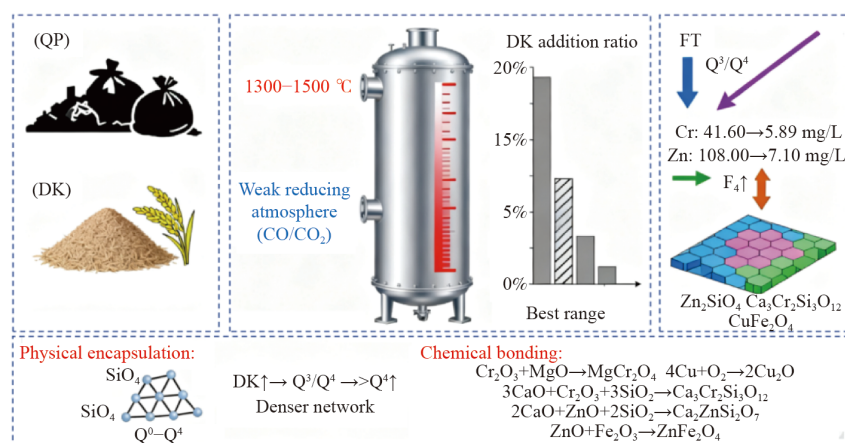


Investigating the migration mechanisms of heavy metals under silicate polymerization in gasification slag from landfilled municipal solid waste with rice husk addition

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Abstract: Slag gasification represents one of the key technologies for the clean and resource-oriented treatment of organic solid wastes with high heavy-metal content, such as landfilled municipal solid waste. Using rice husk as a silicon source, this work investigated the effect of rice-husk addition on the vitrification of landfilled-waste slag and the solidification behavior of heavy metals. The



results demonstrated that the incorporation of rice husk (5%–10%) lowered the flow temperature of the slag and effectively suppressed heavy-metal leaching. Specifically, the leaching concentrations of Cr and Zn decreased from 41.60 and 108.00 mg/L to 5.89 and 7.10 mg/L, respectively. The active SiO₂ introduced by rice husk promoted the formation of highly polymerized siloxane networks (Q³, Q⁴), which inhibited gasification-induced migration of heavy metals and enhanced physical encapsulation of them. Concurrently, it facilitated the incorporation of Zn²⁺, Cr³⁺ and Cu²⁺ *via* substitution or solid-solution mechanisms into stable mineral phases such as Zn₂SiO₄ and CuFe₂O₄, immobilizing them in garnet and spinel structures. This process increased the residual fraction and reduced the bioavailability of the heavy metals. The present study elucidates the mechanistic role of rice husk in modulating slag structure and heavy-metal immobilization, thereby provides a theoretical basis for the clean and efficient co-treatment of landfilled waste and biomass through a “treating waste with waste” strategy.

Key words: landfilled municipal solid waste; rice husk; heavy metal immobilization; gasification slag; ash slag

Over the past 40 years of reform and opening-up, the historically stockpiled landfilled municipal solid waste (MSW) in China has exceeded 8 billion tons^[1]. Its high moisture content (30%–50%)^[2], high ash content (>50%), and complex speciation of heavy metals^[3] make it difficult for traditional incineration-landfill coupling processes to achieve waste reduction, harmless treatment, and resource

recovery simultaneously. Pre-treated (sun-dried and screened) landfilled waste exhibits a calorific value of approximately 18–29 MJ/kg^[4], which is comparable to that of coal^[5]. The overall thermal efficiency during gasification exceeds 75%^[6], rendering MSW suitable as a feedstock for gasification. The application of waste gasification technology to process the vast stockpile of landfilled waste

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with complex composition enables the complete resource recovery of waste^[7]. This not only provides a novel approach to addressing environmental issues but also opens new pathways for developing alternative energy sources.

The composition of municipal solid waste is complex, with common waste items such as spent batteries, electronic products, paints, and cosmetics containing heavy metals^[8]. Heavy metals are generally defined as metallic elements with a density $>4.5 \text{ g/cm}^3$. Elements such as mercury (Hg), cadmium (Cd), chromium (Cr), nickel (Ni), copper (Cu), lead (Pb) and zinc (Zn) emitted from MSW incineration plants are classified as heavy metals^[9]. It is widely acknowledged that the heavy metal content in waste tends to increase with landfill age^[10]. During the collection, transportation, and treatment of waste, heavy metals migrate and transform *via* physical transport, chemical dissolution, and biological leaching^[11]. Their migration and transformation during incineration follow an evaporation-condensation mechanism^[12]. Slagging gasification, a treatment technology capable of simultaneously achieving vitrification of organic solid waste ash and efficient resource recovery^[13], converts waste into syngas (H_2/CO) at high temperatures ($>1200^\circ\text{C}$). The resulting slag possesses vitreous characteristics, offering the dual advantages of nearly complete dioxin decomposition ($>99.9\%$) and physical encapsulation of heavy metals^[14].

Studies indicate that the chemical composition of slag derived from landfilled MSW is typically dominated by CaO and SiO_2 . However, a high content of alkaline oxides (e.g., CaO , MgO) can disrupt the three-dimensional silicate network structure, leading to an increased degree of slag polymerization (indicated by a higher NBO/T ratio) and reduced viscosity. If the SiO_2 content in the slag is insufficient or the alkaline component content is excessive, the silicate network struggles to form a stable vitreous structure^[15]. The migration of heavy metals during the melting process is jointly controlled by temperature, atmosphere, and the silicate-oxygen structure of the slag^[16]. As shown in Table 1, heavy metals exist in diverse speciations. Volatile elements such as Pb, Zn and Cd tend to release as chlorides within $1000\text{--}1200^\circ\text{C}$ range^[17], whereas Cr, Ni and Cu predominantly remain in the slag as oxides^[18]. Consequently, the risk of heavy metal re-release from the slag and the insufficient degree of silicate network polymerization remain the core bottlenecks hindering its engineering application. Furthermore, the complex composition of landfilled waste and significant fluctuations in ash composition^[19] result in inherently low SiO_2 content ($<30\%$). This leads to a low degree of polymerization of the $[\text{SiO}_4]^{4-}$ tetrahedra in the slag, causing breaks in the silicate chains and the formation of non-bridging oxygen atoms, which in turn provides weak binding sites for heavy metals.

Biomass addition can improve slag performance through a triple mechanism involving thermochemical conditioning, provision of a reducing atmosphere, and supplementation of silicon sources. Among various agricultural wastes, rice husk, a by-product of rice processing with an annual global output exceeding 120 million tons^[21], is also a significant

component of the actual discharge of township MSW^[22] and is typically regarded as a low-value waste^[23]. However, rice husk possesses a large specific surface area^[24] capable of capturing heavy metal ions *via* physical adsorption^[25]. Its ash contains $>90\%$ amorphous SiO_2 ^[26] and abundant silanol groups^[27], making it an ideal active SiO_2 source. It reacts with CaO and Al_2O_3 present in the landfilled waste, regulating silicate phase transformation and network reconstruction^[28] during heat treatment to form highly polymerized Q^3 and Q^4 siloxane structures (Si-O-Si). Surface oxygen-containing functional groups of rice husk-derived biochar can undergo chemical reactions with heavy metal ions, forming stable surface complexes^[29]. Acting as a fluxing agent^[30], it facilitates the formation of a dense vitreous network structure^[31]. Theoretically, this dual mechanism of “structural encapsulation” and “chemical bonding” can enhance heavy metal immobilization^[32]. However, excessive rice husk may reduce slag basicity, thereby inhibiting silicate reconstruction and exacerbating heavy metal volatilization. Currently, the coupling relationships among the rice husk blending ratio, melting temperature, and heavy metal immobilization efficiency, along with the microscopic regulatory mechanisms governing silicate polymerization behavior, remain critical scientific questions requiring breakthroughs in this field.

Given the current situation that landfill capacities are nearly exhausted and siting new landfills is challenging, a gap still remains in the comprehensive treatment of landfilled waste. This study focuses on the gasification of the typical landfilled MSW. By regulating the rice husk blending ratio ($0\text{--}20\%$) and the melting temperature ($1300\text{--}1500^\circ\text{C}$), the migration patterns of typical heavy metal elements (Zn, Cr, Cu) in the gas-slag phases were systematically investigated. Methods such as XRD, Raman spectroscopy, and FT-IR are employed to analyze the evolution of the degree of silicate network polymerization (NBO/T ratio, Q^n distribution). The study aims to explore the correlation mechanism among rice husk blending, silicate structure, and heavy metal immobilization efficiency. The objective is to provide a theoretical basis for resource utilization and heavy metal risk control in the slagging gasification process of landfilled waste with rice husk, while also to offer new insights for the thorough remediation of landfill hazards *via* the principle of “using waste to treat waste.”

Table 1 Melting and decomposition temperatures of common heavy metals in municipal solid waste^[20]

Element	Melting point and decomposition temperature/ $^\circ\text{C}$			
	chloride	oxide	sulfate	simple substance
Cu	620	1326	<200	1083
Pb	501	888	1170	327
Cr	1150	2435	173	1900
Hg	277	>400	—	39
Zn	283	1975	1700	419.6
Cd	568	1980	1000	321
Ni	1001	1980	31.5	1555

1 Experimental

1.1 Experimental materials

Landfilled municipal solid waste (MSW), collected from a river remediation project in Qingpu District, Shanghai, with a landfill age of approximately 15 years, was used as the primary research object and labeled as QP. It primarily consisted of 67.15% plastics and rubber, 21.21% textiles, and 8.94% wood/bamboo, among other components. Compared to other MSW^[33], QP had a higher content of plastics and rubber, exhibiting characteristics similar to landfills both domestically and internationally, predominantly featuring humus soil mixed with plastics^[34]. Rice husk specimens were collected from fresh seasonal harvests in the Northeast region. The husks were dried at 60 °C for 5 h, subsequently pulverized and sieved to isolate particles within the 100–120 mesh size range.

Following standards CJ/T 313—2009, the waste underwent collection, sorting, crushing, washing, and drying. Its physical composition is shown in Figure 1. In accordance with national standards GB/T 212—2008 and GB/T 476—2008, proximate analysis, ultimate analysis, and calorific value analysis were performed on two raw materials: freshly harvested rice husk (DK) from Northeast China and the landfilled waste (QP). The analyses were conducted using a Changsha Kaiyuan 5E-MAC III infrared rapid coal analyzer, a Vario MACRO elemental analyzer, and an oxygen bomb calorimeter. The results are presented in Table 2. The rice husk exhibited relatively high ash and volatile matter contents of 14.56% and 67.52%, respectively, aligning with the typical characteristics of agricultural and forestry biomass ash^[35].

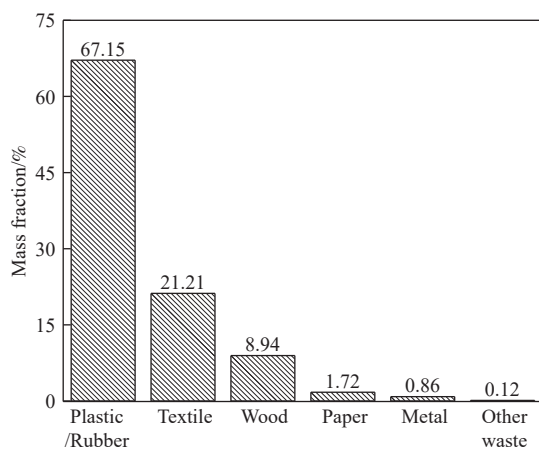


Fig. 1 Physical composition of municipal solid waste (QP)

The QP sample showed ash and volatile matter contents of 20.17% and 75.86%, respectively, approaching the ash composition of lignite^[36]. As plastics are the key factor determining the calorific value of waste, this sample exhibited a relatively high calorific value^[37]. Additionally, the chlorine content of QP was determined to be 1.15%, which is considered a high level^[38].

QP was placed in a muffle furnace, and programmed temperature ashing was performed following standard GB/T 28731—2012: initially, the temperature was raised from room temperature to 30 °C within 10 min; subsequently heated to 250 °C over 50 min and held for 60 min; then heated to 550 °C over 60 min and maintained at that temperature for 2 h. After sample preparation *via* pelletizing, the chemical composition of the QP ash was determined using X-ray fluorescence spectrometry.

To investigate the immobilization effect of minerals in the low-temperature melting zone (Figure 2) on Cu, Zn and Cr, fresh rice husk samples were mixed with QP at weight ratios of 5%, 10%, 15% and 20% to achieve different primary mineral phases at high temperatures. The five samples with varying rice husk additions were labeled DK00, DK05, DK10, DK15 and DK20, respectively. Their chemical compositions are shown in Table 3. The highest component in all five ash samples was SiO₂, with its content significantly increasing as the rice husk addition ratio increased. Contents of CaO, Fe₂O₃ and MgO continuously decreased, while that of Al₂O₃, K₂O and Na₂O changed gradually. The content of Cl and SO₃ showed fluctuations. The silicon-to-aluminum ratio (S/A) of QP was relatively high at 3.08, consistent with the characteristics of typical MSW ash.

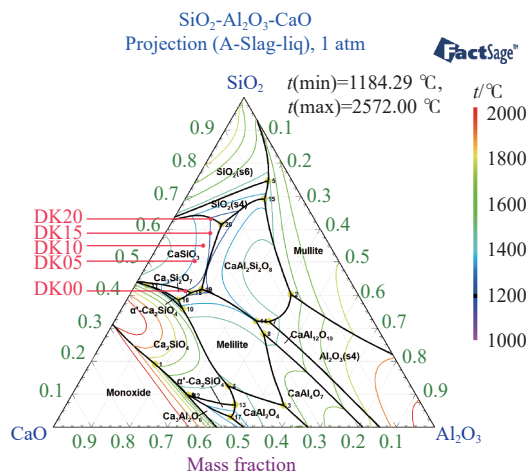


Fig. 2 Ternary phase diagram of the SiO₂-Al₂O₃-CaO system

Table 2 Proximate and ultimate analyses and calorific value of sample

Sample	Proximate analysis $w_d/\%$			Ultimate analysis $w_d/\%$					$Q_{HHV}/(\text{MJ} \cdot \text{kg}^{-1})$
	<i>A</i>	<i>V</i>	FC	C	H	O	N	S	
DK	14.56	67.52	17.92	40.65	5.25	39.07	0.45	0.02	15.63
QP	20.17	75.86	3.99	57.79	7.18	14.48	0.14	0.24	29.43

Table 3 Chemical composition of ash samples with different rice husk additions

Sample	Composition w/%										
	SiO ₂	Al ₂ O ₃	CaO	Fe ₂ O ₃	SO ₃	MgO	K ₂ O	Cl	TiO ₂	Na ₂ O	others
DK00	28.7	9.33	28.4	6.65	6.29	2.96	1.54	6.8	7.37	0.77	1.19
DK05	35.88	9.18	24.96	4.98	9.11	2.19	1.54	3.78	3.16	0.78	4.44
DK10	39.92	9.03	24.14	4.84	6.8	2.14	1.78	3.4	3.63	0.7	3.62
DK15	43.05	8.62	21.68	4.50	6.72	2.07	1.87	4.26	2.94	0.67	3.62
DK20	46.85	6.96	19.59	3.84	7.4	1.74	1.85	5.01	2.52	0.63	3.61
DKH	91.59	0.71	1.59	0.35	0.59	0.38	4.10	—	0.03	0.12	0.54

As shown in Figure 2, DK00 (without rice husk addition) is located near the boundary between dicalcium silicate and calcium silicate. With increasing rice husk addition, its composition gradually shifts towards the upper right.

1.2 Slag sample preparation

To obtain quenched slag samples required for analyzing mineral composition at specific temperatures, a measured mass of ahs sample was placed in a corundum crucible. A CO₂ flow rate of 200 mL/min and a CO flow rate of 300 mL/min were set to ensure a weakly reducing atmosphere^[39] during the experiment. The crucible was pushed into the heating zone of the furnace at 1200 °C, then heated to the specified temperature at a rate of 5 °C/min. It was held at 1300, 1400 and 1500 °C for 5 min each, then rapidly removed and quenched with liquid nitrogen. After cooling, the bottom of the corundum crucible was cut off, and the slag was chiseled out with a hammer and ground to a particle size of less than 100 µm for characterizations.

1.3 Sample analysis

After ball milling, the mineral composition of the ash/slag was analyzed using a Dutch PANalytical D/MAX2550-VB/PC XRD instrument (Cu target, 40 kV/40 mA, scanning range 10°–80°) combined with Jade 6.5 software. Following standard GB/T 219—2008, ash fusibility characteristic temperatures (DT, ST, HT, FT)^[40] were determined under a weakly reducing atmosphere using a Changsha Kaiyuan 5E-AFIII ash fusion temperature tester. Thermodynamic simulations of the SiO₂-Al₂O₃-CaO-Fe₂O₃-MgO-K₂O-TiO₂-Na₂O system from 800–1500 °C (at 50 °C intervals) were performed using FactSage software to predict mineral phase evolution and solid/liquid phase content. The network structure of the slag quenched at 1400 °C was characterized by Fourier Transform Infrared Spectroscopy (FT-IR, Nicolet iS50, KBr pellet method, 4000–400 cm⁻¹, resolution 1 cm⁻¹) and Raman spectroscopy (Thermo Fisher Scientific instrument, 455 nm laser/3 mW, 300–1300 cm⁻¹, resolution 1 cm⁻¹, PeakFit Gaussian fitting). Additionally, the micro-morphology and elemental distribution of the ash/slag were analyzed using a Hitachi SU-1510 SEM-EDS.

1.4 Quantitative analysis method for heavy metals

Based on USEPA Method 29, heavy metal content in ash samples was determined using a microwave digestion and ICP-MS. Approximately 0.1 g of sample was weighed into a polytetrafluoroethylene digestion vessel, followed by the addition of 4 mL concentrated nitric acid. Microwave digestion was performed at 280 °C until the solution became clear. The digestion vessel was then placed on a 150 °C graphite hotplate for acid evaporation until 1.0–2.0 mL remained. After cooling, the solution was transferred to a 50 mL volumetric flask and diluted to the mark with distilled water. Heavy metal concentrations in the digestate were analyzed using ICP-MS.

The enrichment and retention characteristics of heavy metals in the slag were characterized by the Enrichment Ratio (ER) and Retention Ratio (RR), with the volatilized portion in the gas phase represented by the Volatilization Rate (1-RR). The yield of ash converted to slag is defined as Y , and the weight loss rate (1- Y) is the percentage mass loss during the ash heating process. The calculation formulas are as follows:

$$ER = \frac{C_x}{C} \quad (1)$$

$$Y = \frac{m_x}{m} \times 100\% \quad (2)$$

$$RR = \frac{C_x m_x}{C m} = ER \times Y \quad (3)$$

where C_x is the heavy metal concentration in the slag (mg/kg); C is the heavy metal concentration in the ash (mg/kg); Y is the yield of ash converted to slag (%); m_x is the mass of the slag sample after melting (g); m is the mass of the ash before melting (g).

1.5 Solid leaching toxicity analysis

The solid leaching toxicity analysis was conducted according to the GB 16889—2008 standard (using the HJ/T 300—2007 acetic acid buffer solution method). The corresponding ash/slag samples were dried at 105 °C for 2 h to determine moisture content (Note: These dried samples were not used for the leaching experiment). Leachant selection: 5 g of sample was weighed, mixed with 96.5 mL of pure water, and stirred for 5 min to measure pH. If pH <

5, leachant #1 was used (5.7 mL glacial acetic acid + 64.3 mL 1 mol/L NaOH diluted to 1 L, pH = 4.93 ± 0.05). If pH > 5, 3.5 mL of 1 mol/L hydrochloric acid was added, heated at 50 °C for 10 min, and pH was measured again. If pH < 5, leachant #1 was used; otherwise, leachant #2 was used (17.25 mL glacial acetic acid diluted to 1 L, pH = 2.64 ± 0.05). Leachant #1 was used in this experiment. The leachant was added at a liquid-to-solid ratio of 20:1 based on the moisture content, followed by end-over-end agitation (30 ± 2 r/min, 18 ± 2 h). After standing for 20 min, the mixture was vacuum-filtered through a 0.45 μ m membrane filter. The filtrate was treated with 4–5 drops of 1:1 nitric acid and stored refrigerated. Heavy metal concentrations in the leachate were determined using ICP-MS and compared against the limit standards of GB16889—2008.

2 Results and discussion

2.1 Effect of rice husk addition on the ash fusion behavior of landfilled municipal solid waste

The ash fusion temperatures of the mixed samples are related to the amount of the same biomass added^[41]. Since rice husk itself is a lightweight biomass with low density and large volume, an excessively small blending amount can easily lead to uneven combustion^[42]. Therefore, an incremental addition of 5% for investigation was employed in this study. Figure 3 shows the ash fusion temperatures of samples with different proportions of rice husk addition under a weakly reducing atmosphere. It can be seen from the figure that as the proportion of rice husk increases, DT, ST, HT and FT are all lower than those of the original sample. FT shows a trend of first decreasing and then increasing, while DT, ST and HT show a more pronounced decrease at 5% and 10%. DK05 exhibits the lowest values for all four fusion characteristic temperatures, with an FT of 1213 °C. Combined with Table 3, the SiO₂ introduced by 5% addition of rice husk reacts with the abundant CaO, Fe₂O₃ and K₂O in the waste ash, forming low-melting eutectic phases such as anorthite (CaAl₂Si₂O₈) and KCl/K₂SO₄, significantly lowering the initial melting temperature. When the addition ratio exceeds 15%, an increase in melting point occurs. This is attributed to an excessively high silicon-to-aluminum ratio, causing low-melting silicates like anorthite to form a viscous liquid at high temperatures, hindering ash flow and leading to secondary slagging^[43]. Additionally, SiO₂ forms a high-melting quartz phase (melting point 1713 °C), diluting the flux concentration. Meanwhile, the absolute amount of alkaline oxides decreases, which is insufficient to generate an adequate amount of low-temperature eutectic melt.

Thermodynamic simulations calculated the solid and liquid phase fractions and mineral compositions of the five ash samples under equilibrium conditions from 800–1500 °C in a weakly reducing atmosphere, with results shown in Figure 4.

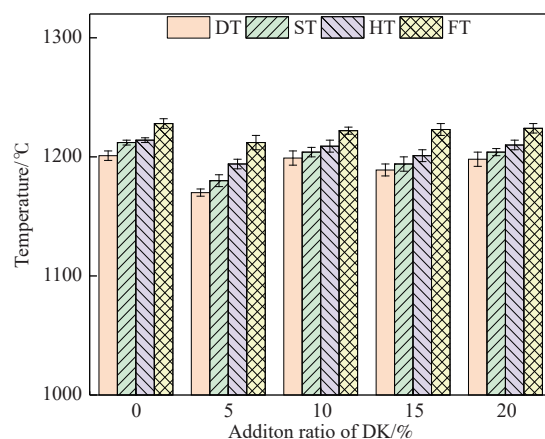


Fig. 3 Ash fusion temperatures of samples with different proportions of rice husk additives

The results indicate that DK05 contains KCaCl₃ and FeS chloride/sulfide eutectics at 800 °C, which can lower the DT. At 1150 °C, the co-melting of anorthite and fayalite causes the molten slag phase to surge to 71.98%, contributing to a lower fusion temperature. For DK00, perovskite with high-melting point remains stable before 1100 °C. At 1200 °C, pyroxene/anorthite still dominate, and due to the relatively high CaO content, it reacts with SiO₂ to form high-melting wollastonite (CaSiO₃). For DK20 and DK15, the flux concentration is diluted due to their excess SiO₂. The higher amount of free SiO₂ and the high-melting quartz network hinder the expansion of the low-temperature melt^[44].

The XRD patterns of the five samples after co-combustion are shown in Figure 5. It can be seen that the minerals contained in the DK00 sample include quartz (SiO₂), calcite (CaCO₃), plagioclase (Ca₂Al₂SiO₇), perovskite (CaTiO₃), etc. As the proportion of rice husk increases, the diffraction peaks of other crystals besides SiO₂ decrease significantly. Both plagioclase (Ca₂Al₂SiO₇) and perovskite (CaTiO₃) have relatively high melting points, which is the reason for the higher ash fusion temperatures observed in Figure 3. As shown in Figure 6, when the temperature reaches 1400 °C, the slags are completely molten because it exceeds the flow temperatures of all five slags. It can be clearly observed that the slags are predominantly amorphous substances, but high-melting silicate minerals such as spinel, cordierite, and åkermanite still exist.

2.2 Effect of rice husk addition on heavy metal distribution in ash/slag from landfilled municipal solid waste

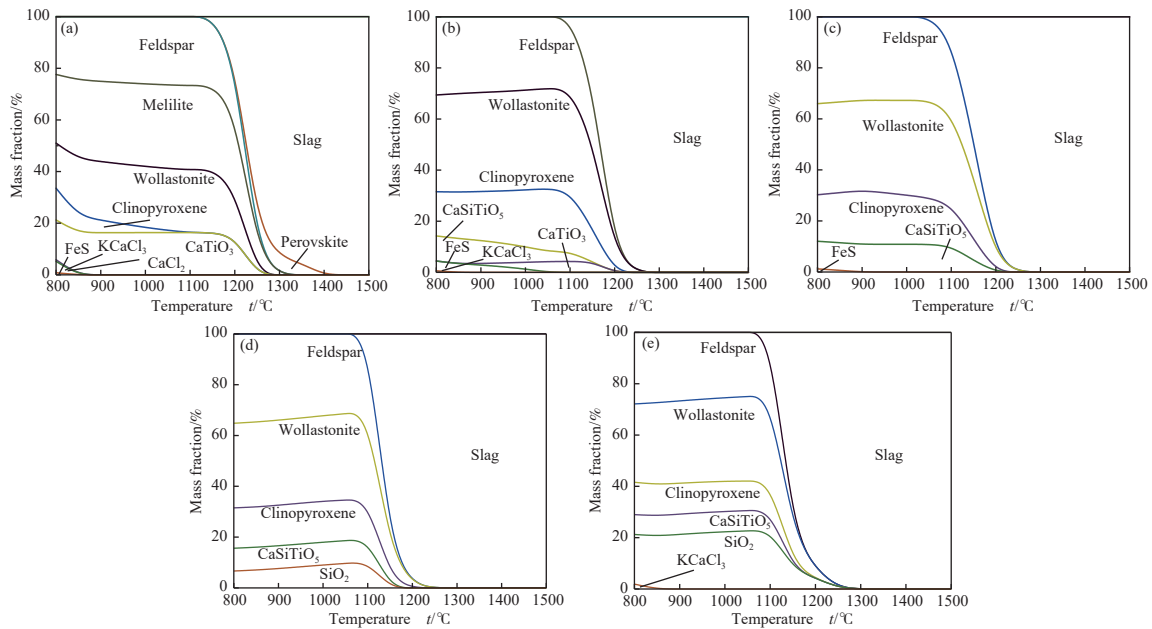
Heavy metal elements migrate to varying degrees during the combustion of landfilled waste and become enriched in the ash/slag. To better understand the enrichment characteristics of heavy metal elements in the solid-phase products, the Relative Enrichment Factor (Re)^[45] is introduced for evaluation. Its expression is:

$$Re = \frac{C_{ash,i} \times A_d}{C_{QP,i}} \quad (4)$$

where $C_{ash,i}$ represents the mass concentration of heavy metal element i in the ash/slag (mg/kg); $C_{QP,i}$ represents the mass concentration of heavy metal element i in the raw waste sample (mg/kg); A_d represents the ash content (as received basis) of the raw sample (%). Here, i represents heavy metal elements Pb, Cd, Cr, Zn, Cu, Hg, As. The contents of several heavy metal elements and their relative enrichment factors are shown in Table 4. Cu and Zn have higher contents due to their widespread pollution sources^[46], while others such as Pb, Cd, Cr, As also have certain contents in the raw material and ash. Due to its low proportion in the overall waste and high volatility, Hg content in ash is extremely low. Benefiting from the high-temperature slagging gasification technology, some heavy metal elements have already formed stable mineral phases^[47] with silicates, aluminates, or iron oxides in the

ash/slag during high-temperature combustion. The relative enrichment factors in the ash/slag follow the order: As < Zn < Cr < Cu < 1.

Table 5 presents the heavy metal leaching toxicity of the five ash/slag samples. It can be found that after adding rice husk, the heavy metal leaching concentrations are all lower than those of the original sample, with DK05 showing the greatest reduction. Since the leaching concentrations of heavy metals such as As, Cd and Hg are extremely low and it is lack of statistically significant differences, the three high-content heavy metal elements: Cr, Cu and Zn were focused in this study. Through three sets (A, B, C) of melting experiments (QP ash with different rice husk ratios, melted at 1300, 1400 and 1500 °C, respectively), the heavy metal content in the slags was detected to explore the effects of melting temperature and rice husk addition on the enrichment and retention behavior of heavy metals in the ash/slag.



(a): DK00; (b): DK05; (c): DK10; (d): DK15; (e): DK20.

Fig. 4 Composition of ash minerals at different temperatures

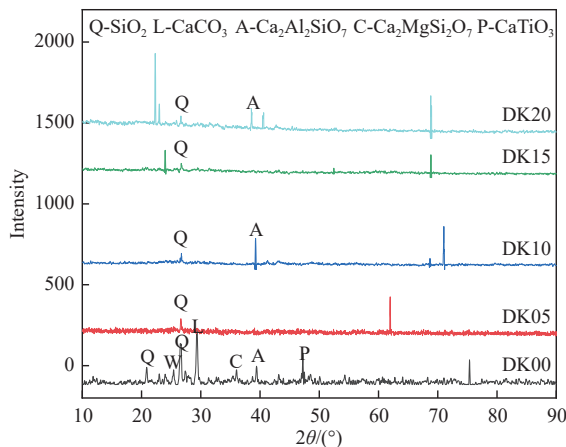


Fig. 5 XRD patterns of ash at 550 °C

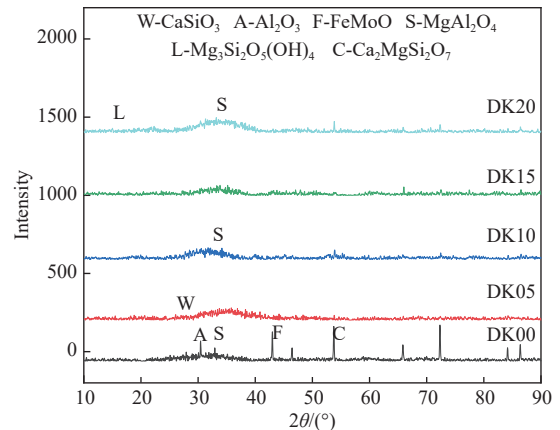


Fig. 6 XRD patterns of ash at 1400 °C

Table 4 Heavy metal content and relative enrichment factor in raw QP and its ash

	Sample	Cr	Cu	Zn	As	Pb	Cd	Hg
Element content/(mg·kg ⁻¹)	QP	11	22	1560	6.8	3.3	1.3	<0.1
	QP _{ash}	19	73	2028	6.1	3.1	1.1	<0.1
Relative enrichment technology(Re)		0.35	0.66	0.26	0.18	0.19	0.17	–

Table 5 Leaching toxicity of heavy metals

Element	Sample	Cr	Cu	Zn	As	Pb	Cd	Hg
Element content/(mg·L ⁻¹)	DK00	41.6	50.0	108.0	1.19	<0.1	<0.1	<0.1
	DK05	16.7	42.9	35.2	0.2	<0.1	<0.1	<0.1
	DK10	6.68	10.8	9.6	<0.1	<0.1	<0.1	<0.1
	DK15	5.89	10.4	7.1	<0.1	<0.1	<0.1	<0.1
	DK20	5.97	18.3	5.2	<0.1	<0.1	<0.1	<0.1

Since increasing temperature promotes the migration of heavy metals to the gas phase^[48], the temperature range with a high weight loss rate usually corresponds to the peak of heavy metal volatilization^[49]. As shown in Figure 7, when the melting temperature is constant, the weight loss rate ranges between 12%–19%, and it decreases as the rice husk addition ratio increases. When the rice husk addition ratio is the same, the difference in weight loss rate at different melting temperatures is relatively small. The high weight loss rate at high temperatures can promote the vitrification of ash/slag, encapsulating heavy metals within the aluminosilicate matrix and reducing leaching toxicity^[50]. This indicates that compared to melting temperature, the rice husk content has a greater influence on the ash weight loss rate, and rice husk promotes the vitrification of the ash/slag.

The Enrichment Rate (ER) characterizes the degree of heavy metal enrichment in the slag^[37], reflecting the relative relationship between its volatilization and mass loss. The results in Figure 8 show that for Cu and Zn elements, when the rice husk addition ratio is fixed, ER decreases with increasing melting temperature. When the melting temperature is fixed, ER increases with increasing rice husk addition ratio. The enrichment rate of Cu mainly fluctuates between 0.4–0.75, and that of Zn mainly between 0.37–0.69. For the Cr element, there is no significant correlation with either the rice husk ratio nor the melting temperature. Due to its high boiling point^[51] and stable chemical forms^[52], enrichment rate of Cr mainly fluctuates around 1.1. Under the same conditions, the enrichment intensity sequence is: Cr > Cu > Zn.

The Residual Rate (RR) characterizes the retention amount of heavy metals in the slag after high-temperature treatment^[53], reflecting their distribution between the solid and gas phases. Figure 9 shows the RR of Cu, Cr and Zn in the slag under different melting temperatures and rice husk addition ratios. The RR of Cu and Zn elements increases with higher rice husk addition ratios and decreases with higher temperatures. At the same rice husk addition ratio, RR changes little with temperature, indicating that rice husk content has a greater influence on RR than melting temperature.

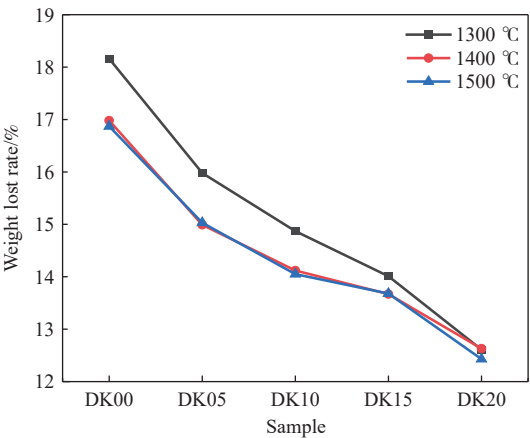


Fig. 7 Effect of melting temperature and rice husk addition ratio on weight loss rate of ash samples

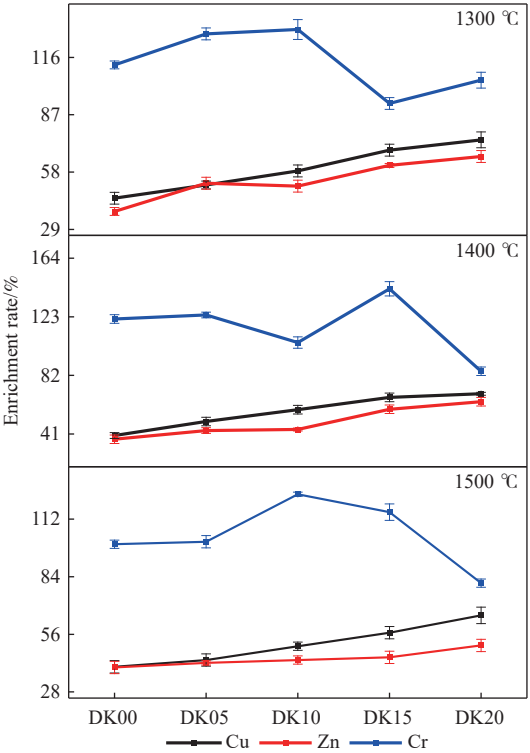


Fig. 8 Enrichment rate of heavy metals in molten slag

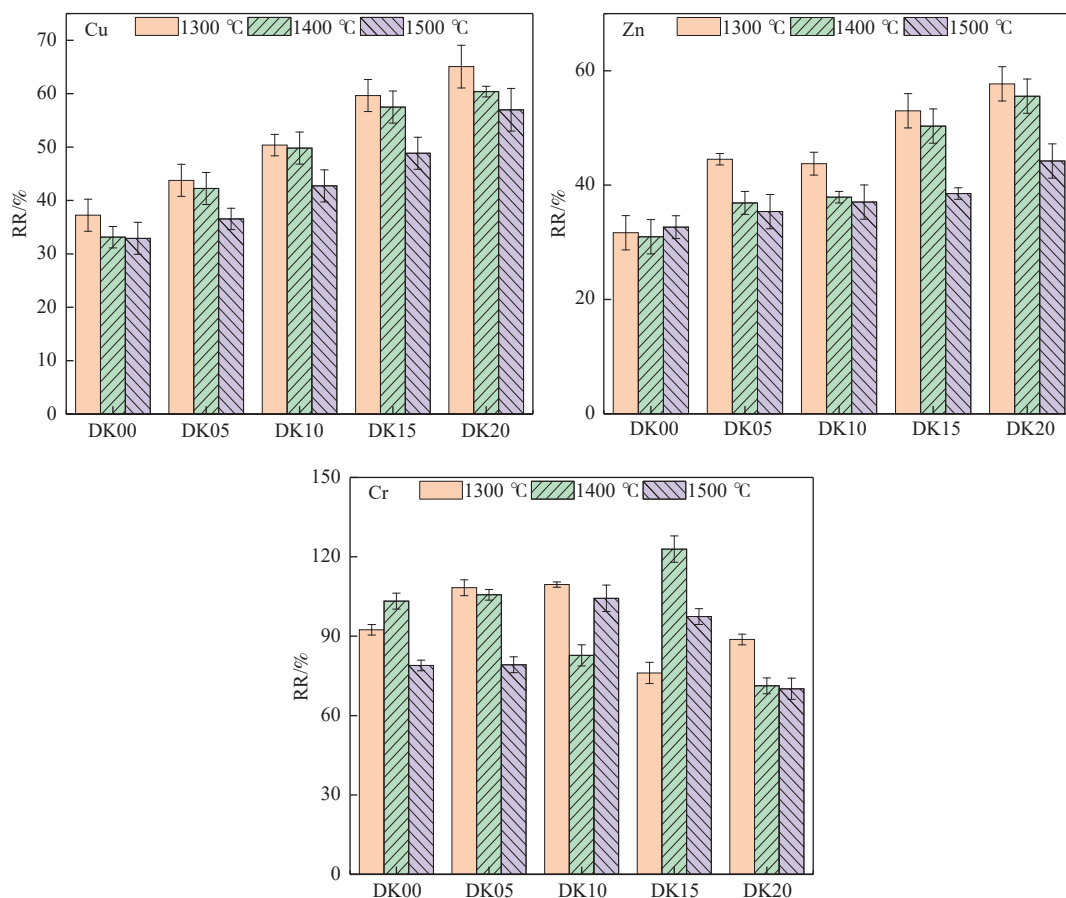
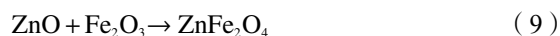
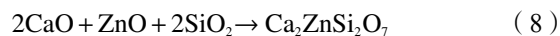
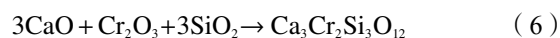
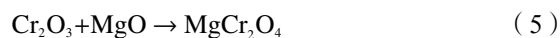


Fig. 9 Residual rate of heavy metals in molten slag

The RR of Cr shows no significant association with either the rice husk ratio nor the melting temperature. The RR ranges for Cu, Zn and Cr are 32%–65%, 30%–58% and 70%–123%, respectively. The distribution of RR range is related to elemental properties. Cu and Zn have good thermal stability and low volatility, while Cr tends to convert into stable oxides and is difficult to volatilize^[54], therefore, all three elements are mainly present in the solid-phase slag.

As shown in Figure 10, thermodynamic simulation calculations reveal that the Cr element tends to be immobilized in garnet ($\text{Ca}_3\text{Cr}_2\text{Si}_3\text{O}_{12}$) during low-temperature processes and tends to be effectively immobilized in the slag through the formation of spinel phases (e.g., MgAl_2O_4 , MgCr_2O_4 , FeCr_2O_4) at high temperatures. The concentration of gaseous CrCl_2 is low, indicating that most Cr is immobilized, which is consistent with the XRD analysis results. The Cu element mainly volatilizes in the form of gaseous CuCl , but the introduction of rice husk significantly reduces its gas-phase concentration, indicating that rice husk has a positive effect on inhibiting copper volatilization. In contrast, the behavior of Zn is relatively more complex. A low proportion of rice husk can suppress its volatilization to some extent, and Zn is immobilized by entering spinels. However, when the addition ratio exceeds 10%, due to the introduction of potassium elements, it promotes the formation of gaseous

double salt chlorides like $\text{KZnCl}_3/\text{NaZnCl}_3$, weakening part of immobilization ability of Zn in silicate minerals such as melilite. The SiO_2 -rich rice husk ash reacts with heavy metal oxides or chlorides to form relatively stable silicate compounds. Relevant reaction formulas are shown below:



The speciation distribution of heavy metals determines their ecological toxicity, and different occurrence forms of heavy metals bring different environmental impacts. According to the BCR method^[55], heavy metals are generally divided into acid-soluble fraction (F1), reducible fraction (F2), oxidizable fraction (F3) and residual fraction (F4). As shown in Figure 11, in the original ash, the occurrence forms of Cu, Zn, and Cr are all dominated by the acid-soluble fraction (F1), but there are some differences. The acid-soluble and reducible fractions are considered bioavailable because they can easily release into the environment^[56], hence called bioavailable forms.

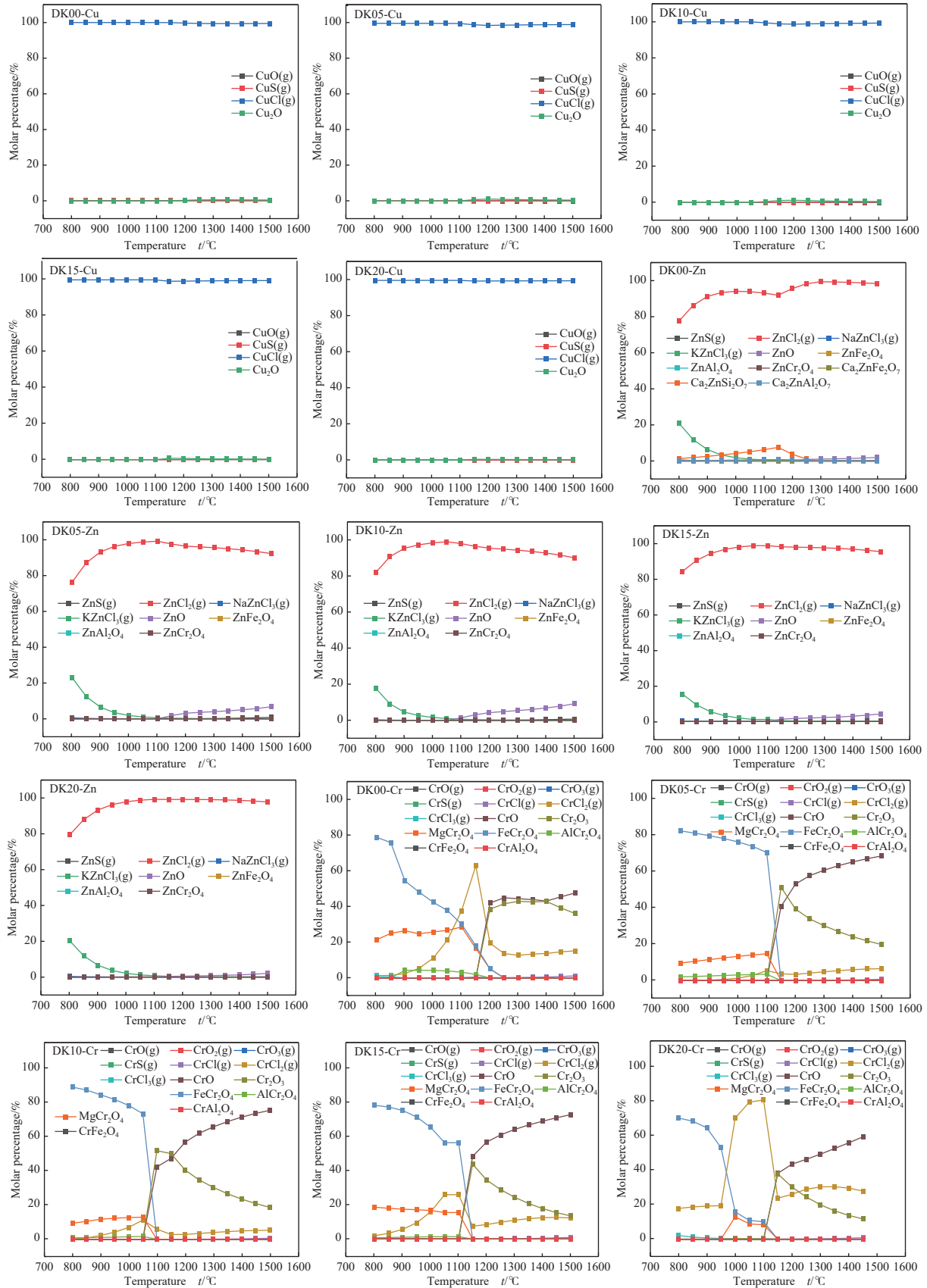


Fig. 10 Effect of rice husk addition ratio on the distribution patterns of heavy metal elements

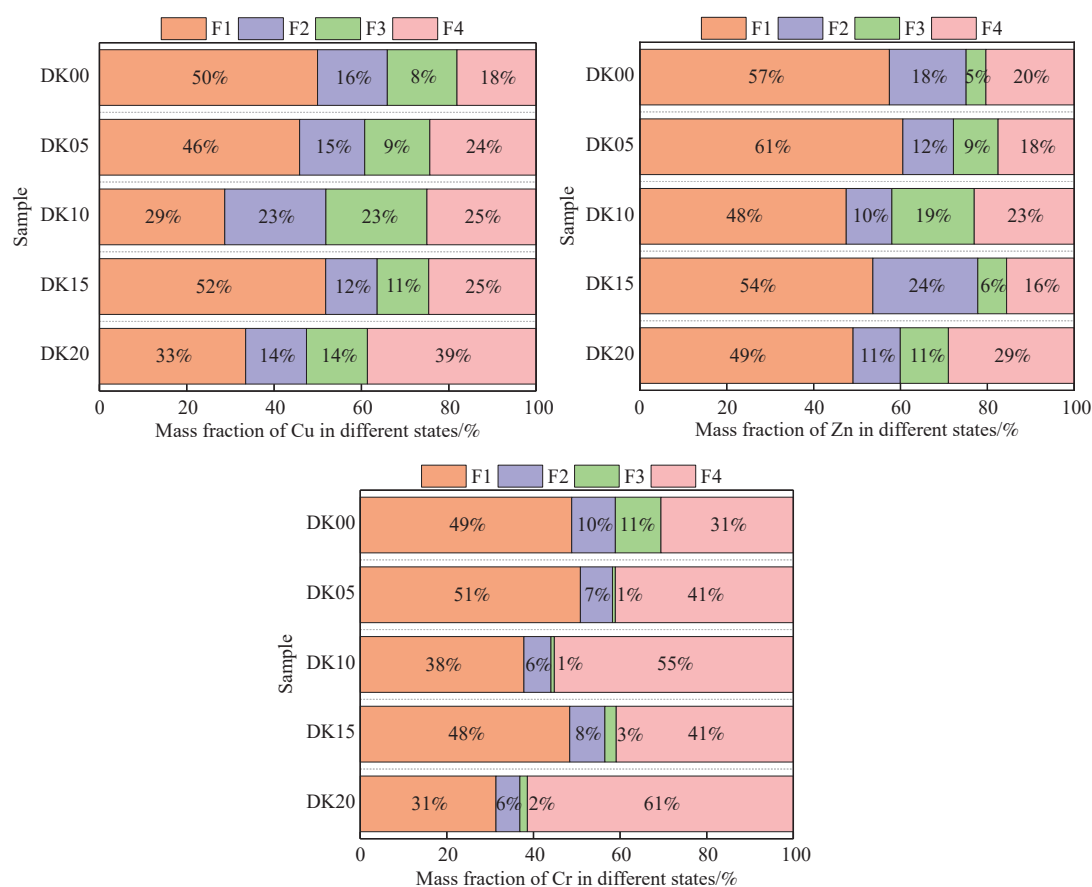


Fig. 11 Occurrence forms of heavy metal elements in five samples at 1400 °C

High-temperature treatment promotes the transformation of heavy metals into the residual fraction^[57], which is stable in nature and difficult for organisms to utilize^[58]. As the proportion of added rice husk increases, Cu, Zn and Cr all exhibit a certain degree of bioavailability and toxicity, but the residual fraction increases for all. The residual fraction is stable and not easily utilized by organisms. It can be seen that the immobilization effect is improved most significantly for Cr. For Cr, the proportion of its residual fraction (F4) continuously increases from 32.9% in the original sample to 61.8% at 20% blending ratio. This is mainly attributed to the similar ionic radii of Cr^{3+} and Si^{4+} , allowing Cr^{3+} to be efficiently immobilized via isomorphism into the newly formed silicate glass network structure. The growth of the residual fraction proportion for Zn is slow when the blending ratio is between 5%–15%, which is attributed to the dilution of fluxes (Fe_2O_3 , Al_2O_3), weakening the formation of spinel phases like zinc ferrite. When rice husk addition reaches 20%, the formation of the more stable willemite (Zn_2SiO_4) becomes dominant, leading to a significant increase in the residual fraction. The speciation distribution of copper is obviously affected by the competitive interference of chlorine. At the chlorine content peak (DK15, 4.26%), the proportion of the residual fraction for copper shows a decline, indicating that the volatilization-recondensation process of gaseous CuCl_2 leads to the enrichment of active forms. Combined with the previous leaching concentration analysis, when the addition

amount of rice husk is 20%, the leaching concentrations of some heavy metals increase. This should be due to the excessive dilution of CaO and Fe_2O_3 content, leading to insufficient formation of garnet and spinel phases. Excess Cr cannot be effectively immobilized in the slag, and the reduced alkalinity weakens the ability to immobilize chlorine, interfering with the speciation of Cu. However, the leaching concentrations of Zn and Cr still show a decrease at the 20% addition level. Based on this finding, the immobilization mechanism of physical encapsulation was further explored.

2.3 Mechanism analysis of heavy metal immobilization in slag by rice husk addition

The core of the slag structure is the network polymer structure composed of silicon-oxygen tetrahedra^[59]. To deeply explore the microscopic mechanism of heavy metal immobilization enhanced by rice husk, FT-IR and Raman spectroscopy were employed to analyze the silicate network structure of the slag^[60]. FT-IR was used to characterize the degree of polymerization in the liquid phase of the five samples at 1400 °C. The strongest absorption band in the 800–1300 cm^{-1} range corresponds to the symmetric stretching vibration of Si–O. The polymerization degree of the $[\text{SiO}_4]$ tetrahedral units is represented by Q^n , where n is the number of bridging oxygens (Si–O–Si) and reflects the

degree of liquid phase polymerization. Q^0 , Q^1 , Q^2 , Q^3 and Q^4 correspond to monomers ($[\text{SiO}_4]^{4-}$, Q^0), dimers ($[\text{Si}_2\text{O}_7]^{6-}$, Q^1), sheet structures ($[\text{SiO}_3]^{2-}$, Q^2), network structures ($[\text{Si}_2\text{O}_5]^{2-}$, Q^3), and three-dimensional networks (SiO_2 , Q^4), respectively. The characteristic wavenumber ranges for Q^0 – Q^4 are 840–890, 900–950, 960–1030, 1050–1100 and 1060–1190 cm^{-1} , respectively. Peaks between 800–1300 cm^{-1} were deconvoluted using Gaussian functions, where the proportion of Q^n is linearly related to the characteristic peak area. Table 6 lists the characteristic structures involved in peak fitting and their center positions. The ratio $(Q^0+Q^1+Q^2)/(Q^3+Q^4)$ (denoted as R) is defined as an evaluation index for liquid phase polymerization degree^[61]. A decrease in the R value indicates polymerization has occurred in the liquid phase. Figure 12 shows that as the proportion of rice husk increases, the degree of liquid phase polymerization improves, with the slag from the 20% rice husk addition sample exhibiting the highest polymerization degree.

Table 6 Characteristic absorption bands and their peak center positions in FT-IR^[62]

No.	Assignments	Wavenumber/ cm^{-1}
1	Si–O stretching vibration of $Q^0(\text{Si})$	~840–890
2	Si–O stretching vibration of $Q^1(\text{Si})$	~900–950
3	Si–O stretching vibration of $Q^2(\text{Si})$	~960–1030
4	Si–O stretching vibration of $Q^3(\text{Si})$	~1050–1100
5	Si–O stretching vibration of $Q^4(\text{Si})$	~1060–1190

As shown in Figure 13, Peak fitting was further performed on the Raman spectra to obtain the proportion of silicon-oxygen structures in the melt. Table 7 lists the characteristic structures involved in peak fitting and their center positions. Similarly, $Q^n(\text{Si})$ ($n=0\sim4$) represents the silicate tetrahedra ($[\text{SiO}_4]$), where n refers to the number of bridging oxygen atoms (BO) in the tetrahedron. A larger n indicates a higher degree of polymerization of the $[\text{SiO}_4]$ units^[63]. This confirms that the addition of rice husk overall increases the polymerization degree of the silicon-oxygen

structure in the slag, achieving physical encapsulation of heavy metal elements. It can thus be seen that when the rice husk addition is less than 15%, Cu and Cr elements are affected by both physical encapsulation and chemical immobilization, leading to decreased leaching concentrations. However, when the rice husk addition exceeds 15%, influenced by chemical immobilization factors, the leaching concentrations of Cu and Cr increase. For the Zn element, the leaching concentration continuously decreases with increasing rice husk proportion.

Combined with the previous thermodynamic equilibrium calculations and heavy metal content analysis, Figure 14 illustrates the mechanism diagram of heavy metal immobilization in gasification slag by rice husk addition. The core mechanism of heavy metal immobilization is shown as follow. On one hand, the active SiO_2 introduced by rice husk promotes the formation of low-melting eutectic phases (e.g., spinel, anorthite), improving slag fluidity and facilitating vitrification. It also significantly enhances the polymerization degree of silicate tetrahedra in the slag, forming highly polymerized silicate network structures (Q^3 , Q^4), thereby strengthening the physical encapsulation capacity for heavy metals. On the other hand, it involves the mineral phase transformation of heavy metals. As the rice husk proportion increases, the proportion of the residual fraction (F4) for Cu, Zn and Cr rises significantly, indicating their transformation from extractable forms to stable forms. Cr^{3+} enters spinel, garnet lattices, or iron oxide phases *via* solid solution; Zn^{2+} is chemically immobilized by entering spinels or replacing Ca^{2+} in silicates to form Zn_2SiO_4 ; Cu forms Cu_2O or silicate eutectic phases.

Table 7 Characteristic absorption bands and their peak center positions in Raman spectroscopy^[64]

No.	Assignments	Raman shift/ cm^{-1}
1	Si–O stretching vibration of $Q^0(\text{Si})$	~850
2	Si–O stretching vibration of $Q^1(\text{Si})$	~900–950
3	Si–O stretching vibration of $Q^2(\text{Si})$	~950–1020
4	Si–O stretching vibration of $Q^3(\text{Si})$	~1100
5	Si–O stretching vibration of $Q^4(\text{Si})$	~1150

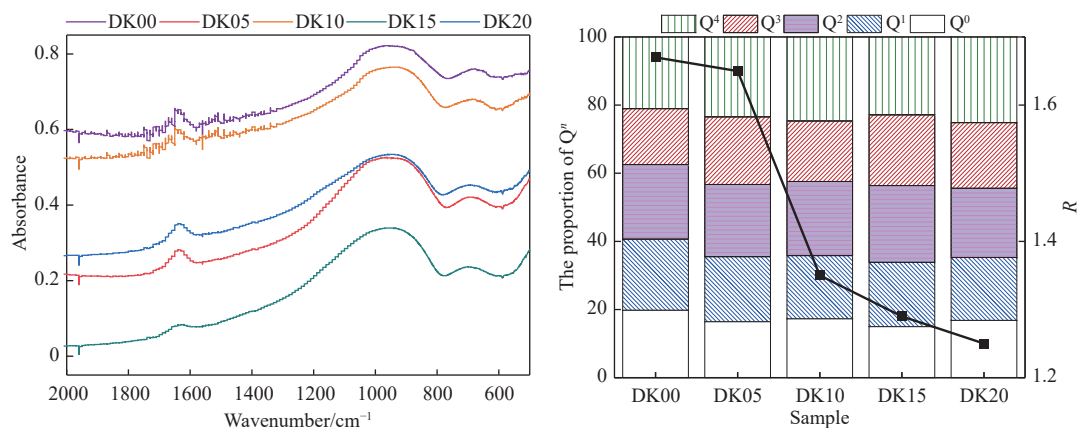


Fig. 12 FT-IR spectra and Q^n distribution of the five samples at 1400 °C

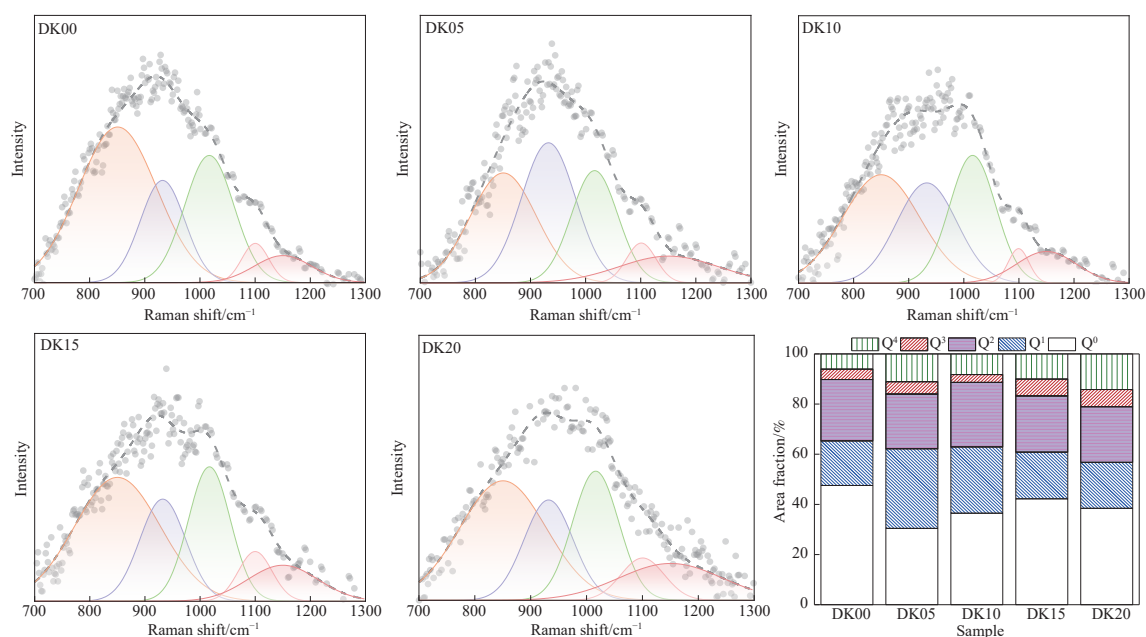


Fig. 13 Raman spectra and Q^n distribution of the five samples at 1400 °C

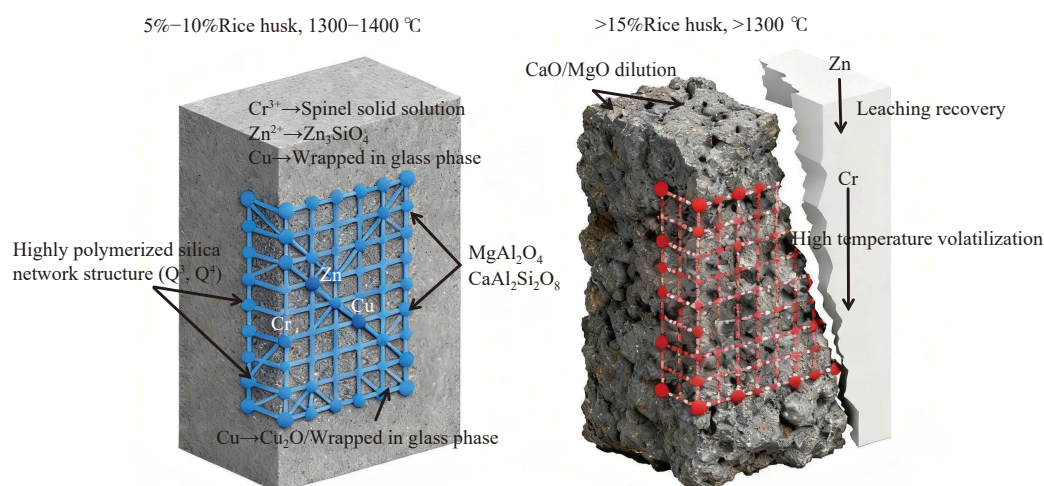


Fig. 14 Solidification mechanism diagram of rice husk addition on heavy metals in waste gasification slag

Leaching toxicity data further show that when adding 10%–15% rice husk, the leaching concentrations of Cr and Zn decrease from 41.60 and 108.00 mg/L to 5.89 and 7.10 mg/L, respectively, indicating that heavy metals are stably immobilized in mineral phases and are not easily extracted by acid.

In summary, the amount of rice husk addition exhibits a dual regulatory effect. An addition of 5%–10% promotes the formation of low-melting eutectic phases, enhances slag fluidity and the degree of vitrification, and strengthens the encapsulation and bonding capabilities for heavy metals. Conversely, an addition exceeding 15% leads to the dilution of alkaline oxides, which inhibits the expansion of the low-temperature melt. Furthermore, the introduced potassium elements promote the formation of gaseous double-salt chlorides such as $KZnCl_3/NaZnCl_3$, partially weakening the immobilization capacity within silicate minerals like

melilite. Simultaneously, the increased FeO content may compete for Cu^{2+} adsorption sites, resulting in an increase of Cu leaching. Additionally, the volatilization of Cu and Zn intensifies at high temperatures (>1400 °C), and their Residual Rates (RR) decrease with increasing temperature. Therefore, for practical solid waste resource utilization, the optimal process conditions are identified as 1300–1400 °C, balancing slag fluidity, heavy metal residual rate, and energy consumption, combined with a 5%–10% rice husk addition. This approach enables the comprehensive optimization of the slag towards high polymerization, low leaching, and high residual rates. These findings provide a theoretical basis for the synergistic treatment of landfilled waste and biomass. The slag co-treated with rice husk possesses the potential for safe resource utilization in applications such as construction materials and road base layers.

3 Conclusions

This study investigated the migration of heavy metals and the polymerization behavior of silicates in gasification slag from rice husk and landfilled municipal solid waste. In the gasification slag of landfilled waste, rice husk, as a silicon source, plays multiple roles in regulating melting behavior, strengthening silicate polymerized structures, and immobilizing heavy metals. The aim is to achieve efficient heavy metal immobilization, slag vitrification, and syngas recovery through the co-gasification technology of landfilled waste and rice husk, providing a feasible solution for the clean and efficient resource treatment of stockpiled waste. The conclusions obtained are as follows:

Adding rice husk (5%–10%) can reduce the flow temperature of the slag, reaching a minimum of 1213 °C. This is mainly attributed to the active SiO₂ introduced by rice husk reacting with CaO and Fe₂O₃ in the waste ash to form low-melting eutectic phases such as anorthite, promoting the formation of low-temperature melts.

As the proportion of rice husk addition increases, the leaching concentrations of Cr, Zn and Cu in the slag significantly decrease. When adding 10%–15% rice husk, the leaching concentrations of Cr and Zn decrease from 41.60 and 108.00 mg/L to 5.89 and 7.10 mg/L, respectively. The amorphous SiO₂ provided by rice husk increases the polymerization degree of silicate tetrahedra in the slag, promotes the formation of highly polymerized silicate networks, enhances the physical encapsulation and structural immobilization capacity for heavy metals, and reduces non-bridging oxygen sites, thereby inhibiting heavy metal leaching.

The volatilization of Cu and Zn is enhanced at high temperatures (>1400 °C), with their residual rates decreasing to 33%–60% and 31%–55%, respectively, as temperature rises. In contrast, Cr, due to its high boiling point and stable chemical properties, maintains a residual rate of 70%–123%. Rice husk promotes the transformation of heavy metals from acid-soluble and reducible fractions to the residual fraction, enhancing their environmental stability. The SiO₂ introduced by rice husk promotes the formation of stable mineral phases for heavy metals, locking them within the crystal lattice and reducing their bioavailability and leaching risk.

Declaration of Competing Interest

The authors declare no conflict of interest.

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稻壳对填埋生活垃圾气化熔渣中硅酸盐聚合作用下 重金属迁移机制研究

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摘 要: 熔渣气化是填埋生活垃圾等高重金属含量有机固废清洁资源化利用的主要技术之一。以稻壳作为硅源, 研究了稻壳添加对填埋生活垃圾熔渣玻璃化及重金属固化行为。结果表明, 添加稻壳(5%–10%)可降低熔渣流动温度, 并有效抑制重金属浸出, 其中, Cr、Zn 浸出浓度分别由 41.60 与 108.00 mg/L 降至 5.89 与 7.10 mg/L。稻壳引入的活性 SiO₂ 促进了高聚合硅氧网络(Q³、Q⁴)的形成, 抑制气化迁移, 增强了对重金属的物理包封作用; 同时促使 Zn²⁺、Cr³⁺、Cu²⁺通过取代或固溶机制形成 Zn₂SiO₄、CuFe₂O₄ 等稳定矿物相, 固化在石榴石、尖晶石中, 提升其残渣态比例, 降低生物有效性。本研究揭示了稻壳在熔渣结构调控与重金属固化的关联作用机制, 为填埋垃圾和生物质“以废治废”清洁高效资源化处理提供理论支撑。

关键词: 填埋生活垃圾; 稻壳; 重金属固化; 气化熔渣; 灰渣

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