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REVIEW

Migration transformation, prevention, and control of typical heavy metal lead in coal gangue: a review

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Abstract
Coal gangue is one of China's main industrial solid wastes, which contains various harmful heavy metal elements, such as lead (Pb). The long-term accumulation of coal gangue causes Pb to migrate to the surrounding environment due to weathering and rain erosion, eventually endangering human life and health with its continuous accumulation in the food chain. This review discusses the migration and transformation process of Pb in coal gangue under different conditions and summarizes the available forms and mechanisms of Pb in coal gangue. The current prevention and control and comprehensive utilization methods of Pb in coal gangue are comprehensively reviewed, and the characteristics of each method are discussed. In order to realize the economical, efficient, and high resource utilization of coal gangue, this paper provides favorable suggestions and support for subsequent in-depth research and the implementation of heavy metal prevention and control measures.

Keywords Coal gangue · Heavy metal · Lead · Migration transformation · Prevention and control

1 Introduction

With the growth of China's energy demand and national economy, the basic energy consumption of coal is increasing, and accordingly, the coal gangue. Coal gangue is the solid waste generated and discharged in coal mining and mineral processing, accounting for 10%–15% of the raw coal production. At present, the reserves of coal gangue have exceeded 7 billion tons in China, primarily occurring in China's bulk industrial solid waste emissions (Zhao et al. 2022; Ma et al. 2021). Compared with other bulk industrial solid waste, the comprehensive utilization rate of China's coal gangue is lower, only accounting for 58.9% (China Resource Recycling Association 2021). A large quantity of coal gangue is stored around the coal mines, and various heavy metal elements in coal gangue will inevitably lead to their migration under the influence of the external environment, such as weathering erosion and rainwater leaching (Gao et al. 2021). The migration of heavy metal elements will not only cause serious environmental pollution but will also accumulate in the food chain, eventually affecting human life and health (Gu and Gao 2017).

In China, coal gangue is commonly disposed of in piles and filling reclamation. Du (2022) pointed out that China's coal gangue is still disposed of in piles as the current disposal method, while future development will aim to comprehensively use landfills combined with reclamation backfill as the disposal method. Li et al. (2021c) proposed that the future disposal of coal gangue should be in line with the ecological civilization construction conditions to promote comprehensive technology use with optimal application and efficiency and low cost. In this paper, the authors summarized the typical heavy metal elements in the soil surrounding different mining areas nationwide (Table 1). It was found that the heavy metal content in the soil surrounding different mining areas and the harm to the environment differed.

Among the heavy metal elements, zinc (Zn) and chromium (Cr) are much greater impact than other heavy metal elements, which has attracted widespread attention. In addition, the minimum Cd content has been studied more extensively due to its high toxicity compared with other heavy metals. However, the maximum, minimum, median, and average lead (Pb) content and coefficient of variation are the midpoints of all heavy metal elements. Pb is one of the five heavy metal elements that are controlled by the Ministry of Ecology and Environment, and the associated mineral

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Table 1 Typical heavy metal element-related contents (mg/kg) and coefficient of variation in soils around mining areas nationwide

Place of origin	Pb	Cu	Cr	Zn	Cd	References
Panzhihua Mining District, Sichuan Province	40.39	48.67	40.25	249.29	0.79	Zhang et al. (2022b)
Yongding Mining District, Fujian Province	34.38	32.04	96.52	73.95	0.13	Lin (2021)
Yangshuo Mining District, Guangxi Province	46.25	68.32	61.97	78.54	3.08	Chen et al. (2022)
Wudang Mining District, Guizhou Province	49.20	31.25	80.50	78.23	0.34	Zhang et al. (2022a)
Pingdingshan Mining District, Henan Province	30.57	24.05	56.24	70.79	0.17	Yang et al. (2019)
Ganan Mining District, Jiangxi Province	106.05	12.02	28.40	67.65	1.13	Qiao et al. (2022)
Tongguan Mining District, Shanxi Province	164.50	37.15	63.30	83.70	0.61	Wang et al. (2022b)
Suizhou Mining District, Anhui Province	16.76	33.57	66.51	63.79	0.39	Su et al. (2021)
Bozhou Mining District, Anhui Province	25.94	31.50	77.75	73.48	0.27	Li et al. (2021a)
Baisan Mining District, Jilin Province	25.78	23.44	72.73	94.06	0.18	Luo et al. (2021)
Zhejiang Province, West Zhejiang Mining District	33.95	70.34	80.14	185.54	2.58	Wang (2021)
East Yunnan Mining District, Yunnan Province	25.99	118.88	206.73	165.39	0.51	Tu et al. (2022)
Xingren Mining District, Guizhou Province	220.23	112.29	180.97	98.47	0.95	Wang et al. (2022c)
Muli Mining District, Qinghai Province	25.07	23.19	68.41	83.19	0.15	Xu et al. (2022)
Lushan Mining District, Shandong Province	23.45	23.23	68.41	83.19	0.15	Li and Li (2018)
Maximum value	220.23	118.88	206.73	249.29	3.08	
Minimum value	16.76	12.02	28.40	63.79	0.13	
Median value	33.95	32.04	72.34	78.54	0.39	
Average content	57.90	46.00	83.52	102.16	0.76	
Coefficient of variation	1.03	0.71	0.57	0.53	1.18	

extraction and disposal industry is also key in the control of Pb. Therefore, more attention should be paid to the prevention and control of Pb in the field of coal gangue disposal.

As a typical heavy element, Pb can be released into the environment from coal gangue, involving the atmosphere, soil, surface water, and groundwater (Yu et al. 2016). Pb is a non-degradable element with long-term toxicity. It is an unnecessary trace element for human life activities and poses serious health hazards to human beings, including damage to the kidneys, liver, nervous system, and basic physiological cell processes (Rajendran et al. 2022). Severe Pb pollution has been found around many mining areas in China, resulting in abnormal blood Pb content, exceeding the standard, in children in the surrounding areas (Liu et al. 2022b). Furthermore, the heavy metal content in different regions of China is higher than the background value of soil. For example, the Pb content in three different areas of the Xiayukou mining area in Shanxi Province is at the severe exceedance level, surpassing the local soil background value of the three areas by 30, 25, and 20 times, respectively (Cui et al. 2021). From this paper, it can be concluded that the impact of excess Pb is quite serious around some mining areas in China.

This review used the Web of Science and Chinese National Knowledge Infrastructure databases and summarized the articles about heavy metals in coal gangue in the past five years (Fig. 1). The figure shows that although the number of articles about Pb in coal gangue in domestic mining areas has increased yearly in past few years, the proportion of Pb is still relatively small and less than 20%. However, studying the migration process of Pb in coal gangue is important for the prevention and control of this heavy metal in coal gangue. Based on the relevant literature, we further summarized the migration process of Pb in coal gangue, including both the occurring form of Pb in coal gangue and the migration mechanism of Pb into the

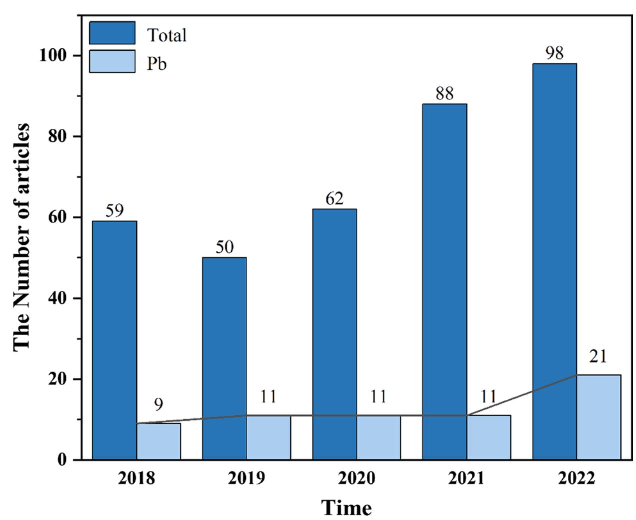

Fig. 1 The number of heavy metal articles related to coal gangue in recent 5 years

surrounding environment. Furthermore, the current methods and measures for the prevention and control of Pb in coal gangue are comprehensively reviewed, and the advantages and shortcomings of various methods are discussed to help and guide the improvement of prevention and control measures of Pb.

2 Migration process and effect of Pb in coal gangue

Figure 2 illustrates the migration process of Pb in coal gangue to the surrounding environment under different conditions. A large amount of coal gangue accumulates in the ground, which causes spontaneous combustion in certain conditions. Pb enters the atmosphere as a harmful gas generated due to spontaneous combustion, further deteriorating the air quality and eventually affecting human health through human respiration and direct contact (Wang et al. 2022a).

At the same time, weathering erosion and rainwater leaching cause Pb in the coal gangue to continuously migrate to soil and water. Pb can be preserved in soil and water in various forms through different physical and chemical processes so that Pb continuously accumulates in the soil surface layer and surface water (Ashfaq et al. 2020; Zhang et al. 2021b). Pb in soil is affected by underground seepage; Pb and soil inorganic salts will be continuously dissolved in water and released into the underground seepage, and infiltrated into the underground aquifer with the flow of water, causing environmental pollution of groundwater (Guo et al. 2021; Strugała-Wilczek and Stańczyk 2016). Since plant roots mostly grow in the soil, the roots will also absorb some Pb with the groundwater and salts, leading to the continuous enrichment of Pb in plants. The continuous accumulation of heavy metal elements in soil and water bodies promotes their enrichment in plants and animals in the food chain, and Pb will ultimately be transferred to the human body through the food chain, eventually causing a direct impact on human health (Sun et al. 2018; Zhang et al. 2021c).

3 Mode of occurrence of Pb in coal gangue

3.1 Classification of Pb in coal gangue

Many studies have shown that the toxicity and hazards of heavy metal elements depend not only on their total quantity but also on their available form (Dang et al. 2020). In 1979, Tessier et al. (1979) classified the available forms of heavy metals into exchangeable state, carbonate-bound state, iron (Fe)-manganese (Mn) bound state, organic-bound state, and residual state using the sequential extraction procedure. In

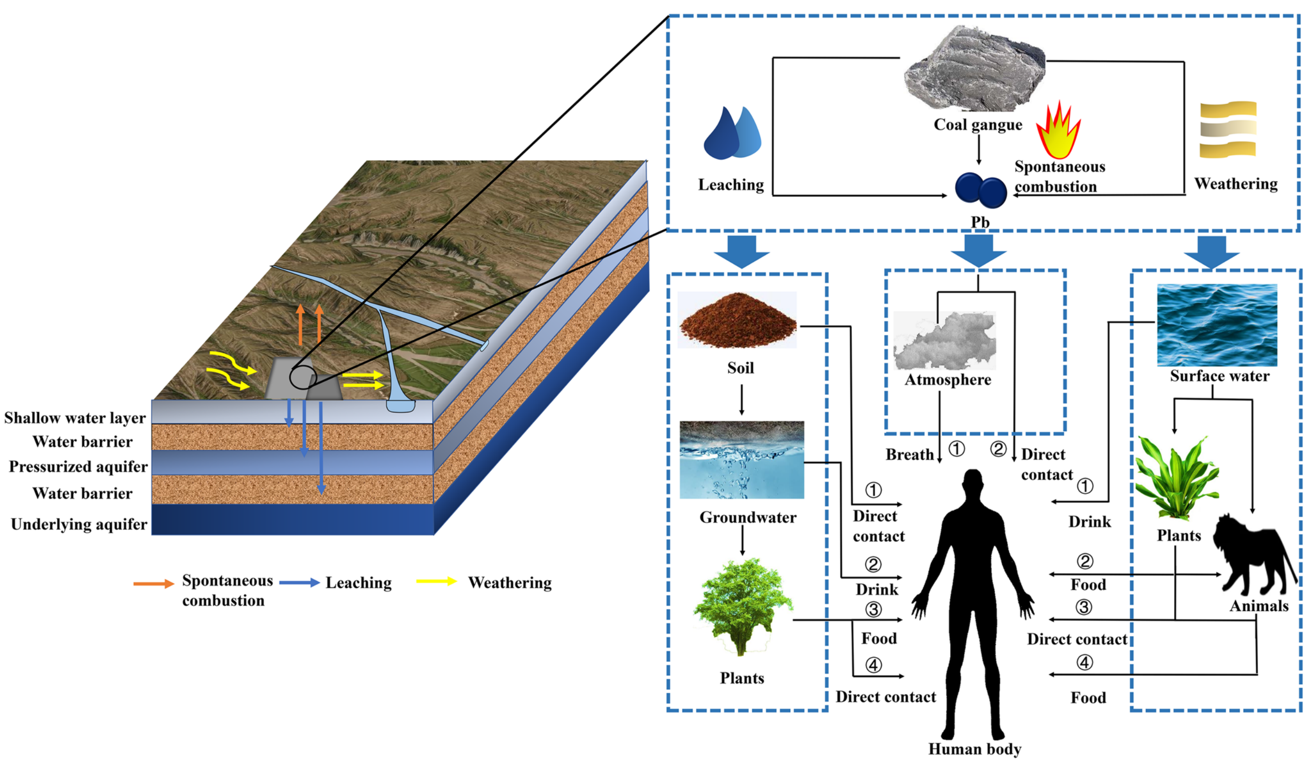

Fig. 2 Migration process and effect of heavy metal elements Pb in coal gangue

1992, the European Community Bureau of Reference (BCR) method proposed by the European Community Bureau of Reference Materials (Quevauviller et al. 1993) classified the morphology of heavy metals into exchangeable, reducible, oxidizable, and residual states. Although these two methods have been improved several times, they are controversial due to their different classification methods, resulting in different classification results (Liu et al. 2018). However, they are currently the most authoritative and widespread classification methods, in which the Tessier method has a more detailed classification of the available forms of heavy metals than the BCR method. The Tessier method was first used in soils and sediments for the classification of heavy metals, and in recent years, it has been applied to sludge, tailings, and coal gangue (Chen et al. 2020). Heavy metal forms classified by the Tessier method as exchangeable state, carbonate-bound state, Fe–Mn-bound state, and organic-bound state easily accumulate and cause pollution in the environment around the coal gangue due to the external environment, whereas the residual state is the most stable available form due to its stable chemical properties and does not easily migrate to the environment (Gutiérrez et al. 2016; Zhang et al. 2020). The available forms and transformation mechanism of Pb in coal gangue are shown in Fig. 3 and Table 2.

3.2 Exchangeable state of Pb

The exchangeable state of Pb in coal gangue usually occurs in the form of Pb^{2+}, $Pb(OH)^{+}$, and $Pb(OH)_4{}^{2-}$ under a neutral and weakly alkaline environment. Exchangeable

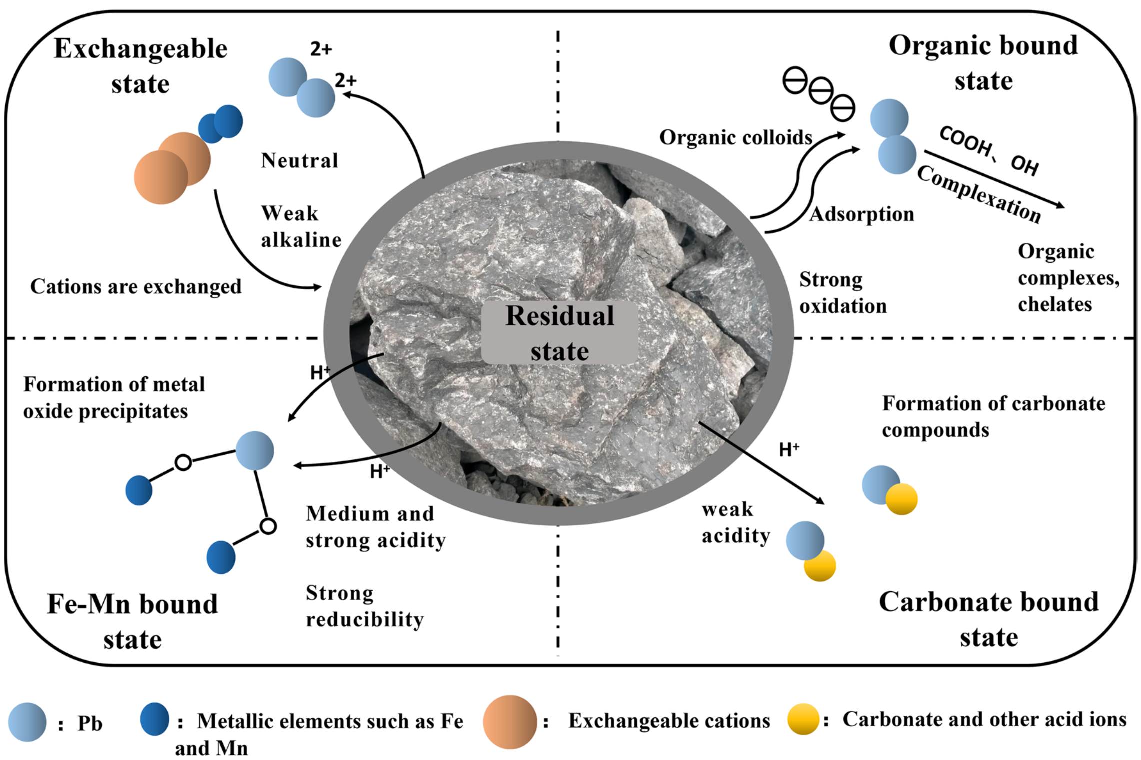

Fig. 3 Modes of occurrence and transformation mechanism of Pb in coal gangue

Table 2 Classification of migration transformation environment, compound form, valence state and activity intensity of Pb in different binding states in coal gangue

Speciation	Migration transformation environment	Compound form	Valence state	Activity
Exchangeable state	Neutral, weak alkaline	Pb^{2+}, $Pb(OH)^{+}$, $Pb(OH)_4{}^{2-}$	Pb(II)	Strongest
Carbonate bound state	Weak acidity	$PbCO_3$	Pb(II)	Strong
Fe–Mn bound state	Medium and strong acidity, strong reducibility	M–O-Pb, M–O-Pb–O-M	Pb(II)	General
Organic bound state	Strong oxidation	PbS, Complexes of Pb, Chelate of Pb	Pb, Pb(II), Pb(IV)	Weak
Residual state	Difficult to migrate and transform	Galena, Leucite	Pb, Pb(II), Pb(IV)	Weakest

state heavy metals exist in clay minerals on the surface of coal gangue. Under most conditions, these heavy metals can migrate, transform, adsorb, and exchange with ions. Moreover, they can easily precipitate in a neutral or weakly alkaline environment. This is the most active state of heavy metal elements in the environment (Huang et al. 2016). The exchangeable state of Pb can easily be replaced with cations of mineral components to form Pb^{2+} (Chen et al. 2017). Coal gangue contains quartz, kaolinite, calcite, feldspar, calcium alumina, and many other mineral components, among which silicon dioxide (SiO_2) has a high content. After weathering, the structure crystals of coal gangue change, and the Si–O bond in the SiO_2 breaks so that the negatively charged oxygen ions (O^{2-}) are exposed to the coal gangue surface in large quantities to form SiOH with OH^- and H^+ in aqueous solution after leaching by neutral or weakly alkaline rainwater. Furthermore, free O^{2-} has strong electronegativity, which can adsorb H^+ in an aqueous solution and generate strong OH^- or SiOH electronegativity (Zhang et al. 2021a).

As one of the mineral components in coal gangue, kaolinite is composed of silica-oxygen tetrahedra and aluminum-oxygen octahedra at a 1:1 ratio. Due to the negative charge of the special aluminum-oxygen octahedral lattice of kaolinite, the surface of kaolinite has strong electronegativity and can thus dissolve in water and react with Pb to precipitate Pb^{2+} (Won et al. 2019). Pb^{2+} reacts with OH^- on the surface of coal gangue to form compounds such as $Pb(OH)^+$ and $Pb(OH)_4{}^{2-}$ (Dai et al. 2020). Since the available form of exchangeable Pb is similar to the natural environment, the compound form is easily soluble in water, causing Pb to have higher mobility; thus, the activity of exchangeable Pb is greater.

3.3 Carbonate-bound state of Pb

The carbonate-bound state of Pb in coal gangue often occurs as lead carbonate ($PbCO_3$) in a weak, acidic environment. The carbonate-bound state is the chemical form of heavy metals formed by carbonate precipitation. The carbonate-bound state of Pb is greatly affected by pH. For example, heavy metals dissolve in the carbonate-bound state in a weak, acidic environment, such as that created by the short-term leaching of coal gangue (Yao et al. 2013). Therefore, mobility and activity increase in this bound state. When the coal gangue is in a weak, acidic environment for an extended period, most of the Pb will be oxidized to lead ion (Pb^{2+}) after weathering and weak acid rain leaching. A layer of lead oxide (PbO) is formed after further oxidation of Pb^{2+} with oxygen. Then, a more active $PbCO_3$ will be formed when PbO further reacts with carbon dioxide (CO_2) in the air (Liu et al. 2022a). As the coal gangue is affected by the acidic environment to different degrees in different geographical conditions, the content of carbonate-bound Pb is often affected by the gangue's particle size, location, and leaching area (Wang et al. 2018).

3.4 Fe–Mn-bound state of Pb

The Fe–Mn-bound state of Pb in coal gangue is often in the M–O–Pb and M–O–Pb–O–M forms in medium and strong, acidic, or strong reducing environments. The Fe–Mn-bound states are compounds formed by heavy metal elements bound on the surface of Fe–Mn oxides or with Fe–Mn oxides, Fe–Mn colloids, or a deposited compound formed by replacing Fe–Mn and other metal elements existing as M–O–Pb as M–O–Pb–O–M (Takahashi et al. 2007). The Fe–Mn-bound state is usually stable under relatively weak acidic conditions but will still migrate and transform under relatively strong acidic conditions or a strong reducing environment. Therefore, the Fe–Mn-bound heavy metals can be easily activated and show instability under moderately strong, acidic conditions (Xu et al. 2006). When coal gangue is leached by acid rain, Pb is often converted to exchangeable and carbonate-bound states in a neutral or weakly acidic environment due to the increased H^+ concentration. However, when the leached coal gangue is in a highly acidic environment, the reducibility in this environment is continuously enhanced due to the increasing H^+ concentration and the decreasing redox potential. The metal oxides with high activity and surface area are easily adsorbed to or co-precipitated with Pb to form metal oxides simultaneously. Pb^{2+} can bond with a single or multiple oxygen atoms, such as multiple bonded metal oxides M–O–Pb and M–O–Pb–O–M (Bo et al. 2017). Therefore, Pb adsorbed on the surface of Fe–Mn oxides can occur in the Fe–Mn-bound state in a reduced environment.

3.5 Organic-bound state of Pb

The organic-bound state of Pb in coal gangue often occurs in the form of PbS, Pb complexes, and Pb chelates under a strong, oxidizing environment. Organic-bound state refers to the type of complex and chelate formed when heavy metal elements react with organic matter, colloids, humic acids, and other organic substances in environmental sediments. It also includes the type of organic sediment formed by the adsorption and precipitation of organic sulfides (Odermatt and Curiale 1991). Organic matter and sulfide present in the soil have strong oxidation potential; however, Pb is a sulfurophilic heavy metal element type, which often exists in soil as PbS under strong, oxidizing conditions. Because of the poor mobility of Pb in soil, the Pb compounds in coal gangue have poor infiltration in soil, and most Pb is adsorbed in soil-solid phases such as organic matter, minerals, colloids, and other soil-solid phases (Zhang et al. 2023). In different valence states, Pb, Pb^{2+}, and Pb^{4+} adsorb to colloids due to their charge attraction and organic matter adsorption. Complexation and chelation reactions occur with carboxyl, phenolic hydroxyl, and alcohol hydroxyl groups in organic matter, and compounds exist in soil as Pb complexes and chelates (Löv et al. 2018; Zhou et al. 2020c). As Pb has a large molecular radius, it often complexes with multiple functional groups simultaneously, increasing the solubility of Pb and enhancing the adsorption capacity of organic matter, making it easier to transform Pb into a more stable organic-bound state.

3.6 Residual state of Pb

As the most stable available form in coal gangue, the residual state of Pb is highly stable and will rarely be released and migrate to the environment; thus, it has little impact on the environment. The residual state of Pb in coal gangue mainly exists as galena and leucite in the lattice of mineral composition, indicating that the residual state of Pb in coal gangue is highly stable in nature and extremely difficult to migrate and transform. As the most stable form of Pb, it can only be obtained under extreme conditions by mixing a strong acid and alkali for extraction (Bhuiyan et al. 2010; Hua et al. 2018). The residual state is the target form of research on the availability of heavy metal elements in coal gangue. Therefore, how to efficiently transform heavy metals in coal gangue from the exchangeable, carbonate-bound, Fe–Mn-bound, and organic-bound states into the residual state is a problem that needs to be solved in future research on heavy metal migration and transformation in coal gangue.

4 Prevention and control of Pb in coal gangue

According to the current disposal measures and properties of coal gangue, the following heavy metal prevention and control mechanism is proposed for coal gangue migration, as shown in Fig. 4. The availability and transformation mechanism of typical heavy metals in coal gangue proposed by Li and Wang (2019) show that the physical and chemical properties, composition structure, and surrounding environment of coal gangue will lead to the migration of heavy metals. Therefore, heavy metal migration, transformation, prevention, and control in coal gangue have been studied, and a series of methods for resource utilization of coal gangue and prevention and control of heavy metal elements have been proposed (Qin et al. 2022; Liu et al. 2021). These methods not only reduce the impact of coal gangue on the surrounding environment and human life but also face the long-term difficulty of dealing with coal gangue so that the limited resources in coal gangue have maximum value and coal gangue has minimal impact on the surrounding area, environmental risks, and human life.

4.1 Coal gangue-based adsorbent for prevention and control of Pb

In recent years, the synthesis of gangue-based adsorbents from coal gangue has become the focus of heavy metal prevention and control. Adsorbents are more suitable for the prevention and control of Pb in water and the atmosphere during its migration. Various comprehensive utilization methods have been adopted to prevent and control heavy

Fig. 4 Prevention and control measures and mechanism of Pb in coal gangue

metal elements in coal gangue, including modified adsorbent and coal gangue-based zeolite (Jin et al. 2022). However, due to the low adsorption capacity and immobilization effect of coal gangue on heavy metals, it is necessary to adopt different physicochemical techniques to activate and modify coal gangue to improve its adsorption ability to heavy metal elements. In addition, natural zeolite is also a common heavy metal adsorbent, whereas aluminum oxide (Al_2O_3) and SiO_2 in coal gangue can provide the main raw materials for zeolite synthesis (Zhou et al. 2020a).

4.1.1 Modified coal gangue

Modified gangue is a type of efficient and economical heavy metal ion adsorbent. The gangue is modified and activated by different activators and reaction conditions with a low activity to achieve specific adsorption conditions (Zhou et al. 2021). Shang et al. (2019) prepared a low-cost sulfhydryl-modified gangue using (3-sulfhydryl) trimethoxy silane to modify the gangue. Many sulfhydryl groups were successfully introduced into the gangue structure, and the maximum adsorption capacity of Pb was 332.8 mg/g. The modified gangue adsorbent with sulfhydryl groups as the active center has great development potential. Studies showed that adsorption and ion exchange are the main mechanisms of heavy metal element adsorption by gangue-based adsorbents. Li et al. (2021b) proposed that ion exchange reactions occur between heavy metal ions and other cations during the adsorption process. Heavy metal ions form a strong affinity with M–O–Si or M–O–Al skeletons, and chemical bonding occurs, which could effectively immobilize heavy metal cations. However, in the process of adsorption and desorption, heavy metal ions cannot be completely ion-exchanged with other cations. When the adsorbent is recycled again, after several times of adsorption and desorption, heavy metal ions and other cations will not be transformed into the same amounts. As a result, the adsorption capacity of the adsorbent will be reduced, consequently reducing the adsorption efficiency of heavy metal elements; this is still a problem of modified gangue adsorbent that needs to be solved in future research.

4.1.2 Coal gangue-based zeolite

The preparation of zeolite from coal gangue can also be used for heavy metal adsorption. Natural zeolite is widely used as a heavy metal adsorbent because of its unique porous structure and large specific surface area (Yuna 2016). The cost of raw materials in the traditional method of zeolite synthesis is relatively high, and Al_2O_3 and SiO_2 are the main mineral compositions of coal gangue. Using coal gangue as a raw material may provide a silicon and aluminum source for synthesizing zeolite; thus, the method of synthesizing high-efficiency zeolite from coal gangue is widely used (Kelm and Helle 2005). Currently, X-type, Y-type, and A-type zeolites synthesized from coal gangue are widely used in heavy metal adsorption. Ge et al. (2020) successfully synthesized Na-X zeolite by alkali fusion and hydrothermal methods using coal gangue as raw material. The adsorption capacity of Pb reached 457 mg/g, which is higher than some natural zeolites previously reported in the literature and is an excellent adsorbent for Pb removal. Bu et al. (2020) synthesized Na–Y zeolite from coal gangue by the alkali fusion process and hydrothermal reaction. After five adsorption and desorption cycles, the removal rate of Pb by the synthesized Na–Y zeolite reached 100%. Qian and Li (2015) used coal gangue as raw material and successfully synthesized pure single-phase highly crystalline Na–A zeolite with high calcium (Ca) ion exchange capacity by in situ crystallization technique. Han et al. (2019) synthesized a new SSZ-13 zeolite using coal gangue under ultrasonic pretreatment conditions using hydrothermal synthesis. However, the research on this gangue-based zeolite system is not yet optimized, and few experimental data exist on this Pb adsorption method. Nevertheless, it is still necessary to improve the related experiments and, thus, the adsorption performance of Pb. In addition, easy access to synthetic raw materials does not equal good economic benefits for the materials, and the production cost of zeolite is still high, which also limits the wide application and development of gangue-based zeolite. Therefore, how to economically and efficiently synthesize zeolite from solid waste as raw material, such as coal gangue, fly ash, and blast furnace slag, has become one of the topics for future research.

4.2 Coal gangue-based geopolymer prevents and controls Pb

In recent years, geopolymer has gradually become one of the directions of coal gangue resource utilization due to its ability to solidify heavy metals, and geopolymer is more suitable for the prevention and control of Pb in soil during the migration of heavy metals. Current research shows that geopolymers can be divided into alkali- and acid-activated materials (He et al. 2022). As a new type of cementitious materials, alkali-activated materials have high compressive strength, durability, and fire resistance properties. Alkali-activated materials are used to remove heavy metals in coal gangue due to their lower carbon emissions and better durability than cement. The process of activation and heavy metal solidification/stabilization of alkali-activated materials often proceeds in the following order: the raw materials are dissolved as precursors after reacting with an alkali solution, during which ion recombination and replacement occur. Gelation of the materials occurs at the later stage of dissolution to form C–A–S–H and N–A–S–H gels, and the gel is continuously solidified and strengthened by continuous crystallization to eventually form crystals (Qian et al. 2022). Due to the late development of acid-activated materials, only phosphoric acid or phosphate is currently used as the activator. The mechanism of action is similar to that of alkali-activated materials, in which the raw materials form P–OH, Si–OH, Al–OH, etc., by phosphoric acid, and gels then form the crystalline phase aluminum phosphate ($AlPO_4$), the amorphous phase Si–O–P, and Al–O–P crystals (Li et al. 2022b). Pb^{2+} is chemically stabilized with Si–O and Al–O in the alkali- or acid-activated gel crystal structure (Guan et al. 2021; Liu et al. 2016). Through this physical solidification and chemical stabilization, the heavy metal cations are immobilized in the gangue base polymer structure, thereby reducing the risk of releasing heavy metal elements into the environment.

4.2.1 Modified coal gangue-based alkali-activated materials

Modified coal gangue-based alkali-activated materials are prepared from coal gangue by modifying alkali activation. Modified gangue alkali-activated materials can be used to immobilize/stabilize heavy metal elements, as shown by a large number of domestic studies. Zhao et al. (2019) prepared alkali-activated materials to stabilize heavy metals from coal gangue, improving the stabilization efficiency of both Pb and other heavy metals. It was demonstrated by morphological analysis, X-ray diffraction, Fourier transform infrared spectroscopy, and scanning electron microscopy that the Pb in the materials was bonded to Si–O and Al–O in the structure by physical solidification and chemical stabilization. Pb was fixed in the structure of the alkali-activated materials, reducing the risk of migration of heavy metal elements into the environment. Since gangue contains many quartz and kaolinite mineral components, it can produce more active silica-aluminum components after activation. Presently, the silica-aluminum content in the system is often increased by adding coal gangue calcined at high temperatures to change the silica-aluminum ratio to improve the performance of materials. However, calcination at a very high temperature will lead to the consumption of a large amount of heat energy and increase costs (Cao et al. 2016). Therefore, how to efficiently control the silica-alumina ratio in different types of gangues and improve the economic benefits of the materials is a future research direction for improving the performance of alkali-activated materials.

4.2.2 Composite coal gangue-based alkali-activated materials

Currently, the main research direction is using other solid wastes and coal gangue for multifaceted synergistic composite coal gangue-based alkali-activated materials. Although the preparation of alkali-activated materials from coal gangue as a single silica-aluminate raw material has been widely studied, the low activity of coal gangue is still a problem that hinders its wide application. To ensure the properties of other solid wastes, the use of coal gangue is necessary to further improve the performance of alkali-activated materials, obtain an efficient and reasonable method to prepare composite gangue-based alkali-activated materials, and use coal gangue on a large scale (Ma et al. 2020). Feng et al. (2021) prepared composite cementitious materials by thermal activation using two solid waste copper tailings and coal gangue as raw materials for compounding. The compounded materials stabilized Pb with a leaching range of 2.2–4.8 mg/L, and the solidification rate was 97.9%. Zhou et al. (2020b) prepared alkali-activated material precursors by mechanical-alkaline activation using red mud and coal gangue as raw materials so that most heavy metal cations in the solid structure of alkali-activated materials were chemically stabilized, successfully immobilizing the structure of alkali-activated materials and Pb was transformed from the highly active morphological fraction to the stable residual state.

Although the activation of coal gangue by high-temperature calcination can improve its activity, the addition of liquid sodium metasilicate (Na_2SiO_3) and sodium hydroxide (NaOH) will lead to higher cost and energy consumption of material preparation (Elzeadani et al. 2022). The extremely high alkalinity of a material will inevitably affect its properties. Higher concentrations of OH^- can lead to excess alkalinity that affects the reaction, while lower concentrations of OH^- cannot provide the required alkalinity in alkali-activated materials. Therefore, a low-energy and low-cost activation preparation method should be established. In addition, the microstructure of alkali-activated materials and the long-term effects of heavy metal leaching are relatively new and have not been thoroughly studied. In addition, the long-term leaching hazard of alkali-activated materials has become a concern that needs to be solved; thus, a more systematic and comprehensive study on the long-term leachability and prevention and control of heavy metal elements is needed.

4.2.3 Coal gangue-based acid-activated materials

Acid-activated materials are a new direction in the current research on geopolymers. The –P–O–Si–O–Al–O– structure of phosphate-based activated gangue geopolymers does not generate alkaline cations during the activation process; therefore, it is not necessary to consider the effect of alkalinity on the materials, and it also avoids competition between alkaline cations and Pb^{2+} in the solidification and stabilization process (Li et al. 2022a). However, due to the recent development of acid-activated materials and the immaturity of the system, the raw materials for acid-activated materials polymers primarily focus on metakaolin, and only a few studies have used solid wastes, such as fly ash, as precursors for acid-activated ground polymers (Wang et al. 2020). Compared with alkali-activated materials, acid-activated materials are more complex than alkali-activated materials due to their reaction mechanism, which is still poorly understood (Moghadam et al. 2019). Compared with solid wastes with different sources and properties, the composition of kaolinite minerals is relatively stable, and the use of gangue and other solid wastes as raw materials for alkali-activated materials will further increase the difficulty of experimental research. If the reaction mechanism of acid-activated materials can be further improved in future research, the use of gangue and other solid wastes to prepare acid-activated materials will increase the prevention and control measures for heavy metal elements and expand the scope of the application of comprehensive utilization of coal gangue.

5 Conclusions and perspectives

The migration, prevention, and control of Pb in coal gangue were discussed and summarized, with the following conclusions:

(1) This paper described the different migration processes of Pb from coal gangue, involving spontaneous combustion, weathering, and leaching, to the environment, which is helpful in understanding the transformation process of Pb under natural conditions. In addition, studying Pb migration and control measures in coal gangue is fundamental.

(2) The available forms of Pb in coal gangue were systematically classified by the Tessier method into exchangeable, carbonate-bound, Fe–Mn-bound, organic-bound, and residual states. The related migration environment, compound morphology, valence, and activity of Pb with different available forms were discussed and compared, and the the migration and transformation mechanism of Pb was considered. This review distinguished between the different available forms of Pb, which is conducive to evaluating the prevention and control effect of heavy metals. Classification of the available forms can explain the occurrence mechanism and migration and transformation rule of Pb.

(3) The National Development and Reform Commission proposed guidance on the comprehensive use of solid waste to encourage coal gangue underground filling, reclamation, and backfill for ecological comprehensive disposal. Adsorbents and geopolymers with coal gangue as raw materials are introduced to activate the physicochemical properties and structure of coal gangue through treatment modification and other target products to prevent and control Pb leaching. Furthermore, the synergistic compounding of multiple solid wastes can achieve better performance, solving the problem of heavy metal leaching in the coal gangue reclamation process and reducing reclamation costs.

(4) Various problems and shortcomings in coal gangue prevention and control methods were pointed out, such as the decrease in the adsorption efficiency of adsorbents, the difference in the performance of alkali-activated material precursors, and the high cost of the material treatment process. In practical applications, the adsorbent needs to further verify the maximum adsorption capacity and long-term safety performance of the material for Pb. It is crucial to analyze Pb migration in the early stage of the reclamation backfill process and monitor Pb to the environment in the later stage.

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Declarations

Competing interests The authors declare that they have no competing interests.

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