RESEARCH ARTICLE



Bibliometric analysis of phosphogypsum research from 1990 to 2020 based on literatures and patents

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Abstract

This research aims to explore the research progress of PG, including basic status, cooperation situation, research fields, and development trends, based on the Web of Science database through bibliometric analysis of publications (articles and patents) from 1990 to 2020. The results show that academic research on PG originated early, but the number of patents grew quickly. China is a global leader in terms of the number of publications and plays a significant role in international cooperation. The knowledge of PG has remained concentrated in the fields of natural radioactivity, cement paste backfilling, soil, crystal morphology, and synthetic gas. However, academic hotspots focus on the microstructure of chemical processes and various environmental impacts; patents and hot technologies are based on the production of refractory materials, ceramics, surface materials, cement mortar, and composite materials. The academic frontiers of PG will be centered on exploiting the methods of recovering rare earth elements from PG, the conditions of ion solidification/stabilization in PG, the impact of reaction conditions on product quality, and the reaction mechanism at the micro-level. The frontiers of patents need to focus on the improvement of manufacturing equipment, new wall materials, and chemically modified polymer materials. Envisaging the number of articles and patents to be published in the future, architectural research has a large room for improvement. This paper conducts an in-depth analysis of PG and provides information on the technological development prospects and opportunities, which is helpful for researchers engaged in PG management.

Keywords Phosphogypsum · Bibliometric · Knowledge map · CiteSpace · Technology life cycle · S-curve

Introduction

The phosphorus chemical industry refers to a chemical subindustry that uses phosphate ore and sulfur as raw materials to produce phosphorus. The products of the phosphorus chemical industry have been widely used in various industries, including national defense, cutting-edge science, and daily

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supplies (Zhang and Zhang 2018). Phosphogypsum (PG; mainly CaSO₄·2H₂O) is a by-product of the wet process of phosphoric acid production (Beretka et al. 1993). According to estimates, the production of 1 ton of phosphoric acid (calculated as P₂O₅) produces approximately 5 tons of PG, which can be discharged, stored, or reused (Chen et al. 2018). With the expansion of the phosphorus chemical industry, piling up on the ground is currently the main disposal of PG, which not only occupies a large amount of land, but also causes severe environmental pollution (Pérez-López et al. 2016; Mohammed et al. 2018; Moreira et al. 2018). Nowadays, approximately 20–30 billion tons of PG is produced annually worldwide, which poses tremendous pressure on environmental protection and creates a huge challenge to its recycling (Pérez-López et al. 2016).

Because PG has the characteristics of large output, concentrated distribution, and complex composition, the current disposal method involves both storage and utilization (Wang et al. 2020), as shown in Figure 1. After pretreatment, PG



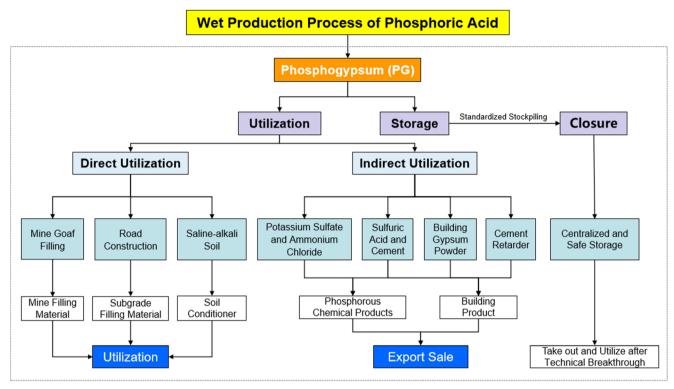


Figure 1 Main treatment and utilization methods of PG

can be directly used as a final product such as mine filling materials, roadbed materials, and soil conditioners (Gu et al. 2020). In addition, PG can also produce raw materials such as sulfuric acid co-production cement, construction gypsum powder, cement retarder, and other building materials indirectly (Chen et al. 2018). At present, many scholars researched the environmental impacts and treatment technologies of PG, such as the radiological and elemental hazards (Attallah et al. 2019; Qamouche et al. 2020), the behavior of heavy metals (Guerrero et al. 2021; Vasconez-Maza et al. 2021), optimum design of PG storage yard (Millan-Becerro et al. 2020), and pretreatment to remove impurities (Zhang and Zhang 2018). However, most studies depend on the preferences of experts in a certain field. To a certain extent, this individual and subjective preference leads to the lack of systematic and quantitative summary of this field, making it difficult to understand the overall structure of the intellectual landscape of PG from a macro-perspective.

Bibliometric analysis is an effective tool for quantitatively analyzing academic literature through mathematical statistics techniques. It has been widely used in various disciplines to explore the distribution of articles in a particular field of countries, institutions, and topics (Peng et al. 2018). More importantly, the combination of literature and patent results is conducive to identifying the relationship between fundamental and applied research and exploring the direction and content of technological development to better understand the direction of future theoretical and technological innovation (Zhang

et al. 2020a, b). However, to the best of our knowledge, only a few studies have researched the knowledge structure of PG using bibliometric analysis (Cánovas et al. 2018; Chernysh et al. 2021). These scholars discussed the management scheme of PG from different perspectives, which laid the foundation for our research.

This study employed bibliometric analysis to quantitatively and qualitatively discuss publications in the field of PG from 1990 to 2020 based on articles and patents. More specifically, we explored (1) the current status of the PG domain, such as the distribution of annual output, disciplines, journals, countries, and institutions; (2) the performance of countries, institutions, and individuals in the cooperation network; (3) the foundation, hot areas, and frontiers of PG study using the co-citation, co-occurrence, and burst value of articles and patents, respectively; and (4) the technology life cycle curve and future development trend of each subfield. This empirical research supplements the traditional narrative-based literature review quantitatively and provides comprehensive and scientific guidance for future studies.

Data acquisition and research methods

Data acquisition

The data in this study were downloaded from the Web of Science. Scientific literature was retrieved using the Science



Citation Index Expanded (SCIE) and Social Science Citation Index (SSCI), and the patent data were obtained from the Derwent Innovation Index (DII).

SCIE and SSCI are commonly used databases in bibliometric analyses (Liu and Liao 2017). They cover more scientific and authoritative publications than other databases and provide information, such as citations, keywords, and references for subsequent in-depth bibliometric. DII, the integration of the Derwent World Patents Index® (DWPI) and Derwent Patents Citation Index® (DPCI), is a web-based patent information database published by Thomson Derwent and Thomson ISI (Lv et al. 2011), where the adequacy, reliability, and sufficiency of the data can be fully guaranteed. As one of the major features of the DII database, the Derwent manual code (DMC) is the proprietary code of patented technology classifications used to identify patents (Wang et al. 2011).

We took "Title = (phosphogypsum)" as the retrieval strategy, and the time span was defined as "1990-2020" (retrieval date October 20, 2020). In order to improve the quality of retrieval results and avoid the interference of subjective reviews, the literature type was limited to "article." In total, 748 articles and 2617 patents met the selection criteria. Subsequently, through manual screening by experts in the field, 719 articles and 2580 patents were selected for further analysis.

Research methods

Bibliometric analysis

Bibliometrics is a quantitative model that analyzes the number of external features of documents (academic documents and patents) and references through mathematical statistics to reveal specific topics (Fahimnia et al. 2015; Zupic and Cater 2015). In this study, we used frequently used bibliometric indicators, such as the number of documents, number of citations, and h-index, to measure the distribution of PG research strength. Knowledge mapping is an important bibliometric process that can represent the status quo of a field and visualize the results (Sun and Zhai 2018). CiteSpace, visualization software coding in the Java programming language, was developed to draw a knowledge map of scientific literature and show the development trend in the technical field (Chen 2004, 2006). CiteSpace provides a variety of functions for drawing knowledge maps. Keyword co-occurrence maps can reflect hotspots in an area, and burst keywords (keywords frequently cited within a time period) represent cutting-edge topics (Liang et al. 2017).

Social network analysis

In recent years, the frequency and importance of scientific cooperation have increased. Scientific cooperation has the potential to solve complex scientific problems and is the key to understanding academic exchanges and knowledge dissemination (Sonnenwald 2007). Social network analysis (SNA) reflects social structural relationships through network nodes and links and focuses on the characteristics of group relationships rather than individuals (Kim et al. 2011). Ucinet is well-known and commonly used software for processing social network data (Vital and Martins 2009). After importing the cooperation data of countries, institutions, and authors obtained in bibliometric analysis into Ucinet, the accompanying software Netdraw was used to visualize the cooperation network. Subsequently, Ucinet calculated the degree centrality of each node to reflect the importance of each subject in the network (Dai et al. 2020).

Technology life cycle theory

The technology life cycle describes the entire process of a technology from the origin, development, applications, and entry and exit of the market (Liu and Wang 2010). The Scurve of the logistic model is commonly used to simulate the technology life cycle of articles and patents (Haupt et al. 2007), in which, logistic analysis, a tool in the Loglet Lab 4 software, was applied to decompose the growth and diffusion patterns into S-shaped logistic components, such as saturation, growth time, and midpoint, to evaluate the development trend of technical fields (Meyer et al. 1999). Saturation is the maximum utility value generated by a certain technology. Growth time is the time required for a technology to produce a utility value from 10 to 90% of the maximum utility value. The midpoint is the turning point for growth and maturity (Franses 1994).

Result and discussion

The basic status of the PG study

The annual trends of PG publications by countries

Figures 2 and 3 plot the annual trends of articles and patents of PG, respectively, from 1990 to 2020. In general, the development of PG-related studies is increasing. In terms of the total amount, the number of PG patents (2580) is far greater than that of articles (719). Although only a few patents were granted before 2006 (none before 1993), the number has increased significantly rapidly since then. Considering the output of each country, it is noteworthy that China publishes a large proportion of both of articles and patents, especially after 2010. Studies have shown that the annual output of PG in China has reached five hundred million tons (Zeng et al. 2021), thus encouraging domestic PG research.



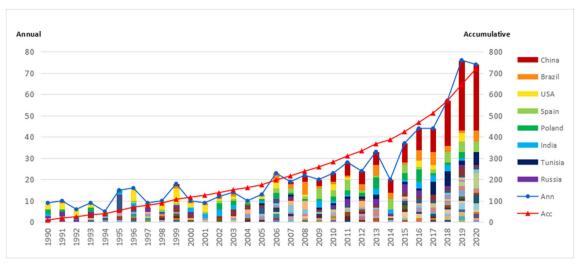


Figure 2 Annual and cumulative articles in PG resource utilization

Tables 1 and 2 show the details of the publications of the top ten countries in terms of PG research output, such as the total number of documents, total citations, citations per article, and records per capita. In addition, the h-index of each country was calculated to supplement the comprehensive judgment of literature quality. Although China has absolute advantages in terms of total article numbers, total citations, and h-index, it does not perform well in terms of average citations per publication and articles per capita. Other countries, such as Spain, India, Brazil, and the USA, have significant performance indicators that reflect the quality of papers. These countries have made many contributions to research on the properties and utilization of PG (Campos et al. 2017; Lambert et al. 2018; Luis Guerrero et al. 2020; Nayak et al. 2018; El Zrelli et al. 2019; Ruiz Canovas et al. 2018). Overall, academic research on PG is scattered around the world. In terms of patents (as shown in Table 2), Asian countries, such as China, Russia, Kazakhstan, and Belarus, are more prominent. Studies have indicated that the main phosphate mine producers and phosphate fertilizers are located in the USA, the former Soviet Union, China, Africa, and the Middle East (Tayibi et al. 2009). Therefore, the large production of PG in these countries has prompted them to accelerate the process of resource recovery through innovative technologies.

The subject categories of PG publications

Based on the subject classification of the ISI Journal Citation Report (JCR), Figures S1 and S2 depict the top ten subjects of PG articles and patents, respectively. Environmental science and ecology (254), engineering (248), and materials science (144) are the top three disciplines of PG articles, whereas chemistry (2422), materials science (1439), and polymer science (918) are the top three subjects of PG patents. The distribution of subject categories suggests the high priority of environmental, chemical, constructional, and agricultural issues in the PG field. The subject area of the articles pays more attention to environmental studies, whereas patents are more focused on chemical material issues.

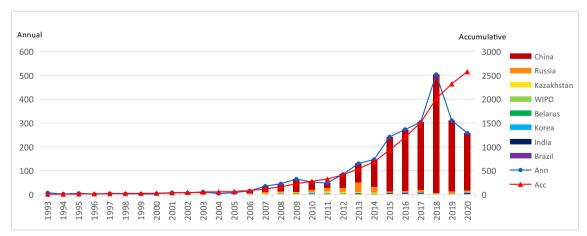


Figure 3 Annual and cumulative patents in PG resource utilization



Table 1 Articles in the field of PG resource utilization by country/region

Rank	Country/ region	TA	TC	TC without self-citations	SP%	ACPP	ACPP without self- citations	h- index	Population	Records/population (10 ⁻⁸)
1	China	151	2084	1535	21.00%	13.80	10.17	24	1,397,715,000	11
2	Brazil	56	974	878	7.79%	17.39	15.68	18	211,049,527	27
3	USA	55	748	701	7.65%	13.60	12.75	17	328,239,523	17
4	Spain	54	1089	908	7.51%	20.17	16.81	21	47,076,781	115
5	Poland	45	449	391	6.26%	9.98	8.69	12	37,970,874	119
6	India	37	731	702	5.01%	18.94	18.17	16	1,366,417,750	3
7	Tunisia	31	297	262	4.31%	9.58	8.45	9	11,694,719	265
8	Russia	29	109	102	4.03%	3.76	3.52	5	144,373,535	20
9	Egypt	27	459	433	3.76%	17.00	16.04	12	100,388,073	27
10	Canada	24	426	392	3.34%	17.75	16.33	12	37,589,262	64

Data source of population: World Bank. https://data.worldbank.org/

TA total articles, TC times cited in Web of Science Core Collection, SP% share of publications (719), ACPP average citations per publication

The distribution of published journals of PG articles

The 719 articles were published in 260 different journals. Table S1 shows several parameters, such as the number of articles, journal impact factors, h-index, country of origin, and number of citations, of the top 10 journals in terms of published PG articles. The journal with the highest number of publications is *Construction and Building Materials* (40 articles), accounting for 5.56% of the total publications, which is 1.38 times that of the second-ranked *Journal of Hazardous Materials* (29 articles) and 1.66 times that of the third-ranked *Journal of Environmental Radioactivity* (24 articles). It can be seen that these are the main carriers of literature in the PG field. In terms of the indicators that measure the quality of journals (impact factor, JCR Partition, h-index, citations), half of the journals are located in the first zone of JCR, indicating that these articles are representative.

The distribution of institutes of PG publications

The top 20 institutions that contributed the most to the total output of PG publications are presented in Tables S2 and S3. Among the top 20 institutions in terms of articles, Universidad de Huelva ranked first, with 41 publications accounting for 5.7% of the total, followed by Kunming University of Science and Technology with 32 articles and the University of Sevilla (28). Although the Council of Scientific and Industrial Research (India), Instituto de Pesquisas Energéticas e Nucleares (Brazil), and University of Alberta (Canada) do not rank high in the gross output, they perform significantly well in terms of indicators, such as ACPP without self-citations and h-index.

Table S3 shows the top 20 assignees, types, and countries with patents of PG. Guizhou Kailin Group Co., Ltd. had the greatest number of patents (208), accounting for 8.06%. The other institutions with a high number of patents are Guizhou

Table 2 Patents in the field of PG resource utilization by country/ region

Rank	Country/region	TP	SP%	Population	Records/population (10 ⁻⁸)
1	China	2241	86.86%	1,397,715,000	160.33
2	Russia	175	6.78%	144,373,535	121.21
3	Kazakhstan	32	1.24%	18,513,930	172.84
4	WIPO	31	1.20%		
5	Belarus	18	0.70%	9,466,856	190.14
6	Korea	15	0.58%	51,709,098	29.01
7	India	13	0.50%	1,366,417,750	0.95
8	Brazil	11	0.43%	211,049,527	5.21
9	Poland	9	0.35%	37,970,874	23.70
10	Uzbekistan	9	0.35%	33,580,650	26.80

TP total patents, SP% share of patents (2580)



University (143) and Kunming University of Science and Technology (74). As for the type of institutions, enterprises and universities are the main forces in the research and development of PG patents. Among the top 20 assignees, seven are in Guizhou Province, three in Beijing, and two in Hubei and Henan, with significant regional differences due to the origin of PG being mainly produced in southwest China. Unlike the distribution of articles, almost all of these institutions are in China. This is because China has been increasing the investment in solid waste management to make significant progress in PG reutilization in recent years (Tayibi et al. 2009).

The cooperation situation of PG study

Social network analysis of academic research groups

To clearly demonstrate the cooperative relationship in academic research on PG, we selected countries, institutions, and authors with more than five articles successively and then used the Ucinet software to conduct co-occurrence analysis on social networks.

In Figure S3, each node represents a country, and the dot size represents the degree centrality, which reflects the influence and importance of the country in the network. The thickness of the line between the nodes indicates the number of cooperation between countries. Table S4 lists the top ten countries by degree centrality and their corresponding number rankings. The degree values of France, the USA, Tunisia, China, and Spain are all more than 20, indicating that these countries have important positions in the academic research of PG. It is obvious from the thickness of the connection lines in Figure S3 that France and Tunisia and the USA and China are two pairs of countries that have a large degree of cooperation. France and Tunisia have cooperated in the fields of economy, trade, security, science, and technology. Moreover, coastal industries in the Gulf of Gabès in southeastern Tunisia caused hundreds of millions of tons of untreated PG to be discharged into the open sea, causing serious environmental problems (El Zrelli et al. 2018; Sinfort et al. 2019). The USA and China are major world powers and have collaborated in many scientific research areas. It is worth noting that Saudi Arabia and Uzbekistan are among the top ten countries in Table S4, even if they rank low in the total number of published articles. This phenomenon shows that countries with a small volume of publications may also play a role in international cooperation in specific areas.

Consistent with the method described above, we analyzed the cooperation network of institutions with more than five articles. As shown in Figure S4 and Table S5, Universidad de Huelva is the node with the largest degree of centrality and has a strong cooperative relationship with the University of Sevilla, University of Cadiz, and Consejo Superior de Investigaciones Cientificas (CSIC). These institutions are

located in Spain, with a large number of articles and are the main research institutions for PG academic papers. Another obvious cooperating group comes from China, which includes Guizhou University, Chongqing University, Chinese Academy of Sciences, and other institutions. In general, cooperating organizations belong to the same country because neighboring institutions have more opportunities to communicate owing to the geographical location and the local economy.

Figure S5 and Table S6 show the author's cooperation network and the degree centrality of the top ten authors, respectively. There are two main author cooperative groups and several sub-cooperative groups as shown in Figure S5. Similar to the content in Table S5, the main author cooperative groups were from Spain and China. Ma Liping from China is ranked first, with a degree centrality of 71, accounting for 7.87% of the total degrees, and is the author with the highest academic influence of PG. The authors ranked second and third are Yang Jing from China (with a degree centrality of 44, accounting for 4.88%) and Perez-Lopez, Rafael, from Spain (with a degree centrality of 33, accounting for 3.66%), respectively. These top-ranked authors led two major collaboration networks, the Chinese and Spanish groups. In addition, there are some scattered uncomplicated cooperation networks in the map, which shows that the academic research on PG is extensive at the individual level and the cooperation between individuals is close in a narrow range.

Social network analysis of applied research group

To further explore the cooperation of PG application technologies, we also used the Ucinet software to plot the cooperation of the assignees and the inventors of the patents in Figures S6 and S7. There are only a small number of institutions in the assignees' cooperation network as shown in Figure S6. Guizhou Guifu Ecological Fertilizer Co. Ltd., Guizhou Institute of Technology, and Guizhou Kexin Chem and Metallurgy Co. Ltd. are the top three assignees in Table S10, with 38 degrees each, accounting for 24.84% of the total assignees. These three institutions constitute the largest group of cooperation networks. In addition, there are two pairs of partnerships as shown in Figure S6, but compared to the largest, these assignees have less degree centrality. In contrast to the publication of articles, the special feature of patents lies in their exclusiveness and self-protection. Therefore, it is rare for a patent to belong to two institutions simultaneously. However, in actual technology research and development, it is necessary to strengthen exchanges between various institutions, even if they cannot be signed in the same patent.

Figure S7 shows the cooperation network of inventors, which is much more complicated than the map of assignees (Figure S6). The top ten inventors have a high degree centrality greater than 100, which proves that there is a strong



communication and connection between inventors. In general, patents are a vital way to protect intellectual property rights; therefore, cooperation is usually limited to individuals and rarely crosses institutions.

The research area of PG study

Basic knowledge structure of PG

When two articles were cited by another article simultaneously, the two articles constitute a co-citation relationship. Cocitation analysis of references is an important method for comprehending the structure, dynamics, and paradigm developments of a given research field (Zupic and Cater 2015). Figure S8 shows the reference co-citation map of the PG literature with 449 nodes and 1036 links drawn using the CiteSpace software. Each node in the graph represents a cited document, and the size of the node is proportional to the total citation frequency of the related documents. The spectra of different colors show the chronological order of the symbiotic relationship between the cited documents: blue indicates the oldest literature and red indicates the newest. Table S9 lists the top ten articles with co-cited frequency and other details, such as centrality, first author, and publication year. The prominent contributions of the top five co-cited articles in the field of PG were analyzed. The most cited article is a study by Tayibi et al. on the radioactivity of PG published in 2011. Tayibi et al. (2011) studied the stabilization/solidification process of PG as a building material from the perspective of radioactivity and obtained a building material with lower radioactivity by mixing PG with a polymeric sulfur matrix. The second most frequently cited article was a study by Rashad published in 2015 (Rashad 2015). The results showed that using a part of PG to replace fly ash in alkali-activated fly ash paste can increase the compressive strength and thermal shock resistance of the material. The third most cited study was by Yang et al. (2009), who used a low autoclave to pretreat the PG and prepared a kind of load-bearing wall brick that meets the Chinese standard brick size and can replace the traditional fired clay brick. Fourth was Hentati et al. (2015), which researched the effect of PG as a fertilizer on soil biological toxicity. The next most cited article was from Pérez-López et al. (2016), which explored the environmental impact of PG piles from the perspective of geochemistry. In general, references with centrality values greater than or equal to 0.1 in CiteSpace were considered key publications (Chen 2006). In Table S9, only the third-ranked article, Yang et al. (2009), has a centrality that meets this requirement; therefore, it can be considered a turning point study. In summary, these publications have paid great attention to the application performance and environmental impact of PG or its products from various fields and perspectives and provide the knowledge bases in the field of PG academic research.

To further explore the specific knowledge areas involved in these articles, we used the built-in LLR algorithm of CiteSpace to cluster these articles with keywords. After discriminating the correlation between the labels obtained by clustering and PG research, we screened nine clusters and displayed their details in Figure S9 and Table S10. Size represents the number of studies contained in each cluster, namely, the measurement of scales. The silhouette scores, an indicator of the value of homogeneity to evaluate clusters, are shown in Table S10. These values are generally greater than 0.8, indicating that the quality of each clustering result is reliable. The mean cited year denotes the average citation time of the documents in the corresponding cluster, which can reflect the time this category has received attention from academia.

It is apparent from Figure S9 and Table S10 that "natural radioactivity," "calcium sulfate," and "soil" are traditional clusters formed before 2010. In general, PG has a high content of natural radionuclides, especially the decay series of U 238 and Ra 226. Owing to these characteristics, PG is considered to be a technologically enhanced naturally occurring radioactive material (TENORM), which has greatly restricted its resource utilization (Moreira et al. 2018). As an inorganic powder, calcium sulfate whisker (CSW), which has excellent mechanical properties, environmental protection, and low price, is used as a high-value-added product of PG. The preparation process of CSW mainly focuses on the hydrothermal method and atmospheric acidification method, both of which can produce products with high aspect ratios and high purity (Sheng et al. 2018). In addition, PG can exhibit a great deal of advantages as a soil amendment in agriculture, such as improving soil physical and chemical properties, supplementing beneficial trace elements, and promoting crop growth. However, the heavy metals and natural radionuclides in PG have prompted studies to pay more attention to agricultural environmental risks (Wang 2020). In addition, information from other clusters shows that the basic academic areas of PG are concentrated in cement backfill materials (cluster 3), the preparation of supersulfated cement (cluster 4), and the extraction of rare earth elements (cluster 12). Furthermore, the crystal morphology of PG (cluster 11) has also received considerable attention (Jin et al. 2020; Lu et al. 2019; Ru et al. 2012). It is worth noting that cluster 10, "carbon dioxide," and cluster 15, "syngas," are unfamiliar research areas. Studies have shown that the products of PG at high temperatures can capture carbon dioxide (Cardenas-Escudero et al. 2011; Msila et al. 2016; Zhang et al. 2020b). This method of synergistic utilization of solid waste resources and carbon emission reduction provides a new perspective of PG recovery. Moreover, some scholars have used chemical-looping combustion to prepare syngas by using lignite as a carbon source, PG as an oxygen source, and water vapor as a hydrogen source, which makes full use of calcium sulfate to realize the utilization of PG (Yang et al. 2017; Yang et al. 2018; Yang et al. 2019).

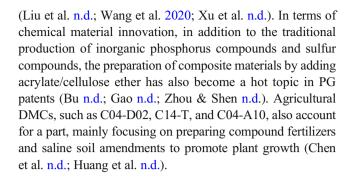


Hot areas of PG research

Figures S10 and S11 plot the co-occurrence network map of PG literature keywords and the DMCs from 2018 to 2020 to reflect the hot areas of PG research. The details of the top 20 keywords and DMCs in terms of frequency and centrality are summarized in Tables S11 and S12.

Obviously, the keywords "phosphogypsum," "waste," "gypsum," "heavy metal," "rare earth element," "hydration," "water," "cement," "soil," and "fly ash" are prominent in the frequency rankings. Centrality values exhibit that "phosphate," "compressive strength," "aqueous solution," "kinetics," "removal," "waste valorization," "sewage sludge," "mortar," "sulfate," and "set retarder" stand out in the rankings. With the information provided in the figures and tables, we divided the academic hotspot research of PG into the following aspects. At the microstructural level, the mechanism of hydration in the chemical process related to PG, the change in crystal structure, and the physical and chemical properties of the product (compressive strength, water resistance, durability, etc.) have attracted attention (Gijbels et al. 2019; Haque et al. 2020; Pinto et al. 2020). These studies can clarify the characteristics of PG to better promote its resource utilization. On the other hand, due to the peculiarity of PGs containing heavy metals and radioactive materials, the environmental impact and safety management of the stacking yard are continuously becoming academic hotspots (Attallah et al. 2019; Lütke et al. 2020; Wang 2020; Tsioka and Voudrias 2020). Furthermore, the utilization of PG focuses on the extraction of rare earth elements (Jalali et al. 2020; Binnemans et al. 2015; Canovas et al. 2019; Canovas et al. 2017), high-compressive strength cement (Huang et al. 2019), the preparation of cement retarders and cement mortar (Gong et al. 2020), sludge dewatering (Dai et al. 2018), and co-production with other solid waste (Wang et al. 2020). Compared with the "Basic knowledge structure of PG" section, although there are some overlaps between hot areas and the knowledge base, the results of keyword co-occurrence in the past 3 years provide us with more particular perspectives of the research directions about PG.

With respect to the DMC frequency, L02-C05 (calcium sulfate cements), L02-A03 (refractories, ceramics, and cement manufacture — shaping, drying), and A12-R01A (concrete, cement, gypsum, mortar compositions, and boards) rank in the top three. As for centrality, A12-R01A (concrete, cement, gypsum, mortar compositions, and boards), L02-D01 (mortars and plasters), A03-A04A (cellulose ethers uses), etc., have high rankings. In general, the results of DMC co-occurrence suggest that patent applications of PG are concentrated in the fields of construction, chemical industry, and agriculture. Specifically, refractory materials, ceramics, cement manufacturing, cement additives, surface materials, and cement mortar are the hot areas of PG patents in construction



Frontier topics of PG research

Burst keywords represent words that are cited continually during a certain period and are considered to be indicators of frontier topics over time. As shown in Tables S13 and S14, we used the CiteSpace software to detect burst keywords and DMCs from 2018 to 2020 to indicate the frontiers of PG. In particular, we placed emphasis on the keywords and DMCs with a longer duration and greater burst strength.

As can be seen from Table S13, the keywords "recovery," "coal," "tailing," "stabilization/solidification," and "temperature" possess higher strengths between 2019 and 2020. Masmoudi-Soussi et al. (2020) and Salo et al. (2020) developed a method for recovering rare earth elements from PG. Owing to the complex process and high cost of extracting rare earths from PG, the industrialization of such methods is still facing challenges and low-cost and high-efficiency rare earth recovery methods still need to be developed. Some scholars have also optimized the methods and conditions of ion solidification/stabilization in PG, which is conducive to the reprocessing and recycling of PG (Li et al. 2019a, b). Temperature, namely, one of the most crucial reaction conditions, plays a pivotal role in the formation of PG products. Exploring the reaction process and mechanism under different temperature conditions will also be the frontier direction of PG academic research (Lu et al. 2020). Besides this, the keywords "mechanism," "durability," and "behavior" indicate that research on the physical and chemical properties of substances at the micro-level will still be focus of PG in the future (Li et al. 2019a, b; Liu et al. 2020).

Likewise, DMCs, such as L02-A08 (refractories, ceramics, cement — manufacturing methods and equipment — testing/control), A12-R07 (building, civil engineering — walls, wall coverings, and ceilings), L02-D13 (mortars and concretes — aggregates), and A10-E05B (chemical modification by carbonization), occupy greater burst strengths during 2019–2020. As patents pay more attention to practicability, part of the development trend of PG patented technology is the improvement of equipment manufacturing, such as innovations in crushing and sorting devices and calcining and drying devices (Zhou et al. n.d.-a, n.d.-b; Zhu n.d.). In the preparation of building materials, innovative wall materials (such as gypsum



plasterboard, gypsum block, cement concrete, and silicate concrete) not only reduce the use of high-energy clay brick to reduce production costs but also solve the problem of large accumulation of PG (Gong and Shi n.d.; Jiang et al. n.d.; Yin et al. n.d.; Zhou et al. n.d.-a, n.d.-b). There are also some DMCs (A10-E21A, A10-E12A, and A10-E05B) in Table S14 indicating the chemical modification of various methods. Since the preparation of PG products involves many chemical processes, it is more appropriate to prepare innovative polymer materials by chemical modification (Shen et al. n.d.; Su et al. n.d.).

Prediction of research trend of PG

In order to quantitatively and accurately predict the growth trend of documents in various fields of PG, we merged articles with similar meanings of subject categories (such as classifying architecture, construction and building technology, and engineering as construction engineering) and summarized them into four categories: constructional engineering, chemical materials, environmental science, and agricultural science. The technical life cycle curve of PG articles, the S-curves, drawn by the Loglet Lab 4 software is shown in Figure S12, where the blue dots in the figure represent the actual cumulative number of PG articles each year, and the blue line is the fitting curve between the actual and the predicted numbers. The specific parameters and growth stage details are listed in Tables S15 and S16. Among the four categories, "constructional engineering" has the highest saturation value (324), which means it will have a broad development space. In contrast, the saturation value of "agricultural science" is only 111, which indicates that PG academic papers in agriculture are not likely to increase drastically in the future. The growth time and midpoint can be used to divide the technology life cycle stages, and the results are summarized in Table S16. From the perspective of the timing of the stages, the four types of technology categories are currently in maturity. The three major categories of "constructional engineering," "chemical materials," and "environmental science" are likely to enter the decline stage in 2033, 2031, and 2033, respectively. The forthcoming decline of the category of "agricultural science," approximately after 2025, stands out the most in Table S16. In summary, in terms of both the number of articles and the stage time, "construction engineering" has the most application prospects followed by "chemical materials" and "environmental science," but there is not much room for the development of "agricultural science."

Similarly, we also counted the annual distribution of the top four categories of the DMCs in the patents, namely, L (refractories, glass, ceramics), A (polymers, plastics), C (agricultural chemicals), and E (general chemicals). Figure \$13 plots the S-curve for each patent technical field. The specific results are presented in Tables \$17 and \$18. It can be seen from the value of saturation that the L category will have the largest number of accumulated patents, followed by the A category, C category, and E category. From the value of growth time, it can be judged that the L category has the longest development time, whereas the A category has the shortest one. In terms of the stage of the technology life cycle

(Table S18), it is considered that the L category is in the development period, the A category is in the decline period, and the C category and E category are in the mature period. Taken together, the future prospect of the distribution of PG patents in L (refractories, glass, ceramics) is the largest, and the development time is still very long. C (agricultural chemicals) and E (general chemicals) are basically the same; however, there is almost no space in the A (polymers, plastics) category to blossom.

Conclusion and prospect

This study conducted a bibliometric analysis of the articles and patents of PG in the Web of Science from 1990 to 2020. The key findings are summarized as follows.

(1) First, the basic status shows that the number of patents for PG is far greater than the number of articles. In terms of the quantity of national output, China has an absolute advantage in both articles and patents, but Spain, Brazil, India, and the USA perform better in a large number of indicators that reflect the quality of the literature. The subjects of the articles paid more attention to environmental studies, whereas the patents were more focused on chemical material issues. The journal Construction and Building Materials ranks first among PGrelated journals. Universidad de Huelva and Guizhou Kailin Group Co. Ltd. are the institutions that produce the most articles and patents, respectively. The distribution of assignees presents a strong geographical characteristic; that is, many are located in Guizhou Province, China. (2) Second, the results of social network analysis suggest that France, the USA, Tunisia, and China play a leading role in the international cooperation of PG. At the institutional and author levels of academic articles, there are mainly cooperative groups represented by Spain and China, respectively. Due to the particularity of patents, the cooperation of assignees and inventors is different from academic articles, namely, little connection between institutions but a lot in inventors. (3) Furthermore, the reference co-citation analysis indicates that the knowledge base in the field of PG research is concentrated in the fields of natural radioactivity, cemented paste backfill, soil, carbon dioxide, crystal morphology, and syngas. According to the results of the co-occurrence analysis of keywords from 2018 to 2020, the academic hotspots are concentrated on the study of the microstructure in the chemical process, the environmental impact of heavy metals and radioactive substances, and application research, such as rare earth element extraction, preparation of cement products, dewatering of sludge, and combined utilization with other solid waste. The cooccurrence results of DMCs indicate that the hotspot of the patents is the production of refractory materials, ceramics, surface materials, cement mortar, and composite materials by adding acrylate/cellulose. Regarding the research frontiers, burst keywords show that the academic frontier of PG is a method of



recovering rare earth elements from PG, the conditions of ion solidification/stabilization in PG, the impact of reaction conditions on product quality, and the micro-level reaction mechanism and physical and chemical properties. On the other hand, the forefront of patents will focus on the improvement of manufacturing equipment, innovation of new wall materials, and chemically modified polymer materials. (4) Finally, the technical life cycle S-curves of the various subfields of the articles and patents indicate that "construction engineering" and L (refractories, glass, ceramics) have the largest application prospects, but "agricultural science" and A (polymers, plastics) have the most narrow development space.

Although many scholars have conducted studies on PG impurity treatment technology, green building material production, and equipment, there are directions for further development in the following aspects. (1) The technology for the large-scale treatment of PG is insufficient. Although a large number of patents have been applied, there are few truly widely promoted patents. Many technologies have shortcomings, such as low promotion value, poor economy, and applicability, and have few advantages in practical applications. To address this, advanced PG production and discharge online quality control technology, low-cost pretreatment technology, and key common technologies for large-scale, high-value-added utilization should be studied urgently. Evaluation methods and systems of economics, applicability, and the scientific nature of different utilization technologies need to be proposed. (2) The increased cost of the PG impurity removal process results in a less competitive market for the final product. In addition, the acidic substances in PG, such as soluble phosphoric acid salts and free phosphoric acid, corrode the processing equipment and increase the cost of equipment corrosion prevention. Therefore, the process principles of pretreatment processes or measures such as neutralization, precipitation, water washing, flotation, grinding, calcination, and admixtures of PG should be studied systematically, and the applicable situations of different pretreatment methods should be compared. (3) At present, the infrastructure of some PG storage yards is not complete, and the anti-seepage treatment facilities and leachate collection devices of most storage yards are not perfect. There is a lack of effective monitoring of groundwater, surrounding rivers, and the soil environment. As a result, with respect to the safety management of PG storage yards, the study of the migration law of leaching pollutants, the historical deposition of PG depots, pollution control, and ecological restoration technology of the yard will become important research directions. (4) The policy system for PG products is insufficient. Although various countries have issued standards and policies for the management of PG, most of the products that use PG as resources are implemented with reference to other standards, which makes it difficult to apply these products on a large scale, and thus, they have low market acceptance. States need to formulate a series of standard systems, improve industrial policies and taxation and fiscal policies, promote the application of PG products in the construction industry, and mobilize the enthusiasm of enterprises.

In conclusion, this study provides an insight into the publications on PG and valuable information for researchers to identify new perspectives concerning potential partners, cooperative institutions, knowledge structure, research hotspots, and frontiers. Although we have obtained many valuable conclusions about the academic and applied research of PG, the key factors restricting the resource utilization of PG are still directions worthy of discussion in the future. Researchers should not only be keen on theoretically innovative research, but also pay attention to the possibility of technological application. Further work needs to be carried out to promote technologies and products with wide applicability and high added value to form a circular economy chain using PG.

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Author contribution C YM: methodology, data collection and processing, writing — original draft, writing — review and editing. C Y: data collection and processing, visualization, writing — review and editing. Y XK: data collection and processing, visualization. L T: writing — review and editing. C IS: supervision. W J: conceptualization, supervision. All authors read and approved the final manuscript.

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