

Release Characteristics of Harmful Trace Elements during Dynamic Leaching and Static Immersion of Coal Gangue in Xinjiang

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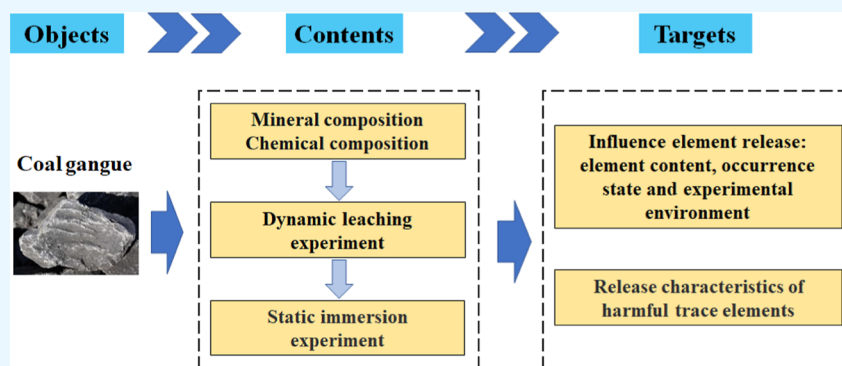
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ABSTRACT: Coal gangue has dual attributes of waste residue and resources. Clarifying the release characteristics of harmful trace elements from the coal gangue can provide a theoretical basis for environmental impact and resource utilization. In this study, the characteristics of harmful trace elements released from coal gangue in Xinjiang during dynamic leaching and static immersion experiments were determined using proximate analysis, X-ray powder diffraction (XRD), X-ray fluorescence spectrometry (XRF), and inductively coupled plasma mass spectrometry (ICP-MS). The results show that (1) the higher the content of harmful trace elements in coal gangue and the greater the concentration coefficient (CC), the greater the release of elements in dynamic leaching and static immersion experiments. The mode of occurrence of trace elements in the coal gangue determines their transport and release. Elements are associated not only with moisture but also with minerals, such as clays, sulfides, and carbonates, which are readily soluble in water. (2) The release of harmful trace elements was inversely proportional to time in the dynamic leaching experiments, and the main reason for the reduction in element release during the late leaching period was the adsorption effect of clay minerals. In the dynamic leaching experiment, harmful trace elements in the surrounding environment continued to accumulate, and static immersion experiments in water showed that harmful trace elements gradually reached dynamic equilibrium. The concentration of most elements in the late stage of the static immersion experiment was lower than that in the early stage, indicating that the environmental hazards of dynamic leaching were greater than those of the static immersion of coal gangue in Xinjiang.

1. INTRODUCTION

Coal plays an important role in fossil energy consumption, and the total coal resources in Xinjiang account for approximately 40% of the total coal resources in China.^{1,2} A hot topic for coal geochemists is the presence of hazardous elements in Chinese coal. The massive accumulation of coal gangue during coal mining has caused widespread concern, and the release of harmful elements from coal gangue has triggered environmental problems such as water and soil pollution, building corrosion, and radiation pollution.^{3,4}

Coal gangue is a gray–black rock with a lower carbon content than the rest of the coal seam during coal formation, and leaching is one of the main pathways by which the toxic elements of coal gangue enter the ecosystem.^{5,6} Harmful trace elements are toxic, carcinogenic, corrosive, and radioactive, and other environmentally hazardous elements are present in coal at levels below 1.00%. Among these, the environmentally

sensitive elements Be, V, Cr, Co, Ni, Cu, Zn, Mo, Ba, and Pb and radioactive elements Th and U are of widespread concern.^{7,8} Ecological vulnerability and serious health risks in mining areas result from the leaching and transport of hazardous elements from coal gangue into water, soil, and other environmental media.^{9,10}

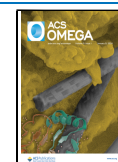
The degree of soil and water contamination by harmful elements leached from coal gangue depends on a number of factors, such as pH, particle size, content, fugitive state, and

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solid–liquid ratio.^{11,12} The pH of the leachate and the gangue particle size have a large influence on the release of harmful elements.^{13,14} The larger the specific surface area of fine-grained minerals in contact with the external environment, the more adequate and rapid the biochemical reactions.¹⁵ Dynamic leaching and static immersion experiments were performed to scientifically understand the impact of leachate on the surrounding environment, and the results of the experiments directly guide coal gangue treatment measures.^{16,17}

The trace element content, modes of occurrence, and mechanism of precipitation from coal gangue can provide basic data on environmental effects and energy utilization.^{18,19} However, the environmental pollution of harmful trace elements from the coal gangue in Xinjiang has rarely been reported, and the elemental release characteristics are not well understood. Therefore, the study of coal gangue mineral composition, chemical composition, modes of occurrence, and release characteristics of harmful trace elements under different experimental conditions provides the necessary theoretical foundation for preventing and controlling environmental pollution and for the comprehensive utilization of coal gangue.

2. MATERIALS AND METHODS

2.1. Sample Preparation and Analytical Methods. The gangue samples were collected from the top and bottom of the coal seams of the Middle Jurassic Xishanyao formation in the Wucaiwan mine area in Zhundong, Xinjiang and the Piliqing mine area in Yili. The gangue samples were collected using the stratified grooving method. Each sample was 10 cm × 10 cm (width × depth), and the samples were quickly placed in plastic bags after sampling to prevent contamination and oxidation. Using national standard GB/T 474-2008, the samples were prepared based on the method of preparing coal samples, and the coal gangue was crushed, sieved into three particle sizes of 880–150, 150–74, and <74 μm, and mixed uniformly in equal quantities to conduct the dynamic leaching and static immersion experiments.²⁰

Coal quality parameters such as moisture, ash yield, volatile matter, and fixed carbon in the gangue were determined using ASTM standards D3173-11, D3174-11, and D3175-11.^{21–23} The minerals in the coal gangue were determined using X-ray powder diffraction (XRD, Bruker D8 Advance). The XRD test range was from 3 to 105° with a scanning speed of 0.2 s/step. A sample of <74 μm gangue was placed in a special cassette for the X-ray diffractometer, compacted, flattened, and then scanned and tested.

The <74 μm coal gangue samples were ashed in a muffle furnace at 815 °C, and the major elemental oxides (e.g., SiO₂, Al₂O₃, Fe₂O₃, CaO, TiO₂, MgO, K₂O, and Na₂O) were determined in the ashed coal gangue samples by X-ray fluorescence (XRF, Bruker S8 TIGER, Germany). The concentration of hazardous trace elements in solution was determined by inductively coupled plasma mass spectrometry (ICP-MS, Agilent 7900) after the disintegration of <74 μm coal gangue samples.

2.2. Dynamic Leaching Experiment. A dynamic leaching experiment was conducted to simulate the migration of harmful trace elements in the coal gangue in a natural environment. Pure water was used as the leaching solution. The instruments were soaked in pure water for 24 h before the experiment and rinsed three to four times with pure water after natural drying. The balance weighed 48 g of gangue samples in an organic glass tube and the top and bottom of the gangue

samples were quartz sand with a thickness of 3 cm. A peristaltic pump controlled the leachate flow rate to 10 mL/h. On the third, fifth, seventh, ninth, and 11th days, 20 mL of the leachate was collected and filtered, and the concentration of deleterious trace elements in the solution was determined using ICP-MS.

2.3. Static Immersion Experiment. The static immersion experiment simulated the change in elemental concentration under the water–rock interaction in a reducing environment, focusing on the reaction between substances. A total of 48 g of gangue samples were weighed on a balance and added to a polyethylene plastic bottle containing 480 mL of pure water, and a mixture was configured with a liquid–solid ratio of 10:1. The cap was tightened, and the mixture was shaken repeatedly. At the beginning of the experiment, on the third, fifth, seventh, ninth, 11th, and 150th days, 20 mL of the supernatant was removed, and an equal amount of pure water was added to the polyethylene plastic bottle, shaken well, and left to stand. The concentration of harmful trace elements in the supernatant was determined by ICP-MS.

3. RESULTS

3.1. Proximate Analyses. Table 1 shows the results of the proximate analysis of the coal gangue. Based on the average ash

Table 1. Proximate Analysis of the Coal Gangue^a

samples	M _{ad} /%	V _{ad} /%	A _{ad} /%	A _d /%	FC _d /%	V _{daf} /%/%
WCW-1	2.16	14.94	82.04	83.85	0.88	94.57
WCW-2	6.47	22.05	50.00	53.46	22.96	50.65
WCW-3	2.07	11.85	81.52	83.25	4.65	72.22
PLQ-1	1.68	7.45	91.17	92.73	0.31	
PLQ-2	1.07	6.94	93.04	94.05	1.07	
PLQ-3	1.80	10.23	87.76	89.36	0.22	97.94

^aNotes: ad, air-dry basis; d, dry basis; daf, dry and ash-free basis; M, moisture; V, volatile matter; A, ash yield; and FC, fixed carbon.

yield ($A_d > 70\%$), the coal gangue was classified as medium–high ash.²⁴ The moisture content (M_{ad}) ranged from 1.07 to 6.47%, the ash yield (A_d) ranged from 53.46 to 94.05%, the volatile yield (V_{daf}) ranged from 50.65 to 94.57%, and the fixed carbon (FC) content ranged from 0.22 to 22.96%. The results showed that the levels of M_{ad} and FC were low, whereas those of A_d and V_{daf} were high.

3.2. Mineral Composition. XRD analysis indicated that kaolinite and quartz were the most abundant minerals in coal gangue, with minor amounts of montmorillonite, magnetite, chlorite, hard anhydrite, and rutile. Figure 1 shows that the mineral composition of the WCW coal gangue is dominated by kaolinite and contains quartz, montmorillonite, chlorite, magnetite, and hard anhydrite. The mineral composition of the PLQ coal gangue was mainly quartz, it also contained kaolinite, rutile, and other minerals. The WCW gangues are richer in minerals and contain more clay minerals than the PLQ gangues.

3.3. Chemical Composition. The contents of the major elemental oxides of the coal gangue samples are shown in Table 2. SiO₂ and Al₂O₃ are the major oxides in the gangue, which accounted for 51.93 and 21.27%, respectively, followed by Fe₂O₃ (1.88%), K₂O₂ (1.57%), TiO₂ (1.36%), MgO (0.62%), CaO (0.39%), Na₂O (0.11%), P₂O₅ (0.047%), and MnO (0.027%). The average content of major elemental oxides in the coal gangue was higher than that in Chinese

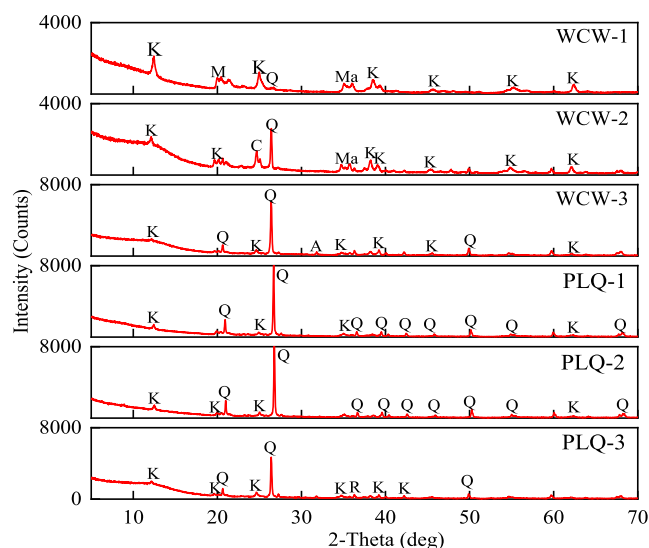


Figure 1. X-ray diffraction pattern of the coal gangue (K-kaolinite, Q-quartz, M-montmorillonite, Ma-magnetite, C-chlorite, A-anhydrite, R-rutile).

coals. SiO_2 and Al_2O_3 content analyses support the results of the XRD chart mineral analyses.^{25,26}

The distribution range of harmful trace element content in the coal gangue (Table 3), the comparative distribution range does not accurately reflect the general characteristics of the study object. The concentration coefficient (CC) presenting the degree of trace element enrichment in coal was proposed by Dai et al.²⁷ The CC were classified into six levels: including depleted ($\text{CC} < 0.5$), normal ($0.5 \leq \text{CC} < 2$), slightly enriched ($2 \leq \text{CC} < 5$), enriched ($5 \leq \text{CC} < 10$), significantly enriched ($10 \leq \text{CC} < 100$), and anomalously enriched ($\text{CC} \geq 100$).

The concentrations of harmful trace elements in coal gangue compared with Chinese coal show that the V and Cr contents were enriched, the Ba and Th contents were slightly enriched, and their CCs were 5.12, 8.72, 2.50, and 2.27, respectively (Figure 2). The elemental contents of Be, Co, Ni, Cu, Zn, Pb, and U were comparable to those of Chinese coal, with a CC range of $0.5 \leq \text{CC} \leq 2$. Only Mo had a deficit with a CC of 0.18.

3.4. Modes of Occurrence of Harmful Trace Elements of Coal Gangue. The state of the elements determines the ease of release and migration of the elements in coal and coal gangue and the complexity of environmental pollution.^{28,29} Cluster analyses were performed using 25 indicators of industrial analysis, harmful trace element content, and major

elemental oxide content to determine the interdependence between variables (Figure 3).^{30,31}

Group 1 includes Mo, U, Pb, Ni, V, Cr, Zn, Th, Be, Cu, and SiO_2 , with pro-iron and pro-sulfur elements, which are found in the clay and sulfide minerals. Elements and moisture (M_{ad}) are related to the elements through the water medium and coal gangue can easily exchange materials. Group 2 includes Al_2O_3 , P_2O_5 , K_2O , Co, Ba, MnO, MgO, TiO_2 , and Fe_2O_3 , which are dominated by lithophilic elements and often form oxides or various types of oxygenates. Fe_2O_3 was mainly sourced from sulfurous iron ore. TiO_2 is a preferrous element that is symbiotic with Fe. Ba and Co are primarily found in clay minerals, sulfide minerals, carbonate minerals, and organic matter. Group 3 includes Na_2O and CaO, which are associated with ash (A_d) and volatile matter (V_{daf}). The Na and Ca were endowed with inorganic minerals and organic matter (Figure 3a).

Group 1 incorporates TiO_2 , Ba, Ni, CaO, MgO, Fe_2O_3 , V, MnO, Co, Cu, Al_2O_3 , Mo, Zn, Pb, Th, U, and P_2O_5 and is dominated by pro-ferric and sulfurous elements, which are predominantly endowed in clay minerals, sulfide minerals, and carbonate minerals; the volatile fractions (V_{daf}) are correlated with the elements. These elements are likely hosted in organic matter. Group 2 contains Na_2O , Be, and Cr, which are mainly lithophilic elements and related to moisture (M_{ad}), indicating that the Be and Cr elements have a certain degree of water solubility and can easily exchange substances. Group 3 elements, SiO_2 , K_2O , and ash (A_d), have a strong correlation, indicating that most are stored in inorganic minerals (Figure 3b).

3.5. Harmful Trace Element Release Behaviors of Coal Gangue. Because of the different contents of hazardous trace elements in the coal gangue, the dynamic leaching effect was compared and judged by the intensity of stage leaching (L_i), which is the ratio of the concentration of the element in the solution to the absolute content of the element in the gangue sample during the leaching stage. The formula for L_i is as follows³²

$$L_i = \frac{C_i \times V}{1000A_i \times M} \times 100\%$$

where L_i , stage leaching intensity, %; C_i , elemental concentration in solution, mg/L; V , volume, mL; A_i , elemental content in sample, $\mu\text{g/g}$; and M , weight, kg. The average elemental release rate in static immersion experiments is the ratio of the elemental concentration in the supernatant to the elemental content in the gangue sample, which reflects the degree of elemental migration.^{33,34}

Table 2. Content of Major Elemental Oxides of the Coal Gangue (%)^a

sample	SiO_2	Al_2O_3	Fe_2O_3	CaO	TiO_2	MgO	K_2O	Na_2O	P_2O_5	MnO	$\text{Al}_2\text{O}_3/\text{SiO}_2$
WCW-1	40.36	35.64	0.43	0.25	1.95	0.31	0.41	0.10	0.030	0.002	0.88
WCW-2	24.71	19.64	0.23	1.10	3.42	0.36	0.17	0.20	0.033	0.003	0.79
WCW-3	49.56	20.85	1.68	0.26	0.52	0.41	1.25	0.13	0.030	0.024	0.42
PLQ-1	68.32	17.22	1.18	0.19	0.69	0.47	2.51	0.09	0.013	0.007	0.25
PLQ-2	69.50	16.47	2.42	0.24	0.68	0.81	2.58	0.06	0.074	0.029	0.24
PLQ-3	59.10	17.79	5.34	0.31	0.92	1.36	2.52	0.08	0.104	0.097	0.30
Ave	51.93	21.27	1.88	0.39	1.36	0.62	1.57	0.11	0.047	0.027	0.48
Chinese	8.47	5.98	4.85	1.23	0.33	0.22	0.19	0.16	0.092	0.015	

^aNotes: Ave and Chinese, the average content of major element oxides in the coal gangue and Chinese coals.

Table 3. Concentrations of Harmful Trace Elements in the Coal Gangue ($\mu\text{g/g}$)^a

sample	Be	V	Cr	Co	Ni	Cu	Zn	Mo	Ba	Pb	Th	U
WCW-1	1.50	118.71	106.70	2.25	34.64	3.60	19.25	1.54	19.15	24.50	9.91	10.20
WCW-2	3.55	247.02	143.37	3.46	58.71	36.03	32.65	1.57	44.45	46.01	27.71	10.48
WCW-3	0.73	46.93	52.74	8.69	12.11	19.77	15.30	0.68	167.58	7.63	4.17	1.65
PLQ-1	2.33	54.18	121.70	2.60	11.31	19.81	24.07	0.49	294.02	11.56	9.30	3.00
PLQ-2	1.25	71.35	80.01	4.60	11.99	23.09	87.96	0.30	291.00	24.95	15.18	3.71
PLQ-3	1.96	106.44	123.00	11.71	33.01	37.91	103.82	0.83	416.05	28.25	15.43	3.76
Ave	1.89	107.44	104.59	5.55	26.96	23.37	47.18	0.90	205.37	23.82	13.62	5.47
Chinese	2	21	12	7	14	13	35	5	82	13	6	3

^aNotes: Ave and Chinese, average concentration of trace elements in the coal gangue and Chinese coals.

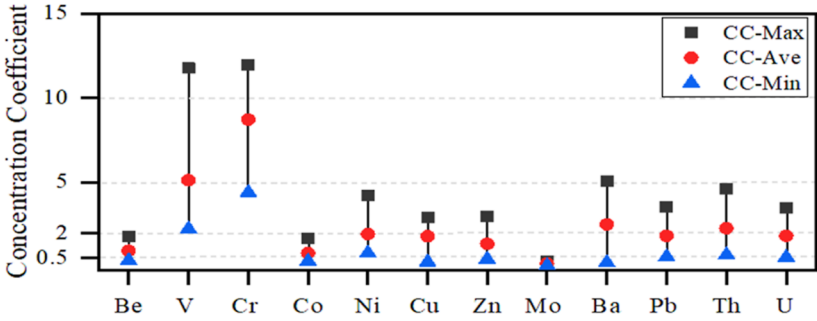


Figure 2. Concentration coefficients of trace elements in the coal gangue.

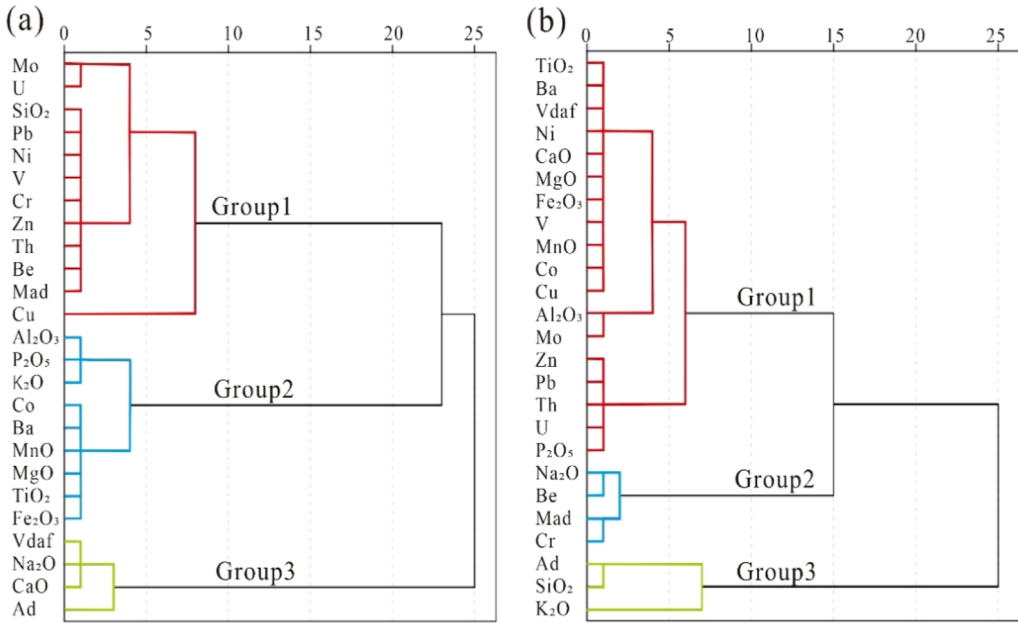


Figure 3. Cluster analysis diagram of the coal gangue in (a) the Wucuiwan mine area and (b) the Piliqing mine area.

The average rate of release of harmful trace elements from the coal gangue is shown in Figure 4. The elemental leaching intensity is $\text{Mo} > \text{Ba} > \text{Zn} > \text{V} > \text{Cu} > \text{Ni} > \text{Co} > \text{Cr} > \text{Pb} > \text{Be} > \text{Th} > \text{U}$ in the dynamic leaching experiment of LWCW-2 and the elemental leaching intensity of LPLQ-3 is $\text{Mo} > \text{Ba} > \text{Zn} > \text{Ni} > \text{Cu} > \text{Be} > \text{Co} > \text{Pb} > \text{U} > \text{V} > \text{Th} > \text{Cr}$. The average elemental release rate of JWCW-2 in static leaching experiments is $\text{Ba} > \text{Zn} > \text{Mo} > \text{V} > \text{Cr} > \text{Ni} > \text{U} > \text{Cu} > \text{Co} > \text{Be} > \text{Th} > \text{Pb}$ and the average elemental release rate of JPLQ-3 is $\text{Ba} > \text{Zn} > \text{Ni} > \text{Mo} > \text{U} > \text{Cu} > \text{Be} > \text{V} > \text{Co} > \text{Th} > \text{Pb} > \text{Cr}$. The WCW-2 and PLQ-3 gangue had high contents of Ba, Zn, Ni, and other elements, and the release rates of Ba, Zn, and Ni in the leachate were high. The above results indicate that

the higher the content of the harmful elements in the gangue, the greater the release of the elements from the leachate (Figures 2 and 4). The dynamic leaching experiments on the concentration and time of harmful trace elements were inversely proportional to the overall performance of a decreasing trend (Figures 5a–c and 6a–c). Static immersion experiments in the harmful trace element concentration change trends are different, with element concentration with the change in immersion time. The elements exchange substances between gangue and aqueous solution (Figures 5d–f and 6d–f). The elemental concentrations detected on the third day were used as the initial concentrations (Figures 5 and 6).

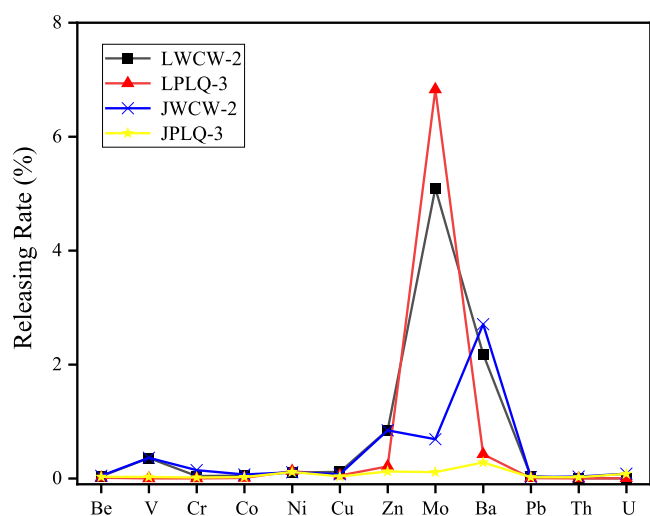


Figure 4. Average release rate of harmful trace elements from the coal gangue.

The elemental concentrations of Ba, V, and Zn decreased rapidly during the dynamic leaching experiment of WCW-2 (Figure 5a). On the 150th day of the static immersion experiment, the Ba and Zn concentrations were higher than the initial concentration, and the V concentration was significantly lower than the initial elemental concentration, returning to the gangue in some form (Figure 5d). The concentrations of Cr, Ni, Cu, Pb, Co, Th, and U in the dynamic leaching experiment slowly decreased over time (Figure 5b,c), and the concen-

trations of Cr, Ni, Cu, Pb, Co, Th, and U in the static immersion experiment peaked at the 11th day (Figure 5e,f).

The elemental concentration decreased on the 150th day, when Cr and Co concentrations were significantly higher than the initial concentrations. The concentration of Mo in the dynamic leaching experiment was significantly higher than that in the static immersion experiment. The fluctuation in the concentration of Be in the dynamic leaching and static immersion experiments was small and stable.

A comparison of the differences in the release of harmful trace elements from the coal gangue in the dynamic leaching and static immersion experiments of PLQ-3 is shown in Figure 6. The dynamic leaching and static immersion experiments in the first 11 days of the Ba and Zn concentration release trend were the same (Figure 6a,d). The Ba concentration was lower than the initial concentration, and the Zn concentration was higher than the initial concentration after 150 days of static immersion. The stable release of V, Cr, Co, Pb, Th, and U was observed in the dynamic leaching experiments (Figure 6b,c). The concentrations of V, Cr, Co, Pb, Th, and U in the static leaching experiments showed rapid accumulation, release, and stabilization (Figure 6e,f). In the dynamic leaching experiment, the concentration of Mo exhibited a rapid decrease, whereas the static leaching experiment showed only a small change in elemental concentration. Be showed stability after rapid release in both experiments.

4. DISCUSSION

4.1. Factors Affecting the Release of Harmful Trace Elements from Coal Gangue. The release of harmful trace elements from coal gangue is related not only to the element

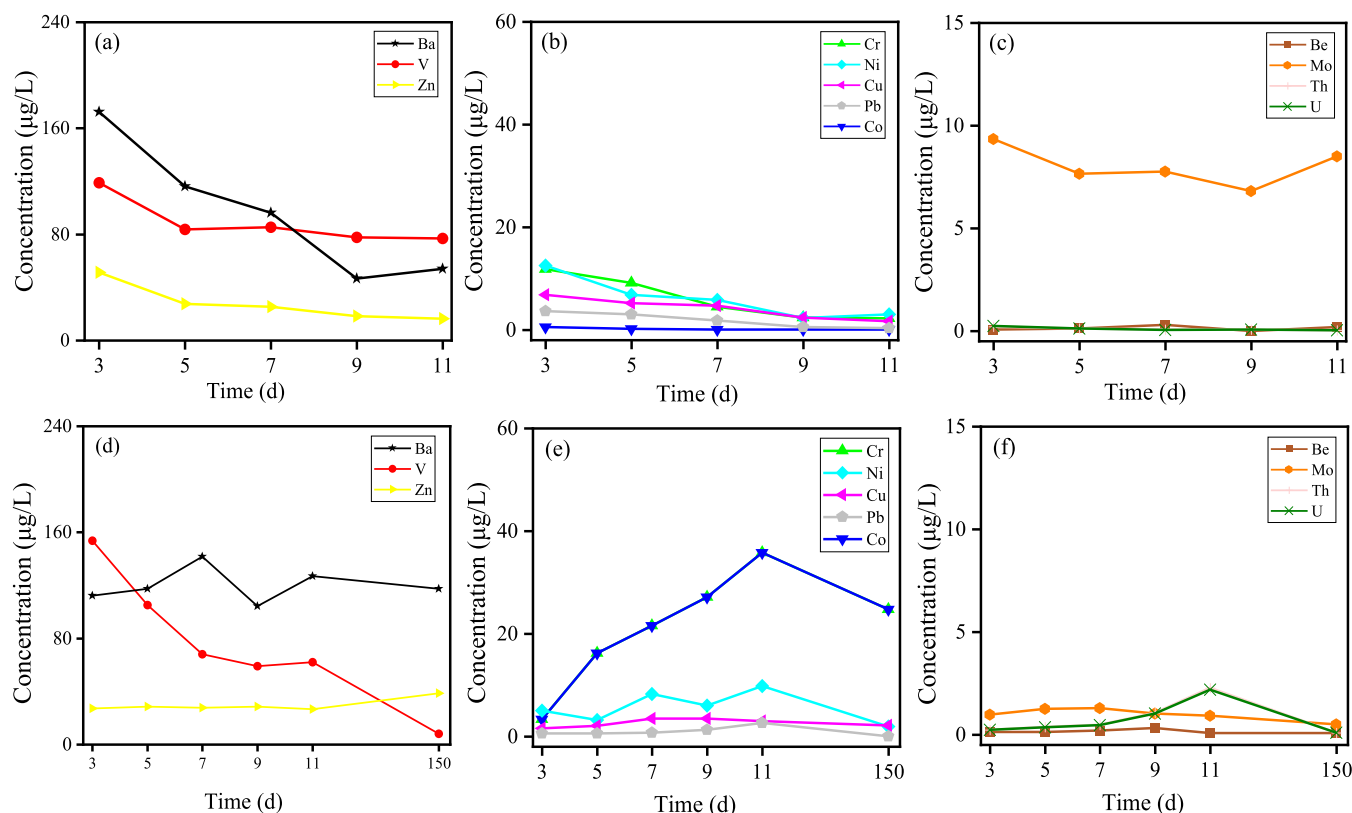


Figure 5. Comparison of the release of harmful trace elements from the WCW-2 gangue. (a–c) Results of the dynamic leaching experiment and (d–f) results of the static immersion experiment.

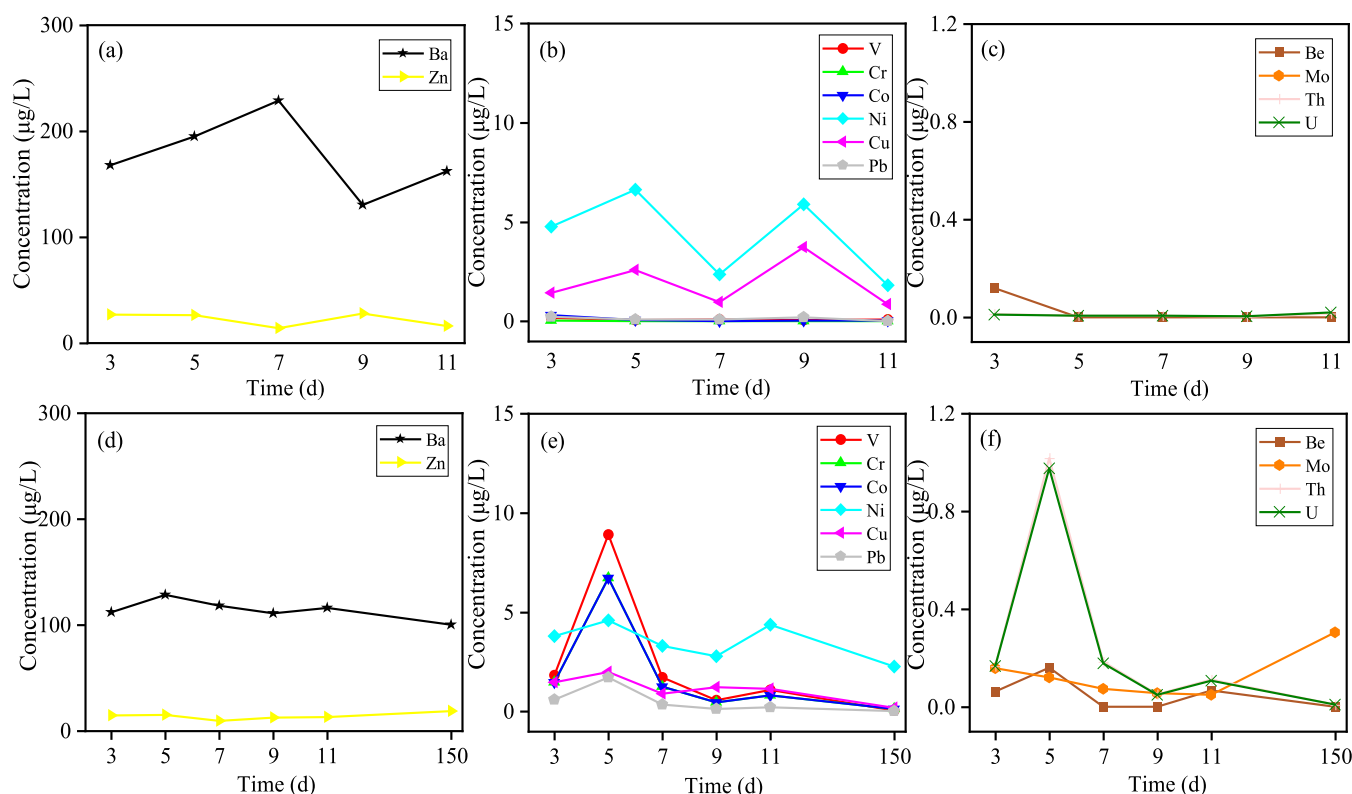


Figure 6. Comparison of the release of harmful trace element from the PLQ-3 gangue. (a–c) Results of the dynamic leaching experiment and (d–f) results of the static immersion experiment.

content and occurrence state but also to the experimental environment. The elemental contents of Ba, Be, V, Zn, Cr, Ni, Cu, Pb, Th, and U in WCW-2 were high and enriched; these elements were primarily stored in clay and soluble minerals. The elemental concentrations in the dynamic leaching experiment decreased with time, and the concentrations of Ba, Be, Zn, Ni, Cu, Cr, Pb, Th, and U in the static leaching experiment were higher than the initial concentration, or the difference was small, indicating that the change in elemental concentration was mainly related to the elemental content and state of existence. In the static immersion experiment, the concentration of V continued to decrease, indicating that the element was in a reducing environment and returned to the gangue in a certain form. The Mo and Co contents in the gangue were low, mainly in clay minerals, carbonates, and other soluble minerals. In the leaching experiments, the concentration of the dissolved Mo was high. In the soaking experiments, the dissolved concentration of Co was high, indicating that the experimental environment significantly affected the release of the elements.

The Ba, Zn, Ni, and Cu contents and CCs in PLQ-3 were high and enriched, and the dissolved concentrations were high in the leaching and soaking experiments, which were mainly related to the content of the elements in the gangue. Be is closely related to moisture and can easily exchange substances through water. In the dynamic leaching and static immersion experiments, Mo was in a deficit state and stored in clay and soluble minerals, and the concentration of released elements was relatively high. V, Cr, Co, Pb, Th, and U were less dissolved in the dynamic leaching experiment, while their concentrations changed significantly in the first 7 days of the

static leaching experiment, and element release was related to the experimental environment.

4.2. Causes of Harmful Trace Element Releases. In the dynamic leaching experiment, the release of harmful trace elements from the gangue was inversely proportional to the leaching time. The release of harmful trace elements from the gangue occurred during the early and middle stages of leaching. XRD, XRF, and cluster analysis showed that kaolinite and other clay minerals in the WCW-2 gangue accounted for a large proportion of the mineral composition, which was more complex, and most elements were related to moisture. The mineral composition of the PLQ-3 gangue was relatively simple, and the release of elements remained relatively stable. An important reason for the decline in the release of harmful trace elements during the late stage of leaching is the good adsorption effect of clay minerals.³⁵

The static immersion experiment, which alternately increases and decreases the concentration of harmful trace elements, is a process of solid–liquid physical and chemical action for achieving dynamic equilibrium. Under long-term immersion, coal gangue is in anoxic condition, and harmful trace elements activated on the surface are rapidly released. On the 150th day, most of the harmful trace elements in the gangue were lower than the initial element concentration, and most of them are lower than the initial element concentration, indicating that the results of the immersion experiment were not the limit of the leaching experiment.

5. CONCLUSIONS

In this study, the influence of element content, storage state, and experimental environment on the release of deleterious trace elements from coal gangue was investigated using

dynamic leaching and static immersion experiments to obtain the release characteristics of harmful trace elements. The main conclusions are as follows:

Coal gangue in Xinjiang is characterized by low moisture content, low fixed carbon content, high ash, and volatile yield. Compared with the average elemental content in Chinese coals, V and Cr were significantly enriched, Ba and Th were slightly enriched, Mo was in deficit, and Be, Co, Ni, Cu, Zn, Pb, and U were enriched to a normal extent. The coal gangue is dominated by kaolinite and quartz minerals, followed by magnetite, gypsum, rutile, and other minerals.

Owing to the high content of harmful trace elements in gangue, the CC is large and the release of elements is greater in dynamic leaching and static immersion experiments. The mode of occurrence of elements in a coal gangue determines their transport and release. Elements are not only associated with moisture but also with minerals, such as clay minerals, sulfide minerals, and carbonate minerals, which are readily soluble in water.

The release of harmful trace elements in dynamic leaching experiments is inversely related to time, and an important reason for the decrease in the elemental concentration is the good adsorption of clay minerals. The concentration of harmful trace elements increases in the solid–liquid interaction and gradually reaches a dynamic equilibrium. The environmental risks from the release of harmful trace elements in dynamic leaching experiments were greater than those in static immersion experiments.

■ ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsomega.3c05736>.

(ZIP)

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Notes

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