

Article

Study on the Synergistic Recovery of Zinc and Iron from Cold-Bonded Briquettes Prepared from High-Zinc Blast Furnace Dust

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Abstract

High-zinc blast furnace dust is a zinc-bearing solid waste generated during ironmaking. Efficient de-zincing and iron enrichment are required for its resource utilization. This study investigated the high-temperature reduction behavior and kinetic transition mechanism of cold-bonded briquettes made from high-zinc blast furnace dust with a small addition of iron ore powder, with particular emphasis on the effects of reduction temperature (1000–1200 °C) and holding time (10–60 min). The results show that reduction at 1200 °C for 60 min can effectively remove zinc and enrich iron. The de-zincing rate reached 92%, and the TFe grade increased to 50 wt.%, achieving the goal of efficiently removing zinc while improving the TFe grade of the reacted briquettes. During the middle and later stages of reduction (1100–1200 °C, 30–60 min), the content of newly formed metallic iron increased, which restored the briquette strength to 524 N after reduction. In addition, the reduction kinetics of the system evolved from interfacial chemical reaction control in the initial stage to three-dimensional internal diffusion control in the middle and later stages. These results provide a theoretical basis and technical reference for the resource utilization of high-zinc blast furnace dust.

Keywords: high-zinc blast furnace dust; cold-pressed briquette; high-temperature reduction; kinetic analysis

1. Introduction

Blast furnace dust is one of the main by-products generated during ironmaking. As shown in Figure 1, according to the World Steel in Figures published in recent years by the World Steel Association, global blast furnace iron production has remained stable at approximately 1.2 to 1.3 billion tons per year [1]. Given that the generation of blast furnace dust accounts for about 1% to 3% of hot metal production, its annual discharge has reached tens of millions of tons [2]. Blast furnace dust has a complex phase composition. It is not only rich in valuable elements such as iron and carbon, but also contains harmful elements such as zinc and alkali metals [3]. If it is stored in the open air or landfilled directly, it will lead to the waste of iron and carbon resources and cause pollution to the ecosystem [4]. If it is directly added to sinter feed for in-plant recycling in order to utilize its carbon content, zinc will continuously accumulate in the blast furnace, resulting in refractory erosion and deterioration of burden permeability [5]. This problem becomes more severe when the zinc content in blast furnace dust is high. Therefore, de-zincing treatment is necessary. In this study, such blast furnace dust is referred to as high-zinc blast furnace dust.



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At present, the industrial treatment of high-zinc blast furnace dust mainly relies on pyrometallurgical processes, such as the rotary kiln process and the rotary hearth furnace process. These processes can efficiently remove zinc by volatilization and recover iron in the form of direct reduced iron (DRI) [6]. However, owing to differences in origin and processing conditions, the zinc content in different types of blast furnace dust varies considerably [7]. For high-zinc blast furnace dust, the relatively low TFe grade of the raw material, together with the retention of gangue components such as silicon oxide and aluminum oxide in the de-zincing residue after reduction, results in a low TFe grade of the residue. As a result, it cannot directly meet the requirements for subsequent use as an ironmaking raw material [8,9] and eventually becomes a secondary solid waste.

To address this problem, some studies have begun to use iron ore powder and zinc-containing dust as mixed raw materials to prepare cold-bonded briquettes for high-temperature reduction. Through room-temperature compaction and binder curing, the briquette process can convert powder materials into cold-bonded briquettes with high strength. This approach can not only reduce dust generation and material loss during direct charging [10], but also has the potential to improve the TFe grade and strength of the de-zincing residue when iron ore powder is added. In this way, the residue can be directly converted into an ironmaking raw material suitable for furnace charging, which is beneficial for improving the resource utilization efficiency of high-zinc blast furnace dust. Mousa [11] and Mantovani et al. [12] found that the addition of iron ore powder can not only improve the particle size distribution and briquetting performance of the mixture, but also increase the TFe grade of the reacted briquettes, thereby improving their subsequent value as ironmaking raw material. The proposed approach of adding iron ore powder therefore provides a possible solution to this problem. Under experimental conditions with a small amount of iron ore powder, studying the preparation of cold-bonded briquettes from high-zinc blast furnace dust and their high-temperature reduction behavior is of great significance for improving the resource utilization of high-zinc blast furnace dust.

However, the reduction behavior and kinetic mechanism of this composite system at a high temperature have not yet been systematically investigated. Therefore, further study is necessary to provide a theoretical basis and data support for the resource utilization of high-zinc blast furnace dust.

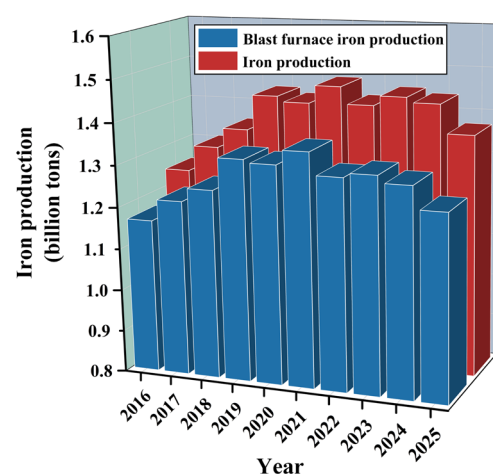


Figure 1. Global iron production and blast furnace iron production, 2016–2025.

2. Materials and Methods

2.1. Raw Material Composition

The high-zinc blast furnace dust used in this study was sourced from a steel enterprise in southern China, and the iron ore powder was obtained from another company in southern China. The chemical composition of the high-zinc blast furnace dust was jointly characterized using X-ray fluorescence spectrometry (XRF) and inductively coupled plasma spectroscopy (ICP), as shown in Table 1. The results indicate that this high-zinc blast furnace dust contains relatively high levels of zinc and iron, reaching 33.79 wt.% and 16.96 wt.%, respectively, classifying it as a high-zinc blast furnace dust. Its carbon content is also relatively high, at 10.01 wt.%.

The main chemical composition of the iron ore powder was determined from the manufacturer's test report and confirmed by repeated measurements in this study. The results were consistent within the experimental error range, as shown in Table 2. The results indicate that the Fe_3O_4 and FeO contents in the iron ore powder are 59.91 wt.% and 28.17 wt.%, respectively, indicating that it is magnetite powder. According to the manufacturer's report, all particles in this iron ore powder are smaller than 0.074 mm.

The high-zinc blast furnace dust was analyzed using a D8 Advance X-ray diffractometer (XRD, Bruker AXS GmbH, Karlsruhe, Germany). The raw material was ground to below 0.074 mm and then prepared by the back-pressing method. Continuous scanning was conducted over a 2θ range of 10° to 80° , with a step size of 0.01° and a dwell time of 0.12 s per step. Phase identification was performed using HighScore Plus software (v. 3.0.5), and the results are shown in Figure 2. The XRD analysis indicates that the main phases in the high-zinc blast furnace dust are zinc oxide (ZnO), hematite (Fe_2O_3), and graphite (C), together with minor quartz (SiO_2) and corundum (Al_2O_3).

Table 1. Main chemical components of high-zinc blast furnace dust.

Components	Zn	K ₂ O	Na ₂ O	C	Cl	Fe	SiO ₂	CaO	MgO	Al ₂ O ₃
wt.%	33.79	1.26	0.55	10.01	4.28	16.96	4.32	2.04	3.58	1.48

Table 2. Main chemical components of iron ore powder.

Components	TFe	Fe ₃ O ₄	FeO	SiO ₂	CaO	Al ₂ O ₃	TiO ₂	MgO	S	P
wt.%	65.25	59.91	28.17	7.98	0.16	0.16	0.01	0.38	0.024	0.0078

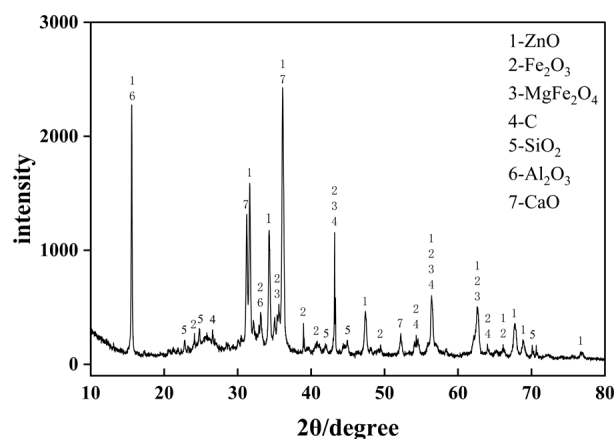


Figure 2. XRD results of high-zinc blast furnace dust.

The particle size distribution of the high-zinc blast furnace dust was determined using 200-mesh and 30-mesh sieves, and its micro-morphology was examined by SEM, as shown in Figure 3. The results show that the particles were mainly concentrated in the 0.074–0.5 mm and <0.074 mm size fractions, with mass fractions of 61.05 wt.% and 27.65 wt.%, respectively. Overall, the high-zinc blast furnace dust had a fine particle size and a relatively narrow size distribution.

In addition, its morphology was complex and varied. Most particles were irregular flakes, some exhibited rod-like forms with intergrown structures, and others appeared as agglomerates formed by fine particles. These irregular particles were intertwined and stacked together, forming a complex microporous structure. During cold compaction, such a structure facilitates particle displacement and rearrangement, plastic deformation, and mechanical interlocking under compaction pressure, thereby increasing the strength of the cold-bonded briquette [13].

The coke powder was obtained from screened material supplied by an ironmaking plant in China. Its proximate analysis results were determined according to the national standard GB/T 212-2008 [14], Method for Proximate Analysis of Coal, and are listed in Table 3. The results show that the fixed carbon (FCd), volatile matter (Vd), and ash content (Ad) of the coke powder were 86.89%, 1.70%, and 11.41%, respectively. To prevent excessive particle size from affecting cold-bonded briquette preparation, the coke powder was mechanically crushed and sieved before use to obtain a uniform particle size below 0.5 mm.

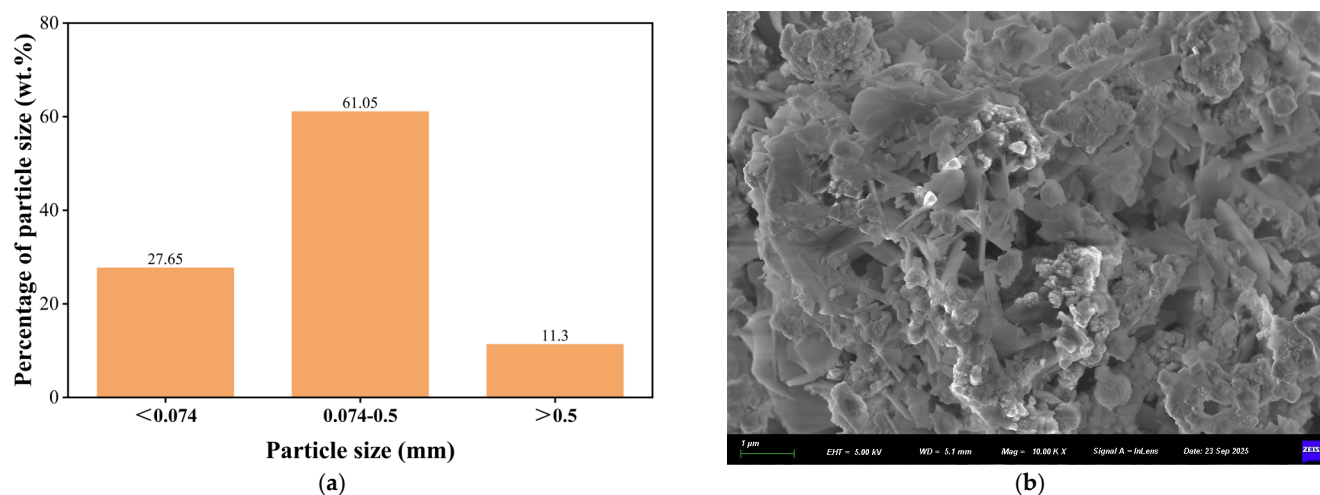


Figure 3. (a) Particle size sieving results of high-zinc blast furnace dust; (b) SEM scan image.

Table 3. Industrial analysis results of coke powder.

Components	FCd	Vd	Ad
wt.%	86.89	1.70	11.41

2.2. Experimental Procedure

2.2.1. Cold-Bonded Briquette Preparation Process

The present study used the cold-bonded briquette process (Figure 4) to agglomerate high-zinc blast furnace dust into briquettes. This process offers the advantages of low energy consumption and significant environmental benefits [15]. In addition, briquettes with high strength can provide a physical framework for subsequent conversion into iron ore products with a certain level of strength.

Based on the preliminary mass balance calculations and forming-mechanics verification, the optimal briquetting parameters were determined as follows: 73 wt.% blast furnace

dust, 9 wt.% iron ore powder, 4 wt.% coke powder, 9 wt.% composite binder, and 5 wt.% moisture, followed by drying at 200 °C for 120 min. Under these conditions, the compressive strength of the briquettes reached 2359 N. This raw material ratio can theoretically raise the TFe grade of the briquettes after reaction to above 48 wt.%.

2.2.2. High-Temperature Reduction Experimental Procedure

Figure 5 shows the schematic of the high-temperature reduction experiment. First, an electronic balance and XRF were used to record the briquette mass and the zinc and iron contents before charging into the furnace. The briquettes were then placed in an alumina crucible, covered with a lid with a small hole, and put into the furnace chamber. The target temperature was set (1000, 1050, 1100, 1150 and 1200 °C), and heating started in an air atmosphere. After the furnace reached the target temperature in 30 min, the holding time was counted, and the sample was kept at that temperature for the specified period (the holding times were set at 10, 20, 30, 40, 50, and 60 min). After the experiment, the furnace was turned off. Once the briquettes cooled to room temperature, they were removed, weighed, and tested for compressive strength. Finally, the zinc and iron contents of the reacted briquettes were analyzed.

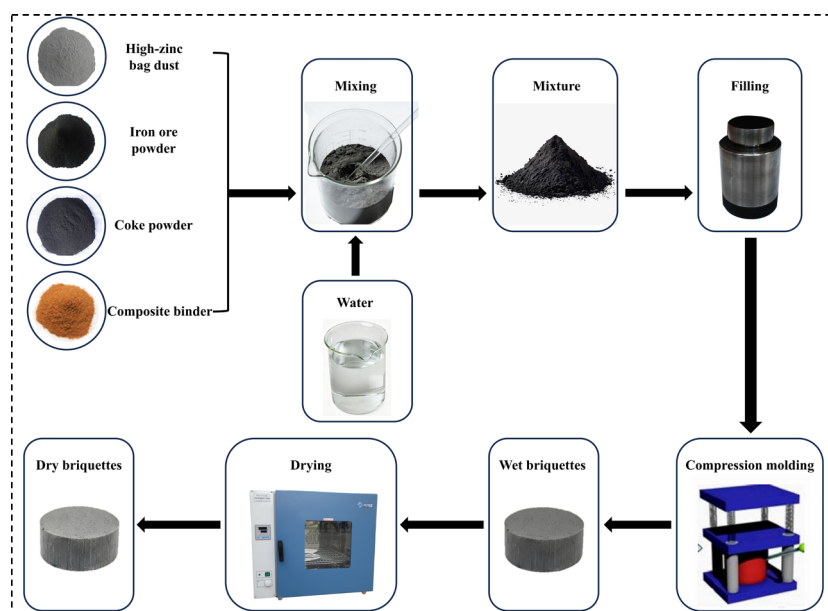


Figure 4. Cold-bonded briquette preparation process.

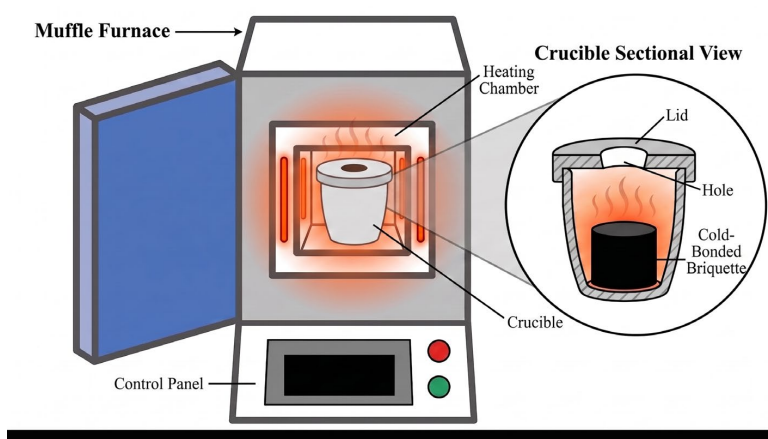


Figure 5. Schematic diagram of the high-temperature reduction experiment.

2.3. Performance Testing Methods

2.3.1. Post-Reaction Compressive Strength

The post-reaction compressive strength of the briquettes was measured using a WDW-20 microcomputer-controlled electronic universal testing machine (Shanghai Bai-ruo Testing Instrument Co., Ltd., Shanghai, China). Since one objective of this study is to use the post-reaction briquettes as iron ore in subsequent ironmaking processes, their compressive strength must satisfy industrial requirements. In related research, 500 N is adopted as one of the strength thresholds for qualified metallized pellets [16]; therefore, we set the target strength for the post-reaction briquettes at above 500 N per briquette. The testing procedure was as follows: ten post-reaction briquettes cooled to room temperature were selected, and the maximum compressive strength before fracture was measured using the universal testing machine. The result was reported as the arithmetic mean.

2.3.2. Mass Loss Ratio

Mass loss ratio is a key indicator of the extent of the reduction reaction, and it is calculated using Equation (1):

$$\omega_{weight} = \frac{m_1 - m_2}{m_1} \quad (1)$$

where m_1 is the mass of the cold-bonded briquette before reaction (g), and m_2 is the mass of the post-reaction briquette (g).

2.3.3. TFe Grade

The zinc content and TFe grade were measured using an M4 Tornado high-performance micro-X-ray fluorescence spectrometer (Bruker Nano GmbH, Berlin, Germany). To obtain representative samples, multi-point sampling was applied to the ten post-reaction briquettes from each high-temperature reduction experiment. A small amount of sample was drilled from the top, center, and bottom of each briquette, yielding a total of 30 sampling points. The collected samples were ground into powder with a particle size smaller than 0.074 mm, pressed into pellets without a binder, and then tested in an air atmosphere. During testing, ten points were randomly selected on the pellet surface for analysis, with an acquisition time of 60 s per point. The final result was reported as the average value to ensure accuracy.

This study adopts the requirements for Grade 5 iron ore products from the current national standard GB/T 32545-2016, Classification of iron ore products. According to this standard, the TFe grade of Grade 5 iron ore products ranges from 48.0 to 58.0 wt.% [17]. Accordingly, the target TFe grade range for the post-reaction briquettes is set at 48.0–58.0 wt.%.

2.3.4. De-Zincing Rate

The de-zincing rate is a core indicator for evaluating the effectiveness of zinc elimination during the reduction process. It is calculated using Equation (2), with the zinc contents in the cold-bonded briquette before and after reaction determined by XRF.

$$\omega_{zinc} = \frac{m_1 \cdot W_1 - m_2 \cdot W_2}{m_1 \cdot W_1} \quad (2)$$

In the equation, m_1 and m_2 represent the masses (g) of the cold-bonded briquette before reaction and the post-reaction briquette, respectively; W_1 and W_2 denote their corresponding zinc contents (wt.%).

2.3.5. Kinetic Models

Under isothermal conditions, the relationship between the reduction degree of a solid reaction and reaction time can be described by kinetic mechanism equations in their

integral form. In this study, nine solid-state reaction kinetic models suitable for cold-bonded briquette reduction, as listed in Table 4, were selected to fit the experimental data.

Table 4. Kinetic models and integral function expressions.

Mechanism Category	Model Code	$G(\alpha)$	No.
Interfacial chemical reaction control	R2	$1 - (1 - \alpha)^{\frac{1}{2}}$	(3)
	R3	$1 - (1 - \alpha)^{\frac{1}{3}}$	(4)
One-dimensional/ two-dimensional diffusion control	D1	α^2	(5)
	D2	$(1 - \alpha) \ln(1 - \alpha) + \alpha$	(6)
Three-dimensional internal diffusion control	D3	$[1 - (1 - \alpha)^{\frac{1}{3}}]^2$	(7)
	D4	$1 - \frac{2}{3}\alpha - (1 - \alpha)^{\frac{2}{3}}$	(8)
Nucleation and growth control	A2	$[-\ln(1 - \alpha)]^{\frac{1}{2}}$	(9)
	A3	$[-\ln(1 - \alpha)]^{\frac{1}{3}}$	(10)
Reaction order control	F1	$-\ln(1 - \alpha)$	(11)

After logarithmic transformation of the Arrhenius equation, the relationship between the reaction rate constant and temperature can be expressed as:

$$\ln k = \ln A - E_a/RT \quad (12)$$

where α is the de-zincing rate (%), k is the reaction rate constant (min^{-1}), A is the pre-exponential factor (min^{-1}), E_a is the activation energy (kJ/mol), R is the gas constant ($8.314 \text{ J/(mol}\cdot\text{K)}$), and T is the thermodynamic temperature (K).

By substituting α into the integral function expression $G(\alpha)$, the reaction rate constant k was obtained. The k values were then fitted to the logarithmic form of the Arrhenius equation (Equation (12)). Plotting the data against $1/T$ yielded a linear relationship (Arrhenius plot), and the apparent activation energy E_a was calculated from the slope ($-E_a/R$). The kinetic model with the highest coefficient of determination (R^2) was identified as the rate-controlling mechanism.

3. Results

3.1. Thermodynamic Analysis

FactSage 7.2 software was employed to calculate the Gibbs free energy change (ΔG) and enthalpy change (ΔH) of the direct and indirect reduction reactions in the cold-bonded briquette system. The results are presented in Figure 6. As shown in Figure 6a,b, the ΔG values of all reactions in the carbothermic direct reduction system decreased with increasing temperature. Compared with iron oxides, the direct reduction of ZnO required a higher initial reduction temperature of 951°C and exhibited a significant endothermic effect, with a ΔH value of approximately 380 kJ/mol . As shown in Figure 6c,d, the thermal effects in the CO indirect reduction system were markedly lower, and the ΔH values of most reactions were below 200 kJ/mol . The reduction of iron oxides could proceed below 569°C , whereas the indirect reduction of ZnO by CO required temperatures above 1308°C .

Based on the thermodynamic calculations presented above, the reduction of iron oxides in the system primarily depends on CO indirect reduction, which offers more favorable thermodynamic conditions, whereas the reduction of ZnO exhibits a pronounced temperature dependence. The overall reduction process requires a high temperature of 1000°C or above. It relies on the carbon gasification reaction to sustain a locally high CO partial pressure and is accomplished through the synergistic effect of direct and indirect

reduction. Accordingly, the high-temperature reduction experiments were conducted at 1000 °C or above. This temperature regime promotes ZnO reduction and iron enrichment, raises the TFe grade of the post-reaction briquettes, and makes them suitable for subsequent use as ironmaking feedstock.

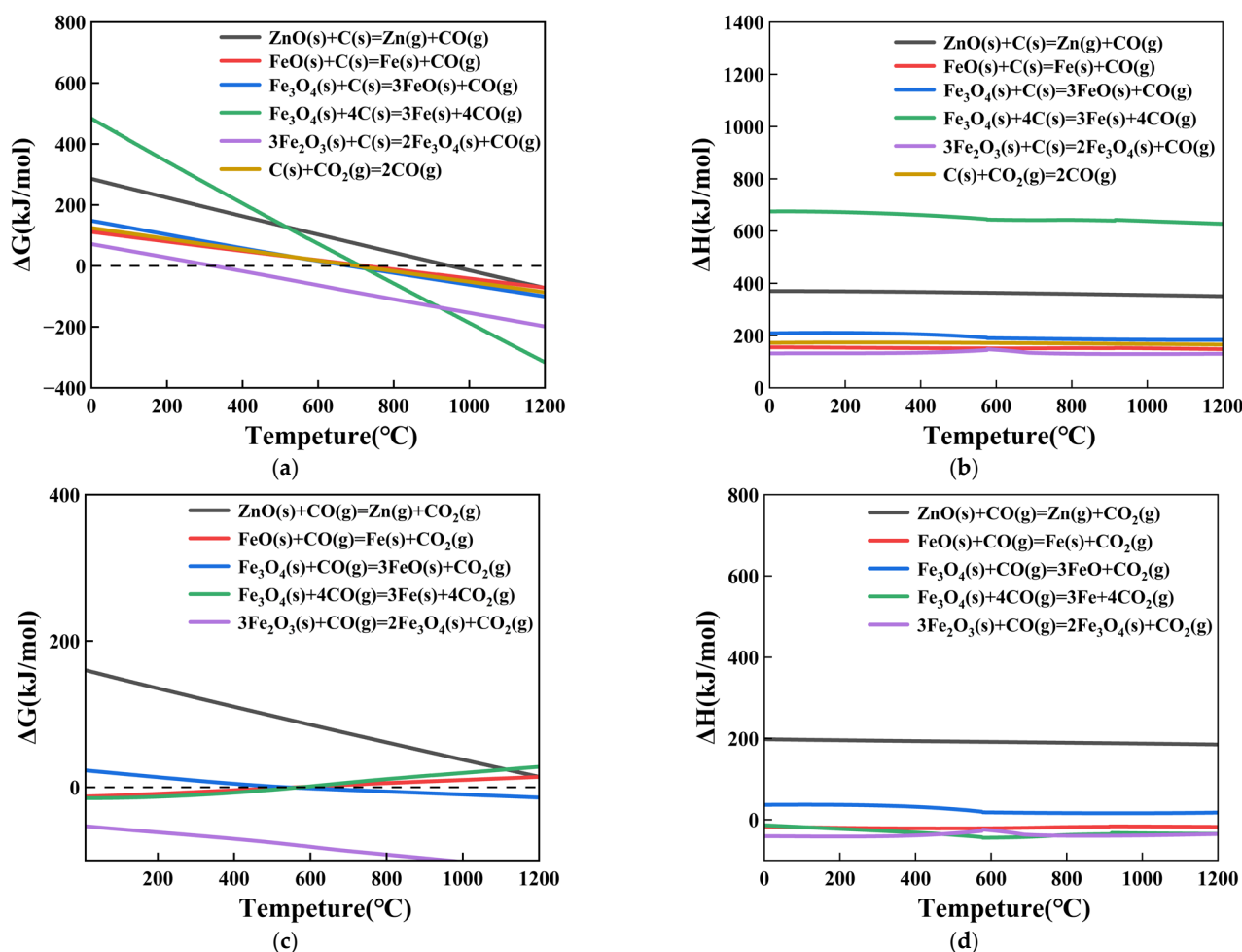


Figure 6. Thermodynamic parameters of the system reaction: (a) Direct reduction—Gibbs free energy change ΔG ; (b) Direct reduction—enthalpy change ΔH ; (c) Indirect reduction—Gibbs free energy change ΔG ; (d) Indirect reduction—enthalpy change ΔH .

3.2. Effect of Reduction Temperature on the High-Temperature Reduction Behavior of Cold-Bonded Briquettes

Cold-bonded briquettes were prepared with the optimal composition: 73 wt.% blast furnace dust, 9 wt.% iron ore powder, 4 wt.% coke breeze, 9 wt.% composite binder, and 5 wt.% moisture. After drying at 200 °C for 120 min, the briquettes exhibited a compressive strength of 2359 N. These briquettes were then placed in a muffle furnace under an air atmosphere and held at different temperatures for various durations. The aim was to investigate the effects of reduction temperature and reduction time on the mass loss ratio, post-reaction compressive strength, de-zincing rate, and TFe grade of the cold-bonded briquettes. Figure 7 illustrates the influence of temperature (1000–1200 °C) on the high-temperature reduction behavior at a fixed reduction time of 30 min. The experimental results show that as the reduction temperature increased from 1000 to 1200 °C, the mass loss ratio rose sharply from 17% to 56%, the post-reaction compressive strength increased from 79 N to 465 N, the de-zincing rate reached 82%, and the TFe grade was enriched to 45 wt.%. Reduction temperature significantly affected the high-temperature reduction

behavior: despite the substantial increase in mass loss, the post-reaction strength did not decline but instead improved. According to findings by other researchers, this likely results from sintering and bonding between newly formed metallic iron particles, which enhances the strength [18,19].

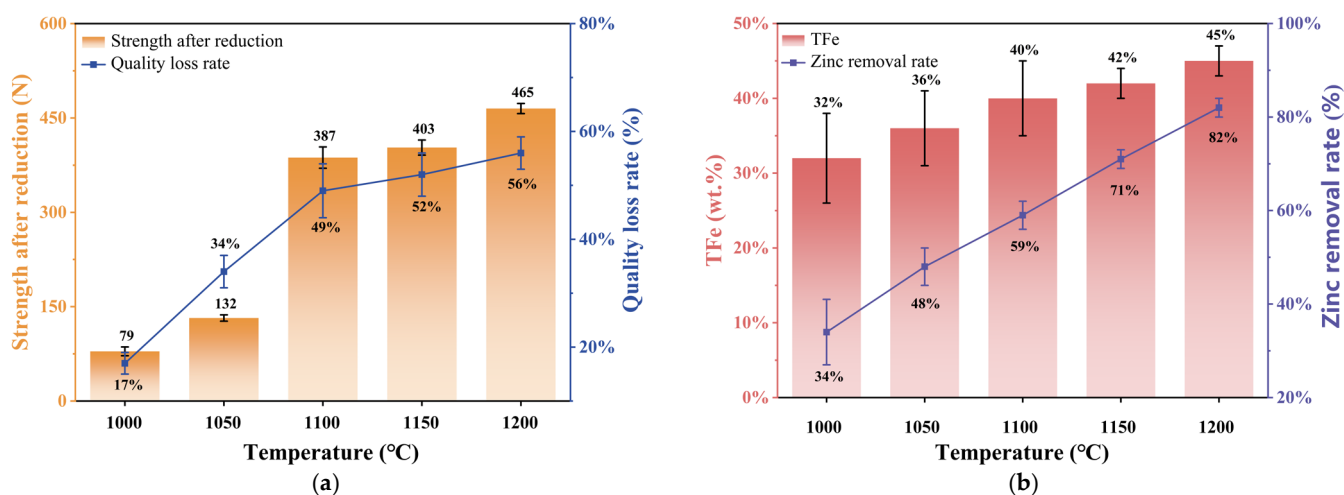


Figure 7. Effects of different temperatures on the high-temperature reduction behavior of cold-pressed blocks: (a) weight loss and strength after reduction; (b) dezincification rate and TFe grade.

3.3. Effect of Reduction Time on the High-Temperature Reduction Behavior of Cold-Bonded Briquettes

Figure 8 shows the effect of reduction time (10–60 min) on the high-temperature reduction behavior of cold-bonded briquettes at a fixed reduction temperature of 1200 °C. The mass loss ratio remained stable at approximately 55%, and the post-reaction compressive strength also leveled off, reaching a maximum of 524 N. The de-zincing rate peaked at 92%, and the TFe grade was gradually enriched to 50 wt.%, thereby satisfying the Grade 5 standard specified in GB/T 32545-2016, Classification of iron ore products. At high temperatures, all reduction behaviors tended to stabilize with prolonged time. This indicates that temperature plays a more dominant role than time in the reduction of cold-bonded briquettes. When the reduction temperature is high, a substantial degree of reduction is already achieved in the early stage. Combined with the previous mass balance calculations, the theoretical maximum weight loss of the briquette during reduction is about 59%. At higher reduction temperatures (1100–1200 °C), a relatively high degree of reduction and weight loss was already observed in the early reaction stage (10–30 min). In the later stage (30–60 min), the reaction rate gradually slowed, and the dezincification rate and weight loss rate increased only gradually.

3.4. Phase Analysis of Reduced Briquettes

Figure 9 presents the XRD patterns of the post-reaction briquettes obtained from the above experiments. The results show that the intensity of Fe diffraction peaks increased progressively with rising temperature or prolonged reduction time. When considered together with the changes in post-reaction compressive strength, this trend suggests that the formation of metallic iron is likely a major reason for the strength recovery [3,20]. In the early stage of reduction, the reduction of iron oxides gradually progressed, producing a large amount of metallic iron within the briquettes. During the middle and later stages, as the metallic iron phase was further enhanced, the degree of metallization in the system steadily increased, and the proportion of newly formed metallic iron in the briquettes rose accordingly. As a result, the post-reaction briquette strength did not collapse; instead, it recovered to a maximum of 524 N.

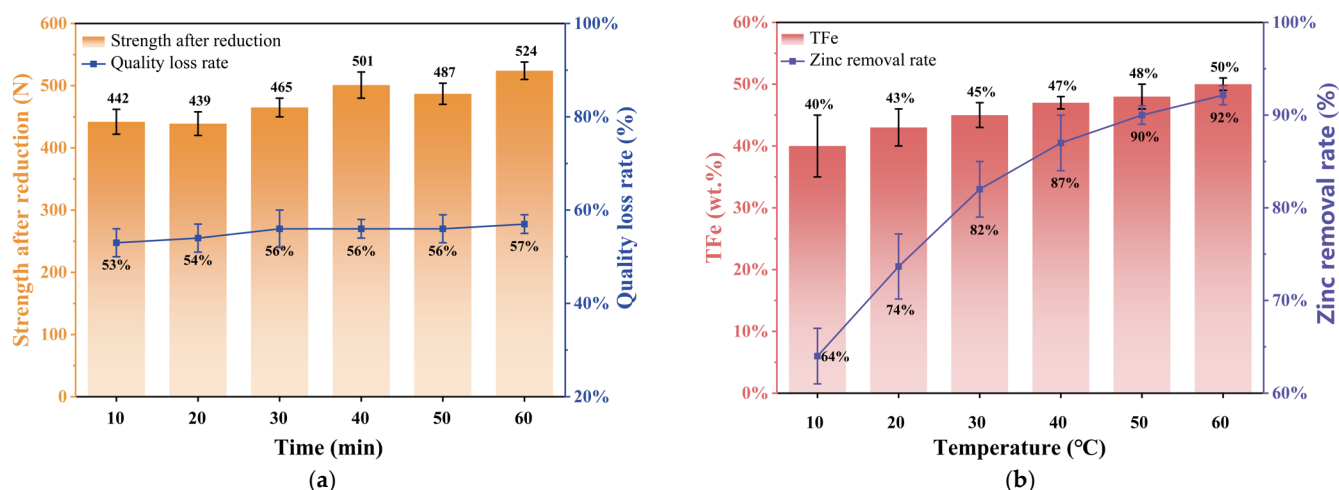


Figure 8. Effects of different holding time periods on the high-temperature reduction behavior of cold-pressed blocks: (a) weight loss and strength after reduction; (b) dezincification rate and TFe grade.

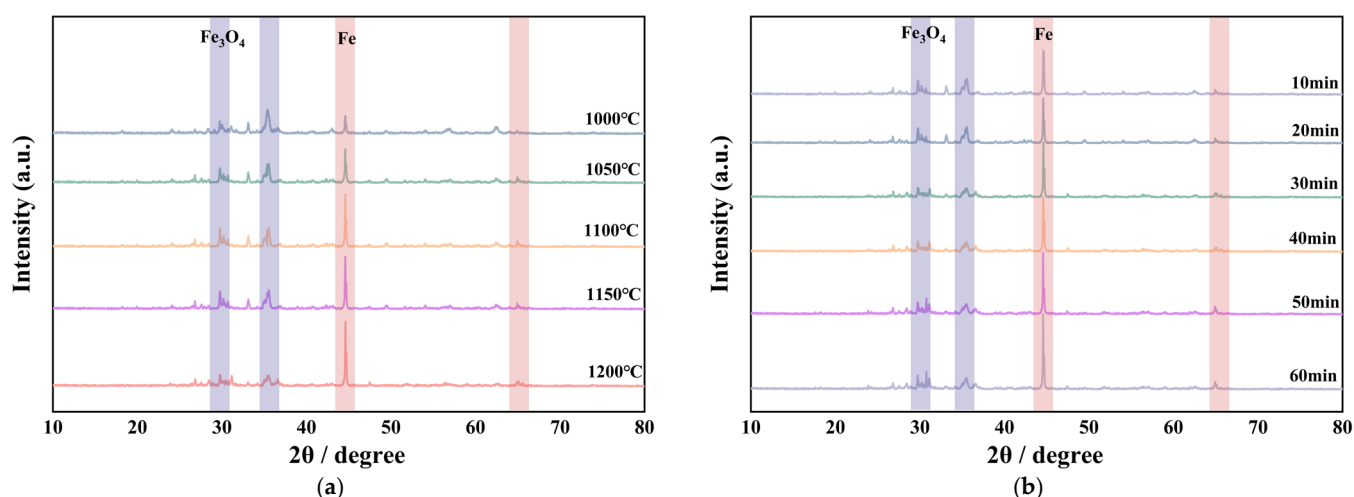


Figure 9. XRD results of the reduced blocks: (a) at different reduction temperatures; (b) at different holding times.

3.5. Kinetic Analysis

The reduction degree–time data obtained from experiments were substituted into the nine kinetic mechanism functions described in Section 2.3.5 for linear regression. By comparing the coefficients of determination (R^2) of the fittings for each model in the temperature range of 1000–1200 °C, the dynamic evolution pathway of the rate-controlling step in the high-zinc blast furnace dust cold-bonded briquette system was determined, and the transitions of the rate-controlling step during the reduction process were analyzed.

Piecewise linear fitting was conducted for each temperature based on the zinc removal behavior across various stages. The 1000 °C data were fitted without segmentation. However, two-stage fitting was applied to the remaining temperatures: 0–40 and 40–60 min at 1050 °C; 0–30 and 30–60 min at 1100 °C and 1150 °C; and 0–20 and 20–60 min at 1200 °C. The fitting results are summarized in Tables 5 and 6.

It is evident that at lower reduction temperatures and during the initial reaction stages, the R3 model yields the highest determination coefficient. This indicates that the system is primarily controlled by interfacial chemical reactions. In the mid-to-late stages, the D4 model achieves the highest determination coefficient, demonstrating that the rate-controlling step transitions to three-dimensional internal diffusion.

Table 5. Results of fitting different kinetic models (1000 °C, 1050 °C, 1100 °C).

Mechanism	No.	1000 °C R ²	1050 °C R ²		1100 °C R ²	
		0~60 min	0~40 min	40~60 min	0~30 min	30~60 min
Interfacial chemical reaction	Equation (3)	98.93%	99.57%	99.57%	98.64%	98.16%
	Equation (4)	99.01%	99.67%	99.67%	98.95%	98.15%
One/two-dimensional diffusion	Equation (5)	96.82%	97.76%	97.76%	97.14%	97.97%
	Equation (6)	94.83%	95.78%	95.78%	96.48%	98.15%
Three-dimensional internal diffusion	Equation (7)	91.94%	92.77%	92.77%	94.36%	98.05%
	Equation (8)	93.91%	94.83%	94.83%	95.94%	98.64%
Nucleation and growth	Equation (9)	96.68%	97.27%	97.27%	98.32%	98.12%
	Equation (10)	93.63%	94.21%	94.21%	96.46%	98.14%
Reaction order	Equation (11)	98.71%	98.25%	98.25%	98.69%	98.01%

Table 6. Results of fitting different kinetic models (1150 °C, 1200 °C).

Mechanism	No.	1150 °C R ²		1200 °C R ²	
		0~30 min	30~60 min	0~20 min	20~60 min
Interfacial chemical reaction	Equation (3)	98.77%	99.87%	99.92%	98.74%
	Equation (4)	99.18%	99.67%	99.88%	98.76%
One/two-dimensional diffusion	Equation (5)	97.68%	97.78%	99.29%	98.02%
	Equation (6)	97.02%	98.98%	96.94%	98.73%
Three-dimensional internal diffusion	Equation (7)	94.78%	99.80%	92.75%	98.41%
	Equation (8)	96.45%	99.89%	95.61%	98.82%
Nucleation and growth	Equation (9)	98.14%	98.91%	99.11%	98.62%
	Equation (10)	95.99%	98.94%	97.38%	98.25%
Reaction order	Equation (11)	99.03%	96.75%	99.01%	98.22%

The fitting trends of the R3 and D4 models at different temperatures are shown in Figure 10. The kinetic fitting results were consistent with the experimental observations obtained from the high-temperature reduction tests. During the initial stage of the reaction, the process was mainly limited by the chemical reaction occurring at the interface between the reducing agent and the oxides. At this stage, the reduction degree inside the briquettes remained relatively low, and the amount of newly formed metallic iron was limited. Therefore, the reaction behavior conformed to the interface chemical reaction model (R3) [21].

As the reaction proceeded, the intensity of the Fe diffraction peaks increased significantly, indicating the formation of a large amount of metallic iron. The newly formed metallic Fe underwent sintering, which refilled the pores generated during the early reduction stage of the cold-bonded briquettes. This process increased the resistance to the outward diffusion of gaseous products formed in the subsequent reaction stages. Consequently, the rate-limiting step gradually transformed into three-dimensional internal diffusion through the product layer, corresponding to the D4 model [22].

Within the 1000–1200 °C range, the activation energy in the interface chemical reaction-controlled (R3) stage was 109.10 kJ/mol. After the process entered the three-dimensional internal diffusion-controlled (D4) stage, the activation energy decreased to 69.78 kJ/mol. This decrease in activation energy provides physicochemical evidence that the system evolved from a “reaction-controlled” regime to a diffusion-controlled” regime. Figure 11 shows the fitting results of the activation energies for the system reaction when it is controlled by the R3 and D4 models, respectively.

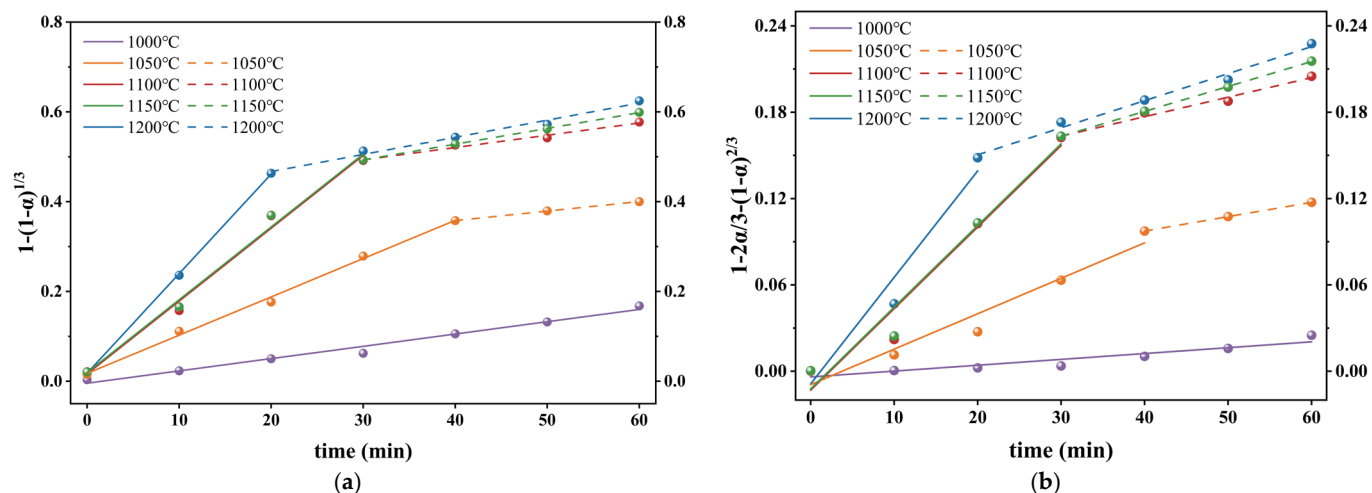


Figure 10. Fitting trends of R3 and D4 models at different temperatures: (a) R3 model; (b) D4 model.

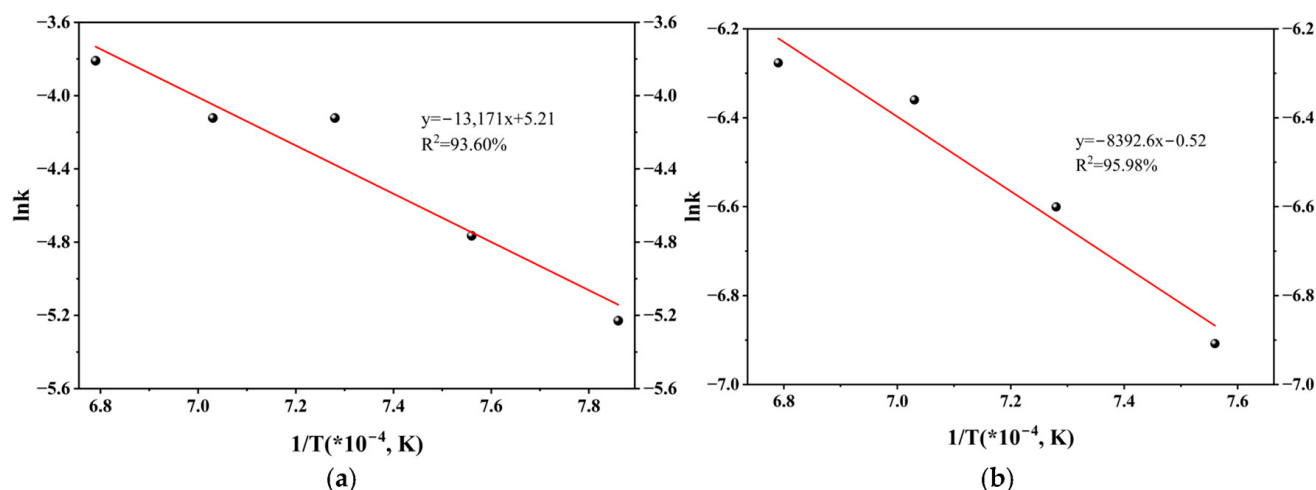


Figure 11. Relationship between T and $\ln k$ under different model control: (a) R3 model; (b) D4 model.

4. Conclusions

- (1) The enthalpy change for the direct reduction of ZnO is about 380 kJ/mol, and its theoretical onset temperature is 951 °C. This reaction is strongly endothermic. In contrast, iron oxides can be reduced gradually at lower temperatures. Based on the thermodynamic analysis, the high-temperature reduction temperature should be above 1000 °C to ensure both the reduction and removal of Zn and the reduction of iron oxides.
- (2) During high-temperature reduction, the briquette strength first decreased and then increased. In the 1100–1200 °C range, the reduction degree of the iron oxides increased in the middle and later stages of the reaction (30–60 min), and the metallization degree of the system continued to rise. The formation of metallic iron may be an important reason for the strength recovery, allowing the final strength of the reduced briquette to recover to 524 N.
- (3) Reduction at 1200 °C for 60 min was the optimal process condition. Under these conditions, the de-zincing rate reached 92%, and the TFe grade was enriched to 50 wt.%, meeting the requirements for Grade 5 products in the national standard GB/T 32545-2016, Classification of Iron Ore Products. This shows that by adding a small amount of iron ore powder, it is possible to efficiently remove zinc while also increasing the TFe grade of the reacted briquette, so that it can directly meet the requirements for use as an ironmaking raw material.

- (4) The reduction process in this system involved a change in the kinetic mechanism. In the initial stage, it was controlled by interfacial chemical reaction, with an activation energy of 109.10 kJ/mol. In the middle and later stages, the formation of newly generated metallic iron increased the resistance to gas diffusion, and the control mechanism shifted to three-dimensional internal diffusion, with an activation energy of 69.78 kJ/mol.

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