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Research Progress on Resource Utilization of Phosphate Tailings

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ABSTRACT

As a non-renewable strategic mineral resource, phosphate ore is the core raw material for phosphate fertilizer production, new material manufacturing and other fields. The high proportion of low and medium-grade phosphate ore in China leads to an annual emission of phosphate tailings (PT) exceeding 10 million tons, with a huge cumulative stockpile and low comprehensive utilization rate. The environmental and safety problems such as water pollution and geological hazards caused by stockpiling have become increasingly prominent. Under the requirements of the "dual carbon" strategy and circular economy development, the transformation of PT from stockpiling disposal to efficient resource utilization has become an inevitable trend. PT is rich in valuable components such as calcium, magnesium, phosphorus and silicon. At present, its resource utilization has formed four core technical paths: recovery of valuable elements, building material utilization, agricultural conversion, and preparation of environmental functional materials, showing good application potential in fields such as bulk consumption, nutrient reuse and pollution control. However, it still faces industrialization bottlenecks such as complex processes, low added value and poor adaptability of soil application. This paper systematically sorts out the current situation of PT generation and disposal, the environmental risks of traditional disposal, and the research progress of various resource utilization technologies, analyzes the advantages and limitations of different technical paths, and looks forward to its development direction of synergization, high-valueization, greenization and intelligence, in order to provide reference for the technological innovation and industrial landing of efficient resource utilization of PT and the construction of a closed-loop industrial chain.

KEYWORDS

Phosphate tailings; Resource utilization; Solid waste disposal; Circular economy.

1. INTRODUCTION

Phosphate ore is the core raw material for phosphate fertilizer production, food processing, new material manufacturing and other fields, and it is a non-renewable strategic mineral resource [1]. China ranks second in the world in phosphate ore reserves and first in output. At present, low and medium-grade phosphate ore has become the main body of phosphate ore mining. A large amount of PT is generated during the phosphate ore beneficiation process. Its stockpiling not only causes resource waste, but also brings environmental and safety risks such as eutrophication, heavy metal pollution and geological hazards [2]. Under the background of the "dual carbon" strategy and circular economy, it is of great practical significance to find environmentally friendly treatment methods for PT to reduce environmental pollution and promote the transformation of PT from stockpiling disposal to resource utilization [3]. At present, the resource utilization of PT has formed a multi-path and multi-scenario technical system. This paper focuses on the full-dimensional technical progress of PT resource utilization, optimizes and sorts out the existing research results, and provides support for subsequent technological innovation and industrial landing.

2. RESOURCE UTILIZATION OF PHOSPHATE TAILINGS

2.1. Current Situation of PT Generation and Disposal

Phosphate ore has the characteristics of non-renewability. Long-term high-intensity mining and low utilization rate of phosphate fertilizer have aggravated the exhaustion of high-phosphate ore [4]. In the process of phosphate ore mining, high-grade and easily mined ore layers are developed first, leading to a gradual decrease in the average grade of phosphate ore and a continuous reduction in the proportion of P_2O_5 with the increase of mining years. The long-term extensive mining mode of "extracting high-grade ore and discarding low-grade ore" and the current situation of low utilization rate of phosphate fertilizer have further accelerated the exhaustion of high-phosphate ore resources. At present, China's proven phosphate ore reserves are 16.7 billion tons, with an average grade of only 16.95%, of which 97.5% are low and medium-grade phosphate ores, and the proportion of high-quality high-grade ore resources is extremely low [5].

Affected by the high proportion of low and medium-grade phosphate ore, 30~40 tons of PT are generated for every 100 tons of phosphate concentrate produced, with an annual emission exceeding 10 million tons [6]. As of 2020, the cumulative stockpiles of PT in China were approximately 1.2 billion tons, with a comprehensive utilization rate of only 10%~17%, which is much lower than the average utilization level of bulk industrial solid waste. A large amount of tailings have been stockpiled in tailings ponds for a long time [7].

2.2. Traditional Disposal Methods and Environmental Risks

Traditionally, mine backfilling is the main disposal method for PT. Long-term stockpiling not only occupies a large amount of land resources and generates high construction and operation and maintenance costs of tailings ponds, but also causes multiple environmental and safety risks. Pollutants such as organic flotation reagents and heavy metals in PT enter water bodies through rainwater leaching, which is likely to induce eutrophication; the tailings pile has poor stability, and landslides, debris flows and other geological hazards are likely to occur in the rainy season [8, 9]. The above problems are contrary to the goals of ecological environmental protection and the "dual carbon" strategy, and the demand for green and resource utilization of PT is extremely urgent.

2.3. PT Resource Utilization Technologies

PT is mainly composed of dolomite ($CaMg(CO_3)_2$) and is rich in elements such as calcium, magnesium, phosphorus and silicon, making it a secondary resource with both industrial and agricultural value. Its resource utilization mainly focuses on four directions: recovery of valuable elements, building material utilization, agricultural conversion, and preparation of new functional materials.

2.3.1. Recovery and Utilization of Valuable Elements

The recovery of valuable elements such as phosphorus, calcium and magnesium from PT is an important way to realize high-value utilization of resources. It mainly adopts metallurgical and mineral processing technologies, including leaching, calcination, calcination-leaching, calcination-carbonation and other processes [10]. Although the traditional pyrolysis process can realize the phase separation of calcium and magnesium, its high energy consumption (>1000 °C) and low separation efficiency have greatly restricted its industrial application [11]. Raiymbekov et al. enriched high-grade phosphate concentrate by selectively leaching carbonates with acetic acid [12]. Ding et al.

treated PT by a combined calcination-phosphoric acid leaching process, which not only enriched phosphate concentrate, but also realized the comprehensive recovery and full utilization of valuable elements such as phosphorus, calcium and magnesium in the tailings, with the recovery rate of phosphorus reaching as high as 99.86% [13]. Anawati et al. prepared calcium hydrogen phosphate dihydrate from PT by a combination of dilute sulfuric acid (0.29 mol/L) leaching and calcium oxide selective precipitation [14]. This path can recover high-value elements and improve the economic value of tailings, but it has problems such as large reagent consumption and complex process flow, which limits its large-scale application.

2.3.2. Building Material Utilization

Building material utilization is the main way to consume a large amount of PT stockpiles, which has the advantages of large dosage and high industrialization level. It is mainly used as filling materials, roadbed materials, geopolymers, cementitious materials, refractory materials and so on.

Some studies have prepared magnesium oxysulfide (MOS) cement by synergistically using calcined PT and acidic wastewater from phosphorous chemical industry. Phosphorus in acidic wastewater can promote the formation of cement hydration phases, significantly improve its compressive strength, water resistance and acid resistance, and simultaneously realize the efficient solidification of phosphorus and fluorine in wastewater. Although calcined PT slightly reduces the cement strength, it can improve the volume stability and simultaneously reduce the waste treatment and cement production costs [15]. Dolomite in PT can be calcined at 700-800 ℃ to prepare light-burned magnesium oxide (LBM) powder, which is used as the raw material for magnesium oxychloride (MOC) cement and MOS cement [16]; dead-burned magnesium oxide can be prepared by high-temperature calcination at 1200-1300 ℃, which can be compounded with bauxite and gypsum to prepare magnesium phosphate cement [17]. Liu et al. have confirmed through research that adding PT as an inert filler into concrete can improve the workability of concrete and effectively increase the slump [18], expanding its application scenarios in the field of building materials.

2.3.3. Agricultural Conversion

From the perspective of agricultural production, PT is rich in plant-available nutrients such as P, Mg, Ca and Si, among which Ca and Mg are key elements for plant growth and photosynthesis, making it a high-quality nutrient resource with great development potential. Using PT as raw material to prepare agricultural fertilizers can not only alleviate the pressure of resource wastes and environmental pollution, but also supplement soil nutrients and improve crop yield. Mixed composting of PT and organic wastes mediated by microorganisms can improve the solubility of insoluble phosphorus and further release the nutrient potential. The fertilizer utilization of PT has become an important research topic in the green development of industry and agriculture [19].

Zheng Jianguo et al. prepared polymerized calcium-magnesium phosphate fertilizer with a polymerization degree of 1-4, a P_2O_5 polymerization rate of more than 80%, and mass fractions of total P_2O_5, CaO and MgO of 50%~60%, 17% and 12% respectively by mixing and calcining phosphoric acid and PT according to the ratio through a high-temperature intermolecular dehydration polymerization process. After pilot-scale production, the industrial path of PT resource utilization was further expanded [20]. Sun Peng et al. added PT into compound fertilizer and systematically studied the physical and chemical properties and field fertilizer efficiency of the modified compound fertilizer. The results showed that PT can accelerate the nutrient release rate of compound fertilizer and improve the absorption of phosphorus, calcium and magnesium by plants, providing field test support for the fertilizer utilization of PT [21].

In addition to traditional fertilizer utilization, the soil application of PT has become a research hotspot, that is, converting tailings into ecological substrates with core soil functions through physical crushing, biochemical modification and other technologies for agricultural production and ecological restoration, a process also known as tailings soil formation [22]. Some studies have adjusted the pH value of acidic soil to neutral by modified phosphogypsum, verifying the application potential of phosphorus-based solid wastes in soil improvement [23]; the single or combined application of PT-based soil amendment with nitrogen-fixing bacteria and phosphorus-solubilizing bacteria can significantly increase the plant height, fresh weight of pakchoi, as well as soil nutrient content and enzyme activity [24]. In summary, PT has formed a variety of agricultural conversion paths including fertilizer utilization and soil improvement and restoration, with significant resource potential. However, PT has inherent defects such as too fine particles, extreme lack of organic matter, lack of aggregate structure, and poor water and fertilizer retention performance, so it cannot be directly returned to the field as a single soil-forming substrate; untreated PT is prone to soil erosion by water scouring, which not only makes it difficult to construct a stable soil structure, but also aggravates nutrient loss and causes secondary problems such as soil compaction, restricting the large-scale landing of tailings soil formation technology.

2.3.4. Environmental Functional Potential of PT

PT has weak alkalinity, excellent ion exchange capacity, and is rich in CaO and MgO, making it widely used in the fields of catalysis and adsorption, and becoming a high-quality carrier of the "treating waste with waste" resource utilization model. Studies have shown that dolomite, the main component of PT, is a natural adsorbent, which has adsorption capacity for heavy metal ions such as Cu(II), Pb(II), Co(II), Zn(II) and organic substances. The adsorption mechanisms include physical adsorption, electrostatic adsorption, cation-π interaction, ion exchange, and complexation with oxygen-containing functional groups such as -OH and CO_3^{2-} [19, 25]. Dolomite can activate peroxymonosulfate (PMS) to degrade tetracycline, with a degradation rate of 80.94% within 60 minutes, which is more than twice that under pure PMS oxidation conditions (37.01%) [26]. Liu et al. prepared sludge-based biochar by co-pyrolysis of sludge and PT. An appropriate amount of PT can improve the specific surface area, micropore volume, low-temperature hydrophobicity and high-temperature aromaticity of biochar, thereby enhancing its adsorption capacity for three polycyclic aromatic hydrocarbons (PAHs): phenanthrene, pyrene, and benzo[a]pyrene [27]. Wei et al. prepared a new type of material by compounding PT, phosphogypsum and starch in a certain ratio, with a maximum adsorption capacity of 31.28 mg/g for phosphate [28].

3. OUTLOOK ON PT RESOURCE UTILIZATION TECHNOLOGIES

In the future, the resource utilization technology of PT should develop towards synergization, high-valueization, greenization and intelligence. It is necessary to focus on breaking through the key technologies of synergistic utilization of PT and multi-source organic solid wastes, relying on organic substrates to make up for the inherent defects of PT such as lack of organic matter and poor water and fertilizer retention, simultaneously realizing nutrient activation of PT and harmless treatment of organic solid wastes, and expanding its application scenarios in fields such as compost improvement, soil remediation and antibiotic pollutant degradation; at the same time, deepen the research and development of green pretreatment processes such as calcination-leaching and targeted modification, improve the efficient and accurate recovery of valuable elements, develop high-value-added products such as high-purity magnesium salts and functional adsorption and catalytic materials, and break through the single limitation of traditional low-value-added building material utilization.

In addition, it is necessary to construct a full-life-cycle environmental risk management and control system for PT, improve the resource utilization product standards and safety evaluation specifications, promote the cross-industry integration of PT utilization with phosphorous chemical industry, agriculture and environmental protection industry, and combine big data and intelligent separation technology to realize accurate component matching of tailings and dynamic optimization of resource utilization paths, so as to help the low-carbon transformation of phosphorous chemical industry and the realization of the "dual carbon" strategic goal.

4. CONCLUSION

As a bulk industrial solid waste generated during the phosphate ore beneficiation process, the resource utilization of PT is a key link to solve the environmental constraints of the phosphorous chemical industry and realize resource recycling and efficiency improvement. At present, a variety of technical paths have been formed around the recovery of valuable elements, building material utilization, agricultural conversion and preparation of environmental functional materials, showing significant potential in improving the economic value of resources, realizing bulk consumption and expanding the application scenarios of "treating waste with waste". However, the existing technologies generally face industrialization bottlenecks such as complex process flow, low product added value and insufficient adaptability of soil application, which restrict their large-scale promotion and application. In the future, we should focus on the development direction of synergization, high-valueization, greenization and intelligence, strengthen the synergistic disposal and nutrient activation of PT and multi-source organic solid wastes, develop high-value-added functional materials, construct an environmental risk management and control and product standard system throughout the whole life cycle, promote the transformation of PT from single disposal to cross-industry collaborative recycling mode, and provide strong support for the green and low-carbon transformation of the phosphorous chemical industry and the realization of the "dual carbon" goal.

CONFLICTS OF INTEREST

The authors declare that they have no conflict of interest.

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