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Ultrasound-assisted activation of zinc powder by antimony salts for the removal of Co and Cd from zinc sulfate solution

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What do these dates mean?

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Abstract

Despite the effective resolution of persistent cobalt (Co) and cadmium (Cd) pollution risks in hydrometallurgical leaching solutions through the alternative use of industrial zinc powder, particularly regarding their potential adverse impacts on subsequent electrowinning quality, ecological environments, and public health, traditional zinc powder purification methods remain inherently limited. Key challenges include surface passivation and particle agglomeration due to zinc powder hydrolysis, which not only diminish zinc utilization efficiency but also elevate the risk of secondary heavy metal contamination. To address these limitations, this study proposes an ultrasonic-enhanced intermittent purification process incorporating two critical innovations: pulsed zinc powder addition to minimize surface passivation and the utilization of ultrasonic cavitation effects to suppress zinc powder hydrolysis. Ultrasonic cavitation induces periodic compression and expansion within the solution via high-frequency pressure waves, generating instantaneous high-pressure/high-temperature microenvironments and micro-jet effects. The former accelerates solid–liquid interface reaction kinetics and destabilizes the zinc powder passivation layer, while the latter disperses agglomerated particles through intense shear forces, thereby increasing the effective contact area of zinc powder and synergistically enhancing reaction efficiency. Experimental findings reveal that compared with conventional purification techniques, this ultrasonic-assisted process achieves significant improvements in multiple dimensions: a reduction in reaction time from 75 to 60min, a 5.4% decrease in zinc powder consumption under equivalent purification efficacy, and an increase in cobalt and cadmium content in enriched residues by 0.47% and 0.17%, respectively, relative to traditional methods. These results indicate enhanced zinc recovery efficiency, improved economic feasibility for subsequent metal recovery processes, and optimized resource utilization. Furthermore, characterization analyses employing X-ray fluorescence spectroscopy (XRF), X-ray diffraction (XRD), laser particle size analysis, and scanning electron microscopy-energy dispersive spectroscopy (SEM-EDS) confirm the dual advantages of ultrasonic cavitation: effective dispersion of particle agglomerates via micro-jet effects and modification of surface reaction kinetics through transient high-pressure/high-temperature microenvironments, significantly improving zinc utilization efficiency. In summary, this research underscores the substantial benefits of ultrasonic-enhanced purification technology in terms of zinc powder utilization, reaction time reduction, zinc powder consumption minimization, and metal enrichment optimization, offering a novel technical pathway for the sustainable and efficient advancement of the hydrometallurgical industry.

Introduction

Under the backdrop of global industrialization and rapid economic development, zinc resources have become a critical element in modern technological systems, finding extensive applications across various high-tech industries [[1], [2], [3]]. Its outstanding physical and chemical properties make it irreplaceable in fields such as electronics, solar energy, chemistry, electroplating, aerospace engineering, and medicine [[4], [5], [6], [7]]. In the electronics sector, high-purity zinc is widely used in the production of efficient batteries, semiconductor components, and circuit boards, serving as a core material for intelligent devices, computers, and communication technologies [8]. The solar energy industry relies on zinc as a key material, extensively applied in the manufacturing of solar cell panels and photovoltaic components, driving the development of clean energy. In the chemical industry, zinc serves as an important raw material for the synthesis of various catalysts and fine chemicals, supporting modern industrial processes [9]. The electroplating industry leverages zinc's excellent anti-corrosion properties to provide durable coatings for steel, automotive parts, and infrastructure, enhancing the longevity and reliability of these materials [10]. In aerospace engineering, the corrosion resistance and high strength of zinc are paramount; high-purity zinc, as a key component in alloy materials, can significantly improve material performance, ensuring stability in extreme environments. Additionally, zinc plays a crucial role in emerging energy storage technologies, medical devices, and advanced optoelectronic systems, such as high-performance batteries, precision medical equipment, and cutting-edge display technology [11]. In recent years, global demand for zinc has shown a consistent upward trend, driven by the acceleration of electronic industry development, advancements in photovoltaic technology, and the expansion of green energy solutions. With technological progress and increasingly stringent environmental standards, the applications of zinc continue to expand, particularly in high-tech materials, emerging energy technologies, and sustainable manufacturing. Its demand is expected to rise further, providing significant support for future industrial growth.

Currently, zinc production processes are mainly divided into pyrometallurgical and hydrometallurgical methods, with the latter dominating global zinc production. Over 85% of the world's zinc output is produced using hydrometallurgical techniques [12]. The conventional hydrometallurgical process includes roasting zinc concentrate to obtain calcine, followed by sulfuric acid leaching to extract zinc into solution, purification to remove impurities, and finally, electrowinning to produce metallic zinc [13]. However, the sulfuric acid leachate from roasted zinc calcine typically contains various impurities such as copper (Cu), cadmium (Cd), cobalt (Co), and nickel (Ni), which can cause numerous adverse effects during the electrowinning stage [14]. Cobalt, with a more positive potential than zinc, tends to co-deposit, disrupting the morphology and structure of the zinc deposit and thereby lowering product quality [15]. Moreover, when Co^{2+} concentration exceeds 1 mg/L, its lower hydrogen overpotential promotes hydrogen evolution reactions, which can lead to anode burning, increased energy consumption, and rising operational costs. Excessive hydrogen evolution due to cobalt deposition can also cause a sharp drop in current efficiency and destabilize the electrolysis process [16]. Cadmium, a highly toxic heavy metal, poses serious environmental and health risks due to its bioaccumulative nature [17]. In zinc electrowinning, elevated Cd^{2+} concentrations significantly affect the quality and purity of zinc, especially in applications with strict cadmium limits such as battery manufacturing and coatings [18]. Additionally, $Cd2^{+}$ can corrode electrolytic cell components, accelerating equipment aging and increasing maintenance costs [19]. Therefore, purification of zinc leachate is a critical step in the hydrometallurgical zinc process, directly influencing the stability of electrowinning, the technical and economic performance of the process, and the quality and competitiveness of the final zinc product. To meet high-purity requirements and minimize environmental impact, optimizing purification processes and improving impurity removal efficiency have become key goals in the development of hydrometallurgical zinc technology. To eliminate the effects of Co and Cd during zinc electrowinning, researchers have developed several purification techniques for sulfuric acid zinc solutions, including the zinc powder–arsenic salt replacement method, oxidation precipitation, solvent extraction, and additive methods. While these techniques effectively remove Co and Cd, they face challenges in large-scale industrial applications [[20], [21], [22], [23], [24]]. For example, the oxidation precipitation method requires precise control of oxidant dosage, reaction time, and temperature. Excess oxidants can cause over-precipitation or negatively affect zinc and other valuable metal ions. Solvent extraction involves volatile organic solvents, posing environmental and safety concerns, as well as increased costs due to solvent recovery and waste treatment. The efficiency of the additive method may be limited by reaction conditions and impurity concentrations, sometimes resulting in incomplete cobalt removal and reduced product purity. The zinc powder–antimony salt replacement method [25] is commonly used in zinc hydrometallurgy to remove Co and Cd from sulfuric acid solutions. In this method, antimony salt promotes the formation of a Co–Sb intermediate compound via electrochemical reactions. At elevated temperatures, this compound alters the deposition potential of Co, and acts as the cathode in a micro-galvanic cell, enabling cobalt removal. During this process, trace impurities are also removed [26]. This method is considered efficient, low-cost, simple, and environmentally friendly. It effectively removes Co with minimal interference to other metals, making it an economically viable and practical approach for cobalt removal in zinc refining. However, challenges arise in practical production. The addition of large amounts of zinc powder to hot sulfuric acid solutions leads to a temperature gradient, reducing the efficiency and rate of the replacement reaction and lowering zinc powder utilization [27]. Moreover, hydrolysis of zinc powder and deposition of reaction products form surface coatings that block Co^{2+} discharge, hinder zinc ion diffusion into the solution, and impede electron transfer in the displacement process, ultimately degrading the impurity removal efficiency [28]. Therefore, improving and overcoming the limitations of the zinc powder–antimony salt replacement method is essential for enhancing the value of nonferrous metal resources and achieving environmentally sustainable development.

Ultrasound refers to microwaves with frequencies between 20 kHz and hundreds of megahertz. In hydrometallurgy, ultrasonic waves can cause the vibration of molecules or atoms, which helps to evenly disperse and stabilize solid particles in the liquid by triggering strong vibration and cavitation effects in the liquid, and prevent agglomeration between particles [[29], [30], [31]]. The uniformity of the reaction is ensured, the efficiency of the reaction and the recovery rate of the metal are improved. For example, Xu [32] et al. used ultrasound to enhance the leaching of valuable metals from zinc oxide dust by tartaric acid and sulfuric acid. The results show that the ultrasonic wave can produce strong microfluidic effect in the liquid, forming small liquid vortex and high frequency vibration flow. This microfluidic effect can promote the movement and collision of particles and solutes in the solution, thus accelerating the reaction. Wang [33] et al. used ultrasound to efficiently recover valuable metals from low-grade zinc residue. When ultrasonic waves propagate in the liquid, microbubbles are generated, and the bubbles shrink rapidly to produce high temperature and high pressure, forming high-speed jets and shock waves, so that the boundary layer is constantly eroded to produce a new reaction interface and accelerate heat and mass transfer. Wei [34] et al. studied the experiments of leaching zinc and germanium from high-silicon zinc oxide dust. The results show that the synergistic effect of the ultrasonic field and sodium lauryl sulfate can significantly improve the leaching efficiency. Ultrasonic waves create an instantaneous high-temperature and high-pressure environment in liquid media through their mechanical and cavitation effects, promoting chemical reactions and increasing the reaction rate.

Based on the above reasons, a method of purifying zinc sulfate solution with the help of ultrasonic assisted antimony salt activated zinc powder was proposed. In this study, the removal effects of Co and Cd by conventional purification process and ultrasonic enhanced purification process were investigated, and the differences in reaction temperature, reaction time, amount of zinc powder and amount of antimony salt were compared between the two processes. The mechanism of ultrasonic enhanced purification of zinc powder was also discussed. The aim is to achieve efficient, economical and environmentally friendly purification, and to provide beneficial reference for the sustainable development of zinc hydrometallurgy industry.

Section snippets

Materials

This zinc sulfate solution is derived from a section of the post-pressing solution after purifying the supernatant obtained from the neutral leaching of zinc calcined products with sulfuric acid in the wet zinc smelting process of a certain enterprise in Inner Mongolia. The pH of the solution is between 5.1 and 5.4, and its main components are shown in Table 1. The zinc powder used in the reaction process is industrial hydraulic atomized zinc powder, prepared by a certain enterprise in Inner …

Results and discussion

The mechanism of removing Co and Cd from zinc sulfate solution by zinc powder purification is that zinc powder acts as a reducing agent and reduces cobalt ions (Co^{2+}) in the solution to cobalt metal through displacement reaction. This process can be expressed by formula (2):[35]$Zn(s)+M^{n+} \rightarrow Zn^{n+} + M\,(s)$where M is the impurity metal ion in the solution, and n is its charge number. In theory, zinc can spontaneously displace cobalt. However, in the actual reaction, the rate of direct replacement of cobalt …

Conclusion

In view of the existing problems in the purification process of Co and Cd, an innovative method of purifying Co and Cd from zinc sulfate two-stage pressing liquid by ultrasonic wave was proposed in this study. By comparing the effect of ultrasonic assisted purification with traditional methods, the study aims to optimize the utilization of zinc powder, reduce production time and cost, and establish a theoretical framework for the advanced purification of neutral leachate supernatant in …

CRediT authorship contribution statement

Xudong Li: Writing – original draft, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Yingjie Xu:** Writing – original draft, Methodology, Data curation. **Ningting Li:** Writing – original draft, Methodology, Data curation. **Bo Yang:** Writing – original draft, Methodology, Data curation. **Hongying Xia:** Validation, Supervision, Software, Resources, Methodology, Funding acquisition, Conceptualization. **Kan Yu:** Writing – original draft, Methodology, Data curation. **Hao Yang:** …

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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