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Review article

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Recycling phosphogypsum in road construction materials and associated environmental considerations: A review



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ABSTRACT

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Phosphogypsum (PG) is a major hazardous by-product of the phosphate industry. The whole world is facing the challenge of increasing stockpiles of PG, which significantly affect safety and the environment. Recycling this material may be an environmentally friendly and safe solution to this challenge. In this paper, we searched the Web of Science database for 2369 articles from 1975 to 2022 using "phosphogypsum" as the keyword. Using the VOSviewer as the literature visualisation tool, density visualisation was performed with keywords as the analysis units, and it was found that the researchers focused mainly on PG applications in construction and radionuclides. Over the years, different routes to PG valorisation have been developed in the agricultural, construction, environmental and energy sectors, and these topics are described in this review. As infrastructure projects, highway engineering is characterised by a large land area and high resource consumption. Therefore, applying PG to road materials can significantly increase the reuse and consumption of PG and reduce PG stockpiles. We hope that PG can be used for road construction on a large scale in the future. This paper focuses on the latest developments in PG as road materials. The most serious difficulty in using PG as road materials at present is that it contains radionuclides and heavy metal impurities, which can be purified by pretreatment methods. However, how to scale it up requires continuous research.

1. Introduction

Phosphoric acid (PA) is a common inorganic acid used in agriculture, food, pharmaceuticals, chemicals, etc. Phosphoric acid is a main raw material for producing agricultural phosphate fertilisers and a major food additive, and it is also used in manufacturing phosphorus-containing drugs in pharmaceuticals. At present, commonly used PA preparation methods include thermal and wet phosphoric acid (WPA) processes. The thermal process refers to the thermal reduction of phosphate ores in an electric furnace to produce elemental phosphorus, while the WPA process produces elemental phosphorus through a chemical reaction between phosphate ores and sulphuric acid. The WPA process is the main industrial process for producing PA, accounting for approximately 90% of global phosphoric acid production (Ober, 2017). However, a large amount of PG is also produced (Canovas et al., 2017). PG is produced in WPA production with sulfuric acid. The mass of the PG produced during phosphoric acid production exceeds that of the product, i.e. 4.5-5.5 tonnes of PG are produced for every tonne of P2O5 produced (El-Didamony et al., 2012).

Phosphogypsum is usually a yellowish-white, light grevish-white or dark grey fine powdery solid. The main component of PG is CaSO₄·2H₂O, which accounts for more than 90% of its total composition. In addition, PG contains phosphorus, fluorine, organic matters and other impurities (e.g. heavy metal elements and nuclides) (Rashad, 2015). These substances negatively affect the environment, especially radium-226, uranium and other uranium decay products contained in PG (IAEA Technical Reports Series No. 475, 2013). The following measures are generally used worldwide to deal with PG: dumping it into the sea, storing it in mines where phosphate-bearing rock is extracted, wet storage, and dry storage. The latter two are the most common approaches (Silva et al., 2022). Phosphogypsum stacked in the open (as shown in Figure 1 (a, b)) severely damages soils, water systems, atmosphere and other environments. Radon-222 exhalation (Rutherford et al., 1994) and hazardous gases containing phosphorus, cadmium and Radium-226 (Marovic and Sencar, 1995) may have a negative impact on the atmosphere, releasing highly polluting substances and spreading pollutants to nearby areas. Since PG is usually transported and disposed of as a water slurry, it is prone to tidal influences, and the elements present in PG may be dissolved and leached (Tayibi et al., 2009). The leached elements can be

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Figure 1. PG storage site (Mianzhu, Sichuan).

deposited into the soils and then diffuse into water systems, severely damaging the environment (Reijnders, 2007).

In recent years, countries and organisations worldwide have been promoting their processes to reduce emissions and protect the environment under the impact of climate change and the energy crisis. China is working towards achieving peak carbon emissions and carbon neutrality; the European Union (Zmemla et al.) is also taking action on its climate and energy goals; the US Environmental Protection Agency (EPA) is developing its plan for power plants to reduce carbon emissions (Tollefson, 2022). China's 14th Five-Year Plan proposes guidance on the integrated use of bulk solid waste. Chinese researchers are supposed to focus more on improving the reuse of solid waste, such as PG, and expanding the ways in which PG can be applied. There is a gap in gypsum supply in the EU due to the Renewable Energy Directive, and PG can be a valuable alternative to natural gypsum and FGD gypsum (Haneklaus et al., 2022). The reuse of PG is very significant in terms of environmental and energy considerations (Wedrychowicz et al., 2019). However, only 15% of global PG resources (Wu et al., 2022a) are used in agriculture, construction and chemicals. The trace amounts of radionuclides in PG have led to slow growth in its reuse worldwide (Willey and Timbs, 2022). Global researchers are currently working to reduce the radionuclide content of PG to a safe level (Arhouni et al., 2022). Most PG is used for traditional purposes, but in recent years researchers have become interested in extracting rare earth elements (REEs) from PG. Rare earth elements are an important strategic resource, widely used in military, metallurgical and petrochemical applications. Technology advances have led to a global shortage of REEs, so researchers have turned to take PG as a source of REEs.

The global consumption of P_2O_5 is predicted to increase to 50 million tonnes by 2023, resulting in a significant increase in global PG production (Costis et al., 2021). The current utilisation rate of 15% is clearly unable to cope with the massive accumulation of PG. To broaden the scope and increase the rate of PG reuse, treated PG can be used as a binder or filler for the pavement or subgrade in highway engineering. The use of PG in highway engineering can simplify the process and increase the reuse rate compared to that in other application areas. In addition, the large work volume and high consumption of road construction can solve the problem of large stockpiles of PG well.

In summary, based on recent research, we summarise the nature, pretreatment methods and resource routes of PG, discuss the radiological hazards of PG, and analyse the current status of PG applications and research in highway engineering. The results of this review provide a basis for future research and the applications of PG in sustainable methods.

2. Method

We searched in detail for works involving PG, with the consideration of all areas of interest, such as solid waste properties, solid waste management, environmental impact and recycling. The literature search was conducted using the world-renowned indexing database Web of Science (http://www.webofknowledge.com/), which compiles peerreviewed publications considered to be of high quality. The Web of Science database was searched from 1 January, 1975 to 3 June, 2022 using "phosphogypsum" as the keyword. A total of 2369 papers were retrieved. Using the VOSviewer as the literature visualisation tool and keywords as the analysis units, we found that "phosphogypsum" and "gypsum" appeared up to 1213 and 280 times, respectively. As a key factor influencing whether PG can be utilised, "radioactivity" appeared 190 times. In addition, "lime", "fly-ash", "cement" and "hydration" all appear more than 100 times. Based on the analysis through the VOSviewer, currently, PG is mainly used in the construction sector, and researchers are concerned about the radioactivity of PG. The analysis results can be verified in the density view, as shown in Figure 2. In the figure, there are also some keywords with a relatively low density, such as "plants", "sulfate" and "rare-earth-elements", which have been relatively little studied but have good promise. These areas will also be covered in this paper.

3. Properties of PG

3.1. Chemical composition

The chemical composition of PG produced in different countries is shown in Table 1. It indicates that the chemical composition of PG varies from one area to another. In general, PG mainly consists of CaO and SO₃, with both accounting for approximately 70% of the total mass. In addition, PG also contains Fe₂O₃, Mg, Al, sulphides and organic matters (Masmoudi-Soussi et al., 2019). During the reuse of PG, impurities in the PG affect the product properties. Singh et al. (Singh, 2005) found that more than 1.0% soluble phosphate adversely affected the setting time and strength development of selenite gypsum. Moreover, the fluoride and organic matters in PG can decrease the compressive strength of gypsum. Min et al. (2007) found that soluble phosphorus and organic matters in PG could prolong the gelling time of gel materials. Furthermore, these impurities in PG have adverse effects on the serviceability of gypsum dihydrate and gypsum hemihydrate. In other words, PG should be pretreated before reuse to remove impurities and prevent the adverse effects on PG-based materials.

3.2. Physical properties

The physical properties of the material influence its application. Phosphogypsum mainly consists of silt-sized gypsum crystals. These gypsum crystals are softly aggregated, and their morphology depends on the sources of the phosphate rock and the reaction conditions of the acid attack (IAEA Technical Reports Series No. 475, 2013). Most PG produced

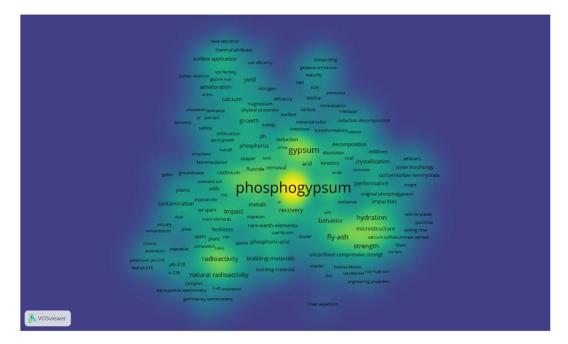


Figure 2. Density view based on the VOSviewer.

Table 1. Chemical composition of PG.

References	Country	CaO/%	SO ₃ /%	SiO ₂ /%	Fe ₂ O ₃ /%	F ⁻ /%	Al ₂ O ₃ /%	P ₂ O ₅ /%	Others/%
(Jie et al., 2021)	China	29.60	41.40	12.20	-	0.24	0.49	0.08	15.99
(Degirmenci et al., 2007)	Turkey	32.04	44.67	3.44	0.32	0.79	0.88	0.50	17.36
(Thanh et al., 2021)	Vietnam	26.74	38.81	10.40	0.22	1.17	0.75	0.27	21.64
(Ajam et al., 2009)	Tunisia	32.80	44.40	1.37	0.03	0.55	0.11	1.69	19.05
(Gaiducis et al., 2011)	Lithuania	34.35	51.11	4.35	0.06	0.50	0.24	2.33	7.06
(Kadirova et al., 2014)	Uzbekistan	36.48	49.89	1.89	-	-	0.08	0.44	11.22
(Rashad, 2015)	Egypt	32.14	34.51	8.82	0.35	0.80	0.29	1.72	21.37

is a fine powder with a particle size distribution mainly ranging from 10 to 1000 μ m (Wu et al., 2022a). The density of PG particles fluctuates between 2.27 and 2.4 g/cm³, and their bulk density fluctuates between 0.9 and 1.7 g/cm³ (Rutherford et al., 1994). Figure 3 (A,B) shows the microscopic morphology of PG, which is mainly in crystalline forms, such as flaky irregularities and rhombuses, with a large number of impurities adsorbed on its surface. Ji et al. tested the fundamental physical properties, such as maximum dry density and optimum moisture content, according to the *Highway Geotechnical Test Procedure (JTG 3430-2020)*. The results are shown in Table 2. After drying and sieving, the gradation of PG is shown in Figure 4(Ji et al., 2021).

3.3. Nuclides

Naturally occurring radionuclides are mainly derived from phosphate rock used in processing (Rutherford et al., 1996). The mined phosphate rock is further processed into the required product by the WPA process, with PG as a by-product. The radionuclides that can occur naturally in the phosphate rock are transferred to PG, leading to the PG containing trace amounts of radioactive elements (Arhouni et al., 2022). The radioactivity is mainly from 238U, 232Th and 226Ra (Saadaoui et al., 2017). During phosphoric acid production, radionuclides are partitioned between the H₃PO₄ and PG phases (Szajerski, 2020). Elemental uranium is transferred

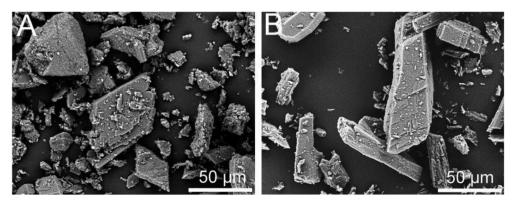


Figure 3. Microscopic morphology of PG (Mi et al., 2019).

Table 2. Physical properties of PG.

Liquid limit/%	Plastic limit/%	Plasticity index/%	Maximum dry density/(g/cm ³)	Optimal water content/%	Relative volume mass
43.2	30.8	12.4	1.285	25.8	2.37

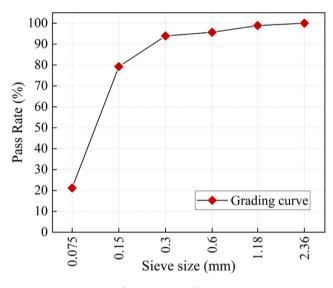


Figure 4. PG gradation.

from the non-mobile fraction in phosphate rock to the bioavailable fraction in PG at a rate of 23% (Perez-Lopez et al., 2007). 226Ra, 210Pb and thorium isotopes preferentially favour PG at 90% for 226Ra (with ore), 100% for 210Pb, and 80% for 232Th and 230Th (Mazzilli et al., 2000). The types and contents of nuclides in PG determined by various countries, regions and organisations are shown in Table3. The radiation

hazard indicators for PG determined by various countries, regions and organisations are shown in Table 4. There are some variations in the types and contents of nuclides in PG produced in various countries, regions and organisations and in the radiation hazard indicators for PG measured by researchers.

4. Pretreatment method of PG

As mentioned above, soluble phosphorus, fluorine, and organic matters in PG have adverse effects on PG reuse. Therefore, pretreatment is required before reusing PG. The adverse substance can be removed using pretreatment methods to improve the utilisation performance of PG-based products. At present, the commonly used PG pretreatment methods can be divided into physical, chemical, and thermal treatment methods.

4.1. Methods for PG pretreatment

4.1.1. Physical method

The physical methods for PG pretreatment can be classified as washing, flotation and screening. Liu et al. (2019) used ionised water to wash PG to purify the PG for cement slurry backfill and improve its filling properties. Water washing can remove a certain number of soluble impurities from the surfaces of PG crystals and greatly improve the workability and mechanical properties of PG-based cemented paste backfill (CPB). After 7, 14 and 28 days of curing, the unconfined compressive strength values of the purified PG-filled CPBs reached 0.838, 1.953 and 2.531 MPa, respectively, which were 0.283, 0.823 and 0.881 MPa higher

l contents of nuclides		

Reference	Countries, regions and organisations	238U	235U	210РЬ	214РЪ	226Ra	228Ra	232Th	234Th	40K
(Shen et al., 2019)	China	$\textbf{28.00} \pm \textbf{13.00}$	$\begin{array}{c} \textbf{2.70} \pm \\ \textbf{0.10} \end{array}$	$\textbf{86.00} \pm \textbf{4.80}$	-	61.00 ± 2.10	-	2.30 ± 0.50	-	$\begin{array}{c} 3.30 \pm \\ 6.00 \end{array}$
(Luca et al., 2009)	IAEA	-	$\begin{array}{c} \textbf{2.86} \pm \\ \textbf{0.46} \end{array}$	150.00 ± 19.00	$\begin{array}{c} \textbf{72.00} \ \pm \\ \textbf{10.00} \end{array}$	100.00 ± 13.00	-	-	$\begin{array}{c} 47.00 \pm \\ 7.00 \end{array}$	-
(Papastefanou et al., 2006)	Greece	19.00 ± 7.00	-	430.00 ± 7.00	-	585.00 ± 2.00	$\begin{array}{c} 3.30 \ \pm \\ 0.10 \end{array}$	3.30 ± 0.10	-	$\begin{array}{c} 13.00 \pm \\ 1.00 \end{array}$
(Msila et al., 2016)	South Africa	31.00 ± 20.00	$\begin{array}{c} 1.00 \ \pm \\ 1.00 \end{array}$	135.00 ± 37.00	-	109.00 ± 18.00	$\begin{array}{c} 404.00 \pm \\ 70.00 \end{array}$	189.00 ± 155.00	-	<100.00
(da Conceicao and Bonotto, 2006)	Brazil	298.00-310.00	298.00– 310.00	-	-	269.00-280.00	-	185.00-206.00	-	60.00– 63.00
(Renteria-Villalobos et al., 2010)	Morocco	100.00 ± 22.00	-	827.00 ± 46.00	-	$\textbf{785.00} \pm \textbf{42.00}$	-	-	-	-
(Rutherford et al., 1995)	United States	4.50	-	340.00-440.00	-	470.00-680.00	-	-	-	-
(Kuzmanovic et al., 2020)	Serbia	-	-	-	-	656.00 ± 4.00	-	3.10 ± 0.90	-	101.00 ± 6.00
(Gezer et al., 2012)	Turkey	-	-	-	-	436.00		9.00	-	13.00
(Alam et al., 1997)	Bangladesh	-	-	-	-	234.00	-	21.00	-	108.00
(Mourad et al., 2009)	Egypt	-	-	-	-	596.00	-	6.00	-	2.00
(Al-Jundi et al., 2008)	Jordan	-	-	-	-	376.00	-	4.00		40.00
(Song et al., 2011)	Korea	-	-	-	-	618.00	-	8.50	-	24.10
(Kovler et al., 2002)	Israel	-	-	-	-	747.00	-	14.00	-	63.00
(Duenas et al., 2010)	Spain	-	-	-	-	740.00 ± 60	-	<8.00	-	$\begin{array}{c} 50.00 \pm \\ 6.00 \end{array}$
(El Zrelli et al., 2018)	Tunisia	1.60 ± 0.16	-	-	-	-	-	$\textbf{0.74} \pm \textbf{0.70}$	-	-
(Diwa et al., 2022)	Philippine	655.80	-	-	-	-		14.50	-	84.80

Reference	(Al-Hwaiti et al., 2014)	(Attallah et al., 2019)	(Joel et al., 2018)	(Wang et al., 2012)	(Bangotra et al., 2018)	(Diwa et al., 2022)	(Boumala et al., 2018)	(Gryb and Thebaault, 2014; Mehboob et al., 2021)	(Khan et al., 1998)
Countries	Jordan	Egypt	Nigeria	China	India	Philippine	Algeria	Saudi Arabia	Pakistan
Raeq (Radium- equivalent activity)/ (Bq/kg)	$\textbf{366.10} \pm \textbf{24.40}$	281.00 ± 8.50	138.44 –391. 10	130.00	125.00	683.10	690.00	68.10	527.00
Iy (Gamma hazard index)/(Bq/kg)	1.20 ± 0.10	$\textbf{0.95} \pm \textbf{0.02}$	0.49–1.36	-	0.47	2.30	-	-	-
Ia (Alpha hazard index)/(Bq/kg)	1.80 ± 0.10	1.16 ± 0.2	0.19–1.21	-	0.18	-	-	-	-
DR (absorbed dose rates)/(nGy/h)	-	-	-	60.00	-	315.30	297.00	35.20	242.00
Dout (dose rate in outdoor air)	-	0.031 ± 0.001	-	-	-	-	-	217.00-849.00	-
Din (dose rate in indoor air)	337.10 ± 22.50	0.124 ± 0.02	118.73–347.77	-	-	-	-	429.00-1668.00	-
Dannual (annual effective dose)/ (mSv/year)	1.70 ± 0.10	0.155 ± 0.006	0.65–1.77	0.074	0.36	0.39	0.36	0.04	0.234
Hin (internal hazard index)/(mSv/year)	-	-	0.49–1.71	-	0.35	3.60	2.25	2.27–9.04	-
Hex (external hazard index)/(mSv/year)	-	-	0.37-1.06	-	0.35	1.80	1.86	-	-

Table 4. Radiation hazard indicators for PG in different countries, regions and organisations.

than those of unpurified PG-filled CPBs. Mohammad Salem Al-Hwaiti (2015) used a mixture of water, sulphuric acid, mixed acids, and calcium carbonate powder to leach 226Ra from PG during the purification process and the maximum percentages of 226Ra removed from PG were 88.78, 88.5, 86.55 and 84.37.

A more desirable method for PG purification is to combine sulphuric acid leaching with preliminary water washing, which provides high efficiency for PG purification (Lokshin and Tareeva, 2015). Ennaciri et al. (2020) combined sulphuric acid leaching with preliminary water washing and experimentally concluded that the optimum conditions for removing P_2O_5 and F at 60 °C were the acid concentration of 5% and the PG/H₂SO₄ ratio of 1/3, under which the removal rates of P_2O_5 and F were 60% and 95%, respectively.

Although the ball-milling treatment cannot remove the impurities in PG, it can improve the fluidity and strength of PG and solve the problems of the high porosity and loose structures of PG-based gelling materials. Wu et al. (2022d) experimentally found that the ball-milling treatment could effectively reduce the particle size of PG and increase the specific surface area of PG. The ball-milling treatment can effectively reduce the water consumption of normal consistency. With a ball milling time of 50 min, the minimum water consumption of the milled PG is 60.07%, 21.5% lower than that of unball-milled PG. In addition, the ball milling treatment also increased the compressive strength of PG at 2 h. Du et al. (2022) performed the ball-milling treatment on the PG raw ore and stirred it for flotation. The organic matters and microfine-particle mud were removed after reverse flotation. Then, dodecylamine was added to flotation the gypsum concentrate. After flotation purification, the impurity content in PG was significantly reduced, with the soluble phosphorus content decreasing from 0.48 to 0.07% and the PG purity increasing from 73.12 to 94.37%. The properties of the PG products before and after purification also varied. The 2-h flexural and compressive strength of α -HH prepared from PG increased by 46.15% and 79.46%, respectively, and the 3-day dry compressive strength increased by 39.6% compared to the original ore. Although the ball milling method cannot remove impurities directly, purification effects are more satisfactory when the PG is purified by other physical methods after ball milling.

The sieving method is only a good choice when the impurities are grossly unevenly distributed. Sieving can substantially reduce the impurity content. In the wet sieve cyclone treatment method, damp sieving is first conducted on PG using a 300-µm aperture sieve to remove the relatively coarse part (10%–15%) rich in impurities, and then PG is purified by a hydrocyclone. The schematic diagram of this method is shown in Figure 5. Using this method, soluble impurities in PG, such as phosphorus, fluorine, and other organic matters, can be effectively removed. The wet sieve cyclone treatment method has an excellent purification effect. However, its process is relatively complex, and there may be secondary pollution during the process (Singh et al., 1996).

Screening methods are used infrequently, and grossly uneven distribution of impurities is extremely rare. Water washing and flotation methods are commonly used to purify PG. The water washing method can remove most impurities. However, the wastewater after washing must be treated properly to prevent secondary contamination. Flotation is not as effective as water washing, but the water can be reused, making it more environmentally friendly. The ball milling method cannot eliminate the adverse effects of impurities but can effectively change the structure of PG. Therefore, combining the ball milling method with other methods is more effective. In conclusion, physical treatment methods need to be selected according to different engineering requirements.

4.1.2. Chemical method

The solidification and stabilisation of toxic and hazardous substances in PG are achieved by the production, adsorption, and encapsulation of insoluble substances. The purification of PG is achieved by acid-base neutralisation reactions because the pH of PG is 3 (Demirel and Caglar, 2015). Calcium carbide slag is a typical alkaline industrial solid waste and can be used as an alkali neutralising agent for PG. Wu et al. found that the 0.5% calcium carbide slag, as an alkaline conditioning agent, mixed with 0.4% silicate cement, 0.3% polymeric aluminium chloride and CaCl₂ had a significant curing effect on P and F. The leaching toxicity met the requirements of the relevant standard (Wu et al., 2022c), laying a good foundation for PG-based eco-remediation materials and fillers. The use of calcium carbide slag as an alkali neutraliser for PG has the advantages of low economic cost, simple process, and high feasibility. However, researchers found that the reaction rate of calcium carbide slag was higher than that of lime, but the reaction effect was significantly lower than that of lime (Wu et al., 2022b).

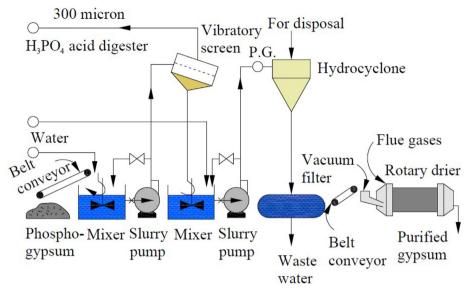


Figure 5. Purification flow chart of the wet sieve cyclone treatment method (Singh et al., 1996).

From an economic point of view, lime purification of PG does not require water, calcination or expensive materials, which can be applicated on a large scale. The chemical treatment based on Ca(OH)₂ reduced the concentration of PG impurities by 80.3% for water-soluble phosphorus and 25.0% for fluoride (Neto et al., 2021). Liu et al. concluded experimentally that 5% Ca(OH)₂ was the optimum level for lime neutralisation of PG and that 1 day was sufficient to complete the neutralisation treatment. By removing the P_2O_5 impurities from the PG, the compressive strength of the cement was effectively increased, while the lime neutralisation accelerated the hydration of the cement and reduced the setting time (Liu et al., 2020a).

Chemical and physical treatments are not independent of each other, but complementary. Applying two or more methods in combination may give more satisfactory results. Wang et al. added lime to the raw PG to remove soluble phosphorus and fluorine and used flotation to purify PG. Reverse flotation was used to remove a large amount of organic matter and fine mud from the PG, and positive flotation to remove the gypsum. The tests showed an increase in PG whiteness from 31.5 to 58.4, a reduction in the percentage of (P_2O_5) from 1.78 to 0.89, a percentage of calcium sulphate dihydrate of 96.6%, and a concentrate recovery of 74.1% (Wang et al., 2020).

Although multiple purification methods will improve purification efficiency and increase purity, it is sometimes difficult to completely remove phosphate impurities incorporated into the gypsum lattice (intergranular phosphate impurities). Incomplete purification will hinder the application of PG. Therefore, removing intergranular phosphate impurities during pretreatment is important for utilising PG in cement. Manjit Singh treated phosphogypsum using a 3-4% aqueous citric acid solution and rinsed it with water for purification (Singh, 2002). This method can partially remove intergranular phosphate impurities with good purification. Thus, using acid solutions to remove intergranular phosphate impurities may be feasible. Citric acid can be used as a cement retarder (Moschner et al., 2009). However, residual citric acid can lead to delayed hydration of the cement, severely limiting the application of PG in the cement industry. Therefore, selecting an acid solution that can remove intercrystalline phosphate impurities without negatively affecting the cement is urgent. Cai et al. experimentally found that H₂C₂O₄ could remove intercrystalline phosphate impurities by destroying part of the crystal structure of gypsum. Under the optimum treatment concentration of 1% H₂C₂O₄, the removal rate of phosphate impurities such as P₂O₅ from PG was 77.7%. The initial and final setting time of silicate cement prepared from PG treated with 1% H₂C₂O₄ was reduced to 155 min and 220 min, respectively. Furthermore, 3d and 28d

compressive strengths of silicate cement prepared from PG treated with 1% H₂C₂O₄ were 20.8 and 44.6 MPa, respectively (Cai et al., 2021).

4.1.3. Thermal treatment method

The calcination pretreatment method has shown good results in removing phosphorus, fluorine, and organic impurities. The method can also convert soluble and eutectic phosphorus into inert impurities with no adverse effects on the properties of PG (Cao et al., 2021). Furthermore, calcination allows large-scale treatment of virgin PG with high efficiency and effectiveness, which is friendly to large-scale industrial production.

The pretreatment of PG by calcination reduces the adverse effects of impurities in PG and changes the gypsum phase. The XRD analysis of Liu et al. showed that PG calcined at 150 °C was mainly composed of dihydrate and part of β-hemihydrate. After calcination at 350 °C, with increasing calcination time, dihydrate was largely converted to β -hemihydrate, and type III hard gypsum appeared. The PG was almost completely dehydrated to hard gypsum, and II-U hard gypsum and II-E hard gypsum appeared at 600 °C-800 °C. In addition, soluble phosphorus was removed by converting soluble phosphorus to insoluble calcium pyrophosphate (CaP2O7) (Liu et al., 2020b). Researchers also prepared supersulphate cement using PG purified at different calcination temperatures. The supersulphate cement prepared from PG calcined at 350 °C has the fastest setting times and the best processing properties, while calcination at 600 °C gives the best early and late strength properties. The temperature selection depends on different requirements for different engineering applications.

PG contains heavy metals, such as arsenic, lead, chromium, vanadium, zinc, and cadmium, which have irregular shapes and various particle sizes (20–50 μ m) and are difficult to be removed completely by conventional debinding methods. Imed Ghiloufi et al. separated heavy metal elements from PG using plasma technology to treat PG by pyrolysis. Both the thermal plasma treatment time and the plasma current magnitude can affect the evaporation degree of heavy metal elements. The results showed that heavy metal elements such as lead, cadmium, vanadium, chromium, arsenic and zinc evaporated completely, and the PG became inert after 40 min of treatment at a plasma current of 160 a. This method offers new ideas for PG decontamination (Ghiloufi et al., 2021). Using electricity as an energy source in the plasma system reduces heat emissions and increases the controllability and flexibility of the working process, making the method safe, efficient and environmentally friendly. Solar energy has been a popular renewable energy source in recent years. Researchers have endeavoured to use the sun's high-quality photothermal resources for the PG industry. Palla et al. performed solar thermal treatment of PG with a Scheffler reflector and used the treated PG as a cement retarder (Palla et al., 2022). It was demonstrated that soluble phosphates such as P_2O_5 were converted to an inert form (e.g. pyrophosphate) in the photothermally treated PG and that the minimum temperature required for PG to be used as a retarder was 350 °C. Compared with the calcination process, the use of solar energy protects the environment, reduces fuel consumption, and saves expenditure, resulting in a double benefit for the environment.

4.2. Application of PG in agriculture

The application of waste PG is currently a hot topic in agriculture, where it can provide a certain amount of calcium and phosphorus according to the composition of the PG. Therefore, waste PG can positively impact plant growth (Hentati et al., 2015). The main current applications of PG in agriculture are to improve soil physicochemical properties, provide trace elements to plants, promote plant growth, and reduce greenhouse gas emissions.

Soil provides plants with the necessary nutrients for growth. Soils in areas such as saline soils are not suitable for plant survival. Soils that contain many soluble salts and exchangeable sodium are defined as saline or sodic soils (Navak et al., 2013). Saline soils are widely distributed and require the necessary amendment. Saline soil amelioration requires an increase in the concentration of Ca^{2+} at the cation exchange point to replace Na⁺ (Liang et al., 1995). The application of PG rich in Ca²⁺ has a good can improvement on saline soils. Huang et al. applied PG as a soil amendment to saline rice soils planted with rice. After 10 years of field trials, the Na + concentration in the 0-20 cm surface layer of PG-amended rice fields was reduced by 42.9%, 61.5%, and 60.9%, respectively; the $CO_3^{2-} + HCO^{3-}$ concentration was reduced by 63.2%; chloride ion concentration was reduced by 75.7%. In addition, the 0-20 cm surface soil organic matter (SOM), fast-acting nitrogen (AN) and fast-acting phosphorus (AP) concentrations were significantly increased in PG-amended rice fields compared with the control. The PG can promote soil desalination during rice cultivation in saline soils, significantly reduce salinization parameters, improve soil fertility, and reduce soil pH (Huang et al., 2022).

Once the extraction of mineral resources is complete, the huge mining areas are abandoned, which is a great waste of land resources. If the mining area is converted into arable land, the area of arable land can be greatly increased. However, plant growth is difficult in mining areas due to the coarse texture and low fertility of the soils. Guéablé et al. found that 65% of PG in greenhouse trials can increase crop yields by enriching the substrate with phosphorus. It can also be used on a large scale by mixing PG, phosphorus sludge (PS), sewage sludge (SS), mining topsoil (Antonick et al.), and phosphate waste rock (Gueable et al., 2021). The method can improve the utilisation of waste while protecting the environment to create a secondary economy.

Trace elements are essential for the growth and development of plants. Sulphur is an essential nutrient and one of the key macroelements necessary for plant growth. Plants cannot absorb sulphur directly from the soil in the form of SO²₄. However, adding PG can readily absorb sulphur from the soil. The range of sulphate levels measured during the experiments of Lipsa Panda et al. ranged from 23.45 to 20.37 mg/L, while the sulphate levels of the PG and biocalcium phosphogypsum composite (BPC) amended soil ranged from 44.68 to 36.96 mg/L and 35.35 to 31.75 mg/L (Panda et al., 2022). Therefore, sulphur is present in the soil in a leachable form for plant growth (Peng et al., 2020). Furthermore, phosphorus is one of the essential micronutrients for plants. The application of a soil amendment of PG mixed with lime by Bossolani et al. increased nutrient concentrations, especially P, Ca, and Mg, and decreased Mn concentrations in the soil and plants. These effects will eventually be reflected in increased plant yields (Bossolani et al., 2021).

Michalovicz et al. found that PG dosages between 4.0 and 6.1 Mg/ha increased crop yields by 11%, 10%, and 10% for maize, barley, and wheat, respectively. The calcium and sulphur provided by PG also benefit plant nutrition and increase crop yields (Michalovicz et al., 2019).

In recent years, researchers have focused on binding PG to microorganisms. Tian et al. reported that phosphorus was more likely to be rapidly released from PG with the addition of Aspergillus niger. In the absence of Aspergillus niger, PG only released 28 mg/l phosphorus. However, PG released 128 mg/l phosphorus with the addition of Aspergillus niger. The PG in the soil also rapidly increased soil P content (22.2 mg/kg) (Tian et al., 2022). The use of PG by Aspergillus niger is a viable way to increase soil phosphorus content, especially to reduce the consumption of phosphate resources and promote their sustainable use. Furthermore, using PG and plant growth to promote rhizobacteria (PGPR) for agricultural purposes is a good option to improve soil properties and increase crop yields. Three field trials were conducted over two growing seasons by Khalifa et al. The results showed that application of PG (9 t/ha) significantly increased chlorophyll and carotenoid content, antioxidant enzymes, microbial communities, soil enzyme activity, and nutrient content, increased maize plant productivity, and improved the negative impact of salinity on maize plants (Khalifa et al., 2021). Microorganisms grow and multiply quickly, and combining them with PG in agriculture can increase crop yields while reducing damage to the ecosystem and protecting the ecological balance.

Plants emit CO₂ during growth and worldwide plant emissions also contribute to global warming. Wu et al. conducted field trials and laboratory cultures of PG applied to winter wheat for two consecutive years. The results showed that compared with the control, soil N₂O emissions in the first year of treatment with PG were reduced by 5.8%-29.9%, CO₂ emissions were reduced by 2.5%-6.6%, while in the second year, N₂O emissions were reduced by 20.6%-30.4% and CO₂ emissions were reduced by 6.2%-9.6%. During laboratory incubation, N₂O emissions decreased by 18.9%-28.5%, and CO₂ emissions decreased by 33.5%-50.1% (Wu et al., 2018). The reason for the results is that PG can reduce greenhouse gas emissions from wheat soils by inhibiting soil cellulase and sucrase activities. The finding can be used to reduce greenhouse gas emissions from agriculture to prevent increased global warming, thus reaping significant economic and environmental benefits.

In conclusion, PG has great application potential in agriculture. However, it is important to determine the effectiveness of specific applications of PG in agriculture and the associated problems, especially environmental problems. Furthermore, PG has some disadvantages and must be mixed with other materials.

4.3. REEs recovery from PG

The REE is a global strategic resource composed of seventeen chemical elements such as lanthanide, scandium, and yttrium. Due to its unique electronic structure and special properties, it has become an essential element group for developing high-tech and cutting-edge technologies (Dushyantha et al., 2020). It plays a vital role in magnets, catalysts, biology, new materials, new energy, and atomic energy (Peiravi et al., 2021). China controls more than 90% of the world's REEs production (Blissett et al., 2014). In addition to the surge in China's domestic REEs consumption in recent years, the quota for REEs exports has been reduced. Therefore, finding new REEs sources has been a focus of scientists from all over the world. With the deepening of research, PG has attracted the attention of researchers worldwide. The PG has a low content of REEs, with the total amount in the mass range of 0.343-0.637% (Grabas et al., 2019). However, PG is a potential source of REEs due to its high annual production (Ramirez et al., 2022). With the development of science and technology, the leaching rate of REEs continues to increase. At present, there are two main methods, i.e., chemical and biological methods, for extracting REEs from PG.

Currently, researchers mostly use inorganic acids as leaching agents to leach REEs in PG. Strong acids, such as nitric acid and sulfuric acid, have attracted the attention of researchers because of their high leaching and recovery rates of REEs. Cánovas et al. used HNO3 (above 80%) or H₂SO₄ as a leaching agent. The leaching rates of REEs with the addition of 3 mol/L HNO3 and 0.5 mol/L H2SO4 were 63% and 46%-58%, respectively. It can be seen that nitric acid is the best leaching agent. When the reaction time was increased from 2 h to 8 h, the leaching efficiency and the impurity release rate of the two leaching solutions increased by 8% and 6%, respectively. Adding the chelating agent DTPA (diethylenetriaminepentaacetic acid) can increase the leaching rate of REEs from 13% to 22% (Canovas et al., 2019). In addition, Hammas-Nasri et al. leached REEs in PG with dilute sulfuric acid and leached PG samples twice with 10% H₂SO₄ at 60 °C with a liquid-solid ratio of 1.3:1. The mixture was stirred continuously for 1 h-2 h. Then, the solid and liquid were separated for evaporative crystallization. Finally, a sample with about 86% REEs was obtained (Hammas-Nasri et al., 2016).

Although the leaching rate of a strong acid is high, it will increase the acidity of PG and affect the comprehensive utilization of PG. Therefore, researchers also focus on the use of weak acids to leach REEs. With different concentrations of leaching agents, Gasser et al. found that the efficiency of leaching total Ln-Y with the citric acid solution was better than leaching total Ln-Y with the citric acid solution with boric acid or malic acid solution. When the solid mass ratio of citric acid volume to PG was equal to 2.0, about 40.9% of Ln-Y was leached from 1.0 mol/L citric acid solution after a retention time of 15 min at 298.0 K. The ratio of acid volume to solid mass ratio of PG remained unchanged, and at 358 K for 5 min and at 358K for 15 min, after 1.0 mol/L citric acid solution leaching for 1 cycle, a total of 53.3% Ln-Y was leached from PG samples. The results showed that the maximum leaching rate was 83.4% after three leaching cycles. Analysis of different Ln-Y in the final results showed that the 1.0 mol/L citric acid solution had a higher leaching selectivity of 89.4% for Er, 88.2% for Ce and 81.8% for La, and a lower leaching selectivity of 71.9% and 40.7% for Pr and Y (Gasser et al., 2019).

Although the leaching rate of REEs using the strong acid leaching method is relatively high, the strong acid can dissolve toxic substances and pollutants, which is unfavourable to the environment. Bioleaching is to redox the target through the redox characteristics of microorganisms in life activities, thus effectively extracting the desired substances. Biological extraction of REEs in PG can solve the adverse effects of traditional leaching methods on the environment. Plant extraction has the advantages of fast growth, large biomass, high pollutant concentration in harvestable parts, strong pollution tolerance, and good adaptation of plants to cultivable environments (Ali et al., 2013). Jalali et al. found that bacteria-plant coupling increased the concentration of REEs in sunflower antenna parts. Compared with abiotic controls, Ce, La, Nd, and Y concentrations increased by 4.4, 38.3, 3.4, and 21 times, respectively (Jalali et al., 2020). Antonick et al. used phosphoric acid, sulfuric acid, commercial gluconic acid, and a bioleaching agent (the main component is gluconic acid) to leach PG artificially doped with REEs. Then, concentrations of cerium, neodymium, samarium, erbium, and ytterbium were studied. For all elements except yttrium, sulfuric acid was found to leach REEs the most and phosphoric acid the least. Compared with gluconic acid, the effect of bioleaching agents consistently outperformed that of gluconic acid (Antonick et al., 2019). The biological method is widely used with the advantages of simple process, low energy consumption, and environmental protection. The application of biological methods to extract REEs from PG is still in the development stage and has broad prospects.

In addition to conventional chemical and biological methods, other efficient REE extraction methods include ion exchange and high gradient magnetic separation. Researchers attempted the leaching of REEs from PG, where mechanical grinding, ultrasonic impact, and resin ion exchange processes were used to improve the leaching rate (Rychkov et al., 2018). The specific process is as follows. PG was allowed to react with sulfuric acid at concentrations of 5 g/L to 30 g/L to produce a suspension, which was transferred into a dissolver for a 2-h grinding. After leaching,

the mixture was filtered. Then, 40 cm³ of sulfuric acid cation exchange resin was added to the filtered leaching solution under ultrasonic impact for 2 h. After that, the resin was separated, and the suspension was filtered. Next, a 400 g/L sodium nitrate solution was added as the eluent, and the REE carbonate enrichment was precipitated from the eluent through the ammonium carbonate solution. An REE content of about 50% (equivalent to 97%–99% of Ln_2O_3) could be achieved. The content of PG produced worldwide varies, and researchers must select appropriate extraction schemes according to the actual situation to maximise the REE extraction rate.

REEs are important strategic resources. Under conditions such as decelerated mining and export restrictions, REEs can be extracted from PG, which increases their production and the PG reuse rate and promotes environmental protection and sustainable development.

5. Application of PG in road engineering

5.1. Soil stabilisation and amendment

The engineering properties of soils, the main material for road base construction, are crucial. In road foundations, sand and gravel soils make good fillers due to their excellent properties when compacted. Although certain soils (e.g., chalk and special soils) are not suitable for direct use as road base fillers, they can be stabilised in advance for improved strength and water sensitivity. Lime or cement stabilisations are common soil stabilisation methods (Kumar et al., 2015), which also have disadvantages such as high water-to-ash ratio and high cost. Therefore, when strengthening soft soil roadbeds, their chemical composition must be analysed first. Adding PG can reduce the amount of cement used while satisfying the same strength requirements, thus reducing construction costs. Degirmenci et al. (2007) found that the unconfined compressive strength of unstabilised soils was lower than that of stabilised soils. Cement with PG was significantly more effective than fly ash with PG for mixing modified soils. Shen et al. prepared a new lime-fly ash-PG binder for semi-rigid road subgrades, the optimum formulation of which was 8-12% modified lime, 18-23% PG, and 65-74% fly ash. Soils stabilised with the new lime-fly ash-PG binder had higher early strength than soils stabilised with lime-fly ash, lime, or cement, and granular soils stabilised with the new binder have higher long-term strength than typical road subgrade materials (Shen et al., 2007). Mineralogical analysis revealed that the chemical reaction between PG and the cementitious material (calcium aluminate hydrate) forms calcium alumina. In the meantime, the increased contents of calcium and PG positively affect the strength development of cement stabilised soils at full cementation after 28 days. The produced calcium alumina tends to swell, increasing the density and strength of the structure. However, excessive PG may increase the volume and cause structural damage and strength loss.

Treating waste with waste is an ideal way of recycling that increases waste reuse, protects the environment and conserves resources. Treated waste can be used to improve the soil for road construction. Dredging is conducted annually throughout China to maintain good drainage conditions of water systems and improve water quality. However, dredged silt is a soft soil with low compressive strength. The effects of PG on the unconfined compressive strength and early strength of cement stabilised dredged soils were investigated experimentally by Zeng et al. The experimental results showed that the unconfined compressive strength of cement-stabilised soils with PG contents of 1.4%-8.6% was about 31.7 to 9.4 times higher than that of cement-stabilised soils without PG. In particular, the early strengths of PG-doped stabilised soils were 1.3 and 2.1 times higher after curing for 3 and 7 days, respectively, compared to those without PG (Zeng et al., 2021). Microscopic analysis revealed that PG accelerated the volcanic ash reaction between lime and fly ash, forming AFt that fills the small pores in the binder. Therefore, road base materials with lime-fly ash-PG binder had excellent performance. Gu et al. (2021) found that PG, slag, and cement with a weight ratio of 6:3:1 mixed with 5% micro silica fume (MSF) and 3‰ hydroxypropyl

methylcellulose (HPMC) showed the best overall performance with a compressive strength of 28.8 MPa at 60 days and a softening coefficient of 0.9. At the optimum mix ratio, the weight of PG and slag was approximately 90% of the total, indicating that this waste recycling method can significantly increase the reuse and consumption of solid waste.

Expansive soils swell and soften with water and shrink with water loss. With poor scour resistance, expansive soils are unsuitable for direct use in highway engineering. Therefore, Oumnih et al. Oumnih et al. (2019) stabilised expansive soils with a mixture of lime and gypsum, concluding that the plasticity and swelling of the stabilised soils were significantly reduced. After cost analysis, the most economical combination was 8% lime and 8% gypsum. The reduced plasticity index indicated improved water stability of the soils and subsequently improved water resistance of the roadbed soils. Kumar et al. (2015) found that the dry bulk weight and optimum moisture content increased in the bentonite containing 8% PG and 8% lime. The increase in dry bulk weight indicated a lower water content and increased water stability of the stabilised soils.

As mentioned above, PG is mainly mixed with diatomaceous ash or cement to improve the strength and water stability of subgrade soils. Although cement-PG mixes provide better stability to the ground, there are disadvantages. Mousa et al. (Mousa and Hanna, 2013) found that impurities in PG can enhance its retarding effect on cement and reduce the early cement strength development rate mainly because they lowered the pH of the mixture and slowed down the reaction of the volcanic ash. Gu et al. (2021) revealed that adding 5% micro silica powder reduced the retarding effect of PG effectively and increased the 3-day compressive strength by 170%. In addition to the 3‰ hydroxypropyl, adding methyl cellulose also retarded the hydration reaction. Therefore, adding accelerators to the mix can improve the performance of stabilised soils and overcome their shortcomings, making their engineering properties more suitable for subgrade construction.

5.2. Applications in modified asphalt and asphalt mixes

Asphalt is a common pavement material to protect highway structures such as road foundations. PG is often used in asphalt modification or applied to asphalt mixtures. In asphalt modification, sulphuric acid mixed with PG was used to modify asphalt, and the results indicated that adding 10 wt.% PG activated with 0.5 wt.% sulphuric acid significantly improved the rheological response of the resulting asphalt at high temperatures (Cuadri et al., 2014). In addition, the modification was less effective when common gypsum of the same concentration was used instead of PG. FTIR tests show no C–O–P band in specimens without sulphuric acid, as shown in Figure 6, confirming that sulphuric acid is the "bonding" promoter.

Cuadri et al. also used PG in foam asphalt as a foaming agent. With a long storage time, no heating requirement, and good rolling shaping performance, foam asphalt makes an excellent road material. After mixing the molten base asphalt with 10 wt% of the foaming agent, a 0.5 wt% sulphuric acid was added into the mixture to start the foaming. The foaming process starting upon adding the sulphuric acid confirmed that sulphuric acid could be a 'facilitator' for releasing phosphorus and structural water from PG (Cuadri et al., 2021).

Despite their high strength and good toughness, wear resistance, and temperature resistance, the applications of calcium sulphate whiskers (CSWs) are limited other than serving as the reinforcing and toughening agents for materials such as rubber. High-quality anhydrous CSWs were produced in an autoclave-free hydrothermal system prepared with a 20 wt% H₂SO₄ solution, a 1 to 5 wt% maleic acid, and a PG to solution ratio of 1:60 (Tan et al., 2017). CSWs can be used in highway engineering, such as asphalt modification, to improve the performance of modified asphalt, thus greatly increasing the utilisation of PG and CSWs and promoting solid waste reuse. The effects of CSW types and contents on the performance of two asphalt binders were investigated, which concluded

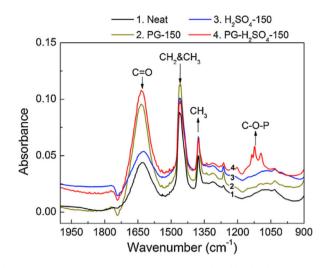


Figure 6. FTIR spectra of pure bitumen with a modified binder from 2000 cm^{-1} to 900 cm^{-1} (Cuadri et al., 2014).

that the CSW contents of 7%-11% improved the properties of all CSW-modified asphalts in terms of high-temperature performance and deformation resistance (Fan et al., 2019). Guan et al. used calcium sulphate whisker fibres (CSWF) to reinforce asphalt mixtures and evaluated their performance with Marshall tests, wheelbase tests, low temperature bending Marshall tests, wheelbase tests, low temperature bending tests, water sensitivity tests, and fatigue tests. The test results showed that CSWF improved the high-temperature stability, low-temperature crack resistance, and water stability of the asphalt mixes. The asphalt mixes showed the best performance at the CSWF content of 0.4 wt% (Guan et al., 2019). Wang and Fan optimised the process parameters of CSW-modified asphalt by determining the optimum mixing time, mixing temperature, and developing temperature. The experimental results identified the optimum process parameters as stirring time of 32 min, stirring temperature of 175 °C and developing temperature of 175 °C. In addition, the effects of the three process parameters on the properties of the bitumen were also investigated, which revealed that mixing time had more significant effects on the softening point, the needle penetration at 25 °C and the ductility at 10 °C than the other two factors (Wang and Fan, 2021).

In addition to modified bitumen, PG can also be applied to asphalt concrete (Foxworthy et al., 1996). applied PG-based slag aggregates to asphalt concrete binder mixes. Based on the freeze-thaw and stability tests results, PG slag aggregates showed good weathering and disintegration properties. Based on the Marshall tests, an optimum asphalt content of 5.7% was determined. In addition, the toxicity of the aggregate was within the range specified by the US National Environment Agency. However, the indirect tensile strength and elasticity modulus of the asphalt concrete with PG slag aggregates were slightly below those of conventional aggregate mixtures. Therefore, the dynamic creep and permanent deformation potential of the modified asphalt concrete are lower than that of conventional asphalt concrete mixtures.

5.3. Application to road base pavement materials

Acquiring conventional road base fillers such as soil and stone could damage the environment. Stabilised and cured PG can be used to replace traditional road materials, thus reducing rock and soil mining and protecting the environment while increasing PG utilisation for sustainable development. A mixture of 46.5% sand, 46.5% PG, and 7% cement was prepared, and its mechanical strength was discussed (Zmemla et al., 2022). The prepared mixture was subjected to conventional compressive and tensile strength tests as well as Young's modulus test, the results of

which indicated that the early and final compressive strengths of the stabilised material were higher than those of conventional road subgrade materials, reaching 2200 and 3500 kPa at 28 and 360 days, respectively.

Fly ash and lime are traditional road materials, and mixing fly ash and lime with PG produces a road material that may have more desirable properties. The suitability of fly ash-lime-PG composites in road pavements was verified using composites of fly ash +8% lime +2% PG, where the unconfined compressive strength, splitting tensile strength, maturation durability, and load-bearing ratio were determined after 28 d of maintenance, concluding that the composites met the criteria for unconfined compressive strength, splitting tensile strength, maturation durability and had a high load-bearing ratio (Rakesh Kumar and Vaibhav, 2016).

Aggregates used as semi-rigid road base materials require good gradation. Therefore, large amounts of sand and gravel were added to meet the gradation requirements. The leaching mass loss of a mixture consisting of 6% PG + 6% lime +12% fly ash +76% crushed stone was 1.8% lower than that of a typical lime-fly ash-crushed stone mixture, in which the compressive strength after wet and dry cycling increased by 57.8%, and the dry shrinkage was significantly lower (Zhang et al., 2020). Moreover, the PG-lime-fly ash-crushed stone mixture had good volume stability and environmental stability.

Composites containing 50% PG were found to have an unconfined compressive strength of 1.08 MPa, and replacing 30% of the lime and fly ash with slag further increased the strength to 3.2 MPa, which met the subgrade foundation strength durability criteria (Mashifana et al., 2018). Researchers have used citric acid to chemically treat the waste PG prior to the experiments, which effectively reduced the radionuclides in PG.

Reflection cracks in asphalt pavements, mainly caused by shrinkage cracks in the subgrade, have become one of the most common pavement diseases. In order to compensate for the shrinkage of the stabilised subgrade, a new slow setting slight-expansive cement was prepared as the road base course binder (RBCB) with an experimentally derived optimum composition of 45% fly ash, 45% silicate cement clinker, and 10% gypsum (Li et al., 2019). The RBCB had a 28-day expansion rate of 4‰, which could compensate for the cement shrinkage and improve the volume stability in wet and dry conditions. The strength of RBCB stabilised aggregates was 15% higher than that of P.S 32.5 stabilised aggregates with the same cement content, thus saving 10-15% of cement. Moreover, RBCB effectively improved the splitting strength of the base material. Compared with ordinary cement stabilised aggregates, RBCB stabilised aggregates had a lower dry shrinkage ratio, thus reducing 82% of reflective cracks in semi-rigid subgrade pavements and extending the service life of the road.

Sludge from wastewater treatment is an extremely complex nonhomogeneous body consisting of organic matter, inorganic particles and colloids. It has high water and organic matter contents and decays easily, producing an awful smell. The rational use of sludge has attracted the interest of many researchers. A composite binder has been prepared by mixing PG, fly ash, and acetylene sludge and applied to rural road construction, where the optimum PG content was experimentally determined as 15%, and the negative effects of soluble phosphate and phosphoric acid disappeared in PG composed of fly ash and acetylene sludge (Qiao et al., 2010). Oil-bearing sludge is a type of mixed oil-earth-water waste. Xiao et al. (2019) used cement, fly ash, and silica fume as binders and PG as the stabiliser to assess the feasibility of oil-bearing sludge as a road base material based on the performance index unconfined compressive strength. Their test results showed that adding 20% binder (cement: fly ash: silica fume = 1:0.7:0.8) and PG to the oil-bearing sludge improved the 28-day compressive strength of the cured specimens. The binder and PG mixture also significantly reduced the leaching rate of heavy metals. Moreover, the PG-stabilised oil-bearing sludge mixture had sufficient strength and good water stability, freeze-thaw resistance, and volumetric stability. PG can also serve as a stabiliser for road base construction and an aggregate for road base fillers. Analysis of the mix indicates that hydration is the main source of filler strength, with the consumption of C3S, C2S, C3A, and C4AF producing hydration products, such as AFt, which are used to solidify particles and fill the pores to improve the overall strength of the filler. The use of organic mixtures improves the overall performance of the road base filler, and the power and water stability are significantly enhanced. Copper tailing sands and steel slag are solid wastes, while oil-bearing sludge contains emulsified petroleum hydrocarbons, heavy metals, and solid particles. The integrated use of multiple wastes is in line with the aim of treating waste with waste and is a practice that protects the environment.

6. Conclusion

This paper summarises the pretreatments of PG, its impact on the environment, and its resourceful usage and focuses on three applications of PG in road engineering. PG in road engineering is a successful way to improve its utilisation. This review presents ideas to address two main issues, namely, improving PG utilisation and protecting the environment. Before applying PG to highway engineering, a safety analysis must be conducted. The focus of further research should be the impurity treatment methods and safety analysis of PG. The application of PG in highway engineering is still at the preliminary stage, where most studies have only analysed the effect of PG on the performance of road bases or pavement materials. However, few studies have focused on its durability or analysed its long-term effects on soil and water resources. Therefore, more relevant studies are required to promote further applications of PG in highway engineering.

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