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Research article

Utilization path of bulk industrial solid waste: A review on the multi-directional resource utilization path of phosphogypsum

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ABSTRACT

Phosphogypsum is one of the hottest issues in the field of environmental solid waste treatment, with complex and changeable composition. Meanwhile, phosphogypsum contains a large number of impurities, thus leading to the low resource utilization rate, and it can only be stockpiled in large quantities. Phosphogypsum occupies a lot of land and poses a serious pollution threat to the ecological environment. This paper mainly summarizes the existing pretreatment and resource utilization technology of phosphogypsum. The pretreatment mainly includes dry method and wet method. The resource utilization technology mainly includes building materials, chemical raw materials, agriculture, environmental functional materials, filling materials, carbon sequestration and rare and precious extraction. Although there are many aspects of resource utilization of phosphogypsum, the existing technology is far from being able to consume a large amount of accumulated and generated phosphogypsum. Through the analysis, the comparison and mechanism analysis of the existing multifaceted and multi-level resource treatment technologies of phosphogypsum, the four promising resource utilization directions of phosphogypsum are put forward, mainly including prefabricated building materials, eco-friendly materials and soil materials, and new green functional materials and chemical fillers. Moreover, this paper summarizes the research basis of multi field and all-round treatment and disposal of phosphogypsum, which reduces repeated researches and development, as well as the treatment cost of phosphogypsum. This paper could provide a feasible research direction for the resource treatment technology of phosphogypsum in the future, so as to improve the consumption of phosphogypsum and reduce environmental risks.

1. Introduction

Phosphogypsum is one of the main by-products produced in the production of phosphoric acid in phosphorus chemical enterprises (Gu and Chen, 2020; Kovler, 2012). As one of the main strategic resources in the world, especially in agriculture and military industry, phosphoric acid is produced from the treatment of apatite with sulfuric acid (H_2SO_4). The production of PG is very tremendous in worldwide (Zhang et al., 2012a), because phosphorus chemical industries are widely distributed all over the world. Approximately 1–3 tons of PG is generated per ton of apatite consumed for an annual worldwide production about 258 Mt in 2018 (Saadaoui et al., 2017). Over 280 Mt of PG output every year in the world is currently being discarded at coastal or else and stored at production sites as stockpiles (Maazoun and Bouassida, 2018), with about 80 Mt emitted in China (Ding et al., 2019). The familiar formula of the production of PG can be written as Eq. (1).

$$Ca_{10}(PO_2)_6F_2 + 10H_2SO_4 + 20H_2O \rightarrow 6H_3PO_4 + 10(CaSO_4 \cdot 2H_2O) + 2HF^{[7]} \quad (1)$$

As environmental issues have become the focus of attention in the process of social development, PG has become one of the most important

Abbreviations: PG, Phosphogypsum; REE, Rare earth elements; MTE, Metal trace elements; α-HH, α-Hemihydrate (α-CaSO4·0.5H2O); CTAB, Ammonium bromide; PEG, Polyethylene glycol; PG-SPC, PG-sulfur polymer cements; HH-CSWs, Calcium sulfate hemihydrate whisker; CSWs, CaSO4 whiskers; α-CSH, α-Calcium sulfate hemihydrate; SRB, Sulfate reducing bacteria; SDBS, Sodium dodecyl benzene sulfonate; nHAp, Hydroxyapatite nanoparticles; AFt, Ettringite; FA, Fly ash; BCP, Biphasic calcium phosphate; CLG, Chemical Looping Gasification; BMED, Bipolar membrane electrodialysis; CMS, CO2 mineral sequestration; CCUS, Carbon capture, utilization, and storage; CCS, Carbon capture and storage; C–S–H, Calcium-Silicate-Hydrate.

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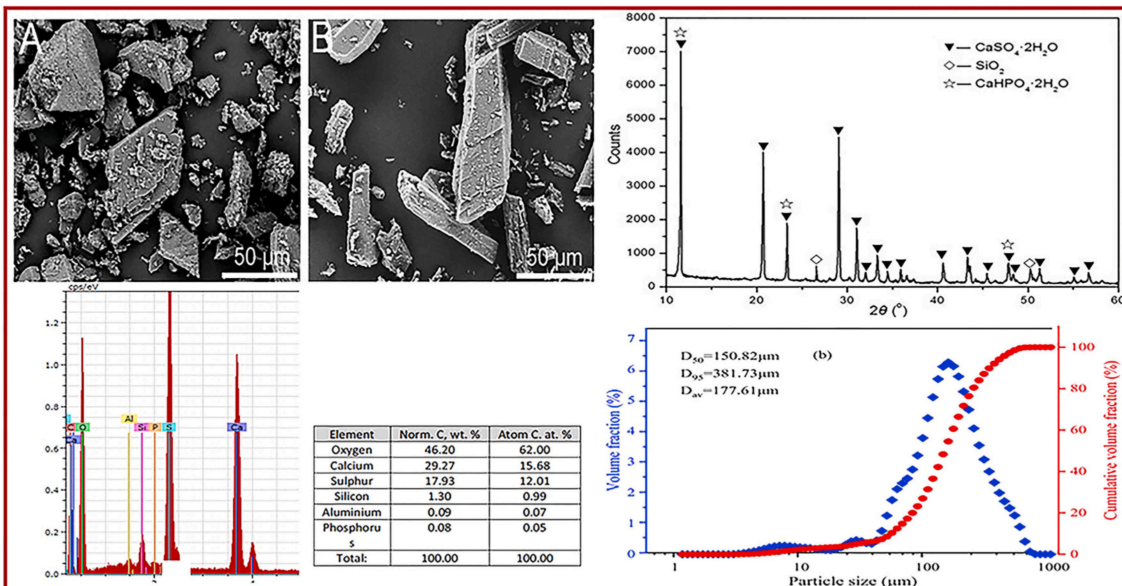
striking spots of environmental protection problems (Wang et al., 2018; Ngsbragaa, 2018; Ben Chokriet al., 2017; Perez-Lopez et al., 2016; V-M et al., 2021; D, 1977; Lieberman et al., 2020; Liu, 2100). The global resource utilization rate of phosphogypsum is less than 15%. With the continuous decline of phosphate rock grade, the production of phosphogypsum increases year by year (Tayibi et al., 2009). The main components of phosphogypsum are $CaSO_4{\cdot}nH_2O$ (n = 0, 1.5, 2), mixed with calcium phosphate in various forms, silica and a broad range of other impurities such as iron oxide (Fe_2O_3), magnesium (Mg) and aluminum (Al), sulfides, organic matter and metal trace elements (MTE) (Hammas-Nasri et al., 2016; Masmoudi-Soussi et al., 2019; El Zrelli et al., 2019; Kuzmanovic et al., 2020). These impurities mainly exist in phosphogypsum in the form of lattice doping, atom replacement, surface adsorption and interstitial filling. The impurities and the lattice morphology seriously restrict the resource utilization of PG (Szajerski, 2020). Presently, research on the resource utilization field of PG primarily centers on the following aspects: preparing cement coproduced sulfuric acid (Rosales et al., 2020), preparing building materials (Wang et al., 2020a), soil conditioner (Samet et al., 2019), and preparing high-strength gypsum (Lu et al., 2019a), preparing $CaCO_4$ (Chen et al., 2020a), calcium sulfate whiskers (CSWs) (Tan et al., 2017), agricultural fertilizer (Dias et al., 2010; Abril et al., 2009; Crusciol et al., 2016), soil amendments (Kassir et al., 2012; Hentati et al., 2015; Li et al., 2015), and so forth (Canovas et al., 2019). Especially, many works revealed the enormous opportunities related to the use of PG in different application fields. Different production methods and equipment of PG could lead to subtle changes in the composition and properties of PG (Meskini et al., 2021; Min et al., 2008a; Ajam et al., 2009; Huang and Lin, 2010; Gaiducis et al., 2011; Shen et al., 2014; Bouchhima et al., 2013; ZCKA et al., 2014; Rashad, 2015; Zhao et al., 2015; Hua et al., 2016; YouqiangHuang et al., 2016; Li et al., 2017). As a result, large-scale utilization rate of PG is low, and a series of environment problems are caused by long-term accumulation, which seriously restricts the development of phosphorus chemical enterprises, so that the utilization of PG is a quite important problem (Ennaciri and Bettach, 2018; Wdrychowicz et al., 2019).

To sum up, the treatment of PG is always the key problem of solid waste treatment. However, new key technologies and field that could be used to consume PG on a large scale are still required, aiming to further improve the rate of resource utilization. This paper reviewed the research status of different high-added value utilization and harmless treatment of PG, and the advantages and disadvantages of PG treatment technology were analyzed and compared. Finally, the effective development prospect of PG treatment was proposed to solve the problem of massive stacking and production of PG.

2. The basic characteristics and properties of PG

With the development of science and technology, more and more characterization methods of materials are applied to PG to detect the composition, physical and chemical properties, and so on.

Many research literatures illustrate the main components and basic properties of PG, as shown in Fig. 1. The basic characterization shows that the main components of PG are $CaSO_4{\cdot}2H_2O$ and SiO_2, accounting for more than 90%, and some toxic and harmful elements, including phosphorus(P), fluorine(F), arsenic (As), chromium (Cr), cadmium (Cd), metal trace elements (MTE), and rare earth elements (REE). The particle size distribution of most PG produced is mainly in the range of 10-1000μm, and it belongs to fine powder. The moisture content of PG is 5%–28%, which results in the agglomeration of fine particles and reduces the generation of dust. The micro morphology of PG is mainly flake irregular, rhombic and other crystal morphology, and a lot of impurities are adsorbed on its surface. The basic research on PG provides theoretical support for its subsequent resource utilization research.



composition	SO_3	CaO	SiO_2	P_2O_5	Fe_2O_3	Al_2O_3	MgO	F
Content(wt.%)	40.86	29.82	9.43	2.04	0.13	0.24	0.055	0.64
composition	Na_2O	K_2O	MnO	Free H_2O	Crystal H_2O	Acid insoluble	/	/
Content(wt.%)	0.043	0.086	0.002	5.38	4.27	8.42	/	/

Fig. 1. SEM-EDS, XRD, XRF and particle size distribution characterizations of PG (Mi et al., 2019; Gong et al., 2020a; Wu et al., 2020).

3. Pretreatment technologies and application of PG

3.1. Pretreatment technologies of PG

At present, the pretreatment methods of PG mainly include ultrasonic pretreatment, microwave treatment (Kavitha et al., 2013), thermal treatment (Ségalen et al., 2015), chemical conditioning and water leaching (ColomboPalumboJZet al., 2014).

3.1.1. Wet pretreatment methods

For a long time, the majority of science and technology workers have conducted a lot of researches on the pretreatment of PG (Singh, 2002), and mainly focused on the following aspect, namely water washing method that can remove a large number of impurities such as P, F and heavy metals adsorbed on $CaSO_4·2H_2O$. However, PG is slightly soluble in water, the washing liquid belongs to secondary wastewater, which is easy to cause secondary pollution and increase the treatment cost of secondary pollutants. The traditional flotation method can remove organic matter suspended in PG slurry naturally adding flotation agents, but fails to thoroughly remove P and F. Besides, the method is slow and inefficient. Lime was added to the PG, the soluble P and F in the lime and PG reacted to form insoluble P and F, and the precipitation method reduced the influence of soluble P and F on the building materials. However, after a long time, insoluble P and F will slow dissolve out, which cannot fundamentally reduce the influence of P and F on PG (Reijnders, 2007; Wang et al., 2020b; Gijbels et al., 2020; Mashifana, 2019). Obviously, there are still many problems to be solved in the wet pretreatment of PG. In order to reduce the treatment cost of PG and improve its resource utilization efficiency, new pretreatment technologies still need to be developed.

3.1.2. Thermal pretreatment methods

The thermal treatment method is the main technology for PG dehydration and stimulating gelling activity (Cao et al., 2022a; Ma et al., 2020). Thermal treatment is the best method in reducing the content of organic matter and removing volatile impurities (Liu et al., 2020; Contreras et al., 2015; Yang et al., 2011; Azouazi et al., 2001). Azouazi et al. (2001)found a reduction in the average radionuclide leaching rate from 26.4% to 6%, with thermal treatment at 800 °C. Ren et al. (Contreras et al., 2015) tested different thermal treatment conditions and reported a significant decrease in the content of soluble P and F. Cesniene (Cao et al., 2022b) evaluated PG thermal treatment at 400 °C, 600 °C and 800 °C for 1 h each to obtain anhydrous calcium sulfate. Singh and Garg (2005a) thermal treated PG at 1000 °C for a period of 4 h–5 h to make anhydrous calcium sulfate to be used in polymerized flooring composition. Min et al. (2008b) used a temperature of 135 °C for 3 h in the thermal treatment process. Değirmenci (Degirmenci, 2008) prepared hemihydrate gypsum at 150 °C for 2 h. Canut et al. (Cavalcanti Canut et al., 2008) prepared anhydrous gypsum at 160 °C for 1 h. Garg et al. (2016) prepared anhydrous calcium sulfate by calcining PG at temperatures of 150°C–160 °C for 4 h. Bumanis et al. (2018) thermal treated PG at the temperatures of 100 °C, 120 °C, 140 °C, 160 °C, and 180 °C for 4 h to obtain a binder material. Liu et al. (2020) tested different conditions to PG thermal treatment: 0.5 h, 1 h, 1.5 h, and 2 h of residence time on kiln and temperatures of 150 °C, 350 °C, 600 °C, and 800 °C. The authors stated that the increase in the thermal treatment temperature and time resulted in the conversion of soluble phosphorus impurities ($2CaHPO_4.2H_2O$) into calcium pyrophosphate ($Ca_2P_2O_7$), with an insoluble and harmless substance. Rodrigo H. Geraldo et al. (2020) evaluated the temperature of 150 °C with residence times on the kiln of 1 h and 2 h, and they found that adequate parameters can obtain considerable $HH\text{-}CaSO_4$ content with lower energy consumption. The main bottleneck of thermal treatment is that it needs high temperature, thus leading to high energy consumption. Moreover, under high temperature, many volatile components volatilize and need to be treated with tail gas, which increases the process complexity and treatment cost.

3.2. The application of PG in building materials

With the rapid development of the construction industry and the continuous improvement of people's living standards, the demand for building materials is also increasing. Most building raw materials come from natural sources and destroy the ecological environment. Due to the huge storage and output of PG and the special properties of PG, it can be used as a substitute for building raw materials. The by-product PG is often used as a building material (Rashad, 2017). The following mainly introduces several main utilization directions of phosphogypsum in the field of building materials.

3.2.1. Preparation of α-HH from PG

α-Hemihydrate ($\alpha\text{-}CaSO_4·0.5H_2O$, α-HH) is also called high-strength gypsum, and it is a high value-added cementitious material with superior workability, remarkable mechanical strength and excellent environmental performance (Jiang et al., 2016). Furthermore, it has been widely applied in the chemical and construction industry (Ma et al., 2018). Preparation of α-HH using PG as raw material is an important utilization approach. A traditional method to synthesize α-HH is in an autoclave under high temperature and pressure. However, the autoclave method is strictly limited by its special equipment and safety issues (Li et al., 2013). Recently, the solution method was proposed to prepare α-HH under normal condition, and NaCl, KCl, $CaCl_2$, $MgCl_2$, Na_2SO_4 and their mixed salts were used as a crystallization agent (Guan et al., 2012; Guan et al., 2009; Shen BHG and Fu, 2009). Van Driessche et al. (Van Driessche et al., 2012) found the recrystallization of PG at under-saturated conditions (Stawski et al., 2016), and the experimental evidence has revealed the dissolution and re-nucleation mechanism of PG (Ossorio et al., 2017). Saha et al. (2012) also demonstrated that the transformation of α-HH to PG was a multistep particle formation model, where an amorphous phase forms first, followed by the transformation into a crystalline product, and the addition of citric acid significantly delays the reorganization to PG crystals. The morphology of α-HH is one of the most important factors affecting mechanical properties. Generally, α-HH particles with a low aspect ratio possess better workability and mechanical strength than that of needle-like or higher aspect ratio crystals (Duan et al., 2017). Therefore, the preparation of α-HH with a special morphology of crystals is very important in the industry, and the morphology of α-HH can be effectively regulated by crystal modifier. Currently, the crystal modifiers mainly include inorganic salt, organic acid, surfactant, alcohols and macromolecule. Extensive studies have proved that the organic acid can suppress the crystal growth of α-HH along the c-axis. For example, the short hexagonal prism α-HH prepared from PG in glycerol-water solutions could be obtained by adding succinic acid (Guan et al., 2017a, 2017b; Li et al., 2018, 2019; Wang et al., 2017a; Tan et al., 2020; Yang et al., 2016a; Chen et al., 2018; Lu et al., 2019b), (Ma et al., 2018). With the continuous development of the research on the preparation of α-HH from PG, many methods and processes have emerged. As displayed in Table 1, a variety of preparation methods and effects of α-HH are proposed.

Although the preparation of α-HH from PG is the best way of high value-added resource utilization, its cost and energy consumption are relatively high. Combined with the huge accumulation and output of PG, it is unable to achieve a large amount of consumption.

3.2.2. The application of PG in construction and building field

PG is widely used in the field of building materials. The following aspects are discussed: ①Application of PG in retarder (Akın Altun and Sert, 2004; Huang et al., 2020; Singh, 2003; Garg et al., 2009; Kuryatnyk et al., 2008). ② Waterproof material (Wang et al., 2020c). ③Burn-free bricks and gypsum blocks (Zhou et al., 2012, 2016a, 2020; Yang et al., 2009; Zielinski, 2015), the mechanism of two-step hydration process (Zhou et al., 2014a; Zhou et al., 2016b; Zhou et al., 2014b; Lz et al., 1016). ④Road material (Li et al., 2020; Silva et al., 2019; Tsioka and Voudrias, 2020; Moussa et al., 1984; Saleh and Rahman, 2016; Singh and Garg, 2005b; Verbeek and Plessis, 2005; Lopez et al., 2011), (Rashad, 2015). ⑤Self-leveling mortar (Wang and Jia, 2019; Yang et al., 2016b). ⑥Foam concrete (Brencis et al., 2011; del Río-Merino et al., 2022). ⑦Non-autoclaved aerated concrete (Colak, 2000; Rubio-Avalos et al., 2005; Bazelová et al., 2010; Brencis et al., 2011; del Río-Merino et al., 2022; Huang et al., 2013; Umponpanarat Wansomet al., 2015; TianYan et al., 2016; Lin et al., 2013). ⑧Haydraulic binder (Diouri et al., 2022), (Kuryatnyk et al., 2008). It is a kind of functional material, while its dosage is small. Facing a large amount of PG, its contribution rate seems to be relatively weak.

No matter what kind of materials PG is used for, its mechanism of action is similar, as illustrated in Fig. 2.

The utilization mechanism of PG in the field of building materials mainly includes: (1) dissolution crystallization. PG has a slight solubility in water, especially that salt solution can enhance the solubility. The dissolution process destroys the lattice structure of PG, releases impurities, and then recrystallizes to form pure calcium sulfate-based materials. (2) According to the principle of dehydration rehydration, the main component of PG is calcium sulfate dihydrate that has stable properties and no gelling activity. However, it can remove 1.5H_2O to form β, α-HH, with excellent performance in all aspects. As an important building material, it can remove 2H_2O to form anhydrous calcium sulfate and remove most harmful impurities of dehydration at the same time. Rehydration can form high performance building materials.

The consumption of PG accounts for a large proportion in the field of building materials, but with the enhancement of public awareness of environmental protection, PG building materials are resisted. Compared with natural materials, most people are not willing to accept solid waste based building materials, and the market of PG based building materials becomes a constraint. Only by changing the public's awareness of solid waste and the government's strong support can the utilization of PG in the field of building materials have a qualitative breakthrough.

3.3. The application of PG in preparation of chemical raw materials

The main components of PG are Ca, S and O source. More than 80% of the content can be used as special raw materials for chemical reaction. In many studies, the application of PG as Ca and S source as raw material is introduced in detail.

3.3.1. Production of H_2SO_4 from PG and co-production of cement

The technology of producing H_2SO_4 and cement with PG as main raw material has matured research and application. The apparent solid-solid reaction between CaS and PG has been widely applied in the production of sulfuric acid and cement (Wang et al., 2020d). López FA et al. studied the microencapsulation of PG into a sulfur polymer matrix to prepare a new type of PG-sulfur polymer cements (PG-SPC) to be utilized in the manufacture of building materials (López et al., 2011). Sichuan Wu et al. prepared iron-rich calcium sulfoatablluminate cement, with gypsum as the sole calcium oxide source and its incorporation into mineral phases (Wu et al., 2021). Although the research of this technology has been relatively mature, it still has various disadvantages, such as high preparation cost, high energy consumption and gaseous pollution, so most of its application has been discontinued, and relatively cheap and safe treatment technology needs to be developed.

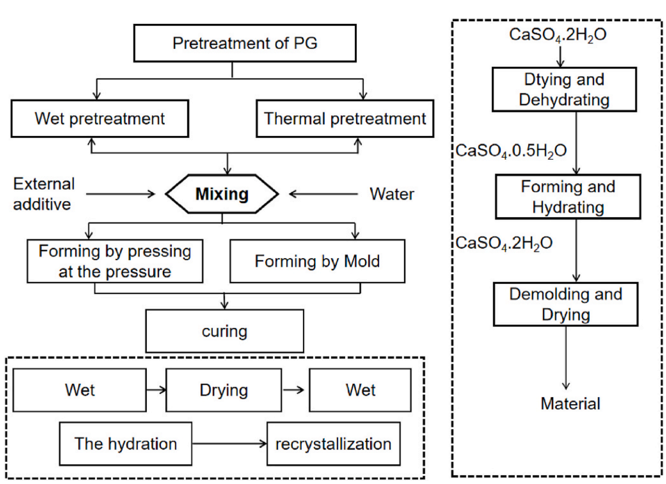


Fig. 2. Preparation process and mechanism of building materials.

3.3.2. Preparation of calcium sulfate whiskers from PG

The main component of PG is crystalline calcium sulfate ($CaSO_4{\cdot}nH_2O$, n = 0, 0.5, 2) (Kovler, 2012). Therefore, the preparation of ($CaSO_4$ whiskers) CSWs from PG is a feasible way for high value-added utilization of gypsum solid waste and reduction of resource waste (Zhang et al., 2012a), and it is also one of the most valuable research directions for the resource utilization of PG. CSWs has a lot of advantages, including high temperature resistance, chemical corrosion resistance, good toughness, high strength, easy surface treatment, and strong affinity with polymers such as rubber and plastics (Perez-Lopez et al., 2016). If the PG produced every year in the world can be prepared into the CSWs for resource utilization, it can not only solve the problem of the fate of a large number of PG, but also produce good economic effect.

So far, there are many methods for crystal preparation, including solution growth, melt growth and vapor growth, as shown in Table 2.

In addition to the above synthesis methods, biochemical activation method can enhance the nucleation ability in the crystallization process. The common hydrothermal synthesis methods are exhibited in Table 3.

Although CSWs has high economic value, its preparation cost is high, time is long, and output is low, so it is necessary to investigate a new process of PG whisker preparation with low cost and high yield.

3.3.3. The application of sulfur and calcium source in PG

PG contains a lot of S elements that can be used to prepare sulfur-containing chemical raw materials and pyrolysis, and the utilization of PG will decompose gradually from water to calcium sulfate when it is heated in inert atmosphere. The transformation to CaS was completed at 1080 °C (Msila et al., 2015), and reductive decomposition of PG had high S concentration coal to SO_2 in an inert atmosphere (Zheng et al., 2011). Liping Ma (Zheng et al., 2018) researched on thermal decomposing properties of PG with Fe addition under multi-atmosphere control, proposed a series of technical process for the decomposition of PG with Fe addition, such as K_2SO_4. The pure K_2SO_4 is colorless crystal, and the appearance of agricultural K_2SO_4 is light yellow. K_2SO_4 is a prominent water-soluble potassium fertilizer. Gan Z et al. proposed an efficient methodology via the co-reaction of K-feldspar and PG for the extraction of soluble potassium salts. The reaction parameters were systematically investigated with the highest S recovery ratio of about 60% (Gan et al., 2016; Ennaciri et al., 2019; Abu-Eishah et al., 2000). In addition, ammonium sulfate preparation from PG waste (Kandil et al., 2016) and chemical looping gasification of PG, as an oxygen carrier (Jing et al., 2019), were the utilization way of sulfur source in PG. The utilization of S source in PG has broad application prospects. At present, there are many researches on the preparation of K_2SO_4, while the mature production process has not yet been formed, and it is still in the laboratory research stage. Therefore, the research on the utilization of sulfur source needs to be further promoted.

The research on the utilization of Ca source in PG involves a wide range of aspects, including the resource utilization of PG, the formation of Ca based cementitious materials in building materials, pure Ca chemicals in chemical raw materials, etc. (Hammas et al., 2013)

3.4. The application of PG in agriculture

PG contains a lot of S, Ca, P and other metal components, many of which are essential for plant growth. However, because of its strong acidity and corrosiveness, it cannot be directly used in soil, but after harmless treatment, it can be used as saline alkali soil, alkaline soil and non-food crop soil. PG, as sulfate source for sulfate-reducing bacteria (de Oliveira Melgaco et al., 2020), had positive effects on the bacterial richness and diversity, and negative effects on the fungal richness and diversity. PG changed the composition and succession of the microbial community by influencing different physiochemical properties during various composting stages where the pH was the main explanatory factor (Lei et al., 2021).

PG belongs to acidic waste and contains trace plant growth elements. Guerrero JL et al. used PG to improve salt marshes soil (Guerrero et al., 2019). The salt-marsh sediment produces a "barrier effect" for the 238U series natural radionuclides coming from the PG stacks decreasing rapidly their activity concentration with depth (Papaslioti et al., 2020) effect of PG and poultry manure on aggregate-associated alkaline characteristics in bauxite residue (Xue et al., 2020), and the main reaction equations are shown in chemical formulas (2) - (9).

$$Ca_3Al_2(SiO_4)(OH)_8+4H_2O\rightarrow 3Ca^{2+} + 2Al(OH)_3+6OH^- + H_2SiO_4 \tag{2}$$

$$CaCO_3+H_2O\rightarrow Ca^{2+} + HCO^{3-} + OH^- \tag{3}$$

$$Na_8Al_6Si_6O_{24}(CO_3)(H_2O)+22H_2O\rightarrow 8Na^+ + 6Al(OH)_3 + 6H_4SiO_4 \tag{4}$$

$$CaCO_3+H_2O \rightarrow Ca(OH)_2 + H^+ +HSO_4^- \tag{5}$$

$$CaSO_4 \rightarrow Ca^{2+} +SO_4^{2-} \tag{6}$$

$$3Ca^{2+} +4OH^- + 2Al(OH)_4^- \rightarrow Ca_3Al_2(OH)_{12} \tag{7}$$

$$Ca_3Al_2(OH)_{12}\rightarrow 3Ca(OH)_2 + 2Al(OH)_3 \tag{8}$$

$$Ca(OH)_2 +CO_2 \rightarrow CaCO_3 + H_2O \tag{9}$$

Based on natural soil covers with organic amendment, these findings propose the possibility of using naturally-occurring SRB in the PG for bioremediation strategies (Castillo et al., 2012). Due to the variety of harmful components and potential harm forms in PG, the utilization of PG in agriculture is limited.

3.5. Preparation of environmental functional materials

PG has significant physical properties, so it has broad prospects in the preparation of environmental functional materials. Zhao L used sodium dodecyl sulfonate and modified PG to prepare the exclusive adsorption material for copper, and the removal rate is as high as 99.23% (Zhao et al., 2020). As the raw material for the preparation of adsorption materials, PG has broad application prospects, but its preparation cost, secondary pollution, waterproof performance and other factors need to be considered (Kadirova et al., 2014). Some researchers used phosphogypsum and clay to store radioactive element uranium (VI), and its adsorption capacity is 0.09 mol/kg, which indicated that it has good effects and has a certain utilization prospect in radioactive material treatment (Syczewski et al., 2020). PG was utilized to prepare hydroxyapatite nanoparticles (nHAp) via microwave irradiation technology, and the adsorption behaviour of F onto the nHAp derived from PG was investigated to evaluate the potential application of this material for the treatment of the wastewater polluted with F. The results indicated that the nHAp derived from PG can be used as an efficient adsorbent for the removal of F from aqueous solution (Zhang et al., 2012b). The utilization of PG for the preparation of porous sound absorbs material (Ma et al., 2020). Gong prepared Nano-$CaSO_4$ from PG, and this method is economical and applicable for the comprehensive utilization of PG (Gong et al., 2020b). Although the application of PG in the preparation of environmental functional materials has a high utilization prospect, the research on the occurrence mechanism, migration and transformation law of many toxic and harmful substances in PG is still not clear, and there exists the risk of secondary pollution to the environment.

3.6. The application of filling materials

The technical feasibility and economic rationality of various PG comprehensive utilization schemes are based on the effective control of PG impurity composition and moisture content. Through the analysis of the resource application of PG in engineering and agriculture, it is difficult for the existing resource technology to achieve the purpose of large-scale efficient resource utilization of PG, and it is hard to have good economic value. The existing chemical enterprises have not achieved the commercialized output and harmless discharge of PG waste residue. Since PG contains a large number of impurities with complex components and there is still a risk of leaching after generating resource products, pretreatment is required before the comprehensive utilization of PG. According to different components and utilization directions of PG, different treatment and disposal methods were adopted, so as to realize the modification of PG composition and structure and improve the resource application performance and economic value of PG (Rong et al., 2020).

The research and development applications of PG comprehensive utilization products should be inclined towards the direction of large dosage, high added value and many ways. The application in cement retarder, building gypsum cementitious materials and new building materials will be the main direction. Paying attention to the impurity removal and modification of PG can lay a solid foundation for PG to replace natural gypsum, effectively expand the application field and improve the content of PG ingredients. Therefore, the project mainly modifies PG in advance and solidifies its harmful impurities, so that it can be used as ecological restoration materials. Combined with the actual situation of the project, it can be used as pit filling. Most importantly, it can not only solve the problem of massive stockpiling of PG, but also meet the purpose of secondary utilization of PG in the future (Zhang et al., 2021).

3.7. The application of CO_2 capture

The emission of greenhouse gas carbon dioxide rises sharply with the accelerated development of economy. As a major environmental problem faced by mankind, the greenhouse effect caused by excessive carbon dioxide emission seriously threatens the earth's environment on which mankind depends. In view of the technical and economic difficulties in the development and application of new energy, the existing energy structure is difficult to change significantly in the short term, and carbon dioxide emissions will rise sharply. Therefore, the solidification of carbon dioxide and related resource regeneration have attracted close attention, with broad development prospect. The main methods of carbon reduction are solidification transformation and separation and purification. Solidification and transformation technology includes carbonate product generation and storage technology. At present, Tianjin Institute of Industrial Biotechnology uses carbon dioxide to synthesize starch. Separation and purification technologies include solvent absorption method, adsorption method, membrane separation method, low-temperature distillation method, and newly developed carbon dioxide recombination method, electrochemical method, biological carbon dioxide recovery technology and hydrate method. Although the above technologies have made great progress, they generally have the disadvantages of high energy consumption and high investment. Meanwhile, there are some problems in the reaction process to be further solved. The threats of global warming and climate change provide a new driving force for carbon dioxide separation technology (Zhang et al., 2020).

At present, the research on the carbon neutralization of PG is shown in Fig. 3, mainly including A-F carbon solidification mechanism, in which A mechanism mainly treats waste with waste, mixes other solid wastes with phosphogypsum, dissolves and carbonizes, so as to achieve the synergistic solidification effect of CO_2; B mechanism is mainly the synergistic carbon solidification of minerals, such as potassium feldspar; C mechanism is mainly the mechanism of plant synergistic carbon solidification, and mainly uses the photosynthesis of plants, the absorption of roots and the secretion of hormones; D mechanism is mainly the excitation of basic chemical reagents; E is mainly solidified and stabilized by the generated CAS after thermal decomposition. Compared with D mechanism, F mechanism increases the pretreatment stage. The above six carbon curing mechanisms are the most common methods at present. Due to the huge output of industrial solid waste, carbon neutralization with industrial solid waste has become a research hotspot (Kumar et al., 2020). As the main component of PG is crystalline calcium sulfate ($CaSO_4.2H_2O$), the treatment of industrial carbon dioxide, containing tail gas with waste gypsum, is an important way of high value-added utilization of waste gypsum solid waste, thus reducing resource waste and carbon emission reduction. On the other hand, it is the most valuable research direction of resource utilization of waste gypsum solid waste. If China uses gypsum waste to treat industrial carbon dioxide containing tail gas for resource utilization, it can not only solve the fate of a large number of waste gypsum solid waste, but also achieve the goal of carbon emission reduction and produce good economic value. What's more, it can improve the earth's environment with good social and economic benefits. Most importantly, it has deeply practiced the concept that green water and green mountains are golden mountains and silver mountains (Gan et al., 2016)' (Romero-Hermida et al., 2017).

3.8. The application of extracting rare earth elements

A growing interest has emerged towards resource development and processing of rare earth elements (REE) that have become indispensable elements in human daily life (Goodenough et al., 2016), (Virolainen et al., 2019). Depending on the type and source of the PG, the REE content ranges between 0.05% and 2% (Binnemans et al., 2015a; Walawalkar et al., 2016). Between 2020 and 2025, the global demand for REE will gradually increase, which could make the source of REE a key factor to be considered as the solid waste or the search for alternative sources (Wang et al., 2017b; Binnemans et al., 2015b). PG is an example of such environmentally harmful wastes containing REE (70%–85%) (Binnemans et al., 2015b; Aly and Mohammed, 1999). Previous studies proved that leaching PG with acids creates favorable conditions for the migration of REE from the solid phase to the leach solution (Kurkinen et al., 2021), (Costis et al., 2021; Brueckner et al., 2020; Gasser et al., 2019). As one of the hot solid wastes all over the world, PG contains a variety of rare and precious heavy metals. With the rapid development of the industry, the market demand for rare and precious heavy metals is gradually increasing. Extracting rare and precious heavy metals from solid wastes has become an important way for high value-added treatment of solid wastes as one of the main sources of rare and precious heavy metals. While reducing the development of natural

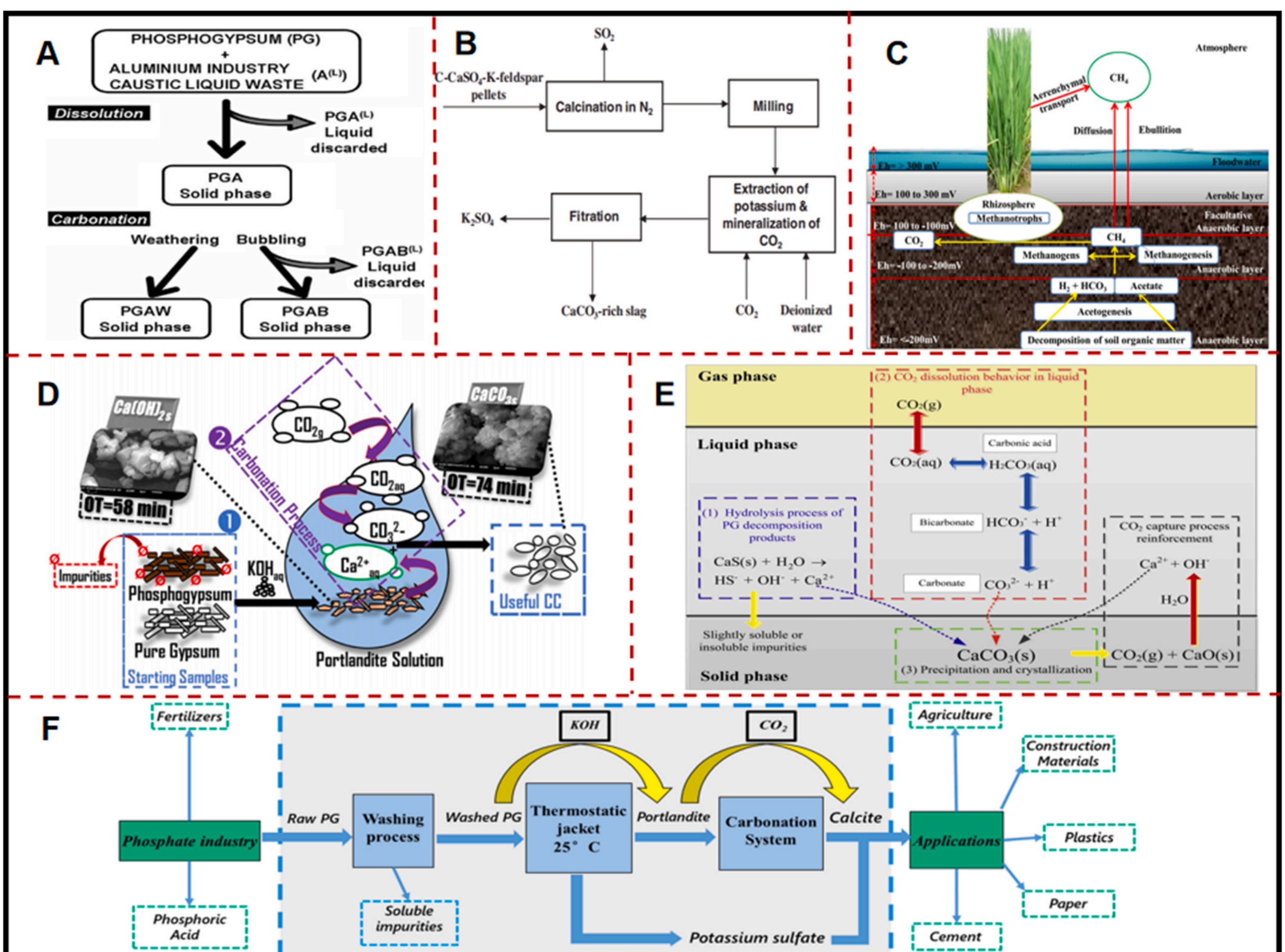


Fig. 3. Process and mechanism of carbon dioxide solidification (Zhang et al., 2020).

resources, environmental protection is realized. The PG leaching solution is long-term precipitation and surface runoff, which carries out long-term flushing of PG and transfers a large number of rare and precious heavy metals to the leaching solution. The leaching solution is continuously transpiration and volatilization in the atmosphere, thus realizing the high concentration enrichment of REE in the leaching solution (Andreevet al., 2014; Kurkinen et al., 2021; Plyatsuk et al., 2020; Gupta and Krishnamurthy, 1992; John et al., 2018; Qi and Qi, 2018; Zheng et al., 2019), and the main reaction equations are shown in chemical formulas (10) and (11) under acidic conditions (Z et al., 2015).

$$2REEPO_4+3H_2SO_4 \rightarrow REE_2(SO_4)_3+2H_3PO_4 \quad (10)$$

$$Th_3(PO_4)_4+6H_2SO_4 \rightarrow 3Th(SO_4)_2+4H_3PO_4 \quad (11)$$

Carbonate and bicarbonate salts are preferred as precipitation agents. The precipitation takes place in ambient conditions. Depending on the time of the reaction, either hydrated REE carbonates (Equation (12)) or REE hydroxy carbonates are formed (Paul et al., 2018; Chi et al., 2003).

$$2REE^{3+} + 3SO_4^{2-} + 6NH_4^{-} + 3CO_3^{2-} \rightarrow REE_2\,(CO_3)_3 + 6NH_4^{+} + 3SO_4^{2-} \quad (12)$$

The decomposition of the phosphates from monazite with caustic soda can be summarized with the following Equations (13) and (14) (Zhang et al., 2016; Sadri et al., 2017).

$$REEPO_4 + 3NaOH \rightarrow REE(OH)_3+Na_3PO_4 \quad (13)$$

$$Th_3(PO_4)_4+12NaOH \rightarrow 3Th(OH)_4+4Na_3PO_4 \quad (14)$$

The common extraction technology of REE is displayed in Fig. 4. The main steps are as follows. After the PG was washed by water or solvent, the solid components of PG were dried at a certain temperature, and the components of washing solution can be recycled. This process was called pretreatment stage. Solid PG was digested by leaching agent. Segmented digestion can be set according to the demand. After valuable elements were leached, solid-liquid separation was carried out and the liquid part was collected. This process was called dissolution of REE. The liquid part used special adsorption materials to purify and separate REE, while the liquid part can be recycled. This part is called REE purification and separation. The whole process realized the extraction of REE. In addition, ion-exchange resins, leaching agents, and eluents were systematically investigated for the recovery of REE from PG by a resin in leach process. Based on the equilibrium batch experiments performed with and without an ion-exchange resin, it is not necessary to break the gypsum structure to recover REE (Voncken, 2016; Virolainen et al., 2019; Omodara et al., 2019; Riddle et al., 2021), (Costis et al., 2021).

Although there have been a lot of studies on the extraction of rare and precious metals from PG, the existing technical means cannot be used for low-cost extraction due to its high cost and low content of rare and precious metals, which leads to the failure of industrial application.

3.9. Other application fields

PG prepares biphasic calcium phosphate (BCP) (Mohamed et al., 2014), and hydrothermal synthesis of nanocrystalline hydroxyapatite is from PG waste (Bensalah et al., 2018). Syngas can be obtained through a properly designed lignite Chemical Looping Gasification (CLG) of PG oxygen carrier (Yang et al., 2017), and the main reaction equations are shown in chemical formulas (15) - (22).

(a) Lignite gasification

$$C_{5.2}H_{4.36}O_{4.26}N_{0.096}S_{0.034} \rightarrow 1.0492C + 2.0738H_2+4.0609CO+0.013186CH_4+0.031969H_2S+0.04798N_2+0.034H_2O(g)+\text{Other substances} \quad (15)$$

$$C + H_2O(g) \rightarrow CO + H_2 \quad (16)$$

$$CO + H_2O(g) \rightarrow CO_2+H_2 \quad (17)$$

$$CO2+H_2O+2CH_4 \rightarrow 3CO+5H_2 \quad (18)$$

(b) PG reduction

Main reaction:

$$CaSO_4+4C \rightarrow CaS+4CO \quad (19)$$

$$CaSO_4+4CO \rightarrow CaS+4CO_2 \quad (20)$$

$$CaSO_4+4H_2 \rightarrow CaS+4H_2O(g) \quad (21)$$

$$CaSO_4+4CH_4 \rightarrow CaS+4CO+8H_2 \quad (22)$$

Of course, a hot research in recent years, the resource research direction of PG does stay in the above aspects. Here, we will not discuss it one by one.

4. Main resource utilization mechanism of PG

In the process of resource utilization of PG, there are the following main mechanisms (As shown in Figs. 5 and 6.). First, multi physical strengthening of dissolution and decomposition in the crystallization mechanism. PG dissolves in water or solvent. Ultrasonic, temperature, electromagnetic, stirring speed and other physical conditions will accelerate the destruction of PG crystal lattice, accelerate its dissolution, and release rare and precious heavy metals, phosphorus, fluorine and

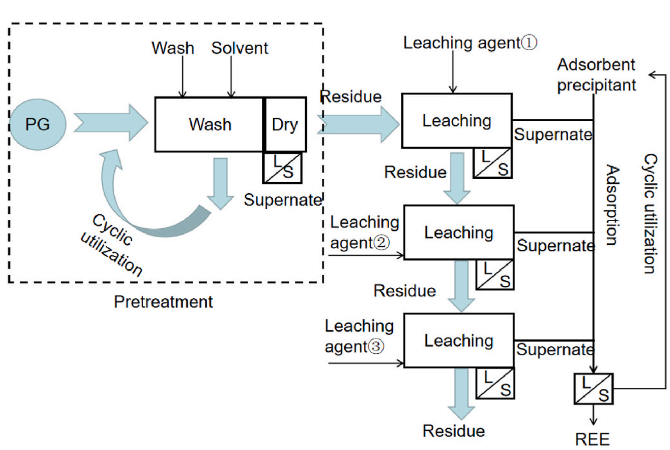


Fig. 4. Extraction technology of REE.

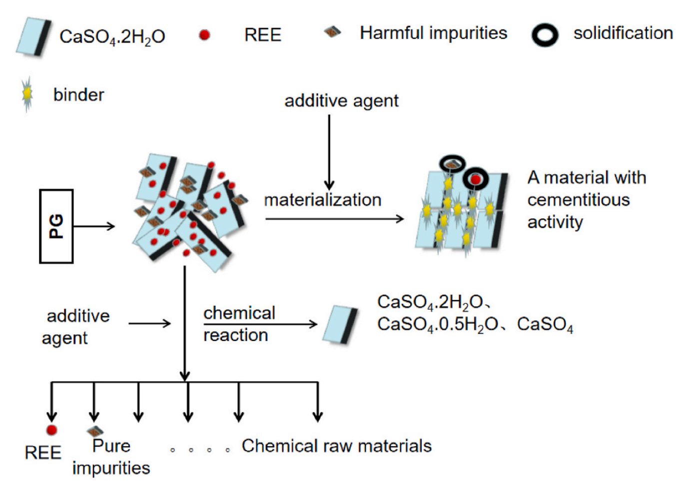


Fig. 5. Resource utilization mechanism of PG.

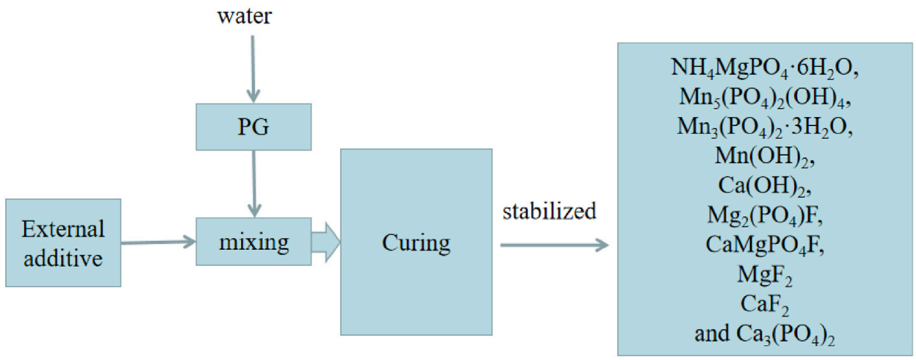


Fig. 6. The solidification mechanism of PG (Chen et al., 2020b).

other impurities. The released substances react with the added solvents to form new and different kinds of substances, including rare and precious heavy metals, chemical raw materials, calcium based and sulfur based pure chemicals. Combined with the research reports of peers at home and abroad, it is not difficult to infer that the crystallization mechanism mainly dissociates the elements (atoms, molecules and ions) that make up the crystal that strictly controls the growth conditions, and re combines them under the driving of chemical potential, so that multiple physical fields can provide different forms of energy and conditions. Therefore, there are multiple physical field coupling effects that can regulate the crystal growth and crystal transformation of PG and improve the controllability of gypsum products. When an electric field is applied during the dissolution and crystallization of PG, the electrons released by the electric field can make PG produce a large number of high-energy molecular excited states and secondary electrons. A series of physicochemical changes will occur in the excited states and secondary electrons with higher energy, such as chemical bond fracture, suprathreshold ionization, suprathreshold dissociation, bond softening, coulomb explosion, the reorientation of molecules in strong field, and the formation of new excited states. When the magnetic field is applied, they are directionally distributed under the action of the magnetic field due to the uniform paramagnetic and diamagnetic substances in PG. When ultrasonic field is applied, the energy provided by ultrasonic can change and break the original lattice shape of waste gypsum. When the temperature is applied, it can provide the energy required for gypsum crystal growth and crystal form. Secondly, as shown in Fig. 5, PG decomposes and reacts with external additives to form precipitates, complexes and gelling active substances, which improves the stability, plasticity and gelling activity of PG. At the same time, the toxic and harmful impurities in PG are solidified and stabilized in the form of coordination, capsule wrapping and adsorption, as shown in Fig. 6. The heavy metals, phosphorus and fluorine mainly form hydroxide and normal acid salt precipitation ($NH_4MgPO_4 \cdot 6H_2O$, $Mn_5(PO_4)_2(OH)_4$, $Mn_3(PO_4)_2 \cdot 3H_2O$, Mn $(OH)_2$, Ca $(OH)_2$, $Mg_2(PO_4)F$, $CaMgPO_4F$, MgF_2, CaF_2 and $Ca_3(PO_4)_2$), which reduces its release efficiency under normal environment and realizes the long-term solidification and stabilization effect.

5. Conclusion and future prospects

As one of the main industrial by-products. PG has become one of the hot spots in the research of bulk solid waste, mainly including multi-directional building materials, preparation of chemical raw materials and other aspects, because of its huge output and storage. However, due to the special properties of PG, it is still unable to consume a large amount of PG to reduce the environmental risk of PG. The following research directions of PG are proposed.

(1) Research on the sustainability of PG cycle can be improved by the combination of electrodialysis and bipolar membrane (Monat et al., 2020). The result is a cleaner and more circular process, as NaOH can be recycled for dissolving PG, while H_2SO_4 can be reintroduced into the industrial phosphoric acid production or used to leach valuable metals out of the PG. However, to assess the feasibility of BMED integration, its industrial application research needs further exploration.
(2) Prefabricated building materials. With the continuous improvement of people's living standards, many works of art and crafts can be seen everywhere in our daily life. PG is mainly composed of calcium sulfate, which has good plasticity and can be prepared into a variety of process exhibits. The rheological property control of PG is an important technical means for PG to be used in prefabricated building materials. It has broad research value and significance to improve the utilization rate and consumption of PG, especially in 3D printing of PG.
(3) New green functional materials. PG contains more than 80% calcium sulfate dihydrate that is a green material. The main harmful components in PG are phosphorus, fluorine and heavy metals. The harmful components are solidified and will not release and migrate with time. On this basis, green environmental protection materials with special functions are prepared.
(4) Ecological and environmental friendly filling materials and soil utilization. With the continuous development of mineral resources, there are many mines on the earth's surface, resulting in many geological disasters. Backfilling after harmless treatment of PG not only solves the problems of environmental risks and geological disasters, but also solves the problem of massive accumulation of PG. At the same time, facing the world soil barren, grain crops reduce production. In order to solve the problem of people's livelihood while solving the PG destination, phosphorus source is used in PG as fertilizer of food crops. However, the low cost harmless treatment of PG has been the limiting factor of large-scale treatment of PG.
(5) Chemical filler. The inexpensive chemical filler can obviously enhance the mechanical properties of rubbers, plastics, adhesives and papers, with great potential to become ideal reinforcing materials.

The large-scale processing technology of PG is not a problem that can be solved by a single technology, but a variety of technologies for collaborative disposal. On the other hand, the comprehensive and multi-level integrated processing technology has become the main research hotspot of PG consumption.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jenvman.2022.114957.

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