

Progress in Phosphorus Removal and Utilization from Converter Slag

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Abstract

Phosphorus, an irreplaceable non-renewable resource essential for agriculture and industry, holds significant economic value. However, global reserves of high-grade phosphate ores are rapidly depleting, making the search for suitable alternatives an urgent priority. Concurrently, the surging demand for steel driven by rapid global industrialization has positioned converter steelmaking—a pivotal process in steel production—as a critical industrial activity. Currently, converter steelmaking accounts for 80% of global steel production, with developed nations generating approximately 200 million tons of steel slag byproducts annually. This massive slag output not only occupies vast land resources but also poses environmental risks. Yet, steel slag is rich in phosphorus, representing a valuable secondary phosphorus resource. Thus, the effective recovery of phosphorus from converter slag is key to addressing phosphorus scarcity. This paper focuses on advanced technologies for efficient phosphorus recovery from converter slag, systematically summarizing global innovations in phosphorus removal from phosphorus-containing steel slag. It reviews metallurgical processes for phosphorus extraction via pyrometallurgical and hydrometallurgical methods, providing a critical research foundation and directional guidance for achieving high-efficiency phosphorus recovery in industrial applications.

Keywords

Converter Steelmaking; Dephosphorization; Pyrometallurgical Dephosphorization.

1. Introduction

With the rapid development of the economy, the ferrous and non-ferrous metallurgical industries have made significant contributions to heavy industrial economic growth. The continuous annual increase in production of various steel products in the steelmaking industry has resulted in massive quantities of metallurgical solid waste generated by industrial development. Steel slag, a primary solid waste in metallurgical processes, can be categorized into electric furnace slag, open-hearth furnace slag, and converter slag, with a production rate equivalent to 8%-15% of crude steel output. Currently, global annual production of steel slag by-products exceeds 200 million tons, with cumulative stockpiles surpassing 2 billion tons. In countries like China and the United States where converter steelmaking accounts for over 80% of production, converter slag constitutes more than 70% of total steel slag emissions, making its treatment and utilization the focal point of comprehensive steel slag management.

Given its rich content of CaO and FeO, the optimal utilization of converter slag lies in its internal recycling as metallurgical flux or raw material within metallurgical processes. When substituting limestone as sintering flux, converter slag can improve sinter structure, enhance sinter quality, reduce sintering speed, and lower fuel consumption. As blast furnace flux, it replaces quicklime and dolomite

while reducing flux consumption and coke ratio. When employed as hot metal desulfurizer, it accelerates desulfurization speed, minimizes iron loss, and facilitates slag removal. Additionally, the iron content in slag can be recycled, thereby reducing iron ore consumption^[2]. However, these utilization methods remain confined to internal metallurgical recycling with low efficiency rates, primarily constrained by the slag's high phosphorus content. Phosphorus segregation at grain boundaries during solidification and heat treatment adversely affects steel's mechanical properties, causing phosphorus contamination that deteriorates weldability, low-temperature ductility, and toughness. High-quality steel requires extremely low phosphorus content, yet external treatment methods risk losing valuable phosphorus resources. Therefore, converter slag dephosphorization has become a critical research focus. Although international scholars have conducted extensive experimental studies, no industrially scalable process suitable for large-scale steelmaking applications has been established^{[34][35]}. Existing dephosphorization techniques either involve prohibitive equipment costs or fail to meet enterprise-standard phosphorus removal rates, presenting persistent limitations. Mere addition of slag to sintering or steelmaking processes would result in sluggish resource utilization, leaving most converter slag unprocessed, leading to cultivated land occupation and pollution of groundwater, soil, and air^[18].

Recent advancements in converter slag dephosphorization processes focus on modifying slag mineral phase structures through physical and chemical methods to enhance phosphorus removal efficiency. This paper provides a comprehensive analysis of existing dephosphorization technologies, examining their advantages, limitations, and effectiveness under practical conditions, with particular emphasis on mechanistic processes and required environmental parameters. Finally, future prospects for dephosphorization technologies are proposed from multiple perspectives, establishing a research foundation for the resource utilization of phosphorus-containing steel slag.

2. Present Situation and Application Field of Phosphorus Supply in the World

Phosphorus, a non-renewable resource, is crucial for global agriculture and industry. As an essential element in manufacturing numerous vital materials such as batteries, pharmaceuticals, and food products, it serves as a raw material in technologically advanced sectors like semiconductors and pharmaceuticals. Notably, Nattorp et al.^[3] highlighted that high-purity phosphoric acid derived from high-grade phosphorus has become one of the most critical components in mRNA vaccines and PCR (polymerase chain reaction) tests for combating COVID-19, garnering worldwide attention. However, high-grade phosphate rock deposits are rapidly depleting, with projections suggesting potential exhaustion within a century. Consequently, identifying alternative phosphorus resources has become imperative for sustainable phosphorus supply. Converter steel slag, both in quantity and quality, represents a robust secondary phosphorus resource. Its advantages as a phosphorus substitute include relatively high phosphorus content, abundant availability, stable phosphorus speciation, minimal contamination risks, and thermochemical properties akin to primary phosphate ores.

Therefore, recovering phosphorus from alternative sources to meet future demand is critically important. Currently, sewage sludge and converter steel slag represent the two most significant secondary phosphorus resources. Globally, phosphorus recovery from converter steel slag has gained increasing attention in recent years as a vital alternative resource. Phosphorus is primarily extracted from high-grade phosphate ores, which typically contain 28-40% P_2O_5 by mass^[3]. In some countries, phosphorus obtained from phosphate ore extraction falls short of meeting P_2O_5 consumption demands, while substantial crude steel production generates massive quantities of converter slag. Thus, converter steel slag emerges as another crucial secondary phosphorus reservoir. Notably, phosphorus ranks among the most detrimental elements to steel quality in steel production. To date, numerous methods have been explored to remove or recover phosphorus from steelmaking processes.

Steel, the most widely used metallic material in society with the largest consumption volume, is projected to see continuously rising global demand in the future. In modern steelmaking processes, approximately 100 kg of converter slag is generated per ton of crude steel produced. Consequently,

the phosphorus enriched in converter slag during steelmaking is substantial and will further increase with growing steel output. Notably, low-phosphorus iron ores currently used in steelmaking are also facing depletion. The value of converter slag as a secondary phosphorus resource will thus be significantly enhanced^[1].

Due to the diversity of raw materials and complexity of smelting procedures in steel production, the chemical composition of steel slag is highly heterogeneous. Generally, most steel slags can be classified into three categories: basic oxygen furnace slag, blast furnace slag, and electric arc furnace slag. Converter slag, the most extensively studied type, constitutes approximately 70% of China's steel slag production. The primary components of steel slag include CaO, SiO₂, Fe₂O₃, Al₂O₃, and MgO, with trace amounts of MnO, P₂O₅, and TiO₂. The calcium, silicon, and phosphorus nutrients in steel slag benefit plant growth, while its high-temperature calcination enhances the solubility of these elements, making them more bioavailable. This qualifies steel slag as a high-quality mineral fertilizer. Furthermore, steel slag demonstrates potential in construction industries for cement and concrete production due to its chemical similarity to Portland cement, containing cementitious mineral phases such as tricalcium silicate and dicalcium silicate, which exhibit hydration processes comparable to conventional cement.

In wastewater treatment, steel slag shows promise owing to its large specific surface area, porous structure, and cost-effectiveness. Surface modification techniques can enhance its adsorption efficiency for pollutants. Recent studies have extensively explored steel slag applications in treating various wastewaters, including mining effluents, industrial wastewater, and dye-contaminated water. Beyond these applications, steel slag exhibits significant potential in marine engineering, ceramic manufacturing, and CO₂ sequestration technologies.

3. Research Progress on Phosphorus Removal from Phosphorus-Containing Steel Slag in Converter

The specific pathway for phosphorus recovery and comprehensive utilization in converter slag lies in separating the phosphorus-enriched C₂S-C₃P solid solution from other mineral phases in the slag. Numerous researchers have investigated the physicochemical differences between the C₂S-C₃P solid solution and coexisting phases in slag, proposing various separation strategies. Additionally, enhancing the enrichment of P₂O₅ within the C₂S-C₃P solid solution is critical for improving phosphorus separation efficiency from steelmaking slag, which has attracted substantial research attention. In practical applications, pyrometallurgical and hydrometallurgical methods are predominantly employed to extract phosphorus-containing mineral phases from converter slag. Pyrometallurgical approaches involve high-temperature reduction processes where suitable reductants are added to recover phosphorus and iron from slag. Current studies demonstrate the exceptional potential of these methods for elemental phosphorus recovery. Hydrometallurgical techniques, on the other hand, focus on selectively dissolving phosphorus from slag using tailored solutions to precipitate phosphoric acid or phosphorus compounds. This strategy leverages the higher aqueous solubility of the C₂S-C₃P solid solution compared to other mineral phases, enabling its separation from the slag matrix.

3.1 Pyrometallurgical Dephosphorization and Phosphorus Recovery from Steel Slag

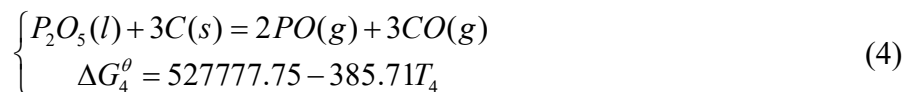
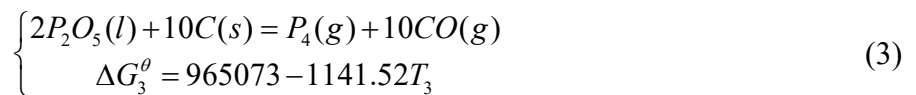
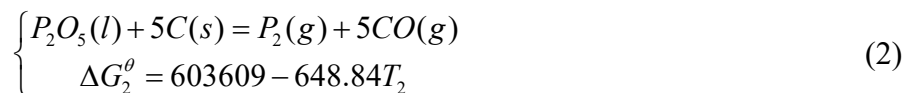
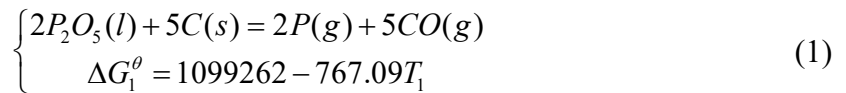
The reduction method refers to processes that remove phosphorus from converter slag using reductants such as coke. Currently, this approach encompasses two established methods: carbothermic reduction and silicothermic reduction.

3.1.1 Carbothermal Reduction Method

The carbothermic reduction method utilizes coke as a reductant to remove phosphorus from converter slag through reduction reactions^[37]. The reaction mechanism is illustrated in the figure below. Numerous experimental studies by scholars have concluded that the reduction method is the most

efficient approach for phosphorus removal from converter slag, achieving a dephosphorization rate of approximately 85%, with 30%–40% of the phosphorus volatilized as vapor^[5].

The reaction for phosphorus volatilization via carbothermic reduction in converter slag, under standard conditions, involves the reduction of pure P₂O₅ by carbon to form gaseous P₂, as shown in the following equation:



During carbothermic dephosphorization, most phosphorus in the slag is reduced by coke and volatilized as phosphorus vapor, while a small fraction remains unreduced and retained in the slag. To enhance phosphorus removal efficiency, scholars have conducted systematic investigations into the influence patterns of key parameters such as basicity, FeO content, carbon equivalent, bottom-blowing gas flow rate, and smelting duration on the reduction-based dephosphorization process.

Liu Yaqin, Lin Lu, et al.^[4]comprehensively evaluated converter slag dephosphorization experiments under various conditions and summarized the influence patterns of key factors in carbothermic reduction, including “basicity”, “FeO mass fraction”, “carbon equivalent”, and “bottom-blowing gas flow rate on phosphorus removal”. They synthesized optimal dephosphorization rates achieved at different temperatures across experiments, concluding that “dephosphorization efficiency increases with rising temperatures”. Additionally, experimental results from Guo Ruihua, Wang Shuhuan, et al.^{[5][6]} demonstrated that slag dephosphorization rates decline with increasing basicity. At a basicity of 1.0, the dephosphorization rate reached 90%, whereas it dropped to approximately 10% at a basicity of 2.0.

Dong et al.^[11] investigated the effects of slag basicity”, “graphite dosage”, “temperature”, and “crucible type” on phosphorus reduction and identified optimal conditions. They found that reducing slag basicity from 1.49 to 0.50 suppressed manganese reduction but enhanced phosphorus reduction rates. By monitoring phosphorus content in phases over time at varying temperatures, they determined optimal reaction temperatures based on equilibrium conditions and reaction kinetics. Their study also revealed that graphite dosage moderately influenced P₂O₅ reduction behavior, while crucible type significantly affected phosphorus reduction rates and distribution ratios.

Zhao Chenglin et al.^[13]explored different reaction modes using coke powder as a reductant to identify optimal process parameters. Their results revealed that “dephosphorization rates rose continuously with increasing carbon equivalent and temperature”, achieving over 30% improvement when carbon equivalent increased from 2.0 to 3.0. Zhou Chaogang et al.^[12]focused on “gasification dephosphorization mechanisms”, particularly analyzing the relationship between “bottom-blowing gas flow rate” and dephosphorization efficiency under varying carbon equivalents. They reported a

maximum dephosphorization rate of 42% at 2× carbon equivalent and 300 m³/h bottom-blowing flow rate.

The above studies directly employed coke to reduce phosphorus in converter slag under a reductive atmosphere, generating volatile phosphorus for recovery. In contrast, Ai Liqun et al.^{[14][15]} adopted a microwave preheating followed by carbothermic reduction approach to recover phosphorus and iron from slag, yielding highly effective results. Their experiments demonstrated that microwave heating significantly enhances phosphorus removal, achieving 80% dephosphorization rates across slags of varying compositions, with carbon equivalent and temperature identified as the most critical influencing factors.

To further refine this process, Ai Liqun, Lü Yan, et al.^[16] investigated phosphorus removal efficiency under different reduction conditions for four slag types. They found that particle size critically affected heating efficiency and dephosphorization performance, with 0.150-0.300 mm being optimal. Additionally, 3× carbon equivalent delivered the best results, balancing heating rates and dephosphorization efficiency without excessive energy consumption. These advancements highlight the potential of integrated thermal and microwave-assisted methods for scalable, high-efficiency phosphorus recovery from converter slag.

3.2 Physical Method

3.2.1 Magnetic Method

Phosphorus-containing slag primarily consists of oxides of calcium, iron, magnesium, silicon, aluminum, manganese, and phosphorus, which can form distinct phases. These phases exhibit varying physicochemical properties under different treatment conditions. Magnetic separation leverages the differing magnetic characteristics of mineral phases in steel slag, applying high-tesla magnetic fields to achieve phase separation. Yokoyama et al.^[19] analyzed the composition of converter slag from domestic steel producers using an electron probe microanalyzer, investigating the forms of phosphorus post-solidification and discussing equilibrium relationships between phosphorus-containing phases. They categorized the phases in slag into four groups:

Phase A: High phosphorus concentration (>10%) with negligible iron content;

Phase B: Composed primarily of Ca and Si, containing trace phosphorus and minimal iron;

Phase C: Nearly pure FeO phase;

Phase D: A mixed phase based on CaO-SiO₂-FeO.

Phase D exhibited paramagnetic properties, while Phases A and B showed diamagnetic behavior. The non-phosphorus matrix phase displayed magnetic susceptibility 100 times lower than phosphorus-containing phases. By exploiting this magnetic disparity under a 2T magnetic field, approximately 50% of phosphorus was recovered via magnetic separation[2]. To validate this process, Yokoyama et al.[19] conducted experiments using a superconducting magnet-based high-field generator, recovering 50% of phosphorus from raw slag, thereby confirming the feasibility of magnetic separation.

Kubo et al.^[20] investigated factors such as slag particle size, solid-liquid ratio, and separation passes on phosphorus recovery efficiency. Their results showed that phosphorus recovery rate decreases with increasing slag particle size under fixed magnetic field strength, as coarse particles often contain mixed phosphorus-rich and matrix phases. Higher solid-liquid ratios improved phosphorus recovery by enhancing the separation of phosphorus-rich phases. Increased magnetic field strength reduced matrix phase contamination, yielding higher phosphorus concentrations in collected fractions. However, weaker fields increased mixed-phase contamination, degrading product quality. Increasing the number of separation passes under strong magnetic fields improved phase separation efficiency. Optimal conditions (32 mm particle size, solid-liquid ratio of 32, 5 separation passes) achieved a phosphorus recovery rate of 62% with near-equal composition in the enriched phase.

These studies utilize differences in physical properties between phosphorus-containing and other phases in converter slag for dephosphorization, with reported phosphorus removal rates typically ranging from 30% to 60%. Critical influencing factors include magnetic field strength, separation passes, and feedstock particle size. While magnetic separation represents an innovative approach, challenges persist due to the intergrowth of phosphorus-bearing phases with other phases in converter slag. Even when successfully separated, the recovered phosphorus-rich phases often contain impurities. Current research gaps include mechanistic studies on phase separation dynamics and phosphorus enrichment during magnetic processing, which are essential for optimizing this method for industrial scalability.

3.2.2 Flotation

The flotation method exploits the physical property that phosphorus-enriched phase particles in molten converter slag have lower density than the liquid phase, enabling the separation of phosphorus-rich phases through upward flotation[4]. Several scholars have conducted research using this approach, exploring dephosphorization rates under varying conditions and the influence of different factors on phosphorus removal efficiency. ONO et al.^[23] designed experiments based on the principle that phosphorus-rich phases exhibit lower density than molten slag. They investigated methods to recover valuable components such as FeO, MnO, and CaO from slag by isolating P₂O₅-enriched dicalcium silicate particles via flotation, while examining the impact of various parameters on dephosphorization rates.

Results indicated that the suspended separation of dicalcium silicate requires slow cooling, with an average cooling rate below 2.0°C/min. Under such controlled cooling conditions, phosphorus-rich phases concentrated at the top, while FeO_t phases accumulated at the bottom. Additionally, oxygen blowing into the molten slag prior to cooling improved separation efficiency. However, challenges related to flotation speed—such as particle agglomeration, positional shifts caused by residual liquid settling, and the dependence of particle size on cooling rates—necessitate further in-depth study.

3.2.3 Supergravity Method

Steel slag, a major byproduct of steelmaking, accounts for 10%-20% of crude steel production. Due to phosphorus content, vast quantities of steel slag are disposed as waste, leading to significant land resource occupation and environmental pollution. Thus, the key challenge lies in separating phosphorus from steel slag and recycling valuable components such as FeO, Fe₂O₃, MgO, MnO, and CaO back into ironmaking and steelmaking processes. Current methods involve modifying steel slag with additives like SiO₂, Al₂O₃, TiO₂, and CaF₂ to enhance phosphorus solubility in enriched phases, followed by magnetic separation^{[7][8][9]}. However, the micron-scale size and embedded distribution of phosphorus-enriched phases hinder effective magnetic separation. Consequently, novel approaches are required to isolate phosphorus-rich phases and advance slag recycling.

The role of supergravity in controlling phase distribution has been established and successfully applied to separate and recover valuable elements from various slags, as well as to remove impurities from alloy melts^[25]. Since phosphorus in steel slag primarily crystallizes in the nC₂S-C₃P solid solution—which has lower density than the residual melt and precipitates earlier during solidification—it is hypothesized that supergravity could enable phosphorus-enriched phase separation. The supergravity method leverages the significant density differences between mineral phases in converter slag, utilizing centrifugal force under hypergravity to separate phosphorus-rich and iron-rich phases for dephosphorization^[4].

Recent studies have employed supergravity to concentrate phosphorus-containing phases in converter slag for recovery. Phosphorus predominantly enriches in the 2CaO-SiO₂ phase, which begins precipitating during slag cooling and solidification. As the precipitated phase exhibits lower density than the residual melt, phosphorus-enriched phases can be separated via supergravity. Li C et al.^[24] applied supergravity separation technology to isolate phosphorus-rich and iron-rich phases from a CaO-SiO₂-FeO-MgO-P₂O₅ melt. Microstructural analysis revealed that phosphorus-rich phases were retained by the filter, while the residual slag primarily consisted of iron-rich phases. This method

achieved a phosphorus recovery rate of 76.67% in the phosphorus-rich slag and an iron recovery rate of 85.02% in the iron-rich slag.

3.2.4 Capillary Tube Method

Miki et al.^[26] developed a novel method to enrich phosphorus from slag using the capillary effect of porous media, leveraging the significantly higher melting point of the phosphorus-rich phase (C_2S-C_3P solid solution) compared to the FeO matrix phase. In their experiments, they investigated the role of capillary action in sintered CaO, Fe_2O_3 , and $CaCO_3$ tablets, confirming that the capillary effect of sintered CaO can facilitate the separation between the phosphorus-containing solid phase C_2S-C_3P and the FeO-rich liquid slag in molten converter slag. The results demonstrated that once the phosphorus-rich solid phase (C_2S-C_3P solid solution) and FeO-rich liquid phase pass through the absorption bed, they can be effectively separated. When sintered CaO particles were added to molten converter slag coexisting with solid and liquid phases, the FeO-rich liquid slag was absorbed into the sintered CaO interior via capillary action, while the phosphorus-containing solid phase C_2S-C_3P remained on the surface of the sintered CaO, achieving solid-liquid phase separation. The study also noted that using calcium carbonate ($CaCO_3$) as an adsorbent could further increase the dephosphorization rate to 91%. However, this method remains in the experimental stage and faces several unresolved challenges, limiting its widespread adoption.

3.3 Research Progress on Wet Dephosphorization and Phosphorus Recovery from Steel Slag

The principle of hydrometallurgical phosphorus recovery from steelmaking slag lies in the higher solubility of the C_2S-C_3P solid solution in aqueous solutions compared to other mineral phases, enabling its separation from the slag matrix, as illustrated in Figure 1. Based on this principle, phosphorus recovery from synthetic, enriched, or practical steelmaking slags has been promoted through leaching with different solutions under varying conditions.

Sugiyama et al.^[31] investigated methods for phosphorus enrichment and recovery from slag. They first studied phosphorus elution using HNO_3 aqueous solutions via batch and continuous washing methods. The batch method achieved 82% phosphorus dissolution from dephosphorized slag within 30 minutes using 1.0 mol/L HNO_3 , demonstrating its suitability for bulk phosphorus extraction from the matrix phase. However, this method lacked selectivity, dissolving all slag components indiscriminately. In contrast, the continuous washing method with 0.05 mol/L HNO_3 dissolved 22% of the slag over 100 minutes, showing stronger selectivity for phosphate dissolution but proving unsuitable for bulk phosphorus extraction. The aqueous solution obtained from the batch method was further treated by adding calcium hydroxyapatite to remove iron under pH=1.5 and t=4 h conditions, achieving near-complete iron removal. Adjusting the solution pH to 7.0 precipitated phosphorus, confirming the feasibility of this approach for phosphate recovery and enrichment.

QIAO Y et al.^[33] explored phosphorus removal from steelmaking slag using an acidic aqueous solution composed of citric acid, NaOH, HCl, and deionized water. Parameters such as leaching temperature, slag particle size, and solution pH were optimized. Results showed that temperature had negligible impact on phosphorus dissolution but significantly affected iron dissolution. Phosphorus dissolution rates increased with smaller slag particle sizes and lower pH values. Under optimal conditions (pH 2.2, 25°C, particle size 75-125 μ m), phosphorus dissolution exceeded 90%, while iron leaching remained at 30%. Post-leaching characterization via XRD and SEM revealed selective dissolution of nC_2S-C_3P solid solutions, leaving iron-enriched residues. Compared to conventional acid leaching, this method avoids strong acids and achieves higher dephosphorization rates than traditional flotation or magnetic separation. Future research will focus on phosphorus recovery from post-leaching solutions for practical applications.

Yang Jinxing et al.^[32] investigated phosphorus dissolution from converter slag using oxalic acid, examining the effects of slag-to-acid ratio, reaction time, solution temperature, and slag particle size. Results indicated a slight increase in phosphorus dissolution with rising temperature, with optimal conditions at a slag-to-acid ratio of 4 and near-equilibrium attainment after 120 minutes. Larger slag particles reduced dissolution efficiency, with the highest rate (52.49%) achieved for particles <48 μ m.

These findings highlight the potential of oxalic acid for selective phosphorus recovery under controlled conditions.

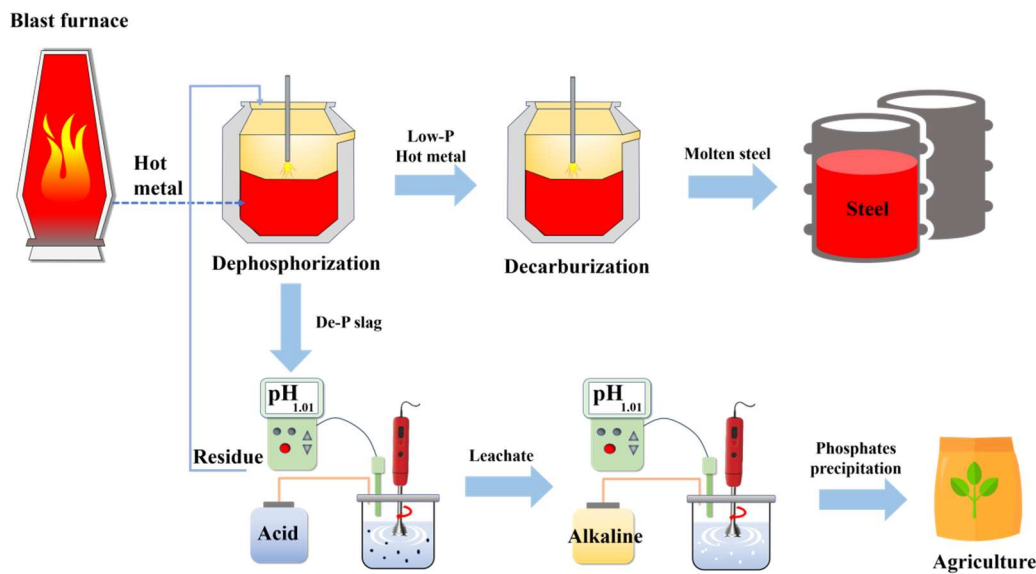


Figure 1. Wet dephosphorization process

4. Conclusion and Prospect

In recent years, the dephosphorization treatment of steel slag has demonstrated significant economic and environmental benefits. Slag contains substantial amounts of reusable components, including phosphorus. Building on previous research, numerous dephosphorization technologies for converter slag have been developed, each exhibiting varying practical removal efficiencies and distinct advantages and limitations. However, both experimental and industrial practices prioritize achieving the highest dephosphorization efficiency at the lowest cost. Against the global backdrop of “carbon peaking and carbon neutrality” goals, enhancing steel slag utilization has emerged as a critical strategy for advancing resource-efficient and environmentally sustainable societal development. High-rate resource recovery from steel slag will undoubtedly contribute substantially to realizing these dual-carbon objectives. Despite the diversity of existing steel slag utilization methods, large-scale industrial implementation remains challenging.

This paper synthesizes global research progress and current practices in steel slag utilization, reviewing the properties of steel slag and the state of processing technologies. Finally, it proposes integrating advanced eco-friendly concepts such as “green steel” and “intelligent steelmaking” into slag treatment processes. These innovations aim to address challenges in recycling and high-value utilization of waste steel slag, plastics, and agricultural/forestry residues, thereby fostering synergistic improvements in national economic, social, and environmental outcomes.

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