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Removal of copper and cadmium from industrial zinc sulfate leachates by ultrasonic enhanced zinc powder

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Highlights

- A method for removing toxic metals by ultrasonic boosting activity of zinc powder was proposed.
- Achieves zinc reduction while maintaining efficacy via pulsed dosing & cavitation-suppressed hydrolysis.
- Acoustic cavitation enhances the grade of metals through particle dispersion and surface activation.

Abstract

While the substitution of industrial zinc powder effectively addresses the persistent risks of copper (Cu) and cadmium (Cd) contamination in zinc hydrometallurgical leaching solutions—particularly their detrimental impacts on electrowinning yield, ecological systems, and public health—this conventional approach has inherent limitations. Key challenges include passivation induced by zinc powder hydrolysis and particle agglomeration, which collectively reduce zinc utilization yield and pose a risk of generating secondary heavy metal pollution. To overcome these limitations, this study proposes an ultrasonic-enhanced intermittent purification process featuring two critical improvements, namely, the implementation of intermittent zinc powder dosing to minimize surface passivation and the introduction of acoustic cavitation to suppress hydrolysis. Experimental results demonstrate that the ultrasonic-assisted process achieves significant advancements compared to conventional purification methods. First, it reduces the operating temperature from 65 °C to ambient conditions and decreases the required reaction duration by 50%. Second, it results in a 26.4% reduction in zinc powder consumption while maintaining purification efficacy. Furthermore, comprehensive characterization through X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), laser particle analysis, and scanning electron microscopy with energy-dispersive X-ray spectroscopy (SEM-EDS) reveals that ultrasonic cavitation generates dual beneficial effects: micro-jet streams effectively disperse particle agglomerates, while transient high-pressure and high-temperature microenvironments modify surface reaction kinetics. These mechanisms collectively enhance zinc utilization yield. Notably, the enriched residue containing 15.48% Cu and 2.75% Cd demonstrates improved economic viability for subsequent metal recovery processes.

Introduction

As global industrialization accelerates alongside economic expansion, zinc has become increasingly integral to modern technological systems. This strategic base metal serves critical functions across multiple industrial domains, including advanced metallurgical processes (particularly galvanization), corrosion-resistant chemical engineering, precision electronics manufacturing, and heavy machinery production, with its global market value exceeding \$45 billion in 2023 [1]. The use of zinc is especially important in the coating of steel and aluminum alloys [2]. Additionally, zinc plays an indispensable role in battery manufacturing, coatings, alloys, glass, and building materials [3], [4]. One of the primary methods for extracting zinc ore is wet zinc smelting, which involves the leaching of zinc from zinc ore using sulfuric acid to produce a zinc sulfate solution. This extraction process frequently results in the incorporation of detrimental impurities, such as copper (Cu) and cadmium (Cd), into the zinc sulfate solution. These impurities can adversely affect the purity and quality of the zinc electrowinning products, as well as pose potential risks to environmental and human health [5], [6]. Consequently, the effective removal of Cu and Cd from the leaching

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However, Cu, recognized as a high-value metal, frequently emerges as a prevalent impurity in the zinc hydrometallurgical process. The presence of Cu ions during zinc refining not only elevates the Cu content in the resultant zinc products but also detrimentally impacts their purity and performance[7], [8], [9]. This concern is particularly pronounced in applications requiring high-purity zinc, such as in electronic materials and precision instruments. Furthermore, during the electrolytic deposition phase, Cu ions tend to be preferentially deposited onto the cathode over zinc ions, resulting in the formation of Cu-zinc alloys. This phenomenon not only diminishes the deposition yield of zinc but also contributes to an uneven electrode surface, thereby compromising product quality [10], [11]. Additionally, Cu ions exhibit significant corrosive properties towards the materials constituting the electrolytic cell. Prolonged accumulation of these ions can lead to a reduction in the operational lifespan of the equipment, consequently escalating maintenance and replacement costs. Cd, a toxic heavy metal, poses both biological toxicity and environmental accumulation risks. Its prolonged presence in the environment can severely affect aquatic organisms, terrestrial animals, and human health [12], [13]. Notably, Cd has a pronounced impact on plant life, with potential transmission through the food chain, thereby exerting significant effects on the ecosystem[14]. Moreover, the presence of Cd ions during the zinc electrowinning process considerably influences the purity of zinc products, particularly in sectors with stringent Cd content requirements, such as battery manufacturing and coatings. Cd also exhibits corrosive effects on the materials of the electrolytic cell, which can accelerate equipment aging and damage [15]. Consequently, the effective removal of Cu and Cd from zinc sulfate solutions is imperative not only for enhancing the quality and production yield of zinc products but also for preserving environmental integrity and ensuring worker health, thus holding substantial economic and environmental significance. In pursuit of mitigating the effects of Cu and Cd during the zinc electrowinning process, researchers both domestically and internationally have developed a range of effective purification and impurity removal technologies for zinc sulfate solutions within the framework of zinc hydrometallurgy. These methods include chemical precipitation, solvent extraction, adsorption, electrochemical techniques, and microbial approaches [16], [17], [18]. While the aforementioned methods demonstrate efficacy in the removal of copper, cadmium, and other impurities from zinc sulfate solutions, they encounter significant challenges in the context of increased industrial production. For instance, the chemical precipitation method generates numerous by-products, and the subsequent separation and treatment of these precipitates is complex, potentially leading to secondary environmental pollution. Similarly, the solvent extraction method involves a complicated recovery and regeneration process, which may result in environmental issues associated with organic solvents. The electrochemical approach is hindered by low current yields and high energy consumption. Additionally, the microbial method incurs high costs related to microbial culture and treatment, coupled with a slow processing rate. The zinc powder displacement method is commonly employed in zinc hydrometallurgy enterprises for the purification and removal of Cu and Cd from zinc sulfate solutions. This technique capitalizes on the differences in standard electrode potentials of metals, utilizing zinc powder to displace impurity ions from the solution, thereby forming insoluble metal precipitates [17]. This method presents several advantages, including operational simplicity and cost-effectiveness, making it suitable for treating zinc sulfate solutions with high impurity concentrations. However, during actual production, this method faces challenges, as the efficiency of the displacement process is affected by various factors such as the particle size of the zinc powder, temperature, and stirring conditions. These factors can lead to issues such as agglomeration or encapsulation of the zinc powder, as well as the potential for secondary pollution and other drawbacks [19]. Consequently, enhancing and mitigating the limitations of the current zinc powder displacement method for the removal of Cu and Cd from zinc sulfate solutions represents a viable strategy for achieving the high-value utilization of non-ferrous metal resources while promoting environmental sustainability.

Ultrasound refers to sound waves that possess frequencies exceeding the upper limit of human auditory perception [20]. Its frequency is usually between 20 kHz and hundreds of megahertz [21]. Because of its propagation in the medium, it will cause the vibration of molecules or atoms, resulting in mechanical effects, thermal effects and chemical effects in the material, and accompanied by strong energy concentration and propagation ability, so it is widely used in the field of hydrometallurgy [22]. For example, Xu et al. [23] used ultrasound to enhance the leaching of valuable metals in zinc oxide dust by tartaric acid and sulfuric acid. The results showed that ultrasound would produce a strong microfluidic effect in the liquid, forming a small liquid vortex and high-frequency vibration flow. This microfluidic effect can promote the movement and collision of particles and solutes in the solution, thereby accelerating the reaction. Liu et al. [24] introduced ultrasound into the chloride-oxidant system to enhance the leaching of platinum. The results showed that ultrasound produced tiny bubbles (cavitation bubbles) in the liquid. These bubbles released huge energy during growth and collapse, resulting in local high temperature, high pressure and strong oxidizing free radicals. When ultrasound travels through a liquid medium, a portion of its energy is transformed into thermal energy, leading to a rise in the local temperature of the liquid. This temperature increase facilitates the leaching process of platinum. Deng et al. [25] found that ultrasound-assisted oxalic acid can significantly enhance the leaching of lithium from waste lithium-ion battery mixtures, which is mainly attributed to the fact that the cavitation effect of ultrasound can not only promote the reduction of particle size and the removal of layered structure, but also promote the full interaction between solid and liquid and the complete reduction of high-valent metals. In order to realize the resource utilization of chalcopyrite, Zhang et al. [26] constructed an advanced oxidation system coupled with ultrasound and ozone, which increased the leaching rate of copper and iron by more than 30%, and reduced the activation energy by 23.40 kJ/mol. What's more, other scholars have introduced ultrasonic waves in the process of solution purification, mainly because the introduction of ultrasonic waves can make the molecules and ions in the solution more active, increase the contact of the reaction interface, and make insoluble substances or impurities easier to precipitate or be removed.

In light of the aforementioned considerations, a novel approach utilizing ultrasonic strengthening of zinc powder for the purification of copper and cadmium from zinc sulfate solution has been proposed. This research investigates the effectiveness of both the traditional purification method and the ultrasonic-enhanced purification technique in the removal of Cu and Cd. A comparative analysis of the two methods was conducted, focusing on variables such as reaction temperature, reaction duration, and the dosage of zinc powder. Additionally, the mechanisms underlying the ultrasonic enhancement of zinc powder displacement were examined. The overarching aim of this study is to achieve a purification process that is efficient, cost-effective, and environmentally

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Materials

The ZnSO₄ solution is derived from the supernatant produced during the neutral leaching of zinc calcine using sulfuric acid, a process employed in zinc hydrometallurgy at a facility located in the Inner Mongolia Autonomous Region of China. The pH of this solution ranges from 5.1 to 5.4. The zinc powder utilized in the reaction is industrial-grade zinc powder, which contains approximately 20% moisture. The zinc powder slurry is formulated as a sulfuric acid solution with a pH of 5.2–5.3, ...

Raw material analysis

Table 1 presents the primary constituents of the zinc sulfate solution utilized in the experiment. The findings indicate that the concentrations of copper and cadmium in the supernatant are notably high, measuring 713 mg/L and 417 mg/L, respectively, whereas the levels of other impurities are comparatively low. Fig. 2(a) and Table 2 illustrate the principal phase and elemental composition of the zinc powder employed for the removal of copper and cadmium, before analyzing the main components of ...

Conclusions

This research demonstrated that ultrasound-assisted purification significantly improved the removal of Cu and Cd from zinc sulfate leaching solutions when compared to traditional methods. The principal findings of the study are as follows:

(1) Under conventional conditions at 65 °C for 60 minutes, the addition of 3.5 times the amount of zinc powder resulted in removal yields of Cu and Cd of 99.84 % and 53.89 %, respectively, with residual concentrations in the solution reduced to 1.18 mg/L for Cu ...

CRediT authorship contribution statement

Kan Yu: Writing – original draft, Methodology, Data curation. **Hao Yang:** Writing – original draft, Methodology, Data curation. **Libo Zhang:** Validation, Supervision, Software, Resources, Funding acquisition, Conceptualization. **Ningting Li:** Writing – original draft, Methodology, Data curation. **Bu Yang:** Writing – original draft, Methodology, Data curation. **Hongying Xia:** Validation, Supervision, Software, Resources, Methodology, Funding acquisition, Conceptualization. **Xudong Li:** Writing – original ...

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper. ...

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