand improves. Nan et al. (2023) revealed that coal gangue can alter soil moisture and aggregate structural stability. They identified that coal gangue content is the primary driving factor influencing soil moisture and plant growth, whereas particle size is the main determinant affecting soil aggregate structural stability, and the optimal coal gangue content is 30%, with a particle size ranging from 5 to 8 mm. Han et al. (2021) pointed out that the application of coal gangue mulch could enhance soil water status through increased water infiltration and reduced cumulative evaporation. Among different mulch models, the most effective one is characterized by a thickness ranging from 8 to

16 cm and particle sizes between 0.5 and 2 cm. The literature indicates that the typical upper limit for untreated raw coal gangue is approximately 20%.

In conclusion, the scientific and rational addition of raw coal gangue can effectively enhance soil properties and facilitate plant growth. Moreover, the harmless treatment of coal gangue prior to application can further optimize its utilization efficiency and improve the overall soil improvement effect.

Problems of direct application of raw coal gangue

The bottleneck problem of directly applying coal gangue as a soil conditioner has emerged, thereby impeding its further development. The relatively stable chemical structure and inert organic carbon of coal gangue result in less active beneficial components, thereby limiting direct absorption of nutrients by plants and microorganisms, leading to poor fertilizer efficiency when utilized as a fertilizer (Zhu et al. 2022). Additionally, the coal gangue with large particle size and low capillary porosity makes it resistant to weathering, which leads to soil structure loosening when directly applied, thus hindering its functions in water holding, fertilizer retention, and internal nutrient release (Du et al. 2020; Wang et al. 2016a). Consequently, it fails to effectively improve degraded soil such as sandy soil. Moreover, the addition of overmuch coal gangue will lead to excessive accumulation of trace elements, sulfur, and salt in the soil, causing environmental pollution and impeding plant growth (Singh et al. 2010). The long-term weathering and rain leaching of coal gangue will generate acidic wastewater, causing the release of toxic and harmful elements and subsequent contamination of soil and underground water (Gao et al. 2021b; Li and Wang 2019). The maximum feasible amount of coal gangue addition can be increased if the harmful substances in it are effectively controlled (Clavier et al. 2020).

To sum up, the direct application of coal gangue to improve degraded soil presents challenges such as low activity of beneficial ingredients, poor water and fertilizer retention performance, environmental pollution, etc., resulting in limited efficacy, restricted application amount and hidden safety risks. Therefore, it is crucial to thoroughly analyze the activity, structural characteristics and pollutants contents present in coal gangue prior to application. Additionally, considering the specific requirements of soil and crops, a rational modification treatment is imperative for producing high-quality and eco-friendly soil conditioners. This approach aims at achieving the harmless resource utilization of accumulated coal gangue while promoting sustainable agricultural development efficiently.

Table 3 Raw coal gangue-based soil conditioners and their effects on soil properties, plant growth and development (CG—coal gangue, EC—electrical conductivity, OM—organic matter)

Used as	Application rate (% w/w)	Plant species	Claimed effects on soil	Claimed effects on plants	Ref
Fertilizer and soil amendment	50%	<i>Alfalfa</i>	The levels of soil OM, total N, total P, total K, available N and available P exhibited significant improvements	The germination of plants initially increases and then decreases with the increase of CG. A moderate addition of CG significantly enhances plant growth (including plant height, root length, fresh and dry weight)	(Du et al. 2020)
Fertilizer and soil amendment	20%	<i>Lyme grass</i>	The soil exhibits enhanced pore structure, water and fertilizer retention capabilities compared to natural soil	The promotion of plant survival and growth was observed, with a 10% increase in seedling emergence rate and an average plant height increase of 53.65%	(Su 2021)
Fertilizer and Soil amendment	10%	<i>Vetiveria zizanioides</i>	It not only improves pH, EC (12.6%), OM (222.7%), N (52.1%), P (55.1%) and K (57.7%) content, but also reduces the bio-availability and mobility of most heavy metals (Zn, Pb, Cd, Cu)	Biomass and plant height increased by about 16.12% and 19.24%. Zn, Pb, Cd and Cu concentrations decreased by about 27.47%, 37.64%, 40.54% and 28.11%, respectively, and Cr concentration increased by 45.30%	(Chu et al. 2020)
Fertilizer and Soil amendment	10–50%	<i>Forage maize</i>	With increasing coal gangue percent in the soil, the amounts of soil OM, total N, P, Zn, Fe, available P, available K, microbial activity, soil microbial population and respiration increased	Plant nutrient content increased significantly. The increase of CG to 10% resulted in a significant augmentation of root and ground dry weight, whereas higher CG content (> 20%) led to a decline in both root and ground dry weight	(Moteszarezhadeh et al. 2017)
Soil amendment	26%	<i>Pachoi</i>	Improve the physical and chemical properties of soil and increase the nutrients required for plant growth process	The germination rate, plant height above ground, and dry weight increased by 23%, 23.05%, and 20.62% respectively. Additionally, the product met the requirements for cleanliness	(Luo et al. 2015)
Soil amendment	20%	<i>Medicago sativa</i>	The EC, pH and alkali-hydrolyzed nitrogen content of saline-alkali soil exhibited a decrease, whereas the contents of total C, total N, total P, and available P demonstrated an increase	The average plant height and biomass exhibited a significant increase of 21.45% and 25.89%, respectively	(Zhang et al. 2021b)
Soil amendment	20%	<i>Alfalfa</i>	Soil microbial biomass, urease, catalase and dehydrogenase activities showed an increasing trend	The plant height and fresh weight of alfalfa were significantly increased by 34.86% and 45.28% respectively	(Kong et al. 2018)
Soil amendment	1–10%	<i>Tomato</i>	The soil OM content can be significantly enhanced, while the soil pH can be regulated effectively. It can also promote soil aggregation and enhance microbial activity in the soil	The germination rate of tomato seeds was enhanced, and the levels of K, Ca, and Mg in tomato leaves were elevated	(Yong et al. 2022)

Table 3 (continued)

Used as	Application rate (%, w/w)	Plant species	Claimed effects on soil	Claimed effects on plants	Ref
Soil amendment	40%	<i>Ryegrass</i>	The soil enzymes were more active, microbial diversity increased, and organic matter utilization and PAHS degradation were promoted	The chlorophyll content and peroxidase activity were increased by 21.69% and 62.44% respectively	(Zhou et al. 2023)
Soil amendment	1–10%	-	Soil dispersity is restrained, and water stability of dispersed soil is improved, making it change from dispersive to non-dispersive. The soil compressive strength and tensile strength were increased by 591% and 192% respectively	-	(Zhao et al. 2023a)
Soil amendment	15–60%	-	The permeability is reduced by 30–80%, the saturation water conductivity is decreased by 25–60%, the dispersity is increased by 130–760%, and the flow water component experiences a reduction of 12–31%	-	(Zhou et al. 2010)

The modification methods of coal gangue

The raw coal gangue requires appropriate modification to improve its performance, activate its beneficial ingredients, enhance its water and fertilizer retention, and mitigate its pollution and biological toxicity. Therefore, this chapter provided a comprehensive overview of the current research status on coal gangue modification technologies. It further examined the impact of various modification methods on soil improvement-related indicators and conducts a comparative analysis to identify their respective advantages and disadvantages.

At present, the common modification methods of coal gangue include mechanical modification, chemical modification, microbiological modification and thermal activation (Han et al. 2022; Lv et al. 2022; Zhang and Ling 2020). As an advanced modification method, hydrothermal method has garnered significant attention in recent years (Chao et al. 2022; Lachos-Perez et al. 2022; Munir et al. 2018).

Mechanical modification

Mechanical modification refers to the physical and chemical changes of the material under the action of mechanical forces, causing the particle size reduction, specific surface area enhancement, particle structure destruction, fracture of inter-particle hydrogen bonds and Al–O–Si bonds of kaolinite, thus promoting the defects or displacement of the lattice or even the amorphous state, and achieving the activation of beneficial elements (Said et al. 2018). Mechanical grinding was employed to modify the structure and enhance the pozzolanic reactivity of coal gangue (Zhao et al. 2022b). It has been observed that during the progressive grinding process, the coal gangue particles undergo sequential stages of particle size reduction, particle agglomeration and aggregate dispersion (Guo et al. 2016), leading to gradual improvements in both dehydroxylation degree and activity of coal gangue (Guo et al. 2009). The ball mill can transform Muscovite into amorphous state at 600 rpm, which significantly enhances the dissolution rate of silicon from 0.23% to 16.05%, as well as boosts the dissolution rate of potassium from 2.62% to 81.39%, rendering it suitable as a silica-potash fertilizer (Liu et al. 2020b). The mechanical modification is characterized by its simplicity of operation and low cost; however, it exhibits limited functionality and activation effectiveness, which fails to eliminate the pollution risk associated with coal gangue. Therefore, it is commonly combined with other modification technologies to enhance activation efficiency and mitigate pollutants (Fig. 3).

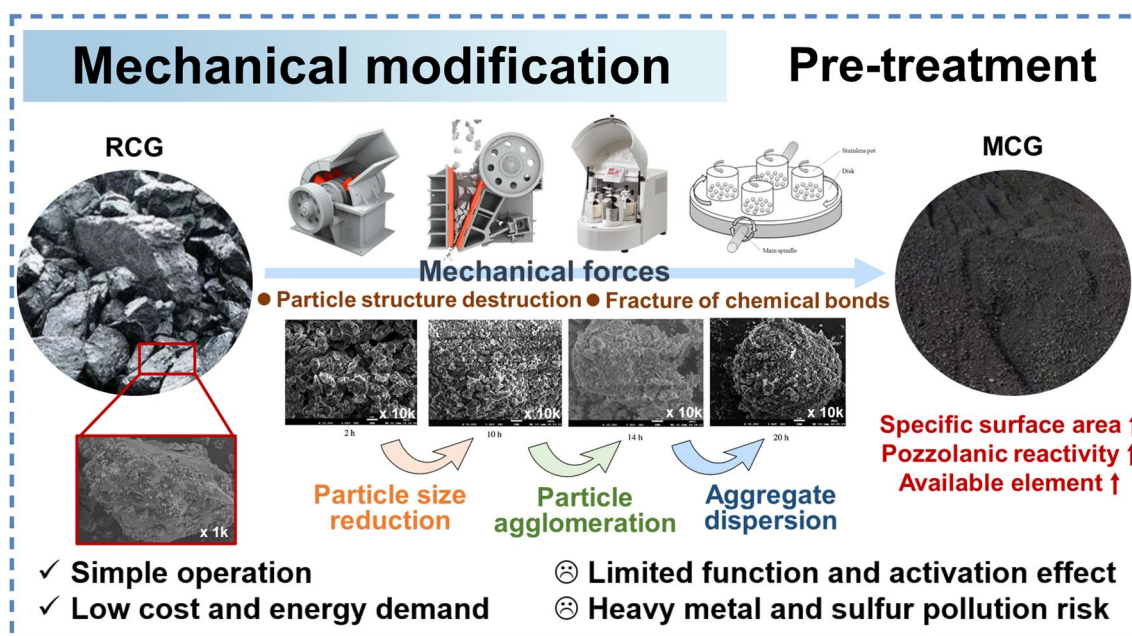


Fig. 3 The process, principle, effects and defects of mechanical modification (Guo et al. 2016; Liu et al. 2020b; Zhao et al. 2021b)

Chemical modification

The chemical modification is to modify the coal gangue with alkaline solution (such as Na_2CO_3 , NaOH , KOH) or acidic solution (such as HNO_3 , HCl). This modification disrupts the chemical bonds and alters the crystal structure and surface activity of coal gangue. Consequently, there is a significant enhancement in pore size distribution, active sites, and adsorption performance of coal gangue (Gao et al. 2015; Qian and Li 2015). As a result, the preparation of functional materials, the removal of impurities and the extraction of valuable components can be realized. Valuable elements can be extracted from coal gangue through heating and HNO_3 leaching, with extraction efficiency of 95.2%, 56.4% and 80.5% for Al, Ga and Li respectively. Additionally, the leaching residue possessed a large BET surface area and high available silicon content that can be utilized as silicon fertilizer (Shao et al. 2022). Alkaline solution destroyed the skeleton of silicoaluminates, so that Si–O–Si and Si–O–Al bonds quickly dissolved into the solution to form $[\text{SiO}_4]^{4-}$ and $[\text{AlO}_4]^{5-}$, thus enhancing gel performance (Cao et al. 2022). The addition of CaO facilitated a reaction between SiO_4^{4-} and AlO_4^{5-} with alkaline $\text{Ca}(\text{OH})_2$, yielding calcium silicate hydrate (C–S–H) and calcium aluminate hydrate (C–A–H), further improving gel properties (Li et al. 2006; Zhang et al. 2021a). The surface of alkali-modified coal gangue exhibited a rougher texture and displayed a distinct pore structure, whereas the acid-modified coal gangue merely present numerous grooves on its surface (Guo et al. 2021). In general, alkalinity is mostly used to activate the gel

properties of coal gangue, while acidity is mainly employed to leach rare earth elements and heavy metals, as well as improve pore structure and adsorption properties simultaneously. Chemical modification entails a straightforward operation with limited activation effects, typically combined with thermal activation (Fig. 4).

Microbiological modification

The metabolic processes of microorganisms are harnessed in microbiological modification to degrade minerals present in coal gangue, facilitating the release of nutrient elements and ultimately obtaining microbial fertilizers (He 2010). The study conducted by Bi et al. (2019) has proved the beneficial role of both arbuscular mycorrhizal fungi (AMF) and phosphate solubilizing bacteria (PSB) in the decomposition of organic matter and subsequent release of nutrients. PSB had the function of converting organic and inorganic phosphorus into soluble forms, which sensibly improved the availability of phosphorus in minerals and promotes plant growth (Benbrik et al. 2020). Silicate bacteria exhibited selectivity towards silicate minerals with distinct crystal structures, obviously promoting the dissolution of silicon and potassium (Lv et al. 2020). Moreover, scholars successfully isolated a strain of *Stenotrophomonas maltophilia* YZ1, which effectively dissolved nutrients and fix Pb^{2+} in coal gangue, thereby prominently increasing the content of available phosphorus, potassium, silicon and reduce the lead released (> 91.1%) in the modified coal gangue (Zhu et al. 2022; Zhu et al. 2023).

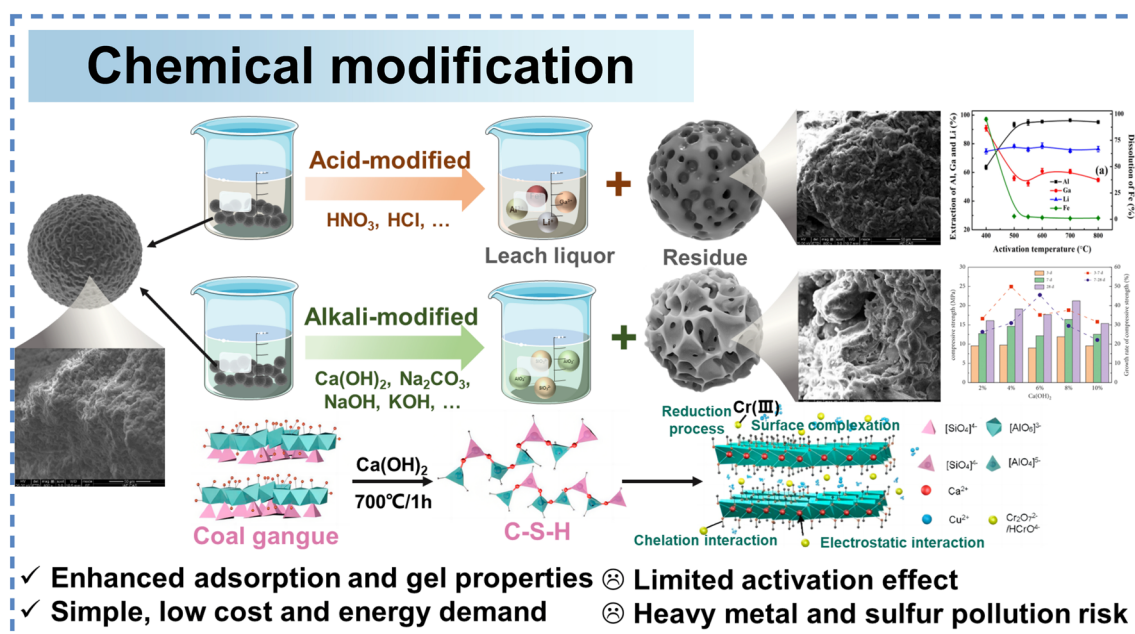


Fig. 4 The process, principle, effects and defects of chemical modification (Cao et al. 2022; Guo et al. 2021; Zhang et al. 2021a)

Organic acids produced through microbial metabolism can effectively enhance the solubilization and activation of mineral nutrient elements (Rezakhani et al. 2019), as well as the adsorption and immobilization of heavy metal ions (Yuan et al. 2017). Therefore, the screening of appropriate and efficient strains is anticipated to facilitate nutrient activation in coal gangue while mitigating its pollution. Due to the intricate composition of coal gangue, its microbiological modification necessitates the screening of a diverse range of functional bacteria for synergistic compounding, which is a complex and time-consuming endeavor and crucial challenge. However, it is noteworthy that the microbiological approach offers advantages such as low energy consumption, environmental friendliness and remarkable efficacy, making it deserving of extensive attention and research (Fig. 5).

Thermal modification

According to the literature, thermal activation is the most normally employed method for modification, which is utilized to enhance the activity of beneficial elements through breaking down polymerized long chains of silico-oxygen tetrahedrons and alumino-oxygen octahedrons at high temperatures (Jabłońska et al. 2017). The key factors influencing thermal activation efficiency are temperature and time. The reported optimal temperature and time for achieving maximum activity of coal gangue vary in different literature sources, which could be attributed to disparities in the composition and structure of the coal gangue (Arribas et al. 2018; Zhao et al. 2022b). It is noteworthy that the optimal

activation temperature for a specific clay mineral may induce recrystallization in another mineral (Zhang and Ling 2020). The presence of clay minerals in coal gangue renders it a potential pozzolanic material upon thermal activation at temperatures up to 600–800 °C. With the temperature rising from 100 °C to 900 °C, coal gangue experiences dehydroxylation and structural transformation, causing a significant enhancement in reactivity and a gradual increase in available silicon content (Li et al. 2016; Lv et al. 2022). The uncalcined coal gangue has a scaly-like layered structure that remains unchanged below 500 °C and becomes porous and irregular at 600–800 °C. This transformation may be caused by the phase transition of metakaolin, dehydroxylation of kaolinite, as well as the reduction of bound water and organic matter (Cao et al. 2016). Excessive calcination above 950 °C causes the active amorphous material to recrystallize into stable mullite, severely reducing the reactivity (Frías et al. 2012).

In addition to conventional calcination, microwave thermal activation has gained increasing attention. The micro-morphology and micro-aggregate effects induced by microwave activated coal gangue conferred it with strong hydrophilicity, water fixation and water retention capabilities (Qiu et al. 2022). Besides, microwave thermal activation altered the mineral composition and gelling properties of coal gangue. The microwave-thermally activated products can serve as auxiliary gelling materials that are conducive to the formation of micro-aggregates (Guan et al. 2021). Thermal activation is an effective, simple and feasible approach for industrial applications; however, its implementation is

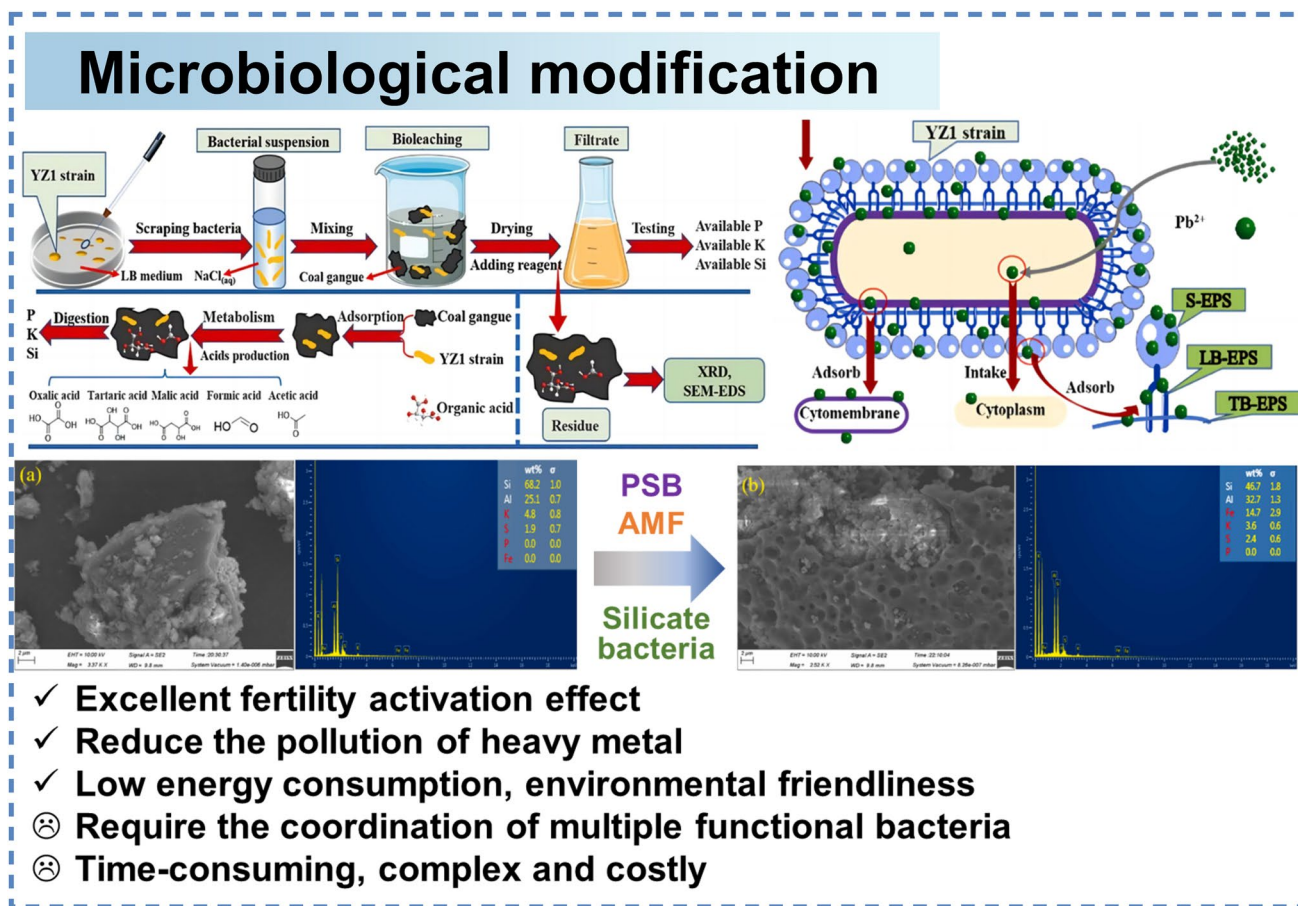


Fig. 5 The process, principle, effects and defects of microbiological modification (Zhu et al. 2022; Zhu et al. 2023)

hindered by its high energy consumption. Additionally, carbonaceous minerals and organic carbon are oxidized and emitted as greenhouse gases during the high-temperature activation process (Huang et al. 2018; Wang et al. 2022b), resulting in a substantial loss of organic matter, and thus observably diminishing the soil improvement value of coal gangue (Fig. 6).

Hydrothermal modification

The hydrothermal modification process takes place within a specialized closed reaction vessel, utilizing an aqueous solution as the reaction medium. Under high temperature and pressure conditions, a sub-critical water state is formed in the vessel (Kumar et al. 2018), promoting multiple conversions of coal gangue including hydrolysis, depolymerization, polymerization, isomerization, dehydration, aromatization, condensation, and other transformation reactions (Lachos-Perez et al. 2022). Because of its splendid activation effect, milder operating conditions and faster reaction rate, it has been widely applied in the field of solid waste disposal and resource recovery. The key parameters for hydrothermal

modification of coal gangue include reaction temperature, residence time, pressure, solid–liquid ratio and reaction medium. Among these factors, temperature is the most critical determinant (Munir et al. 2018). When the temperature reaches the activation energy threshold for the reaction, chemical bond are broken, resulting in a variety of reactions (Sabio et al. 2016).

The commonly employed reaction medium for hydrothermal modification of coal gangue typically involves an alkaline solution, which accelerates the fracture of Si–O–Si and Si–O–M (M: Al, Mg, or others) bonds (Zhao et al. 2023b), thereby enabling the synthesis of novel functional materials with diverse morphology and properties through hydrothermal condensation or gel reaction (Fan et al. 2023; Zhao et al. 2023b). In comparison with chemical modification, the surface of coal gangue appears a higher abundance of basic functional groups and micro-pores after alkaline hydrothermal modification, which brings an augmented specific surface area, a more extensive distribution of pore sizes, and a significantly enhanced adsorption capacity (Jin et al. 2022; Li et al. 2016). The latest research demonstrated that alkaline hydrothermal modification activated nutrients and

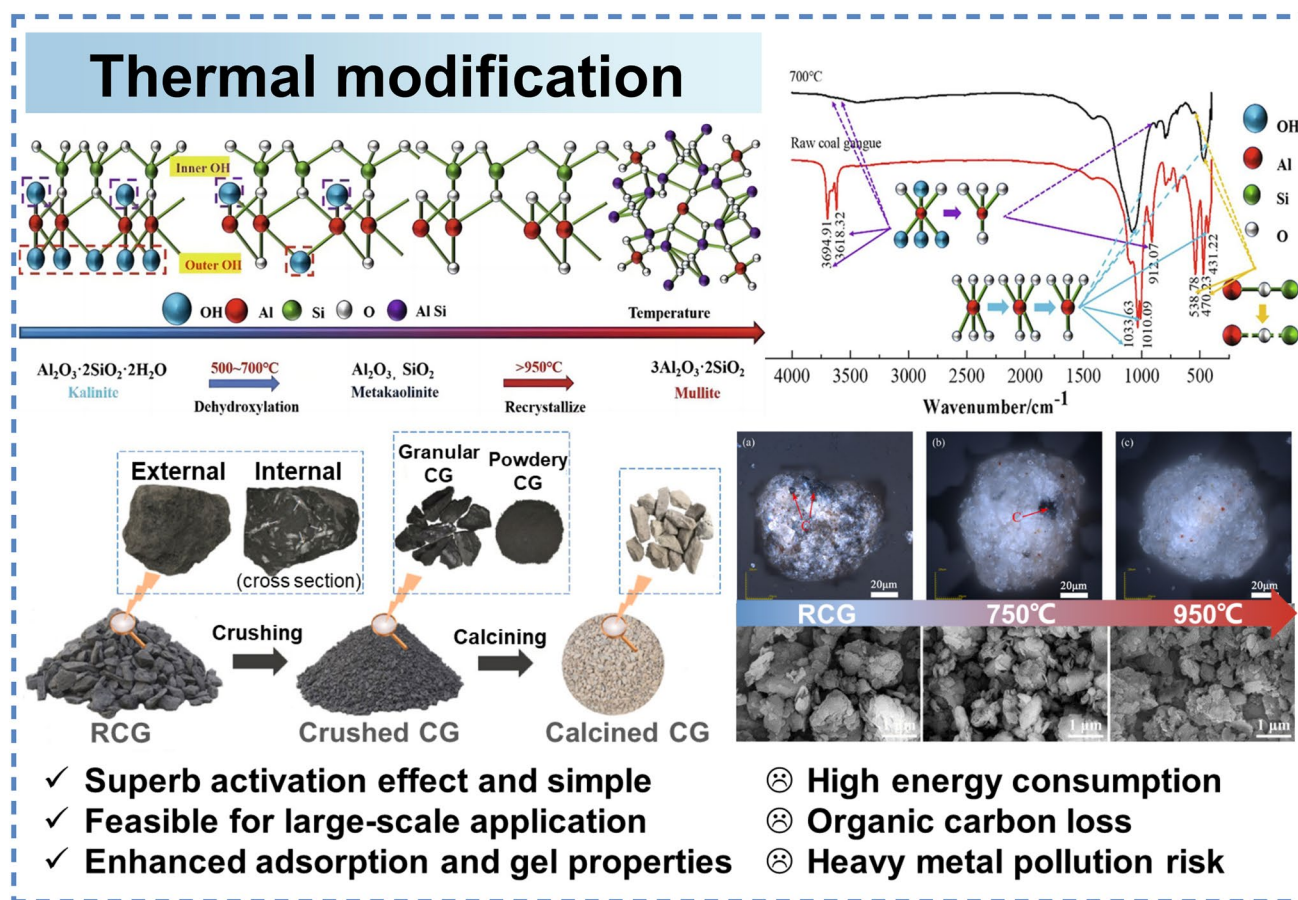


Fig. 6 The process, principle, effects and defects of thermal modification (Shao et al. 2022; Wang et al. 2022b; Zhao et al. 2022b)

removed pollutants in coal gangue simultaneously, yielding a highly active and eco-friendly silicon-based compound fertilizer (Tang et al. 2024). The subsequent sections provided a comprehensive account of the various specific functions associated with hydrothermal modification on soil improvement performance. Despite being a scientific, efficient, and versatile modification technology, the hydrothermal method still remains on laboratory or demonstration scale and has not yet been fully commercialized due to its reliance on expensive and intricate reactors (Munir et al. 2018) (Fig. 7).

Activate beneficial components

A vast body of literature has confirmed that alkaline hydrothermal treatment can effectively activate the beneficial components of silicaluminate minerals, and it is worth noting that red mud and coal gangue primarily consist of silicaluminate minerals. Chao et al. (2022) subjected red mud to hydrothermal conditions at 360 g/L K_2O and 240 °C for 1 h, resulting in solid phase effective K_2O and SiO_2 contents of 12.20% and 18.59%, respectively, meeting the market demand for multi-element compound fertilizer. The

excessively prolonged reaction time caused a decrease in the available silicon content in solid products, which was attributed to the partial overlap between the reaction conditions for aluminosilicate hydrothermal activation and zeolite hydrothermal crystallization, bringing about a tendency for structural stability and reduced activity during the transition from activation to crystallization conditions (Liu et al. 2022). The availability of nutrients in the hydrothermal process was closely correlated with the severity of the hydrothermal reaction. For example, the total phosphorus content in the solid phase increased as the degree of reaction improved, while nitrogen exhibited a significant exponential decrease (Wang et al. 2019). The hydrothermal reaction motivated the gradual degradation of polyphosphate and organophosphate into orthophosphate (inorganic phosphorus), which was more favorable for crop uptake, while ensuring relatively stable phosphorus leaching (Huang et al. 2017). The nitrogen in the raw material underwent polymerization or condensation reactions in the hydrothermal solid phase, forming more stable quaternary ammonium compounds (Xiao et al. 2017). Consequently, hydrothermal methods can be employed to

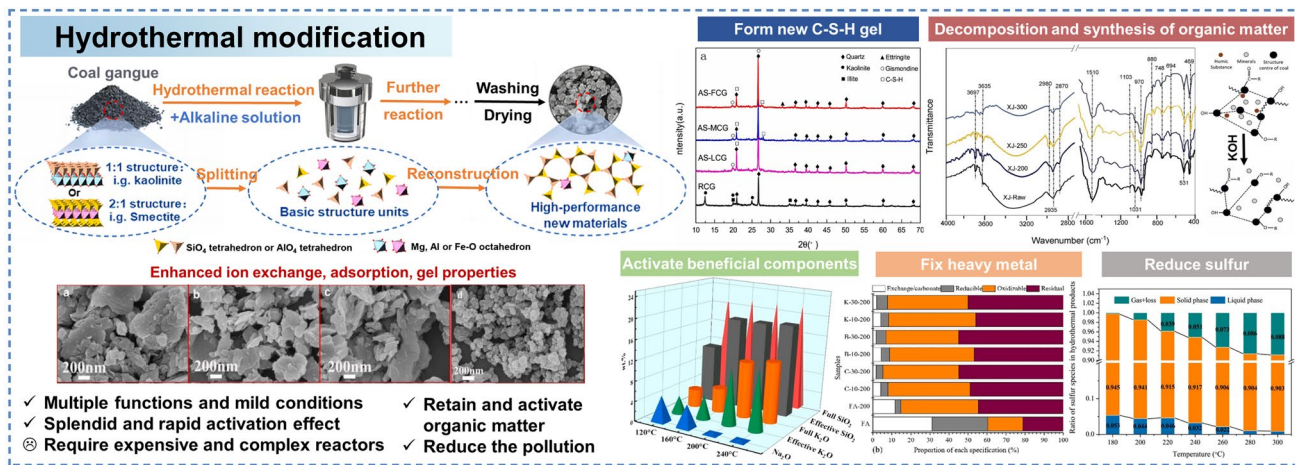


Fig. 7 The process, principle, effects and defects of hydrothermal modification (Chao et al. 2022; Wang et al. 2016c; Zhang et al. 2022b; Zhao et al. 2023b)

regulate the form and properties of nutrients, achieving a balance between plant absorption and soil fertility retention.

Improve the performance of coal gangue

According to the existing literature, the hydrothermal method is a viable way for removing inorganic ions from industrial solid waste and promoting the development of porous structures, while inducing alterations in mineral composition (Bayuseno et al. 2009; Cao et al. 2020). The high-quality zeolite with excellent properties, such as a high cation exchange capacity (CEC) and specific surface area, can be synthesized by alkaline hydrothermal methods. It is capable of stabilizing heavy metal ions and nutrients through cation exchange and adsorption (Li et al. 2020b; Li et al. 2014). Moreover, several literatures have stated the formation of new C-S-H and tobermorite phases from coal gangue after undergoing hydrothermal modification (Cao et al. 2022; Ye et al. 2022). The development of porous structure and clay minerals in these hydrothermally modified products can confer them with robust ion exchange, adsorption, and gel properties, which are positive for soil structure reconstruction, remediation of heavy metal pollution, and maintenance of soil nutrients.

Reduce the pollution of coal gangue

The application of hydrothermal modification has been revealed effective in the treatment of metal-contaminated soil, fly ash, and incineration bottom ash (Chen et al. 2020; Chen et al. 2019). Alkaline hydrothermal processes facilitated the conversion of silicon and aluminum into stable silico-aluminate minerals, thereby immobilizing heavy metals within the lattice structure and impeding their reactivation

(Bayuseno et al. 2009; Shi et al. 2017). In comparison to conventional methods such as chelating agent stabilization and cement fixation technology, hydrothermal treatment offers simultaneous stabilization of diverse heavy metals, along with advantages in terms of simplicity, rapid, efficiency, as well as environmental and economic benefits (Hu et al. 2015; Zhang et al. 2022b). Not only that, hydrothermal modification triggered the destruction, decomposition and transformation of organic sulfur, thereby elevating the stability of sulfur structure in hydrothermal products (Chen et al. 2020; Wang et al. 2016c), which efficiently mitigated soil sulfur pollution and alleviated plant growth inhibition caused by excessive available sulfur in coal gangue.

Maximize the utilization of organic matter in coal gangue

Coal gangue is characterized by a high proportion of stable recalcitrant organic matter (ROM) (Bi et al. 2020). The organic matter underwent a complex process of decomposition and synthesis during hydrothermal processes (Hou et al. 2022). However, the structure and compositional changes of organic matter in coal gangue during the hydrothermal process have not been elucidated. Subsequent research is essential to focus on investigating suitable conditions to regulate the proportion of two organic matters with distinct functions, in order to ensure a sufficient supply of stable macromolecules ROM for promoting the formation of soil aggregates (Qin et al. 2021), and an appropriate labile organic matter (LOM) that can be directly absorbed and utilized by plants and microorganisms (Muqaddas et al. 2019). Additionally, coal gangue contains stable organic matter and humus that can be solubilized in an alkaline hydrothermal environment (Kappler and Brune 1999; Sriramoju et al. 2022). In the future, it is also

considered to extract humus from the hydrothermal liquid phase to further improve the added value of coal gangue.

Combined modification

In practical applications, a combination of multiple modification methods is recommended to achieve optimal effectiveness and maximum benefits. Combined modification methods, such as chemically assisted mechanical milling (Zhao et al. 2021b), thermal activation coupled with chemical modification (Dong et al. 2023), mechanical pulverization combined with microbiological modification (Zhu et al. 2022), chemical reagent facilitated hydrothermal treatment (Jin et al. 2022), hydrothermal method coupled with chemical, mechanical and thermal modification (Li et al. 2010; Li et al. 2020b) have attracted a lot of attention. The combined technology can effectively address the limitations associated with a single technology; however, it necessitates intricate and multifaceted operational procedures.

Potential applications of modified coal gangue

The soil conditioners derived from different modification methods of coal gangue exhibit diverse properties, showing varying degrees of enhancement in soil improvement performance and safety. The potential applications of modified coal gangue soil conditioner are thoroughly analyzed in this chapter.

Used as fertilizers to supply nutrients

The modified coal gangue derived-soil conditioner is anticipated to exhibit enhanced efficacy as a fertilizer compared to the raw coal gangue, owing to the activation of beneficial components. It generally serves as silicon fertilizers, micro-nutrient fertilizers, as well as nutritious supplementary.

The modified coal gangue exhibits a significantly elevated effective silicon content (Chao et al. 2022; Liu et al. 2020b), rendering it suitable as a silicon fertilizer to mitigate transpiration loss, enhance nutrient utilization efficiency, and augment plant resilience against abiotic and biotic stresses, thus enhancing the survival ability of plants in extremely harsh environments (Castro and Crusciol 2013). Furthermore, It has been confirmed that supplementing available silicon can remarkably upgrade soil water availability and holding capacity (Kuhla et al. 2021), enhance soil microbial community and mitigate heavy metal accumulation in plants (Wang et al. 2020; Zhao et al. 2022a).

Micronutrient deficiency in soil may impede agricultural productivity and affect human nutrition either directly or indirectly (Steinnes 2009). The modified coal gangue

contains an abundant amount of available micronutrient. Taking selenium as an example, the activation rate of modified coal gangue towards selenium reaches a remarkable 81.24% (Liu et al. 2020a). In China, it is estimated that approximately 72% of the territory suffers from selenium deficiency. The application of selenium fertilizer derived from modified coal gangue in soil will contribute to enhancing crop resistance and ensuring optimal crop yield in saline-alkali land and arid areas (Long et al. 2019).

Besides, the modified coal gangue can function as a multi-nutrient supplementary, providing moderate available organic matter, N, P, K for the poor soil. While ensuring the provision of essential nutrients for plant and microorganism growth, it effectively prevents nutrient loss. Additionally, modified coal gangue possesses the capacity to absorb nutriment while maintaining them molecular state, thereby facilitating their absorption by crops and effectively improving nutrient utilization rates (Wang et al. 2021a; Zhou and Shan 2008).

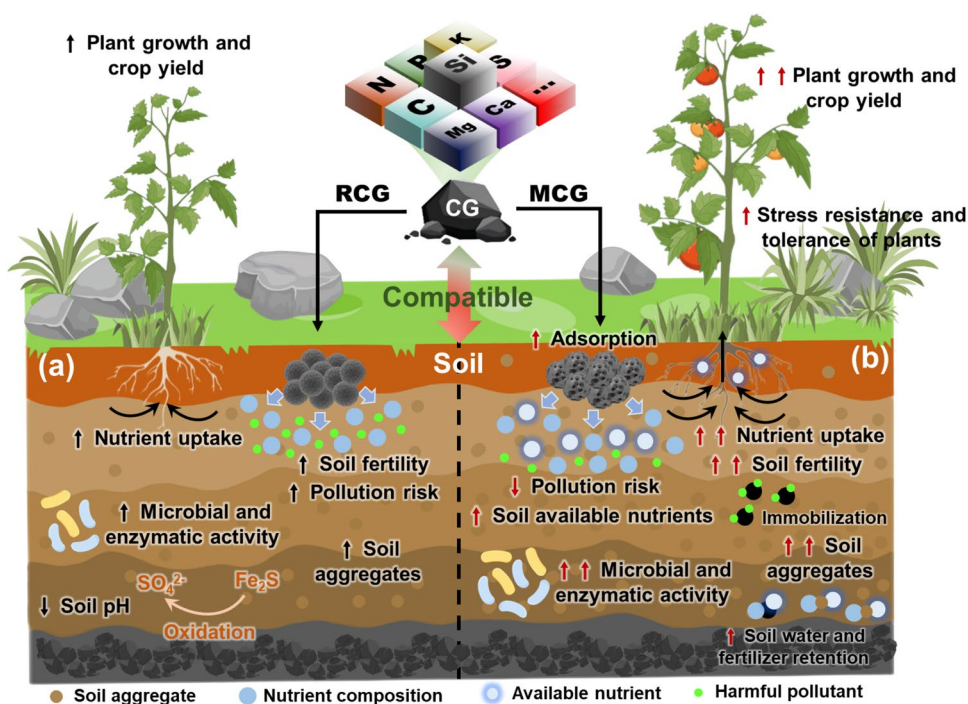
Used as soil amendments to improve degraded soil

The modified coal gangue is expected to serve as an eco-friendly soil amendment with exceptional soil improvement performance, primarily attributed to its enhanced pore structure, water retention capacity, fertilizer retention ability, and environmental safety, as well as remarkable ion exchange, adsorption, and gel properties.

The addition of porous modified coal gangue can improve soil structure, which is conducive to nutrient absorption and respiration of plant roots, and significantly boost the biomass of soil microorganisms (Ananyeva et al. 2013). The abundance of soil microorganisms plays a pivotal role in effectively increasing soil humus content, maintaining the dynamic transformation of nutrients, improving soil fertility, and ultimately promoting plant growth (Blagodatskaya and Kuzyakov 2013).

In addition, the properties of modified coal gangue make it applicable for the remediation of sandy soil and heavy metal-contaminated soil. The activated organic matter in modified coal gangue serves as the foundation for soil aggregate formation, thereby influencing soil structure and porosity and subsequently altering water holding capacity and infiltration capacity of the sandy soil (Gao et al. 2016). After the application of modified coal gangue, the effective Cd content in soil decreased by 21.2–33.9% (Zhao et al. 2021a). The coal gangue-based material modified by (Chen et al. 2023) effectively reduces the available As and Cd by 17.94–29.81% and 14.22–30.41%, respectively. These findings suggest that modified coal gangue can reconstruct sandy soil function and immobilize heavy metals available, rendering it an ideal amendment for remediating sandy soil and co-contaminated soil with multiple heavy metal (Fig. 8).

Fig. 8 Intended benefits and unintended risks of (a) raw coal gangue (RCG)-based soil conditioners and (b) modified coal gangue (MCG)-based soil conditioners on soil properties, plant growth and development



Environmental consideration

The resourceful utilization of coal gangue as soil conditioners, particularly the modified coal gangue, not only facilitates the harmless and resource treatment of substantial quantities of coal gangue, but also realize the restoration of degraded soil, thereby yielding positive economic, social, ecological, and environmental benefits. Additionally, it plays an important role in carbon reduction. The spontaneous combustion, pollution, and soil degradation caused by centralized stacking coal gangue will lead to a decline in surrounding vegetation and reduction of carbon sink capacity. However, the utilization of coal gangue as a soil amendment or fertilizer can effectively restore degraded soil and improve stable ecological carbon sinks such as soil aggregates and phytic carbon (Liu 2023). Su et al. (2010) demonstrated that over the 7-year and 32-year recovery periods of sandy land, soil carbon sequestration in the 0–15 cm layer reached 1.8–9.4 Mg ha⁻¹ and 7.5–17.3 Mg ha⁻¹, respectively. Hence, modified coal gangue exhibits significant potential for enhancing soil quality and promoting soil carbon sequestration in desertification region. According to the carbon footprint analysis conducted by (Ashfaq et al. 2021), the procurement and transportation of raw materials exhibit the highest carbon emissions compared to other stages of utilization. Therefore, employing modified coal gangue for enhancing local degraded soil through in-situ utilization can yield significant reductions in carbon emissions.

It needs to be emphasized that although heavy metal levels in coal gangue generally remain below the threshold,

prolonged usage may result in the accumulation of heavy metals and pose a threat to crop safety. Current research primarily focuses on the influence of modification methods on coal gangue characteristics, with limited attention given to the impact of modified coal gangue on soil and plant growth. It is imperative to closely monitor the environmental impact of modified coal gangue throughout its cradle-to-grave life cycle and ensure both environmental protection and crop safety. In practical applications and subsequent research endeavors, an appropriate modification method should be selected to obtain modified coal gangue with matching performance based on specific local soil improvement requirements, thereby facilitating in-situ treatment of coal gangue while yielding positive environmental, economic, and social benefits.

Conclusions

As one of the largest industrial solid wastes in the world, coal gangue not only poses a pollution risk but also holds immense untapped potential for soil improvement that is often overlooked. The raw coal gangue is cost-effective, easily accessible and compatible with soil. Incorporating less than 20% of raw coal gangue typically optimizes soil structure, enhances soil fertility, and stimulates plant growth. The limited effectiveness and application amount of raw coal gangue are attributed to the low reactivity of beneficial components, inadequate water and fertilizer retention, as well as a high risk of pollution. The modification technologies,

including mechanical, chemical, microbiological, thermal, hydrothermal and combined modifications, are effective to enhance the soil improvement performance while mitigating pollution of coal gangue. This enables the acquisition of high-quality and pollution-free multi-functional soil conditioner. Among them, hydrothermal and combined modification stands out due to its comprehensive and effective functions, making it deserving of attention. The modified coal gangue not only serves as a multi-nutrient source, providing essential nutrients for the soil, but also acts as an effective soil amendment in rehabilitating degraded soils such as sandy soil, heavy metal-contaminated soil. This not only facilitates the conversion of coal gangue waste into valuable resources but also effectively rehabilitates degraded soil and fosters the establishment of a stable ecological carbon sink. This paper provided an innovative approach to achieving clean coal production and promoting sustainable agriculture.

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Data availability The authors declare that the data supporting the findings of this study are available within the paper and its Supplementary Information files. Should any raw data files be needed in another format they are available from the corresponding author upon reasonable request.

Declarations

Competing interests The authors have no relevant financial or non-financial interests to disclose.

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