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# Research Progress on the Recovery of Zinc from Steelmaking Dust by Calcination-Chlorination-Deep Eutectic Solvent Complexation Technology

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## Abstract

**Zinc, as a key raw material for the national economy, has seen a continuous increase in demand in China over the past two decades. In 2018, China's global consumption share exceeded 50%, making the recycling of zinc-containing waste an important way to ensure resource security. Steelmaking dust, a typical zinc-containing waste, had a zinc content of 400,000 to 1.5 million tons in China in 2018. However, traditional acid leaching consumes a large amount of acid and has complex iron removal processes, while ammonia leaching has a low leaching rate and high pollution risks, which have restricted the efficiency of zinc recovery. Low-melting eutectic solvents (DESs), with advantages such as being green and degradable and having an atomic utilization rate of 100%, offer a new direction for hydrometallurgical zinc extraction. The calcium-chlorination-DES composite technology breaks down the $ZnFe_2O_4$ spinel structure at low temperatures with a calcination agent, releasing easily soluble zinc compounds, which are then selectively leached by DES. The hydrogen bond complexation effect of DES enhances the solubility of zinc, achieving efficient separation of zinc and iron simultaneously. The zinc recovery rate reaches 85% to 95%, and the leaching time is reduced by 50%. This paper reviews the theoretical mechanism and research progress at home and abroad of this technology, analyzes the core challenges such as the optimization of calcination agents and the recycling and regeneration of DES, and proposes solutions such as the compounding of calcination agents and multi-stage recycling processes. Looking ahead, the optimization of compound calcination agents and the modification of DES molecules will be key research directions. This composite technology provides a green and efficient path for the resource utilization of zinc-containing waste and is of great significance for ensuring the strategic security of zinc resources in China.**

## Keywords

**Steelmaking Dust; Deep Eutectic Solvents; Calcination Chlorination; Composite Technology; Hydrometallurgical Zinc Extraction.**

## 1. Introduction

Zinc is an important raw material for the national economy. Over the past two decades, China's demand for zinc has been continuously increasing. The proportion of China's zinc consumption in global zinc consumption has risen from 15.18% in 2000 to over 50% in 2018. Although China is also a major producer of zinc, the poor endowment of primary zinc ore resources and the resource depletion problem caused by years of mining have led to China's zinc dependence on foreign sources exceeding 35%, which will have a serious impact on China's resource strategic security to some extent. Currently, recovering zinc from zinc-containing waste materials is becoming an important way to obtain zinc resources. However, zinc-containing waste materials have a wide range of sources, significant quality differences, and are prone to secondary pollution and resource waste during the recycling process, making the regeneration process extremely difficult. Green and efficient approaches will be the key direction for the development of the recycled zinc industry.

Steelmaking dust, as a typical zinc-containing waste material, has become an important source of raw materials for recycled zinc with the rapid development of the steel industry. Based on China's crude steel output of 800 million tons in 2018, the amount of zinc metal contained in steelmaking dust reached 400,000 to 1.5 million tons. How to economically and efficiently achieve selective leaching of zinc from steelmaking dust is a hot issue in the field of zinc recovery. Traditional acid leaching processes consume a large amount of acid and iron easily enters the leaching solution, making the subsequent iron removal process complex. Using the ammonia leaching process developed in recent years, it is difficult to dissolve zinc ferrite, resulting in low leaching rates, high ammonia volatility, and slow diffusion of the leaching agent during the leaching process. Especially when recovering useful minerals from solid waste, the secondary pollution caused by this technology also limits its application. Therefore, in-depth research on the design and synthesis of clean and efficient new solvents, selective leaching behavior, and separation mechanisms is of great significance for hydrometallurgical zinc extraction.

Deep Eutectic Solvents (DESs), as a new type of green ionic liquid, have attracted extensive attention from researchers around the world due to their biodegradability, low cost, and 100% atomic utilization in the synthesis process. They have shown promising application prospects in various fields such as separation processes and chemical reactions. This project aims to introduce DESs as a new type of solvent into the field of hydrometallurgical zinc extraction, achieving clean separation and efficient recovery of difficult-to-separate metal compounds, and providing new ideas and methods for the development and utilization of similar resources.

The calcination-chlorination-Deep Eutectic Solvents (DES) composite technology (referred to as the calcination-chlorination-DES composite technology) is a new type of green and efficient zinc recovery technology. Compared with traditional pyrometallurgical smelting and hydrometallurgical leaching, this technology has significant advantages: calcination-chlorination can completely decompose $ZnFe_2O_4$ at a relatively low temperature, reducing energy consumption; at the same time, DES, as a solvent, not only has good ionic conductivity and leaching selectivity, but also avoids environmental pollution caused by chlorinated reagents. This makes the calcination-chlorination-DES composite technology have broad application prospects in zinc-iron separation and zinc resource recovery.

## 2. Theoretical Basis and Reaction Mechanism

(1)Core Mechanism of Calcination-Chlorination Decomposition of $ZnFe_2O_4$

The process of calcination-chlorination decomposition of $ZnFe_2O_4$ mainly involves the reaction path between calcination products and $ZnFe_2O_4$. Calcination products (such as CaO or $CaCl_2$) react with $ZnFe_2O_4$ at high temperatures, initially separating zinc and iron. Through carbothermal reduction or chlorination reactions, zinc is further released as the soluble $ZnCl_2$. Calcination products react with $ZnFe_2O_4$ to disrupt its spinel structure, effectively releasing zinc components.

(2)Leaching Mechanism of Novel Deep Eutectic Solvents

Deep eutectic solvents (DES) are solvent systems formed by mixing two or more low-melting-point compounds in specific proportions. Their structural characteristics determine their excellent ionic conductivity and good leaching performance. The hydrogen bond network in DES promotes the complexation reaction between metal ions and the solvent, thereby increasing the solubility of zinc ions. The selective complexation mechanism of zinc ions mainly relies on the coordination of carboxyl or hydroxyl groups in the solvent with $Zn^{2+}$. Additionally, the leaching kinetics of DES is mainly diffusion-controlled, with the activation energy of the leaching process ranging from 32 to 45 kJ/mol.

(3)Synergistic Mechanism of the Composite System

In the calcination-chlorination-DES composite system, the role of calcination pretreatment is to disrupt the spinel structure of $ZnFe_2O_4$ and release free ZnO. Subsequently, the DES system selectively leaches ZnO and $ZnCl_2$, while inhibiting the dissolution of iron phases (such as $Ca_2Fe_2O_5$). This process achieves efficient zinc recovery through two-step synergy: first, calcination pretreatment, followed by leaching with DES. Through process coupling, the viscosity of DES can be reduced by the dispersion effect of the calcination agent, thereby enhancing mass transfer efficiency and further increasing zinc recovery.

## 3. Research Status at Home and Abroad

(1)Application Progress of Calcination-Chlorination Technology in $ZnFe_2O_4$ Treatment

IWASE et al. used the Ozawa method and Gibbs free energy calculation method, under $N_2$ atmosphere and in an infrared lamp heating furnace, with an activation energy of $CaCl_2$ at 123 kJ/mol, which eventually dropped to 84 kJ/mol [1]. The chlorination volatilization of zinc increased the reaction rate and reduced the activation energy. It was concluded that the addition of calcium chloride was effective and could synergistically remove zinc, avoiding the traditional addition of carbon. Guo Ting et al. analyzed the microscopic morphology changes using SEM and EDS, including solid-liquid reactions and gas volatilization processes. $ZnFe_2O_4$ particles were wrapped by molten $CaCl_2$, and chlorination reactions occurred at the solid-liquid interface, showing an activation energy of 110 to 175 kJ/mol. The final products would continuously bond and fuse [2].

Chairaksa-Fujimoto et al. used the method of mixed calcination of zinc ferrite with calcium oxide (CaO), under conditions where the molar ratio of Ca to Fe was greater than 1.1 and the temperature was 900°C, converting zinc ferrite into zinc oxide (ZnO) and calcium ferrite ($Ca_2Fe_2O_5$) [3].The results indicated that calcium oxide could effectively separate zinc and iron from zinc ferrite. Jiang Liudong et al. studied the effect of CaO modification on the selective leaching of zinc from zinc ferrite [4]. The results showed that increasing the molar ratio of Ca to Fe had no significant effect on the leaching of zinc and iron from modified zinc ferrite, but it could increase the leaching rate of calcium. As the pH value increased, the leaching rates of iron, zinc, and calcium from modified zinc ferrite all decreased. Increasing the temperature promoted the dissolution of calcium ferrite in modified zinc ferrite, and the leaching rates of iron and calcium both significantly increased. Qi Dexing et al. adopted a method combining thermodynamic calculation and experiments to study the carbothermal reduction of zinc ferrite at different reduction temperatures and carbon-to-oxygen molar ratios [5]. This approach reduced the reaction temperature and carbon consumption, and the CaO restructured phase promoted the complete reaction of zinc ferrite in advance. At 1270K and a carbon-to-oxygen molar ratio of 0.7, Zn volatilized as vapor and Fe was reduced to the elemental form, achieving a good separation effect between zinc and iron.

Gupta et al. conducted an isothermal kinetic study on the thermal decomposition of pure $Ca(OH)_2$ using TGA (thermogravimetric analysis). The results indicated that $Ca(OH)_2$ decomposed by losing water to form CaO and $H_2O$ when heated to approximately 370–500 °C, and the thermal decomposition process could be quantitatively described by the decomposition rate parameters, thereby demonstrating that $Ca(OH)_2$ decomposed before the solid-state reaction with $ZnFe_2O_4$ [6].Sawada et al. observed the thermal decomposition behavior of $Ca(OH)_2$ in a mixed system using TGA combined with XRD (X-ray diffraction) [7].The results showed that $Ca(OH)_2$ decomposed significantly in the range of approximately 350-450°C to form CaO, which was characterized by a significant mass loss on the TGA curve, indicating that the thermal decomposition of $Ca(OH)_2$ was the main step before the solid-state reaction.Boning suggested that the thermal decomposition of $Ca(OH)_2$ was a solid-gas reversible reaction, and the dehydration to form CaO was the dominant process in the 400-600°C range [8]. The study emphasized the influence of factors such as temperature and particle size on this reaction process, providing a comprehensive thermodynamic and kinetic argument for the preferential decomposition of $Ca(OH)_2$ in a thermal system.

Wang et al. investigated the thermal decomposition behavior of calcium carbonate using TGA and DSC (differential scanning calorimetry) [9]. The results indicated that $CaCO_3$ decomposed significantly in the range of approximately 700-900°C to form CaO and $CO_2$, and the decomposition process was significantly affected by particle size, heating rate, and atmosphere. This study provided experimental evidence for the decomposition kinetics of $CaCO_3$ in high-temperature treatment.Rodriguez-Navarro et al. systematically examined the phase transformation and decomposition pathways of $CaCO_3$ during heating using in-situ X-ray diffraction and thermal analysis [10]. The study found that at atmospheric pressure, $CaCO_3$ began to decompose at approximately 750°C and was almost completely decomposed at around 850°C, forming a porous CaO structure, accompanied by significant mass loss and endothermic effects.Borgwardt et al. systematically explored the decomposition kinetics of calcium carbonate and its application in industrial calcination processes in earlier studies [11]. They pointed out that the decomposition of $CaCO_3$ was a reversible gas-solid reaction, and its equilibrium temperature was controlled by the partial pressure of $CO_2$. In actual high-temperature processes (such as cement calcination and metallurgical roasting), $CaCO_3$ was usually completely decomposed at 800-950°C, generating highly reactive CaO.

(2)Research Status of Novel Deep Eutectic Solvents for Leaching Zinc-containing Dust

In the direction of metal dissolution, domestic research has confirmed that $DES_S$ (deep eutectic solvents) have good dissolution performance for ionic metal oxides (such as ZnO, $Cu_2O$, PbO), but are almost ineffective for covalent metal oxides (such as $Al_2O_3$) [12][13]. Based on this selective dissolution characteristic, some teams have conducted experiments on the targeted leaching of metals such as zinc and copper from complex solid wastes (such as steelmaking dust) using $DES_S$, achieving high metal recovery rates [14]. Research on the dissolution mechanism of typical choline chloride-based systems has also initially revealed the diversity of coordination structures and mechanism differences [15][16].

In the field of metal extraction and complex mineral resource development, the selective dissolution characteristics of DESs (deep eutectic solvents) have been fully verified. Ashraf Bakkar et al. successfully extracted zinc from zinc-containing dust with choline-based DES, demonstrating excellent metal enrichment capacity [17]. McCluskey et al. utilized an ionic liquid-assisted DES system to achieve highly efficient copper leaching from chalcopyrite, with a leaching rate exceeding 90%. Relevant studies have shown that compared with traditional acidic leaching systems, the synergistic effect of DESs and ionic liquids significantly enhances the leaching rate and selectivity, effectively breaking through the efficiency bottleneck of traditional systems [17][18].

(3)Research Progress on the Calcination-Chlorination-DES Composite System

Research on the calcination-chlorination - deep eutectic solvent composite system has gradually deepened, and significant progress has been made in the coupled pretreatment and leaching process. DES leaching after calcination roasting can significantly increase the recovery rate of zinc to 85%-95%. Studies on the $CaCl_2$-DES system have shown that the synergistic effect of chlorination and complexation can reduce the leaching time by 50%. However, there are still research gaps in the optimization of multi-calcination compound blends, DES molecular design, and industrial scale-up technology in existing studies.

## 4. Technical Challenges and Solutions

The selection of calcination agents is crucial for the efficient recovery of zinc resources. Choosing the appropriate calcination agent requires considering not only its reaction efficiency but also its cost and environmental impact. Traditional calcination agents such as lime and calcium hydroxide have high costs and certain environmental burdens, so it is necessary to find low-cost and environmentally friendly alternatives. By blending different calcination agents, their reaction performance can be optimized, increasing the recovery rate and selectivity of zinc. For example, researchers can improve the reaction efficiency of the calcination process and reduce the cost of a single calcination agent by combining different metal salts. Additionally, developing calcination agents with lower environmental impact, such as organic calcium salts, may be an important direction for the future.

The recovery and regeneration technology of deep eutectic solvents (DES) remains a challenging issue to be solved. Although DES has high leaching efficiency in the zinc recovery process, its recovery process faces problems such as solvent loss and low regeneration efficiency. To increase the recovery rate of DES, multi-stage recovery processes can be explored, combining membrane separation, extraction, or distillation technologies to reduce solvent loss and improve regeneration efficiency. Moreover, developing more stable and sustainable DES formulations can enhance their durability and reusability in industrial applications.

## 5. Future Research Directions and Prospects

The optimization of calcination compound blends will be an important research direction in the future. By exploring new calcination compound systems, the recovery rate and selectivity of zinc can be further improved. For example, by synthesizing new composite calcination agents, their affinity with zinc can be enhanced, thereby improving the efficiency and economy of the calcination process. By further studying the interaction mechanisms between different metal ions, the formulation of calcination agents can be optimized, potentially achieving more efficient zinc recovery.

Molecular design and functional modification of deep eutectic solvents (DES) are important means to enhance their performance. Through molecular-level regulation, such as adjusting the component ratio in the solvent system and adding functional molecules, the leaching efficiency of DES in the zinc recovery process can be significantly improved. Functionalized DES not only enhances the selective leaching of zinc but also improves its adaptability in complex mineral systems, thereby further enhancing the industrial application potential of the technology.

The trend of mass data in power system provides a basis for load characteristic analysis and prediction model establishment, but the classical load forecasting method can not afford such a huge time and computing resource consumption. The problem of over fitting in large sample set will affect the prediction accuracy. In this paper, a power load forecasting model is built by using the BP neural network model, making full use of the powerful data processing function of Clementine and preventing the over fitting function. The experimental results show that the BP neural network model has good predictability and robustness, and has a certain practical application value.

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