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journal homepage: www.elsevier.com/locate/jmrt**Review Article****Recent research in utilization of phosphogypsum as building materials: Review****G. Murali ^{a,*}, Marc Azab ^b**^a Division of Research & Innovation, Uttarakhand University, Dehradun, 248007, Uttarakhand, India^b College of Engineering and Technology, American University of the Middle East, Egaila, 54200, Kuwait**ARTICLE INFO****Article history:**

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ABSTRACT

The phosphate industry produces a hazardous byproduct called phosphogypsum (PG). A rising global stockpiling of PG poses severe threats to public health and the environment. Fortunately, recycling this material may be a risk-free and environmentally beneficial answer to this problem. It is necessary to examine its current application and future growth prospects in civil engineering to increase the PG utilization rate. This paper summarizes previous research on reusing PG in building materials. This review article is divided into four essential sections. Firstly, the chemical properties of PG and their treatment methods before using PG in any application are reviewed extensively. Secondly, the strength properties of bricks made with PG and their microstructural characteristics through scanning electron microscope (SEM) and X-ray diffraction (XRD) were reviewed. The third part of this article is about using PG as a substitution for cement and studies the setting time, compressive strength, SEM and XRD of cement mortar. The final section is PG-based concrete and studies concrete specimens' workability, compressive strength, SEM and XRD. The last three sections extensively focused on the microstructural characteristics of the building materials. Following a comprehensive literature search, it was determined that PG could be utilized in the construction industry. Several studies have been performed on using PG in building materials such as bricks, concrete and other materials. However, PG utilization in large-scale practical applications needs better social and political awareness. More money is needed for research and development to free the economy and technology from their shackles. The findings of this review will serve as a foundation for further research and practical applications of PG in environmentally friendly processes.

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1. Introduction

Phosphogypsum (PG) is a byproduct of the wet manufacturing process for phosphoric acid and is considered an industrial

waste material. Around 5 tonnes of PG are generated for every 1 tonne of phosphoric acid production [1]. As the phosphorus chemical sector has expanded rapidly in countries like the Middle East, Africa, Russia, the United States, and China, more

* Corresponding author.

E-mail address: murali_220984@yahoo.co.in (G. Murali).

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PG solid waste has been dumped into the environment [2,3]. Between 200 and 300 million tonnes of PG are produced annually worldwide, with about 5.6 billion tonnes already stockpiled. About 85% of all PG is often dumped into massive mounds without being processed, exposing it to occupying a large area of land, weathering processes, causing significant harm to the environment (pollution with chemicals and radiation) (Fig. 1), and particularly along the seaside [2]. About 15% of the phosphogypsum produced is used directly, mostly in fertilizers, soil stabilization measures, and raw materials for the construction industry [2,4]. As a result, several countries have investigated various PG disposal options in light of the growing community outrage over the issue. For example, Japan, which has a severe gypsum shortage, uses PG primarily for gypsum construction products. PG manufactures gypsum board, blocks, brick, and cement in India. Stockpiling is widely practiced in developed nations like the United States and Britain due to the widespread belief that PG will be in high demand soon [5–7]. Storing PG for an extended period can pollute the soil, underground water after rain and surface water, and groundwater when it rains. Also, the PG stockpile field has a dust problem that needs to be addressed to benefit the environment. Additionally, PG's steam release into the atmosphere may be hazardous since it carries potentially dangerous pollutants like radioactive materials, cadmium, mercury, and phosphorus.

PG is a thin powdery solid that can be a range of colors but is most commonly one of three shades of dark grey, light greyish-white, and yellowish-white. $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ makes up over 90% of PG, making it its primary component. As a consequence of the environmental damage caused by PG storage, the question of how to deal with PG has become increasingly pressing, even in more industrialized countries is receiving more attention from the public and the government. Presently, it is common knowledge that one of the most efficient and difficult-to-implement procedures is the resourceful exploitation of PG. Notably, despite civil engineering's promising use of PG, there is an immediate and critical need for the safe treatment and extensive recycling of PG to increase resource usage [9–13]. As a result, this work highlights the primary strategies for PG resource usage (Fig. 2a). The potential of PG as a versatile raw material has recently attracted considerable attention. As a replacement for natural gypsum in the role of setting regulator, PG has been utilized in the

cement sector [14]. The cement and gypsum industries have substituted PG for natural gypsum in setting regulator applications. In addition to its use as a fly ash-lime reaction activator in producing various construction materials, PG has found utility as an agricultural fertilizer or a supplement for soil stabilization [15,16]. Zhang et al. [17] reported that the drainage pipe industry benefits greatly from using PG polymer composite material. In order to get a high concentration of PG in the polymer, it must be organically modified, but even after that, a dosage of 70% PG has good dispersity and compatibility. For the artificial aggregate to have a high PG utilization rate and enough strength, it was suggested that 80% PG, 16% slag, and 4% clinker be used in its preparation. Artificial aggregates based on PG can be used as a partial replacement for natural aggregate in asphalt mixtures and for pavement base layers. After investigating the pavement layers of fly ash (FA) stabilized with PG and lime, experimental studies found that the mixture of FA + 8% lime + 2% PG is suitable for various geotechnical uses, including backfilling and building rail and road embankments [18]. According to Gregory, Saylak, and Ledbetter [19], a solidified mixture of FA, PG, and cement could stabilize a road base. The strength development of stabilized mixes is significantly influenced by the amount of stabilizer used, the dry mix density, and the pH of the phosphogypsum.

Valancius et al. [26] reported that the paint with a lower TiO_2 content and better quality characteristics than the control sample was developed by incorporating different amounts of paint ingredients and PG into the samples. These improvements included resistance to wet scrub, film formation rate, and greater whiteness. Additionally, it can be used in the creation of PG-based eco-friendly paint. Interior building partitions are low-cost and energy-efficient construction materials that can benefit from PG dihydrate's binding characteristics. Sand, quicklime, and PG were used in the production of internal walls by Zolotukhin et al. [27]. The results of the experiments showed that it was possible to create a mixture that could yield building blocks with a softening factor of 0.5–0.9 and a resistance of 5–20 MPa. Natural plaster and PG were used to develop an experimental home's construction [21], as shown in Fig. 2b. The research focused on improving the PG plate production process with a compressive strength of up to 60 MPa [21]. It is possible to use hollow blocks made of PG instead of hollow bricks made of traditional hollow concrete blocks or burnt clay, which is a viable



Fig. 1 – Location for storing PG, for example, (a) Mianzhu and (b) Sichuan [8].



Fig. 2 – (a) PG resource usage in the construction industry [20], (b) Plate [21], (c) Hollow block [22], (d) Wall [23,24], (e) Brick construction [25] and (f) ceramic blocks [22].

alternative. Fig. 2c demonstrates the hollow blocks that may be produced with PG. Because these blocks fulfill the durability, resistance, and thermal insulation specifications, they are suitable for the building's construction. According to Kumar (2003) [28], hollow blocks can potentially substitute hollow concrete blocks, traditional eggs, and clay bricks. Other authors (Zhang et al. [23,24]) have applied the molten PG as a new type of wall (Fig. 2d). Integrating the structure for enhanced mechanical properties and insulating it for increased thermal insulation are two of this in-situ-built wall's most notable qualities. Fired bricks made from a 25% PG, 10% sand, and 65% clay mixture were developed by Ajam et al. [25]. As may be seen in Fig. 2e, in Tunisia, rooms measuring $4 \times 4 \times 3 \text{ m}^3$ were produced using the mixing mentioned above combinations. Radon levels were tracked in the same room over two years and two seasons. The findings demonstrated that weather conditions significantly affect radon levels. The mechanical and physical properties of bricks made with PG were satisfactory. Radon levels in the PG brick room were significantly lower than the recommended 150 Bq/ m^3 maximum. Ceramic blocks, comprised of clay, cement, and PG, are yet another potential application of PG in construction (Fig. 2f). Although Godinho-Castro et al. [29] (2012)

found that substituting 10% PG for cement could yield excellent compressive and tensile strengths, replacements above this level were found to reduce these properties. Hence, the review is useful in encouraging the efficient use of PG in engineering. It helps the phosphorus chemical and construction industries produce cleaner products while saving energy and reducing pollution.

2. Physical-chemical properties and treatment methods of PG

2.1. Chemical composition

Table 1 displays the PG's chemical compositions employed by various researchers. There is some variation in the chemical composition of PG from study to study. PG primarily contains CaO and SO₃, about 65% of the overall mass. Additionally, PG includes organic materials, sulphides, Al, Mg and Fe₂O₃ [30]. When PG is recycled, contaminants in the PG degrade the quality of the final product. A study by Singh et al. [31] indicated that selenite gypsum's strength development and setting time were negatively impacted by more than 1.0%

Table 1 – PG's chemical composition.

Constituent (%)	Na ₂ O	MgO	Al ₂ O ₃	SiO ₂	P ₂ O ₅	SO ₃	K ₂ O	CaO	Fe ₂ O ₃	F ⁻	LOI	Others
[33]	0.07	0.12	0.35	1.37	2.26	56.68	0.12	38.39	0.45	—	—	—
[35]	—	—	0.29	8.82	1.72	34.51	—	32.14	0.35	0.8	—	31.37
[36]	0.51	—	0.43	1.25	3.50	42.19	—	29.05	0.21	—	19.48	—
[37]	—	0.26	4.38	4.86	3.57	30.95	0.41	31.5	—	—	—	—
[38]	0.02	0.03	0.23	3.66	0.45	41.94	0.02	32.31	0.2	—	21.14	—
[39]	—	0.15	0.16	2.95	1.30	49.7	0.043	38.93	0.057	—	—	—
[40]	—	—	0.08	1.89	0.44	49.89	—	36.48	—	—	—	11.22
[41]	—	0.17	0.08	3.62	1.05	42.84	—	31.57	0.05	—	19.88	—
[42]	—	0.5	0.62	6.4	0.7	41.7	—	27.8	0.3	0.8	20.5	—
[43]	—	—	0.24	4.35	2.33	51.11	—	34.35	0.06	0.5	—	7.06
[44]	—	—	0.24	3.74	0.74	46.83	—	39.72	0.3	0.63	7.73	—
[45]	0.05	—	0.38	1.80	1.30	54.94	0.12	41.04	0.27	—	—	—
[46]	—	—	0.11	1.37	1.69	44.40	—	32.80	0.03	0.55	—	19.05
[47,48]	—	0.06	1.09	3.21	1.10	47.3	—	34.52	0.31	0.20	11.01	—
[49]	—	—	0.75	10.40	0.27	38.81	—	26.74	0.22	1.17	—	21.64
[50]	0.50	0.21	0.96	8.34	2.01	39.4	0.15	49.53	3.29	—	4.21	—
[51]	—	0.28	0.46	4.20	1.37	44.82	—	37.86	0.45	—	—	—
[52]	—	—	0.88	3.44	0.50	44.67	—	32.04	0.32	0.79	—	17.36
[34]	0.12	0.29	0.35	2.3	1.05	42.1	0.17	31.64	0.27	0.2	21.19	—
[53]	—	—	0.15	3.51	2.28	44.50	0.07	46.70	2.23	—	—	—
[54]	0.29	0.13	0.40	7.07	1.83	43.12	0.34	29.63	0.30	1.25	—	15.67
[55]	0.21	—	0.85	1.03	0.96	44.01	0.001	31.16	0.012	0.64	21.1	—
[56]	0.17	0.06	1.09	7.50	0.95	37.75	0.60	27.05	0.44	1.83	20.77	—
[57]	—	0.04	0.13	0.37	0.81	53.48	—	38.60	0.03	0.14	6.4	—
[58]	—	0.134	0.054	0.18	—	61.16	—	38.16	0.022	—	—	0.022
[17]	—	0.27	0.16	1.44	0.71	38.30	0.13	57.90	0.78	—	—	—
[59]	5.88	1.95	0.66	3.55	3.39	48.45	0.18	34.96	0.47	—	—	—
[60]	0.17	0.06	1.09	7.5	0.948	37.75	0.6	27.05	0.44	1.83	20.77	—
[61]	0.08	0.246	0.757	3.91	1.05	50.05	0.142	42.26	0.878	0.258	—	—
[62]	0.10	0.33	1.37	7.84	2.47	33.93	1.25	24.01	1.21	5.03	—	>22.13
[63]	0.25	0.15	0.35	9.47	0.83	50.26	0.09	37.84	0.10	0.48	0.11	—
[64]	—	0.15	0.16	2.95	1.30	49.7	0.043	38.93	0.057	—	—	>0.004
[65]	—	0.04	0.13	0.37	0.82	55.48	—	38.60	0.03	0.14	6.40	—
[66]	0.11	0.02	0.13	1.74	1.04	52.94	—	38.87	0.16	0.32	3.97	—
[67]	—	1.36	0.39	1.09	0.01	44.30	—	29.09	0.40	—	22.67	—
[26]	0.14	—	0.18	0.2	1.20	45.91	0.08	—	0.1	0.24	19.55	—

soluble phosphate additions. PG can weaken the gypsum's compressive strength due to the organic materials and fluoride. According to research by Min et al. [32], the gelling time of materials can be increased by the presence of soluble phosphorus and organic substances in PG. Additionally, the functionality of gypsum hemihydrate and gypsum dihydrate is negatively impacted by these contaminants in PG. Up to 56.68% SO₃ can be found in PG from Guiyang (Li et al. [33]), but only 42.10% SO₃ is found in PG from Hubei (Huang et al. [34]). Table 1 shows that approximately half of the tested PG samples lacked sodium oxide and potassium oxide. In addition, Liu et al. (2019a) found that raw PG and calcined PG were not too dissimilar chemically, containing mostly SiO₂, CaO, SO₃, and a small quantity of P₂O₅, Al₂O₃, and other components.

Additionally, after 500 °C calcination pre-treatment, the amounts of P₂O₅, SiO₂, and CaO decreased while the amounts of SO₃ increased. However, raw and calcined PG had drastically different mineral compositions; CaSO₄·2H₂O was the predominant mineral in the former, while CaSO₄·0.5H₂O was the predominant mineral in the latter. In addition, the composition of the PG and the amount of water absorbed varied depending on its source. Depending on the manufacturing method, PG may have trace amounts of P₂O₅ and F⁻ etc. The fundamental reason for the low utilization rate of PG was the differences in physical and chemical qualities between PG and natural gypsum, which are caused by the presence of impurities. The fluorine and phosphorus volatiles created by organic matter were eliminated during the calcination process, along with any soluble contaminants in PG. Consequently, the ignition losses of various PGs are highly variable. According to Ajam et al. [46], CaSO₄ accounts for around 77% of the PG's composition. Adding this lime, which breaks down into micropores around 830 °C, is expected to improve thermal conductivity while increasing porosity and decreasing mechanical strength. In addition, using calcium sulphate enables less energy to be used overall in the brick-making process. However, it may result in a more fluorescence result. A methodical crushing could eliminate this residual effect. There is a negligible amount of the remaining ingredients. Because of its low silica content (1.37 wt %), this PG could reduce the heat required to make bricks. It is recommended that PG undergoes pre-treatment before reusing to avoid further damage to PG-based products.

2.2. Treatment of PG

2.2.1. Sieve technique

The sieve technique is valid only if the contaminants are widely dispersed. A significant amount of contamination can be removed using sieving. Wet sieving with a 300-μm aperture sieve is the first step in the wet sieve cyclone treatment procedure, in which the PG is processed to eliminate the coarser, more impurity-laden fraction (10–15%) before being further refined in a hydrocyclone [8]. Sodium oxide, silicon, and fluorine chemicals were discovered to be highly concentrated in particle size fractions larger than 170 mm in an analysis of a Moroccan phosphogypsum [68]. Particles smaller than 25 μm had a high concentration of organic compounds and phosphate in a crystalline lattice form. Clean gypsum recovery with this approach is estimated at 75% [68]. This approach can

efficiently eliminate soluble contaminants in PG, including fluorine, phosphorus, and other organic substances. The treatment procedure that utilizes a wet sieve cyclone achieves unprecedented purity levels. Nevertheless, its method is somewhat complicated, and secondary pollution may occur while treatment is being carried out [69].

2.2.2. Neutralizing agent

PG can solidify and stabilize harmful and toxic substances by creating, adsorbing, and encapsulating insoluble substances. Because PG has a pH of 3, acid-base neutralization reactions are required to complete the purification process successfully [70]. Slag composed of calcium carbide is an example of an alkaline industrial waste material that may be used as an alkali-neutralizing agent for PG. According to Wu et al.'s findings [71], adding calcium carbide slag of 0.5%, which served as an alkaline conditioning agent, to a mixture consisting of CaCl₂, polymeric aluminium chloride of 0.3%, and silicate cement of 0.4% resulted in a considerable curative impact on F and P. This provided a solid basis for PG-based eco-remediation products and filler by ensuring that their leaching toxicity complied with the specifications of the applicable standard. Alkali neutralizing PG using calcium carbide slag provides many benefits, including cheap additional expenses, a straightforward method, and a high degree of practicality. On the other hand, research showed that the calcium carbide slag's reaction rate was significantly faster than lime [72].

2.2.3. Lime purification

Lime purification of PG is cost-effective since it may be used on a wide scale and does not necessitate expensive ingredients, calcination, and water. Ca(OH)₂ based chemical treatment decreased PG impurity levels of fluoride by 25.0% and water-soluble phosphorus by 25% [4]. Both chemical and physical treatments should be used in conjunction with one another rather than separately. Using multiple approaches at once could lead to better outcomes. Wang et al. [73] could eliminate the fluorine and soluble phosphorus by mixing lime into the raw PG and then flotation. Positive flotation was employed to separate the gypsum from the PG, while reverse flotation was utilized to separate the vast quantities of fine mud and organic debris. The results revealed that the percentage of whiteness in PGs increased from 31.5 to 58.4. Additionally, decreasing (P₂O₅) concentration from 1.78 to 0.89, 96.6% of calcium sulphate dihydrate and 74.1% of concentrate were recovered. PG was washed in 0.5% milk of lime at 20 °C for 5 min (PG: milk ratio of 14%), neutralized, and then dried at 80 °C by Mun et al. [74]. An important role as a binder to granulated blast-furnace slag was also attributed to the PG that had been pre-treated for this investigation.

2.2.4. Washing

Solvable contaminants, such as fluorine and soluble phosphorus, might be successfully removed via dehydration, filtering, and washing. Although the results were clean, the washing process used a significant amount of water and energy, and the secondary contamination produced by the newly created effluent was severe. The cost of releasing the wastewater was also significantly raised because it had to be

treated separately [75]. As a result, this strategy has not been widespread implementation yet. Liu et al. [76] washed the PG with ionized water to remove contaminants and improve PG's filling capabilities in cement slurry backfill. PG-based cemented paste backfill's workability and mechanical qualities can be vastly improved by first subjecting it to a water wash, which will eliminate many solubility contaminants from the surfaces of the PG crystals. Leaching 226Ra from PG was accomplished by Mohammad Salem Al-Hwaiti [77] using a combination of water, calcium carbonate powder, mixed acids and sulfuric acid, and the percentage of 226Ra removed were 84.37, 86.55, 88.5, and 88.78, respectively. High efficiency in PG purifying can be attained with the combination of sulfuric acid leaching and preparatory water washing [78]. Experimental results from a study by Ennaciri et al. [79] on the use of sulfuric acid leaching in conjunction with a preliminary water washing showed that the acid concentration of 5% and the PG/H₂SO₄ ratio of 1/3 were optimal for removing P₂O₅ and F at 60 °C, with removal rates of 60 and 95%, respectively. They also stated that the distilled water washing of PG is an effective method for removing most soluble contaminants, including traces of acids (H₃PO₄, H₂SO₄, HF, and H₂SiF₆). Particle sizes less than 200 mm at 40 and 60 °C have P₂O₅ and F contents of 10 and 20% in purified PG by sulfuric acid. It is essential to consider that the phosphates and fluoride impurities decrease as the concentration of sulfuric acid, the temperature, and the time spent in contact with the substance rise [79]. When the washed PG is put through the purification process, the maximum removal rate of these contaminants is 53% for P₂O₅ and 82% for F, respectively. For granule sizes less than 200 mm, 98.5% of P₂O₅ and 97% of F are removed from washed PG to produce pure PG. Kandil et al. [80] purified Phosphogypsum with H₂SO₄ to produce an insoluble residue. The resulting filtrate is examined with appropriate analytical techniques to evaluate the removal efficiencies of some impurities, such as lanthanides (15.32%), fluoride ions (99.12%), Fe₂O₃ (80.32%), Al₂O₃ (93.22%) and P₂O₅ (98.67%). Citric acid and ammonium hydroxide have been used to treat PG by Singh et al. [81]. PG waste can be cleaned up by first soaking it in a citric acid solution of 3–4%, then washing it in water. There has been little research on conventional acid leaching of rare earth elements from PG [82–84]. According to the available literature [85,86], nitric acid (HNO₃) results in greater leaching efficiency than sulfuric acid (H₂SO₄). While HNO₃ would require an extra acid circulation system, H₂SO₄ is recommended for financial advantages and is already present in phosphoric acid plants [87,88]. Lutke et al. [89] reported that the leaching efficiency was improved by switching to sulfuric acid from citric acid, increasing the acid concentration and temperature, and decreasing the solid/liquid ratio.

2.2.5. Calcining

P₂O₅ in PG was converted into inert, stable phosphate through a calcination process at 800 °C. During this time, volatilization could eliminate a trace quantity of hydrogen fluoride and organophosphorus. Besides, secondary contamination and fluoride volatilization were also avoided by combining calcination with L neutralization [75]. Consequently, despite the

drawback of high energy use, the benefits of this approach are undeniable. Calcination, in particular, can potentially mitigate the negative impacts of hazardous contaminants. However, the calcination process transformed the PG into hemihydrate gypsum and anhydrous gypsum, the former recyclable and useful [90]. Garg et al. [91] used calcined PG at 150–160 °C for 4 h, one of many research that attempted to remove contaminants in PG using calcination. Egyptian PG (90 mm in size) was calcined at 200, 400, 600, and 800 °C at a rate of 10 °C per minute for 2 h by Taher [92]. According to the findings, 600 °C is the best calcination temperature, followed by 400, 200, and 800 °C. PG was calcined at 800 °C for 4 h by Singh et al. [93], 800 and 900 °C for 30 min by Leskeviciene and Nizeviciene [94], 850 °C for 2 h by Rashad [35], 170, 600, 750, 850 and 950 °C for 3 h by Smadi et al. [4]. The best calcination temperatures were determined to be between 650 and 950 °C. Studies showed that as calcination temperature increased, PG's pH value, density, specific surface area, and specific gravity increased while the ignition loss reduced.

2.2.6. Flotation

Flotation is a method that could be used to purge the water of organic contaminants. At the right concentration, the PG and water were introduced to the flotation machinery; the organic contaminants rose to the top and were removed via scraping. A high organic content in PG was successfully treated using this approach. However, the effectiveness of the treatment was low, and the elimination of soluble contaminants was not readily apparent. It was common to practice combining flotation with washing because the water used could be reused [75,95].

2.2.7. Chemical treatment

Salts, alkalis, and acids were frequently used as modifiers or admixtures in PG to adjust the pH or interact with contaminants to produce a precipitating. Sulfuric acid [75], citric acid [96], and lime [97] are just a few examples of alkaline-modified materials that have been introduced to PG and have reacted with soluble fluorine and phosphorus to generate precipitate. This method might process PG of low organic matter concentration and varying quality successfully. Cement retarder manufacturers typically utilize lime neutralization treatment due to its ease of use, relatively inexpensive, precise results, and low harmful emissions. In any case, this approach only partially resolved the issue of fluorine soluble and phosphorus. Whereas the organic components of PG would be unaffected, the fluorine and soluble phosphorus would dissolve after a considerable time. After being treated with a 3–4% citric acid solution in water, the PG can be washed in water to remove any remaining acid [81]. As water-soluble phosphoric acid, ferrate, hydrofluoroaluminate, hydro-fluorosilicate, hydrofluoric acid, and sodium citrate, impurities of phosphates and fluorides are eliminated [81]. Potgieter et al. [68] showed that treating PG-based cemented paste backfill with sulfuric acid or ammonium hydroxide efficiently reduced set retardation. While the aforementioned pre-treatment methods are effective at removing contaminants in PG, the properties of the resulting mixture will vary depending on the mechanism used.

2.2.8. Ball-milling treatment

The ball-milling treatment is not effective in purifying PG, and it has the potential to enhance the fluidity and strength of PG while also providing a solution to the issues of excessive porous and weak structures that are present in PG-based gelling materials. After conducting experiments, Wu et al. [98] reported that the ball-milling treatment could efficiently reduce PG's particle size, increasing the material's specific surface area. Ball milling is an excellent method for lowering the water needed for a typical consistency. Water usage for milled PG drops to 60% at a minimum after 50 min in a ball mill, 21.5% less than for unmilled PG. Additionally, at 2 h, the compressive strength of PG was improved after being subjected to the ball milling treatment. In preparation for flotation, the raw PG ore was ball-milled and stirred by Du et al. [99]. Reverse flotation was used to eliminate the microfine-particle muck and organic materials. The gypsum concentration was then floated with the help of dodecylamine. PG's soluble phosphorus level dropped from 0.48 to 0.07% during flotation purification, indicating a considerable reduction in impurities, resulting in the percentage of pure PG rising from 73 to 94%. Both prior to and following purification, the PG products had distinct differences in their properties. Compared to the initial ore, the 2-h compressive and flexural strengths of the α -HH that was made from PG improved by 79 and 46%, respectively. However, the purification benefits are more satisfying when the PG is filtered through other physical processes following ball milling since ball milling is unable to remove contaminants directly.

In a nutshell, the benefits of chemical treatment from an economic perspective are cheap cost and high viability. It is unusual for significant disparity in the distribution of contaminants, necessitating the adoption of screening procedures. Typically, PG is cleaned by floating or washing in water. Many contaminants can be eliminated by simply washing them in water. However, it is important to cleanse the effluent after washing in order to avoid subsequent pollution. Although flotation is less efficient than water washing, it is more sustainable because the water may be recycled. However, investing in necessary infrastructure and paying to clean sewage significantly raises the pretreatment price. Furthermore, the infrastructure and energy requirements of the calcining process are quite high. Calcination emerged as the most successful treatment approach after being evaluated

alongside the other options (washing in water, floating and chemical treatment) [20]. Specifically, fluoride and organic phosphate contaminants were effectively removed using calcining. At higher calcination temperatures, PG was transformed into anhydrite and calcium sulphate hemihydrate, which have different solubility and activity characteristics [100,101]. Despite these benefits, PG performance must be enhanced and the negative environmental implications eliminated. New admixtures, lower calcination temperatures, and the activation of inert minerals in PG should be the primary target of future calcination research. The ball milling technique can alter the structure of PG, and it cannot completely nullify the impact of any contaminants. Ball milling is most efficient when used in conjunction with other techniques. Therefore, distinct physical treatment procedures should be chosen based on specific engineering needs.

3. Mechanical and microstructural properties of bricks

3.1. Properties of PG-based bricks

Sustainable bricks made from recycled materials offer a viable solution to the issue of waste management and the rising cost of building supplies. There has been research on using PG in solid bricks, and while the outcomes were promising, the product required a lot of compaction energy to generate bricks. Different research prepared various shapes of bricks using the PG according to the applications (Fig. 3). Bricks with an appropriate compressive strength have been made using a combination of calcined PG at a concentration of 20–40% and FA-lime binders, as reported by Kumar [102]. Kumar produced bricks [103] using calcined PG of 10%, lime of 30%, and fly ash of 60%; then, calcined PG (10 and 20%) was used instead of lime. A larger percentage of calcined PG was shown to increase compressive strength. Zhou et al. [104] reported that the compressive strength improved as the amount of PG in the bricks increased; it topped out when the amount of PG was 75%; after reaching this highest, the compressive strength began to drop (Fig. 4a). The increasing trend of compressive strength was reported while increasing the amount of PG from 65 to 75%. The compressive strength of 21.8 MPa was recorded, corresponding to the 75% PG content. After that, a decreasing

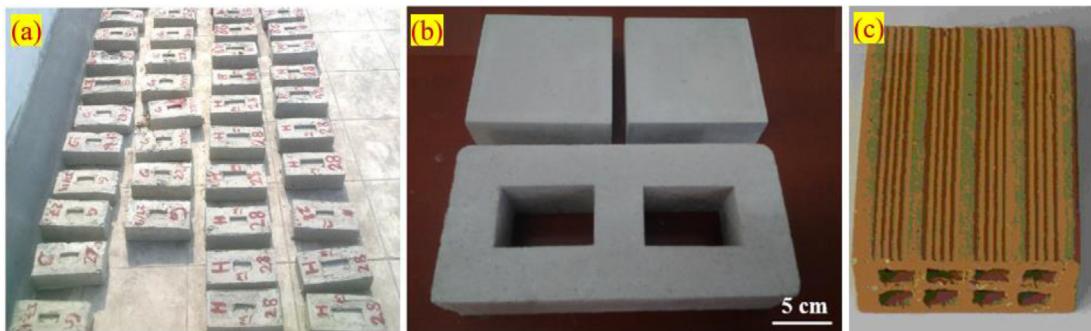


Fig. 3 – Various shapes of brick comprised PG, (a) Dried-out bricks placed in the sun [66], (b) Forms of PG unfired brick, both solid and hollow [105] and (c) Small bricks containing 25% PG [46].

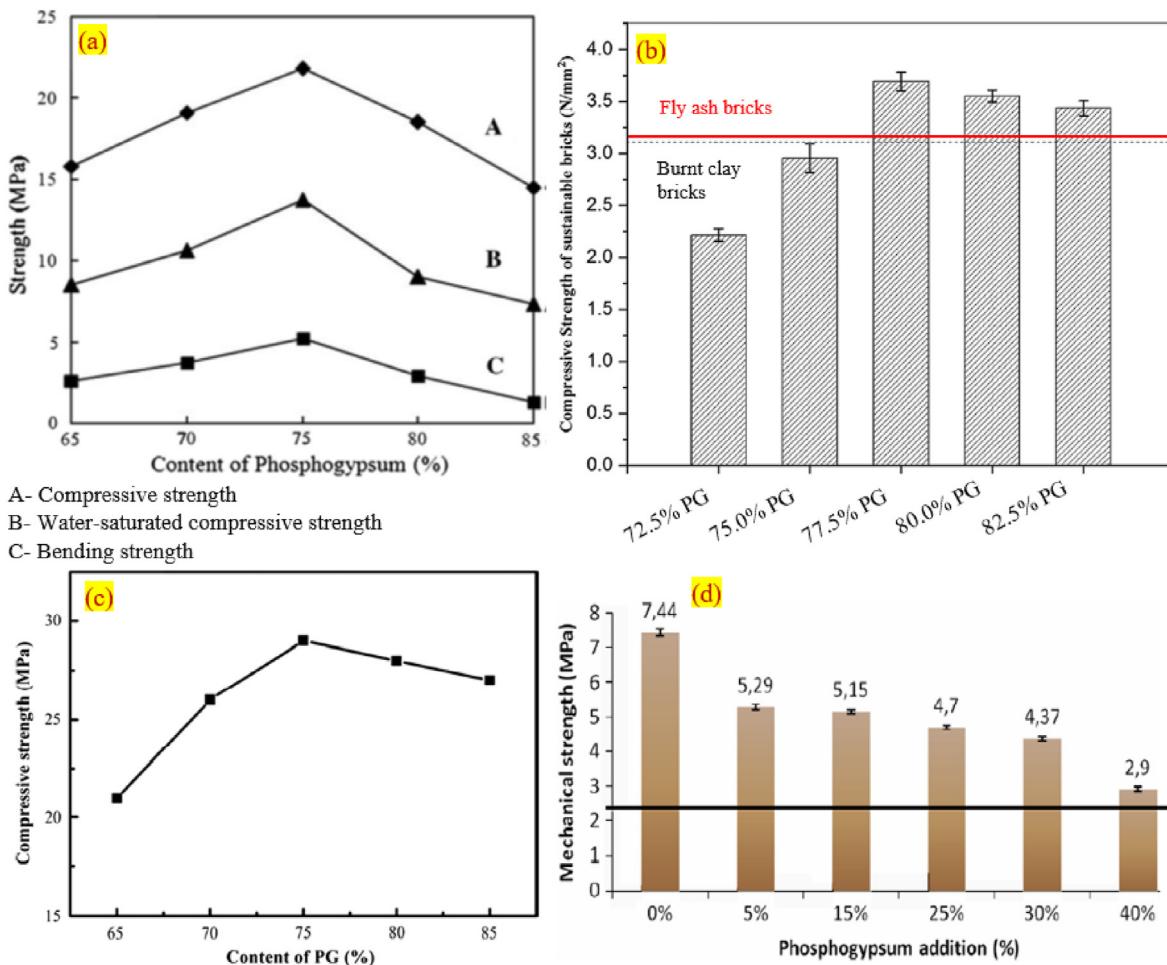


Fig. 4 – Strength of bricks (a) Zhou et al. [104], (b) Raut et al. [66], (c) Zhou et al. [105], (d) Azam et al. [46].

trend was noticed using the amount of PG beyond 75%. This behavior evidenced that the optimum PG content was 75%, leading to the brick's higher compressive strength. Phosphogypsum unfired bricks have high mechanical strength because the gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) crystals recrystallized in situ inside the green brick bodies during the Hydration-Recrystallization process are regular, dense, and interlock crystalline [104]. Conversely, the water-saturated compressive strength also exhibited an increasing strength trend by increasing PG content from 65 to 75%. When PG content is beyond 75% resulting in a sharp decrease in compressive strength, it is shown in Fig. 4a. A compressive strength of 13.7 MPa was measured after being saturated with water, equivalent to a PG concentration of 75%. A 5.2 MPa was noticed regarding the bending strength, corresponding to 75% of PG content. Compared to a bending strength of 75% PG content, the brick comprised 65, 70, 80, and 85% PG content exhibited a lower bending strength. It is essential to highlight that the bending strength of brick with 65% PG content is higher than the 80 and 85% PG content. This behavior indicated that the bending strength is maximum when the PG content is 75%. This exceeded the Chinese standard (JC/T 422–2007) minimum requirements for unfired MU20-grade bricks allowed (20.0, 13.3 and 3.5 MPa). As a result, 75% was decided to be the ideal percentage of PG material for the three

parameters discussed above. Raut et al. [66] developed bricks using PG varied from 72.5 to 82.5%, cement of 15%, and rice husk ash varied from 2.5 to 12.5% and tested. The compressive strength of the brick improves in direct proportion to the amount of PG present in the mixture, that is, up to 77.5% (Fig. 4b), and then it slightly declines beyond that, but the difference detected is relatively minor. Previous research had similar findings [104].

An innovative “Two-step Hydration Process” was presented by Zhou et al. [105] to create a 29 MPa compressive strength of non-fired bricks made with PG, not utilizing any cement and only applying a pressure of 10 MPa during the pressing process. Different combinations of PG (65–85%), lime, and sand content were used. Notably, even at a press forming pressure as low as 10 MPa, the final unfired brick can achieve up to 29 MPa compressive strength after just 7 days (Fig. 4c). The compressive strength of the final unfired bricks is proportional to the press-forming pressure, so increasing the pressure results in a denser green brick and a stronger brick. On the other hand, a higher press-forming pressure typically results in a greater amount of consumed energy and a higher overall manufacturing cost. After being characterized, the grease-removing Tunisian PG was mixed into clay bricks at 0, 5, 15, 25, 30, and 40% were investigated by Azam et al. [46]. After incorporating PG at a rate of 30%, the acquired findings

Table 2 – Summary of the findings regarding the properties of bricks made with PG.

Ref	Proportions	Findings
[107]	Clay is substituted with PG at 5, 10, 15, 20, 25 and 30%.	<ul style="list-style-type: none"> When PG was added at a rate of 30%, the sample density increased to 1.75 g/cm³. Adding PG at a 10% level yielded the highest compressive strength (36.3 MPa). The compressive strength of the bricks was 18.63 MPa after being subjected to 15 cycles at temperatures ranging from 20 to 20 °C, and the weight loss was only 0.029%. The bricks had good durability in humid or cold climates. The mechanical strength of bricks is enhanced when the ideal percentage of PG is incorporated into the brick.
[108]	Additive-1%, Natural sand-29%, FA-15%, lime-15% and autoclaved PG-40%,	
[66]	PG varied from 72.5 to 82.5%, cement of 15% and rice husk ash varied from 2.5 to 12.5%	
[109]	PG content varied from 0 to 40% + Clay additive	<ul style="list-style-type: none"> When more than 30% of the PG is used in the sintering process of bricks at 1000 °C, the external gamma radiation exposure may reach almost 0.4 mSv y⁻¹. PG-based cement binders have been shown to have minimal water absorption and improved compressive strength, making them ideal for usage in the building sector. The bricks' water absorption dropped by 5.1 and 10%, respectively, when 70% and 80% PG were introduced compared to 60% PG. Adding 5–20% of PG increased water absorption by 5–10%. Adding 30% PG led to a reduction in water absorption by 5%. Adding 30% of PG, the bricks could meet all standards. The water absorption coefficient of the bricks was reduced by 26 and 20%, respectively, after 5 and 15% PG were introduced. Once PG was added, the pH level of the bricks fell from 10.5 to 7.5. At 830 °C, the carbonates that served to buffer the acidity vanish, contributing further to the pH drop. The strength decreases steadily up to a 25% PG-mix in bricks, then drops off precipitously at a 40% M-mix. Adding 30% of PG and 30% of FA increased the water absorption of brick by 3.69%. Bricks made with up to 30% PG were found to emit less than one mS of external gamma radiation. Mineralogical investigation reveals that creating a new gehlenite phase at 1000 °C correlates positively with the rise of PG in the brick samples.
[107]	PG-5 to 30% with clay	
[46]	PG-5, 15, 25, 30 and 40% substitute of clay	
[106]	PG-30% + FA-30% + cement 40%	
[109]	PG-0-40%, sintered at 1000 °C after drying.	

demonstrated that the bricks met all specifications. Mini-bricks' mechanical strengths were reduced due to increased PG content, although they were still above industry norms (Fig. 4c). As a precaution against shrinking and considering the scale impact, it has been suggested that 25% PG be used in

place of sand in the brickmaking process. Naik et al. [106] reported that the compressive strength dropped off at 7, 14, and 28 days when PG was combined with fly ash. With PG concentrations between 35 and 45%, the compressive strength declination after 28 days varied between 12 and 49%. Table 2

summarizes the other properties of the bricks made with PG. It is clear from the above discussions and table that the optimum content of PG for manufacturing bricks was different due to the properties of PG and the usage of treated and non-treated PG.

3.2. XRD observation from PG-based brick samples

It can be viewed prudently from Fig. 5a quartz was identified as the primary mineral phase in all brick samples with PG [109]. Results from XRD analysis concluded that as PG levels rise, gehlenite and anhydrite become more common. At the same time, the proportions of mullite, hematite, and quartz in the brick samples reduce as the percentage of PG rises. A few brick samples still had muscovite residue after being heated to 1000 °C. Other brick clays have also been shown to experience the same muscovite phase development [111,112]. When bricks are baked, the mineral phases such as illite and kaolinite that were initially the most abundant in the starting clay are no longer detectable. This phenomenon is caused by the disintegration of their crystalline structure between 450 and 900 °C [113]. The crystallization process causes smaller mullite phases to form at temperatures of 1000 °C. Research also shows mullite can form at temperatures lower than 1100 °C [111,113]. Brick samples with a higher concentration of phosphogypsum have a higher concentration of elements S and Ca and lower concentrations of Fe, K, Si, Al, Mg, and Na. However, clay predominantly CaSO₄ has no special affinity for the disintegration of this mineral at 1000 °C, in contrast to clay containing CaCO₃. In contrast, carbonate clays have the potential to generate larnite (Ca₂SiO₄), wollastonite (CaSiO₃), and other mineral phases [114]. A minor gehlenite (Ca₂Al₂SiO₇) phase has formed, but anhydrous gypsum has mainly maintained its crystalline shape.

The XRD of examined cement pastes containing PG with angular ranges from 5 to 75 2 h is depicted in Fig. 5b. The study [115] found that the hydration products of paste, in which the waste river sludge and hemihydrate PG were the predominant adhesion agents, contained gypsum (CaSO₄·2H₂O), with a highly intense crystal of brushite (dicalcium phospho dihydrate; CaHPO₄·2H₂O). Both waste river sludge and

hemihydrate PG relied heavily on SiO₂ and CaO for their chemical compositions. Samples of cementing paste were found to include numerous non-reacted CaO and SiO₂ (quartz). Further, a few minor peaks in the matrix indicated the presence of ettringite (Group ID 9). It should be noted that the well-formed CSH and ettringite were clearly visible in the tiny peaks of the OPC matrix based on Group ID 16, Al₂O₃, CaO, SiO₂, and CaSO₄ were all hydrated, and their phases formed as a result of a chain reaction.

3.3. SEM from PG-based brick samples

By employing a technique called the Non-immersing Method, semi-hydrated gypsum is formed into the standard bricks used in quality control, and the brick body retains some of the semi-hydrated gypsum [105], which is chemically represented by the formula CaSO₄·0.5H₂O, consisting of a microstructure that is both permeable and disordered (Fig. 6a). As a result, the hydration process binding effect is not entirely released, indicating that the semi-hydrated gypsum is not totally hydrated. This does not improve the brick's 18 MPa compressive strength for seven days. On the contrary, the partially hydrated gypsum (CaSO₄·0.5H₂O) becomes completely hydrated and recrystallizes, becoming dihydrate gypsum (CaSO₄·2H₂O) when the water-immersion procedure is used [105]. In addition, the as-prepared brick specimen also possesses a uniform, compact, and interconnecting microstructure, contributing to its top standard of compressive strength (Fig. 6b). Samples of bricks made with the innovative Hydration-Recrystallization technique and those made without it were examined using SEM, and the outcomes are shown in Fig. 6c–f. The microstructure of green bricks was uneven and porous, and the primary crystalline component was gypsum (Ca-SO₄·2H₂O) (Fig. 6c). The cutting-edge Hydration-Recrystallization technique was used to manufacture the unfired bricks [104]. After being subjected to drying bricks at hot temperatures, the primary crystalline phase of the bricks had transformed into semi-hydrated gypsum (CaSO₄·0.5H₂O), which possessed a significantly less dense and more dispersed crystalline structure (Fig. 6d). Following the immersion of the hot-dried brick in

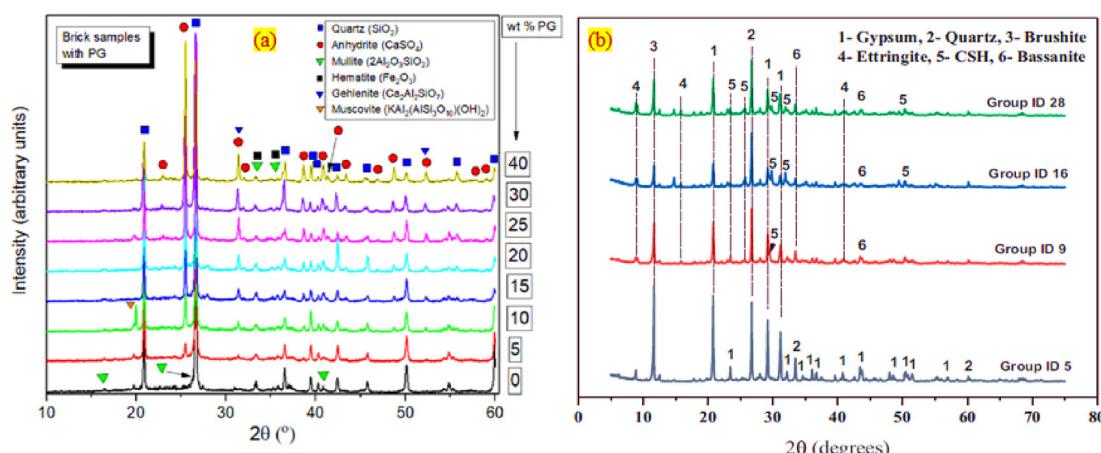


Fig. 5 – XRD analysis from brick sample (a) [109], (b) [115].

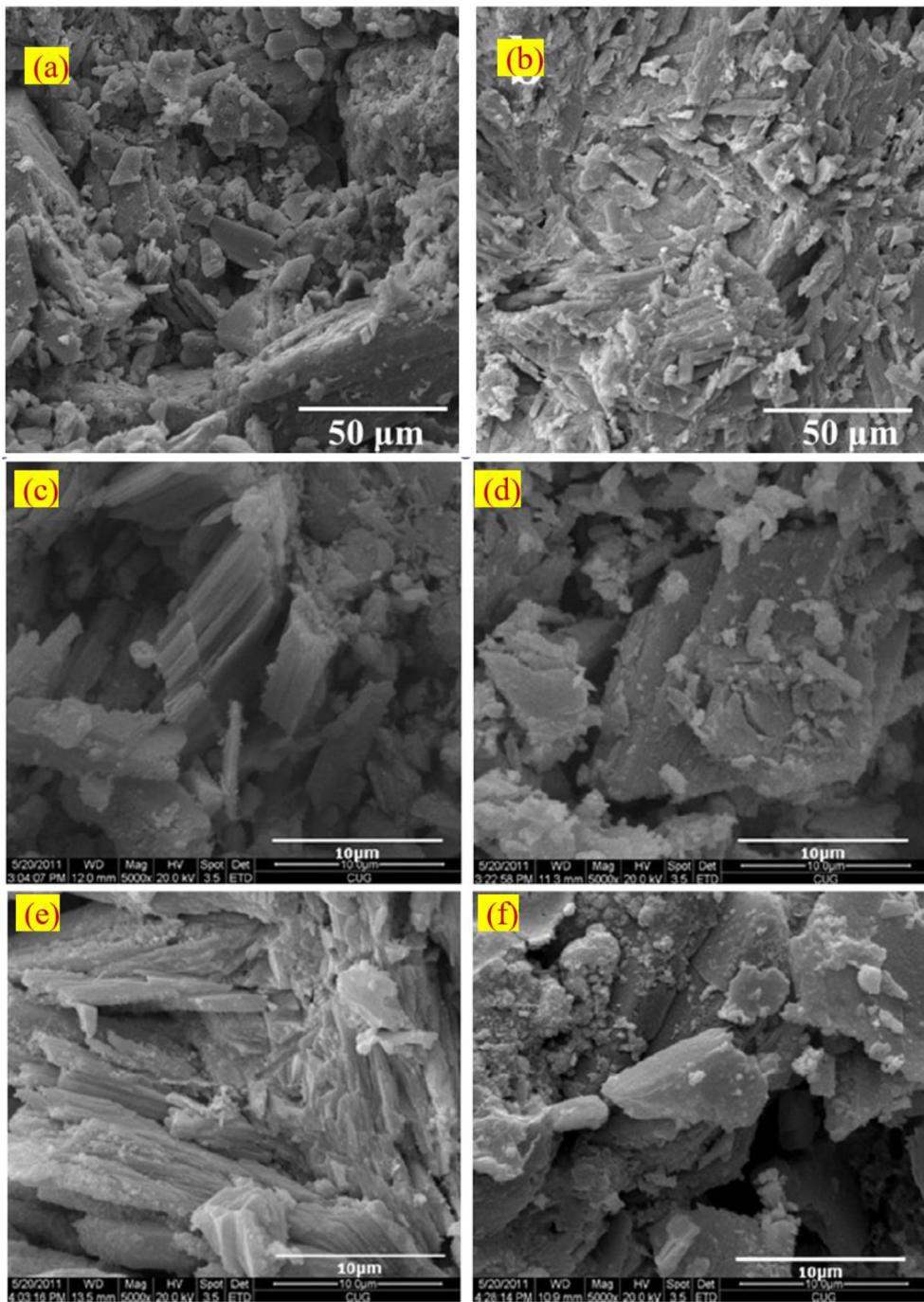


Fig. 6 – SEM (a) Non-immersed process control PG unfired bricks, (b) the finished, unfired PG bricks made with the Two-Step Hydration Method [105] (c) PG green unbricks containing their original gypsum, (d) Fire-dried brick with gypsum that has been partially dehydrated, (e) Bricks made from gypsum that has been recrystallized after being hydrated for 28 days and (f) 28-day-old gypsum-original brick that has not been hydrated or recrystallized [104].

water for recrystallization and hydration, the newly formed gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$) crystals that had recrystallized in situ displayed a uniform compact and interlocking microstructure (Fig. 6e). This phenomenon imparts a high compressive strength of 21.8 MPa onto the 28-day-cured bricks [104]. Whereas the significant crystalline phase of the unfired bricks remained gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$), this was not the case

as they were manufactured by the Non-Hydration-Recrystallization method, which involved neither hot drying nor water immersion. This phenomenon led to the compressive strength being only 9.5 MPa after being cured for 28 days because of their uneven, porous, and insular microstructure (Fig. 6f) [104]. It is clear from the above discussions that the mechanical property enhancement of the

Table 3 – Summary of the setting time of mortar.

Ref	Replacement level of PG (%)	Final setting time (% Improvement)	Initial setting time (% Improvement)	Substitution material
[93]	10, 20, 40 & 80	160, 190, 260 & 480	32, 68, 132 & 144	Cement
[118]	2, 3 & 4	103, 112 & 130	91, 100 & 109	Clinker
[119]	5, 10, 15 & 20	283, 517, 717 & 867	22, 78, 92 & 99	Cement
[120]	5, 10 & 15	300, 500 & 700	21, 77 & 92	Cement
[121]	5, 10, 15, 20 & 25	220, 443, 597, 723 & 729	30, 44, 102, 106 & 107	Cement
[122]	60	9.8	6.37	Slag + cement
[48]	35, 55 & 65	1.5, 12, & 18	4, 0 & 1.38	slag-limestone

PG unfired bricks was primarily due to the hydration-recrystallization mechanism.

4. Mechanical and microstructural properties of cement

4.1. Setting time of binder

The phrase “setting time” refers to the time it takes for cementitious materials to transition from their unhardened, fresh state to their hardened state [116]. The summary of the setting time recorded from the literature for the mortar is demonstrated in Table 3. According to Smadi et al. [93], using PG as a cement substitute extends the time it takes for cement paste combinations to set, both initial and final. Adding 10–80% of PG as cement replacement increased the initial and final setting time by 160–480% and 32–444%, respectively. Various PG concentrations were used in the manufacturing of calcium sulfoaluminate cement by Shen et al. [117]. They observed that the addition of PG increased the setting period. Compared to mixes comprising gypsum of 18%, the 17% PG mixtures took 147% and 54% higher time for the initial and final settings, respectively. Cement pastes were made by Altun and Sert [14] using clinker combined with either natural gypsum or PG. The partial substitution of clinker is made at quantities varying from 1% to 12.5% by natural gypsum or PG. Compared to the ones that included natural gypsum, the ones that contained PG required a significantly longer time to complete and initially set. Coated cement particles with inert elements like calcium phosphate and fluorides while mixing may be crucial because of PG’s significantly longer retarder impact.

Hydraulic binders made from phosphogypsum were tested for the setting time, as depicted in Fig. 7. It was determined which cement formulations and their systems (ternary and quaternary) are appropriate for supersulphated cement. Phosphogypsum dihydrate (Di-PG), PG hemihydrate (α -PG and β -PG), and insoluble anhydrite (A-PG II) made up the majority of the mineral phases employed in the formulation of Hydraulic binders made from phosphogypsum. Additionally, various setting times can be observed. Furthermore, three classifications were developed to comprehend the rate at which Hydraulic binders made from phosphogypsum hardened. Each option is categorized as slow (90–120 min), medium (60–75 min), and rapid (7–45 min). Using a calcination temperature of 800 °C, Fig. 7 reveals that the total setting time for the A-PG II + GGBFS system is 360 min, with an initial

210 min [123]. When calculated at 750 °C, both setting times of system (A-PG II + GGBFS) vary according to the ratio of PG to GGBFS employed. Setting times are at their longest when composed of 70% of A-PG II and 24% of GGBFS. Both setting times are reduced to their smallest possible lengths when 50% of A-PG II and 50% of GGBFS are used [124]. In addition, it is conceivable to ascertain that the A-PG II + Slag + lime system has an initial and final setting time of 259 and 550 min, respectively [74]. The initial and final setting times for the different combinations of the additional admixtures and the recommendation of ASTM standards are also shown in Fig. 7.

Research confirms that adding PG to a mixture increases the initial and final setting times. This rise in the time needed to set was proportional to the increase in PG content. Protection coats of calcium phosphate and calcium fluoride as inactive components created by F^- and PO_4^{3-} on the cement particle surface by mixing may be related to the reasonably long retarder action of PG [129]. The setting time of cement is significantly impacted by impurities in PG, as illustrated by Imez and Erdem [130]. It is possible to reduce PG’s effect as a retarder by employing appropriate treatment options on the PG particle surface. Because several contaminants like P_2O_5 , F^- , alkalis and organic matter were removed during the treatment process, and the treated PG setting time was significantly faster than the untreated PG. Additionally, setting time was reduced by employing calcined PG as compared to uncalcined PG. This might be pertinent to the decrease in fluorides and water-soluble P_2O_5 reported in PG [92]. As a result, PG can be utilized instead of the chemical admixture traditionally employed as a retarder agent, which has positive repercussions for the economy. As a by-product, PG has various potential uses in civil engineering (For example, mass concrete, retaining deep walls, and hot weather), and its retarder effect is one of them.

4.2. Compressive strength

The compressive strength of PG-based cement mortar is discussed in this section. Several literatures have been reviewed; for example, Wang et al. [131] investigated the beta-hemihydrate PG, slag, FA, and alkali activator used to produce the PG-based geopolymers cement. According to the findings, combining a high slag content and a low FA content maximized the compressive strength of PG-based cement. The 28-day strength could be greater than 60 MPa when the ratio of 50:44:6 for the PG, slag and FA was used. Li et al. [44] reported that the cement clinker content was drastically reduced, and the CAH_{10} and C_2AH_8 phases were eliminated

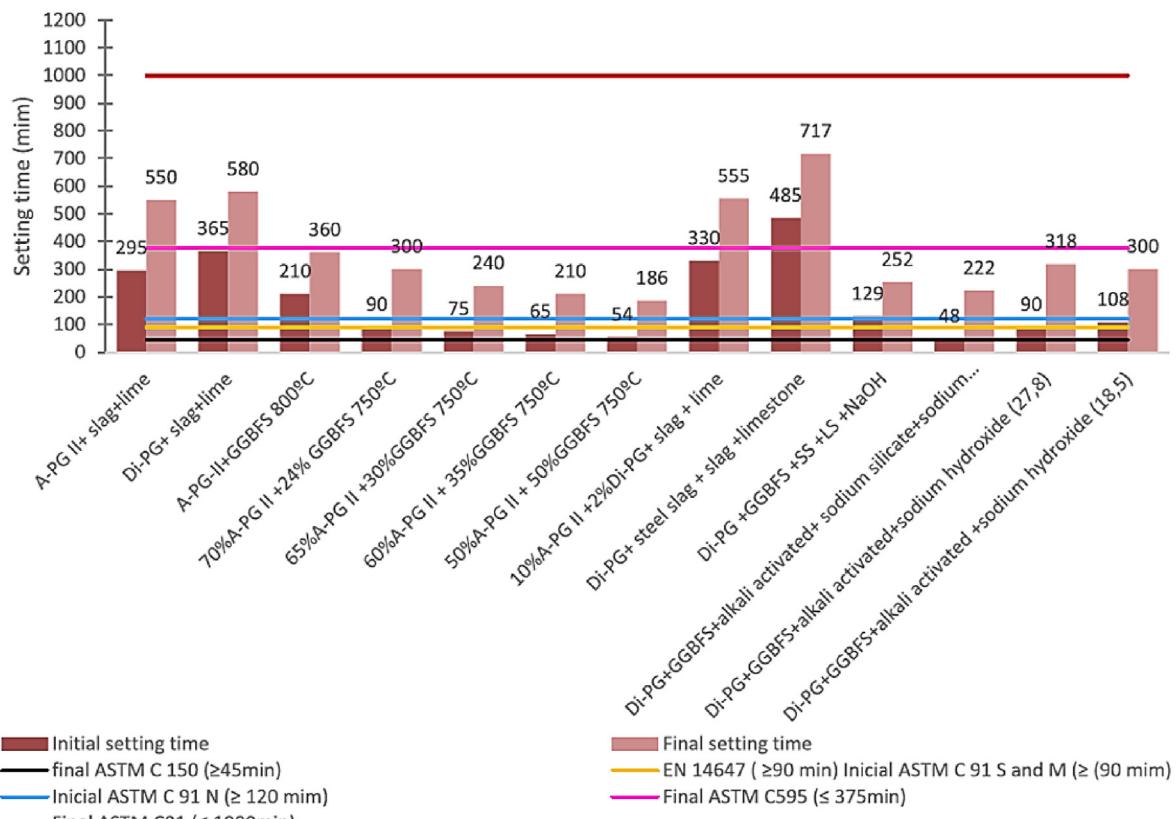


Fig. 7 – Hydraulic cement-based PG setting time [3,125–128].

after some PG was added to the calcium aluminate cement mixture in a 1:1 ratio. Subsequently, a decrease in the hardened paste's compressive strength was noticed due to the formation of AFm [132]. Additionally, calcium aluminate cement's compressive strength was greatly improved by a rise in PG concentration (higher than 5%); however, the increasing rate of compressive strength slowed when the PG content was more than 15%. According to Potgieter et al. [68], the compressive strengths of cement containing PG were lower at ages 2, 7, and 28 days than those of cement containing natural gypsum. Hwaiti also confirmed the identical behavior [133]. Wei et al. [63] reported that the cement samples containing 50, 75, and 100% PG had higher compressive strengths than the reference sample that experienced air curing at 28 days. Adding 75% of PG resulted in maximum compressive strength, which increased by about 19.6% compared to the reference sample (Fig. 8a). In the beginning stages, PG and the soluble phosphate found in PG can potentially reduce the rate at which the intensity phase is generated. Moreover, compressive strength is diminished because of the development of microcracks caused by $\text{Mg}(\text{OH})_2$ expansion [134]. Zhou et al. [135] reported that the magnesium potassium phosphate cement samples exhibited fewer impacts of the magnesium oxide to monopotassium phosphate ratio on compressive strength across the board after curing when PG was absent. Nevertheless, the compressive strength at all ages was negatively affected by the addition of PG, and this effect was exacerbated when PG concentration was raised (Fig. 8b). A noticeable decrease in compressive strength was detected

when the inclusion level was higher than 20% [135]. Fig. 8c depicts the decreasing trend of compressive strength while increasing the concentration of PG on all three days of testing. The least strength was observed in mortar specimens comprising a 5% PG and calcium aluminate cement of 95% (Fig. 8c).

Mortar combinations made of fine aggregate, PG, and clinker were created by Abdelhadi et al. [118]. The clinker content was reduced by 2, 3, and 4% and substituted with PG. The drying process for PG took place at either 60 or 120 °C. The findings showed that the compressive strength decreased with the inclusion of PG when it dried at a temperature of 120 °C. A declining trend in compressive strength at 28 days was 7.36% when 2% PG was added; at 17.17% when 4% PG was added. According to Smadi et al. [93], substituting cement with PG at concentrations up to 100% decreased the compressive strength of mortars. This decline became more pronounced as the PG rating increased. Addition of 10–80% PG, the 28-day compressive strength decreased between 10 and 53% under air curing and between 13 and 71% under moist curing, respectively. Bhaduria et al. [120] revealed a 35.5, 7.6, and 4.3% improvement in the mortar's compressive strength by containing 5% PG as a replacement of cement, correspondingly, for 3, 7, and 28 days. While introducing 10%, PG decreased by 25.4, 31.4 and 18.3%, respectively. In their study, Mun et al. [74] showed that the mortar's compressive strength of mortar made from 91% slag, 8% PG, and 1% slacked lime increased with adding PG concentration up to 12%, beyond which point it began to

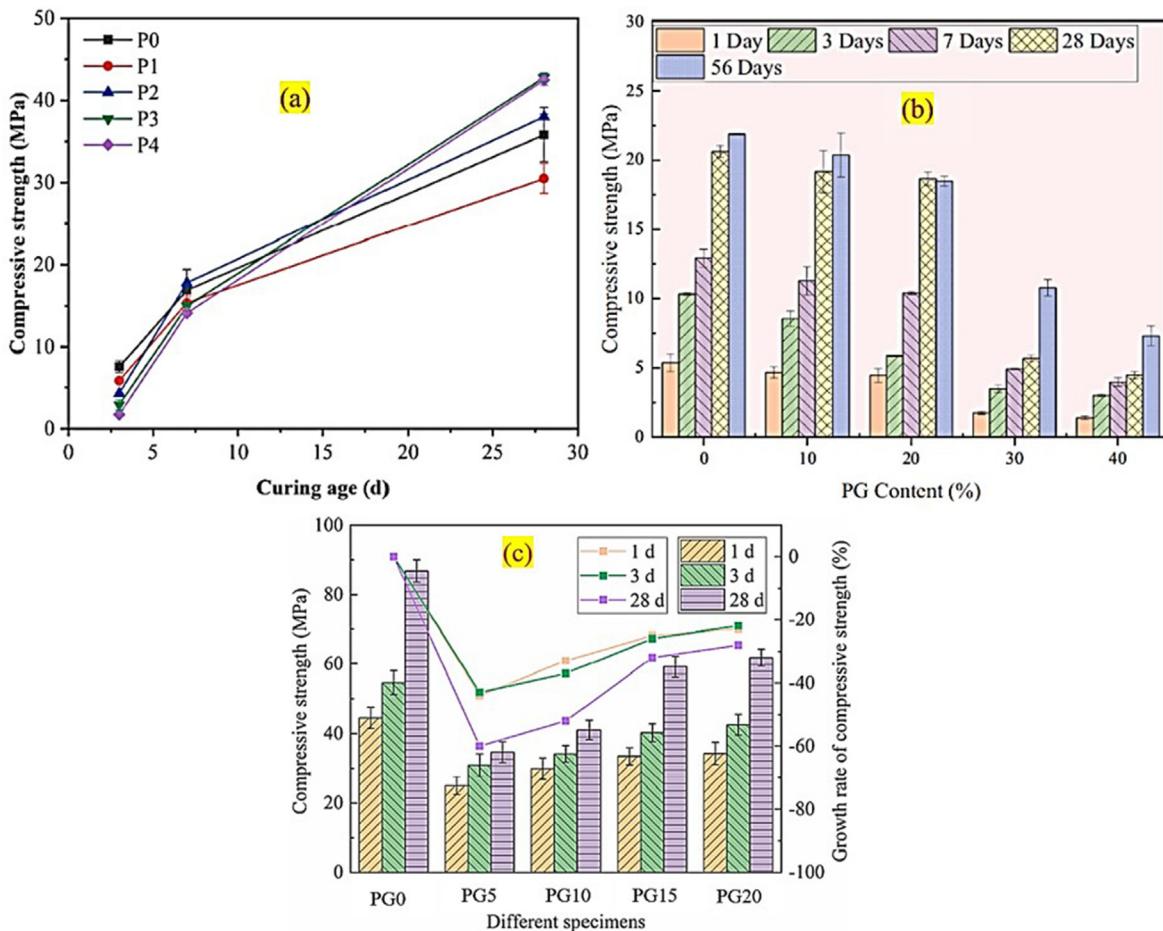


Fig. 8 – Compressive strength observed by (a) Wei et al. [63], (b) Zhou et al. [135], (c) Li et al. [44].

deteriorate. The key findings of other research regarding the compressive strength of mortar comprising a PG are summarized in Table 4.

4.3. XRD analysis from PG-based mortar

Fig. 9 displays the XRD analysis for the hydrated samples at varying PG concentrations observed by Li et al. [44]. In the XRD pattern, we can see that the diffraction peaks of un-hydrated C_2A_S , CA_2 , and CA (with almost negligible hydration) are present. Accelerated hydration of CA results in forming of C_2AH_8 , AH_3 , and CAH_{10} ; these compounds predominated in the PG-free samples (Fig. 9a). There was a complete disappearance of the C_2AH_8 and CAH_{10} , diffraction peaks in the XRD pattern, and the production of AFm when the PG concentration was 5%. Longer curing times resulted in a weaker ettringite diffraction peak, which was correlated to the presence of PG [139,140]. Peaks of ettringite and AH_3 in diffraction diagrams were emphasized due to the presence of PG. Unhydrated $CaSO_4 \cdot 2H_2O$ could be seen as a diffraction peak when the PG concentration was raised to 15%, and this peak intensified further when the PG percentage was raised [44]. The XRD patterns also feature many peaks characteristic of hemihydrate gypsum and quartz [131]. At first glance, Ettringite and dihydrate gypsum were the most prominent hydration

products. Wang et al. [131] reported that the cement was anticipated to yield a non-crystalline geopolymer system resultant, namely $C-S-H$ gels. Existing research additionally highlights the problem of XRD results of compounded PG with slag not scanning to $C-S-H$ gel crystals [48,141].

Hou et al. [143] reported that the micro-experimental evidence suggested that PG added early on enhanced Aft development, $C-S-H$ gels can polymerize and undergo morphological change and C_3S dissolution. With an increase in PG concentration, the quantity of Aft crystal initially climbed, subsequently reduced, and then continued to increase again; the $[SO_4]^{2-}$ produced by its breakdown could promote the activity of FA. It was demonstrated that PG and FA have a beneficial synergistic impact. Fig. 9b demonstrates no discernible difference between the two sets of samples regarding the diffraction peak intensity of C_2S , C_3S , calcium hydroxide, and ettringite [54]. This result suggested that after 28 days of hydration, the cement made with modified PG was virtually identical in hydration degree to cement made with unaltered PG. Gypsum, ettringite, and α - SiO_2 are the primary minerals of phosphogypsum slag cement by Li et al. [142] (Fig. 9c). Hydrolysis of raw materials results in the release of α - SiO_2 and ettringite formation via hydration. In addition, numerous investigations have demonstrated that $C-S-H$ gel is present in phosphogypsum slag cement hydration products

Table 4 – The summary of the findings of PG-based mortar.

Ref	Replacement level of PG (%) and other materials	Findings
[131]	PG- 0 to 50%, Slag- 0 to 50%, and FA – 0 to 50%	<ul style="list-style-type: none"> The specimen was highest after 28 days, when its compressive strength was 45.3 MPa, 50% of the main cementitious material mass, and FA and PG made up 40 and 10%, respectively.
[44]	PG-0, 5, 10, 15, 20% + calcium aluminate Cement 80–100%	<ul style="list-style-type: none"> Compressive strength dropped by 44, 43.6, and 60.1% at 1, 3, and 28 days, respectively, when 5% PG was added to the specimens. Compressive strength increased by approximately 24.9% at 1 day, 30.5% at 3 days, and 71.1% at 28 days for specimens with 15% PG, respectively, compared to 5% PG.
[63]	PG- 0,25, 75 and 100%+ magnesium oxysulfate cement	<ul style="list-style-type: none"> PG extends the final setting time of magnesium oxysulfate cement, decreasing the cement's fluidity by 25–75%. Mortar with a PG content of 50% or more has a higher compressive strength over time.
[53]	PG-60-100%+ GGBFS-0-25% +cement-15%	<ul style="list-style-type: none"> Hemihydrate phosphogypsum composite containing recycled waste glass exhibited a softening coefficient of 0.98, a flexural strength of 7.67 MPa, compressive strength of 42.6 MPa, low drying shrinkage of 378 μe at 28 days, and a lightweight bulk density of 1849 kg/m³.
[136]	PG-1,3,5 and 10% +PC + waste marble + waste clay brick	<ul style="list-style-type: none"> Due to the presence of $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ and phosphorus impurities in the PG composition ($\text{CaHPO}_4 \cdot 2\text{H}_2\text{O}$, $\text{Ca}_3(\text{PO}_4)_2$, $\text{Ca}(\text{H}_2\text{PO}_4)_2 \cdot \text{H}_2\text{O}$, H_3PO_4), the setting times rise proportionally with increasing PG content. Compressive strength dropped by 26.84% after introducing 2% of PG. Adding 4% of PG resulted in an 11.27% reduction after 14 days and 17.17% after 28 days.
[118]	PG-2,3 and 4%+Clinker	<ul style="list-style-type: none"> The highest compressive strength was achieved with a 7.5% PG addition, regardless of the crystal structure of the PG. The strength trended downward when the integration rate went above 7.5%. PG-containing mortars are more resistant to acid corrosion than mortars that only contain OPC,
[137]	PG-5, 7.5, 10 and 12.5%	<ul style="list-style-type: none"> Compressive and flexural strength barely changes as PG content rises from 40 to 55%, but shrinkage, bond strength, and wear resistance are significantly impacted.
[138]	PG-40%+ cement-40%+ natural sand-40%	<ul style="list-style-type: none"> Compressive and flexural strength barely changes as PG content rises from 40 to 55%, but shrinkage, bond strength, and wear resistance are significantly impacted.

[47,144,145]. Dispersion peaks and distinctive crystal phase peaks at roughly 10–35° are visible in Fig. 9c. From a $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio of 3.5 in SY3 to 3.7 in SY6, quartz and gypsum's typical crystal phase peak becomes more prominent. The decrease in Al_2O_3 content prevents ettringite from forming, as evidenced by the weakening of the characteristic phase peak of ettringite near 15° [146]. CaO/SO_3 significantly affected the mechanical characteristics of phosphogypsum slag cement. The effectiveness of an alkaline activator can be improved by increasing its CaO content, which stimulates sulphate activation to some degree and causes significant hydrolysis of phosphogypsum. Ultimately, it encourages the synthesis of large quantities of hydration products, which enhances strength [147].

4.4. SEM analysis from PG-based mortar

Several distinct cement morphologies are depicted in Fig. 10. The C–S–H gels stood out for having a more compact composition than the others, while the microstructure distribution was haphazard in clumps of ettringite and certain gypsum crystals. Moreover, there was also the presence of big

particles that could be gypsum crystals. Current studies of PG reveal that the phosphate ions were physically adsorbed onto the gypsum crystal surfaces. The gypsum crystals' shape was altered because such ions inhibited axial development. Phosphate caused the gypsum crystals to change shape from needle-columns to plates and increase in size [148]. The plate-like appearance of some of the gypsum crystals is shown in Fig. 10a. However, gypsum in needle-like columns can also be found in Fig. 10b. It is possible to see that the needle-columns crystallized ettringite forms (Fig. 10c) a framework by inserting itself into the gel in a cross-sectional manner [44]. Moreover, AFm was produced, which was connected to the amount of sulphate present [139,140]. It was noticed that a significant quantity of columnar ettringite was produced when the percentage of PG in the mixture reached 20% (Fig. 10d), which has a significant amount of AH_3 surrounding it. According to Wei et al. [63], the PG blocks can be broken down into flakes and used for growth (Fig. 10ef). This effect can be explained by the fact that citric acid alters the gypsum crystalline phase's shape. Additionally, it has been noted that soluble P_2O_5 in the form of HPO_4^{2-} degrades the mechanical characteristics of gypsum by turning $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ into semi-flakes [149].

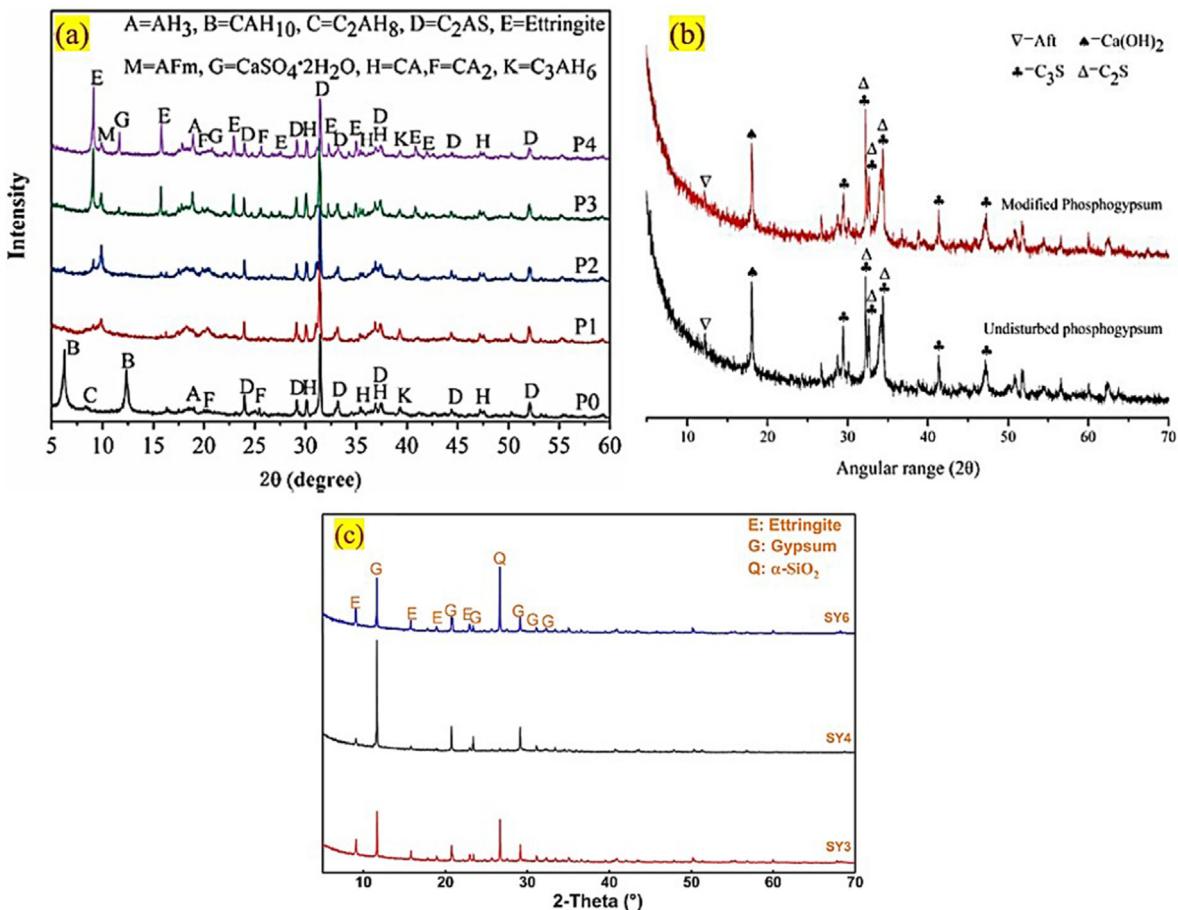


Fig. 9 – XRD analysis from mortar sample (a) Li et al. [44], (b) Li et al. [54] and (c) Li et al. [142].

5. Mechanical and microstructural properties of concrete

5.1. Workability of a concrete mixture comprising PG

In this section, the workability of concrete made with PG is discussed. A study reported that the concrete mixes workability was increased while using 2.5 and 5% of PG as a substitute for cement; however, the workability declined gradually when using 7.5 and 10% of PG [150]. In a study by Sindhuja et al. [151], the authors demonstrated that the concrete's workability decreased when 10, 20, or 30% PG was used as a cement replacement. The concrete workability that has between 5 and 20% PG as cement replacement was shown to decrease by Bagade and Satone [119] Bhadauria et al. [120]. According to Buhari and Raju [152], the concrete workability decreased when PG was used as a cement replacement at 5, 7.5, 10, 12.5, and 15%. Yang et al. [138] developed a mortar mixture for self-leveling, which contains natural sand of 40%, PG of 40%, and cement of 20%. A mixture of 5, 10, and 15% PG was used in place of natural sand. The findings showed that adding PG reduced the initial workability. Increases in PG content were associated with a steeper decline in workability. Because PG has a higher specific surface area than regular sand, this difference in workability may be attributable to this

feature. However, after 20 min, the workability improved with 40–50% PG, and then reduced when 55% PG was added. It is possible that the interplay between morphological and setting-retarded of PG is responsible for the material's increased workability. In contrast, the material's increased specific surface area is responsible for its decreased workability. In preparing the concrete mixture, Huang et al. (2016) used 45% pretreatment PG (with a 281.9 m²/kg surface area), 4% clicker, 2% steel slag, 49% slag and aggregates. When compared to cement concrete, the PG based concrete mixture performed poorly in terms of workability and the height of the slump declined by 36.36%. According to the investigations that were recorded in this area, the workability of incorporating PG has significantly decreased. This decrease became more pronounced as the PG concentration became higher. Workability may have decreased since PG is less dense than sand or cement, which are both common construction materials. One of the drawbacks of employing this material is the decrease in workability that occurs when PG is added to the combination. Whichever the case may be, this issue can be remedied by the utilization of various chemical admixtures.

5.2. Compressive strength of PG based concrete

Zhang et al. [153] examined the mechanical properties of recycled aggregate concrete made with slag, FA and PG. In this

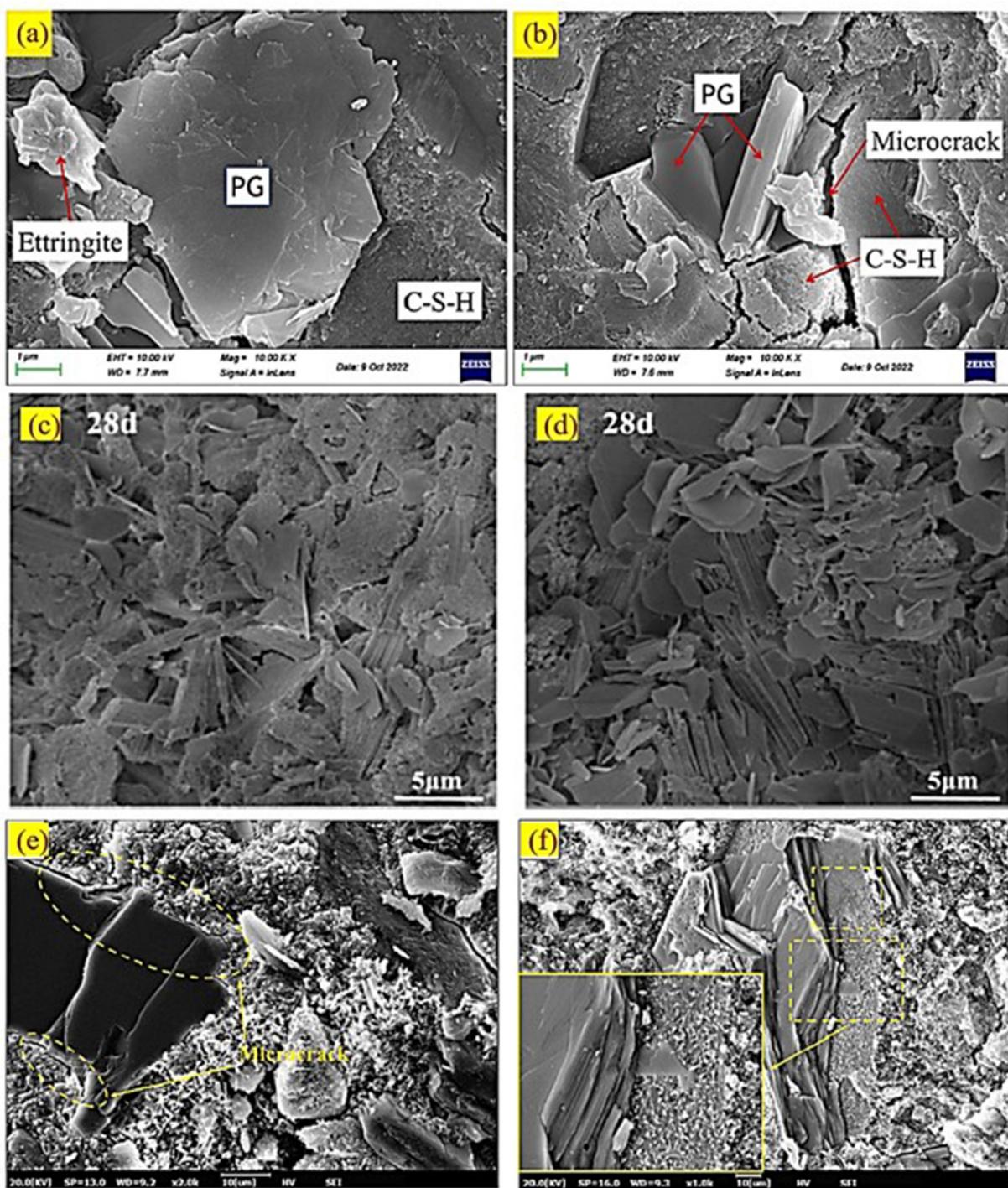


Fig. 10 – SEM analysis of binder (a) and (b) [131], (c) and (d) [44], (e) and (f) The interface between magnesium oxy-sulfate cement and PG [63].

study, the three ingredients are kept as common, and the recycled aggregate replaces the natural aggregate. Findings indicated that the compressive strengths dropped with increasing recycled aggregate replacement rates (0–100%). According to the findings, the specimen's compressive strength was more vulnerable to reduction due to recycled aggregate flaws when the replacement percentage was low. When the recycled aggregate content increased, the PG, FA, and slag reacted with the CH in the old mortar on the recycled

aggregate surface, significantly increasing the ITZ's density while compensating for recycled aggregate flaws. This phenomenon increased the compressive strength of the specimen containing recycled aggregate. Based on these findings, it is clear that PG, FA, and slag with recycled aggregate have a significant impact on specimen compressive strength [153]. The summary of the compressive strength of concrete made with different concentrations of PG is depicted in Fig. 11. Cement replacement with 10% PG in concrete can achieve

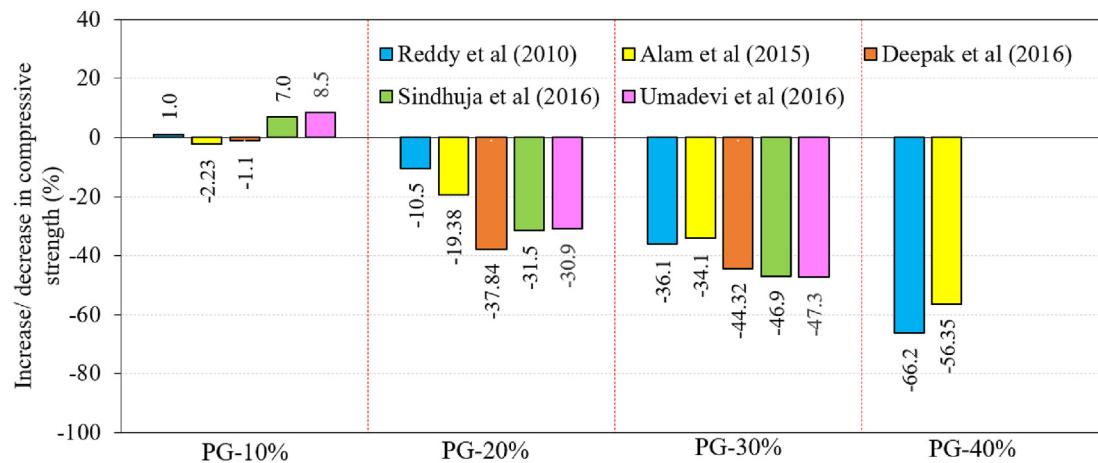


Fig. 11 – Compressive strength of concrete comprising PG as cement substitution [151,154,159–161].

excellent strength, according to the research of Reddy et al. [154], but substituting cement with more than 10% PG led to a significant decrease in strength. When using 5% PG as a cement substitute, Bhaduria et al. [120] showed a 6.4, 12.2, and 7.3% increase in the concrete compressive strength after 28, 35, and 45 days, but using 10%, PG decreased by 24.6, 10.8, and 20.2%. Lou et al. [38] used unprocessed PG (5, 10 and 15%) along with cement (32%), FA (46.5, 51.5, 56.5, and 61.5%), Al powder (1.25%), Na_2CO_3 powder (1.25%) and lime to create a novel autoclaved aerated concrete. The results showed that the observed compressive strength ranged from 4.1 to 7.6 MPa when the PG content varied between 0% and 15%. This fortification of the autoclaved aerated concrete can be attributed to two primary factors; (1) Following autoclaving, the autoclaved aerated concrete's anhydrite content was significantly increased due to the addition of PG, which increased the sample's gypsum content. (2) evidences that after autoclaving, the samples containing PG showed a prominent anhydrite peak, tobermorite and anhydrite, and were both important to the autoclaved aerated concrete's overall effectiveness [155,156]. Fig. 12a illustrates the influence of PG on compressive strength reported by Yang et al. [157]. If the percentage of PG is raised from 0 to 10%, the material's compressive strength rises accordingly. On the other hand, this reduces when the PG concentration is more than 10%. As a result, it may be concluded that PG serves as both an activator and a filler in the system. When the percentage of PG in the binder is higher than 10%, the compressive strength will decrease because the binder's volume will decrease [157]. Tian et al. [158] reported that most of the PG is used as a filler, and whether the sulphate content has a function in activating needs further investigation. CaCO_3 was substituted for PG in the quick lime-GGBFS-cement system to illustrate its function better. The CaCO_3 has a particle size distribution comparable to the PG utilized. The findings suggest a consistent rise in the compressive strength (59–75 MPa) when the percentage of PG changes from 0 to 12% (Fig. 12b). After that, it virtually descends in a straight line from 12 to 60%. The highest strength was recorded at 12% PG concentration which is the optimum strength.

5.3. XRD analysis from concrete sample

XRD patterns for pre-autoclaving samples PG1-PG4 (0–15% PG) are displayed in Fig. 13a. Gypsum, quartz, calcite, portlandite, Mullite, and ettringite are the most common crystalline phases [38]. Specifically, a peak was observed between 26 and 30 $^{\circ}\text{2}\theta$, which is indicative of an amorphous phase, C–S–H gel [162]. The C–S–H gel can chemically bond all of the sample's constituents, resulting in a dense network structure that aids in the material's strength gain [163]. Mullite is the main module in fly ash, so it was found in high concentration in all samples [164]. Prominently, the incorporation of PG stimulated ettringite creation due to the rise of system's residual gypsum content. Ettringite concentrations in cement tended to rise sharply as gypsum concentrations rose, and ettringite crystallization helped speed up the material's early strength evolution [165,166]. Fig. 13b shows the most abundant minerals in the autoclaved samples treated with PG. Since ettringite decomposes between 50 and 110 $^{\circ}\text{C}$, the ettringite XRD peaks vanished in the autoclaved aerated concrete following autoclaving [167]. After autoclaving, the autoclaved aerated concrete contained a high concentration of tobermorite, and their three peaks can be found at 8.1, 29, and 30.1 $^{\circ}$ [168,169]. The compressive strength in autoclaved aerated concrete is susceptible to tobermorite content. Autoclaving the C–S–H gel commonly causes it to transform into tobermorite [155,163].

When the PG level raised from 0 to 30%, the appearance of calcium aluminate sulphate hydrate (AFm) diffraction peaks, as seen in Fig. 14a [157]. $\text{Ca}(\text{OH})_2$ diffraction peaks decrease as PG content rises, disappearing altogether at 30% PG; it demonstrates that adding PG encourages hydration. The $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ diffraction peak disappears below 15% PG. It means all the $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ has been used up, despite the PG concentration reaching an unsafe level of 30%. As a result, the PG fulfills an activator and a filler role [157]. Mixes devoid of PG typically contain mineral phases, such as quartz, calcite, portlandite and a great deal of C–S–H, which are all invisible in XRD patterns (Fig. 14b) due to their amorphous or non-crystalline nature [158]. Ettringite diffraction peaks were present; hence

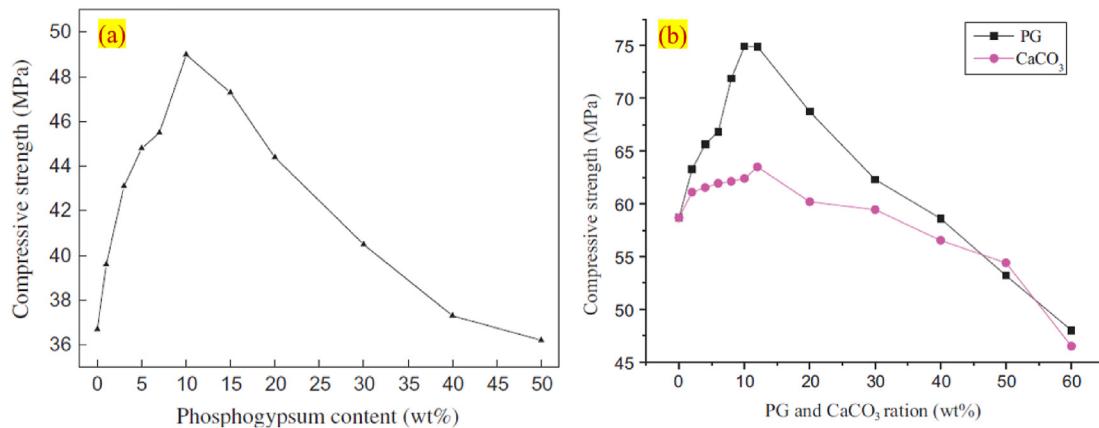


Fig. 12 – Compressive strength (a) Non-autoclaved aerated concrete made with PG [157], (b) Foam concrete [158].

there was a clear distinction between the hydration products of the PG-containing samples (4–12%) and those of the blank category. The primary ingredient in PG is gypsum. Nevertheless, gypsum's diffraction peaks are invisible in the samples with PG concentrations between 4 and 12%. This demonstrates that every gypsum particle participates in the hydration process and transforms into ettringite. The appearance of gypsum peaks at a PG concentration of 50% indicates a surplus of gypsum and that a portion of it does not contribute to the hydration process.

5.4. SEM analysis from concrete sample

Fig. 15ab displays a microscope image of autoclaved aerated concrete that was autoclaved without adding PG. The lamellar portlandite and C–S–H gel are the primary hydration products identified, in addition to discovering several fractured hollow beads. These results proved that the autoclaved aerated concrete without PG was hydrated in a highly alkaline environment, particularly in the foaming phase. The uneven development of bubbles is caused by excessive alkalinity, which speeds up the rate at which the foaming process occurs. An enlarged micrograph of pre-autoclaving PG-treated

autoclaved aerated concrete samples is shown in Fig. 15 cd. The gypsum rods and massive ettringite needles were found with the C–S–H gel and a few lamellar portlandite crystals. Pre-autoclaving the AAC samples with ettringite maintains a consistent level of strength, facilitating effortless specimen release from the mold [155].

Carbonization's impact on the microstructure of cement-PG with GGBFS and FA is shown in Fig. 16 [158]. Fig. 16 (a, d, g), (b, e, h) and (c, f, i) are the morphology of AFT, walls between two pores and surface pores, respectively. Pre-carbonization AFT, depicted in Fig. 16a, anisotropic well-crystallized druse with baculiform and needle-shaped hydration products. As shown in Fig. 16d, nevertheless, when CO₂ causes corrosion in a cement-PG with a GGBFS system, blemishes and flaws on the surface, including stains and pinholes, start to show themselves as the occurrence of corrosion occurring on cement-PG with FA (Fig. 16g) is a significant threat. The flocculent C–S–H gel is seen in Fig. 16b; prior to carbonization, the space between two pores in a wall was filled with unreacted gypsum and acicular ettringite [158]. As a result of carbonization, they have a high degree of stability in the cement-PG with the GGBFS network (Fig. 16e). The cement-PG with FA system corroded acicular ettringite and C–S–H gel (Fig. 16h). Before

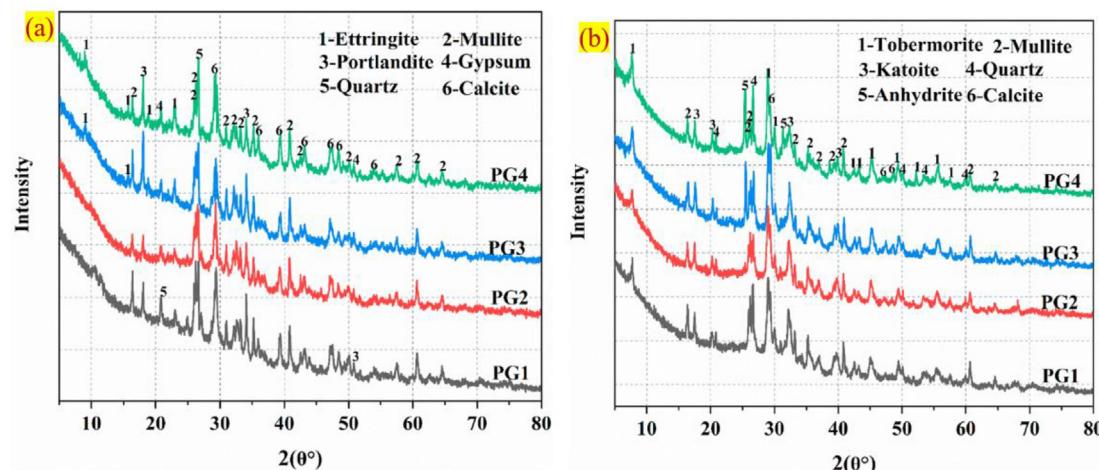


Fig. 13 – XRD analysis from a concrete sample (a) before autoclaving and (b) after autoclaving [38].

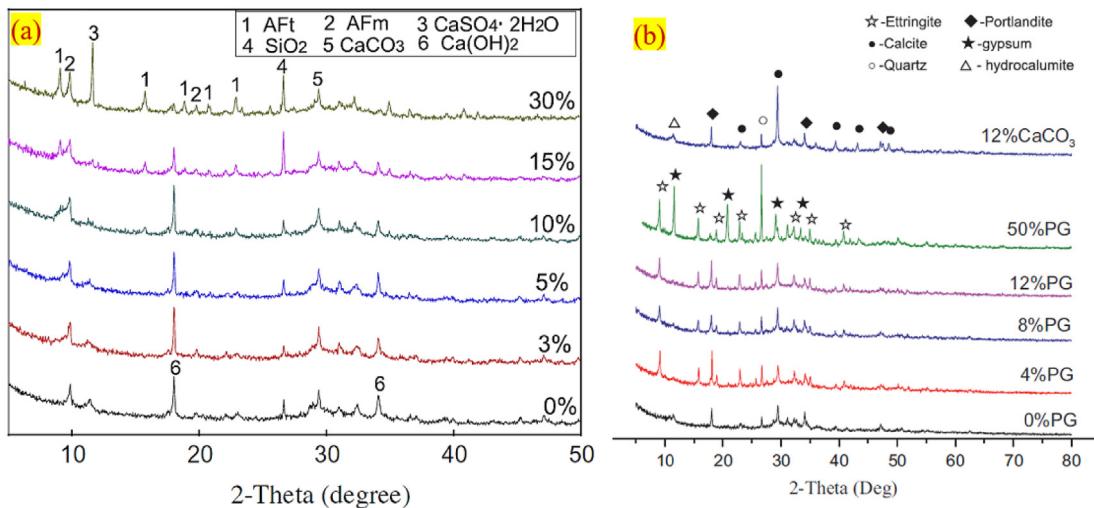


Fig. 14 – XRD analysis from a concrete sample (a) Yang et al. [157], (b) Tian et al. [158].

being carbonized, the surface of the pores was rather smooth (Fig. 16c). However, a rise in pore volume is possible due to gaps allowing for ettringite development. Following carbonization, the cement-PG with the GGBFS system's typically smooth surface pores becomes covered in many wedge

crystals (Fig. 16f). It is also clear that the C–S–H gel was corroded and that less calcite was produced following carbonization in the cement-PG system with FA (Fig. 16i). The FA is an aluminum-silicon compound, which explains this, and the system's calcium levels were low [158].

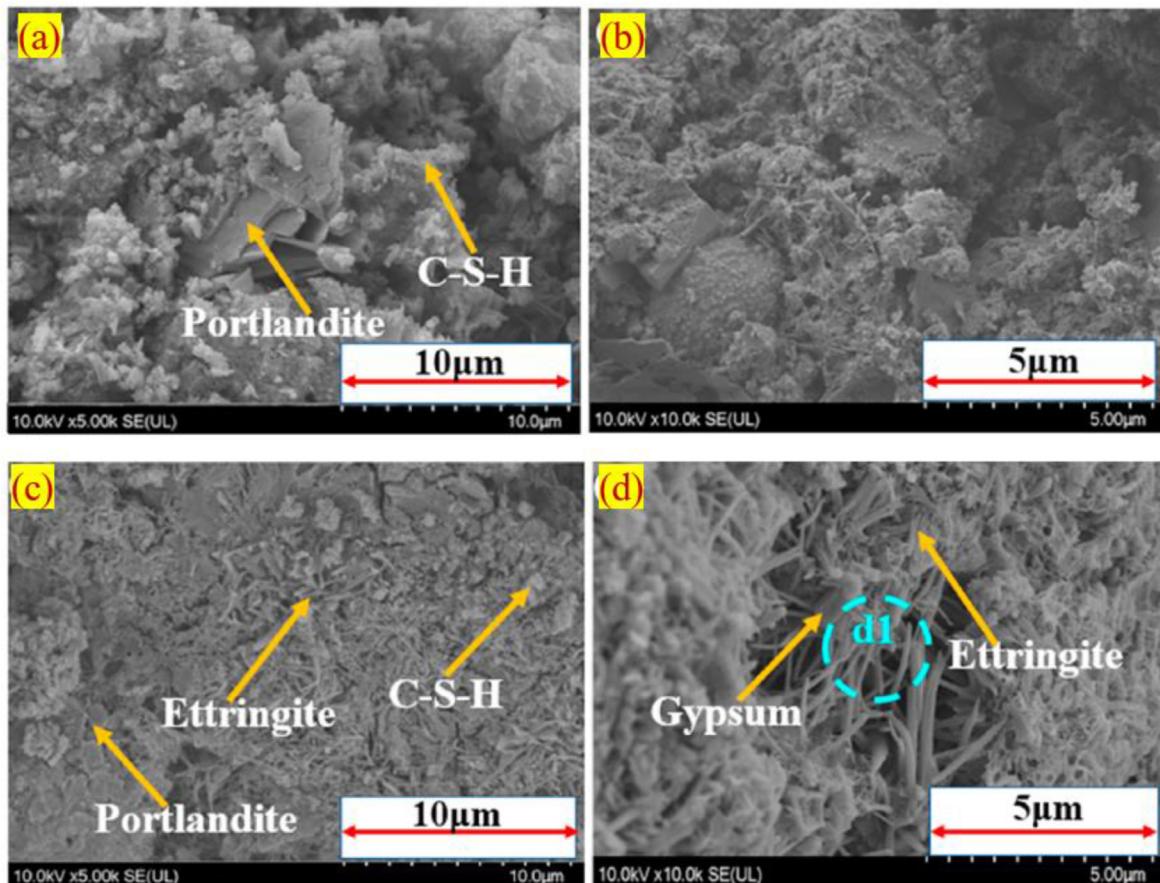


Fig. 15 – SEM analysis from a concrete sample (a–d) [38].

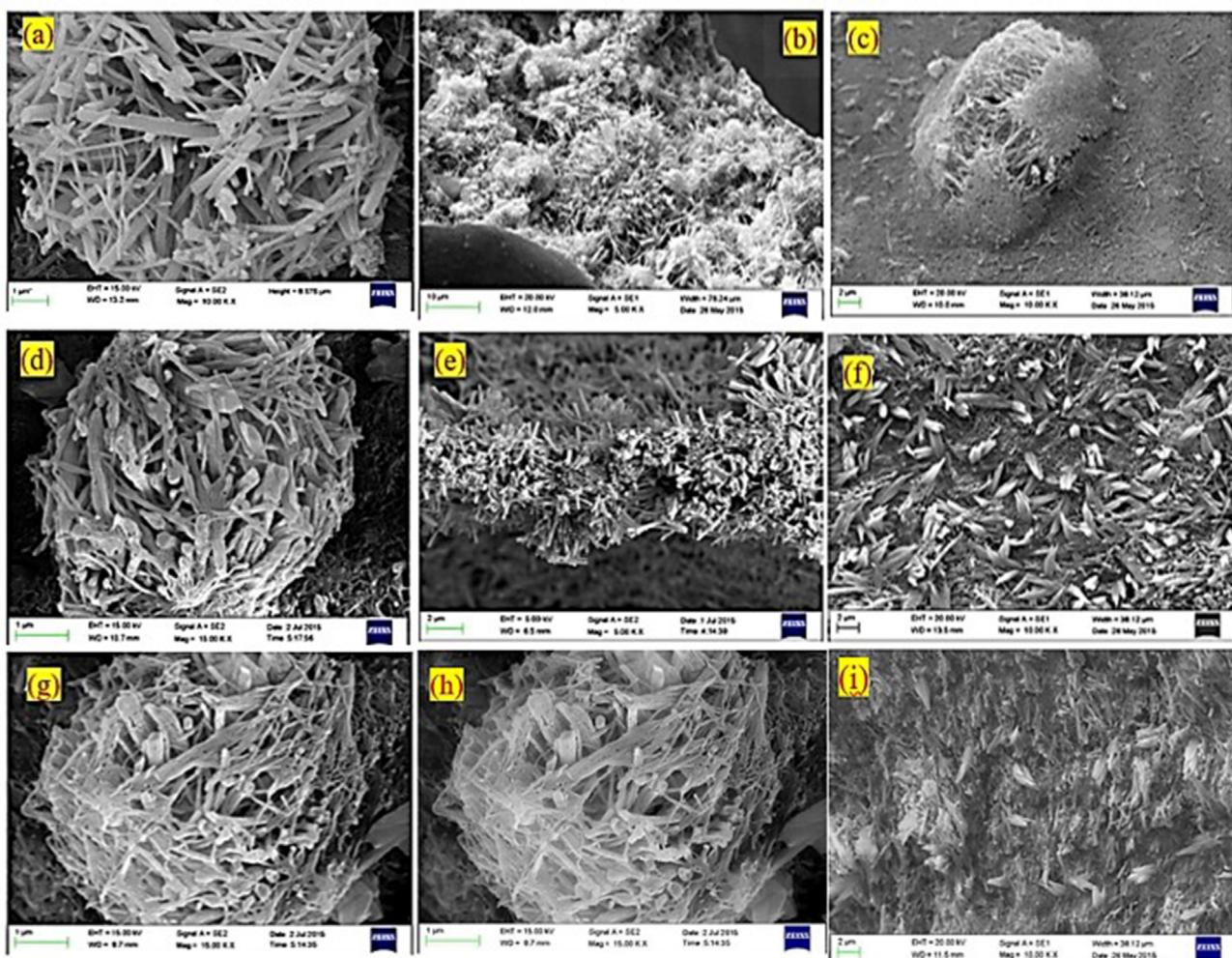


Fig. 16 – SEM analysis from a concrete sample by Tian et al. [158].

6. Conclusions

Public and governmental awareness has been focused on the environmental issues brought on by PG stocking up, including water and soil contamination, dust pollution, and land occupation. Hence, increasing the recycling rate requires identifying and developing new uses for PG. Thus, PG-based building materials, including bricks and raw materials for cement and concrete, are summarized herein. Nonetheless, in order to increase efficiency, lessen damage to the environment, and encourage the interdisciplinary growth of civil engineering's PG-based materials, it is suggested that further study be conducted in the following areas.

1. The CaO and SO₃ make up most of the mass of PG, roughly more than 65%. The minimum and maximum percentages of CaO were 24.0–57.9%, SO₃ were 30.9 and 61.6%, and a small quantity of P₂O₅ (0.01–3.57%), Al₂O₃ (0.054–4.38%), and other components; the impurities in PG lead to subtle but significant variations in their chemical properties.

2. Many methods are available to purify PG, including sieving, neutralizing agents, lime purification, washing,

calcining, flotation, ball-milling, and chemical treatment. After considering alternative treatment methods, calcination was the most effective. In addition, improved calcination combined with other methods may increase pre-treatment productivity while cutting energy use. Other methods, such as calcining temperature reduction, inert minerals activation, and effective admixtures addition, are worthy of further study.

3. Several studies reported that the optimum content of PG was 75% in combination with FA and lime to reach maximum compressive strength and bending strength. Another study reported that the optimum PG content was 25 and 30% combined with other materials. Therefore, the optimum PG content varies using different material combinations, and more studies are required to optimize the PG content. Crushing strength, durability, efflorescence, and radioactivity of bricks are unexamined and could be scope for future work.

4. The compressive strength of calcium aluminate cement mortar increased significantly with increasing PG content (>5%); however, the rate of improvement slowed when the PG percentage was greater than 15%. The calcium aluminate cement's hydration products C2AH8 and CAH10 were eliminated

upon the incorporation of PG. Different studies reported the different optimum content of PG in cement mortar. Further study is required to determine the optimum content of PG with superior compressive strength of mortar.

5. Cement and cement clinker can employ PG as a raw material due to its reactivity with water, leading to various products with varied applications. After then, more research into the reaction mechanism is needed to help the hydration process move forward.

6. The compressive strength of concrete consistently increases up to 12% PG use, according to the research. After that point, it almost immediately begins to fall, contrary to findings from other studies. Therefore, more research is needed to determine the ideal PG percentage for concrete.

7. When PG is added to a concrete batch, the gypsum content increases, leading to a higher anhydrite content in the final autoclaved aerated concrete product. The disappearance of the $\text{Ca}(\text{OH})_2$ diffraction peaks at 30% PG shows that adding PG promotes hydration. Below 15% PG, the diffraction peak for $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ disappears. This line should be a priority in the future.

8. The commercial implementation of the innovative technology can assist in considerably decreasing the environmental effects of waste PG due to their high proportion in the mix designation. Regular encouragement of extensive, eco-friendly, and resourceful PG use is essential, and the creation of novel PG-based materials is essential—for example material reclaimed from PG and processed into aggregate for concrete production, prefabricated plates for the foundation of roads, PG-based wallboard, blocks and tiles, possessing a high PG consumption capacity.

7. Prospects for utilizing PG and challenges

The paradigm shift required to think of waste as a raw material is the first obstacle that needs to be conquered. There has been a considerable discussion about recycling and reusing materials in manufacturing recently, but this trend will not take off unless concrete results are obtained. Phosphate gypsum is used in relatively small quantities during several valorization processes; hence, the product's demands are modest and cannot meet the needs of widespread use. In this regard, producing bricks and concrete, which uses tiny quantities of this substance, offers the most potential uses. Studies on the utilization of PG in the hydration of cementitious systems, eco-friendly concrete, high workability concrete, high-performance concrete and stabilized mortar have the potential to advance the development and adoption of more environmentally friendly construction practices. Widespread application of PG is possible: polymer flooring composition, glass ceramics, artificial stone, tiles, plasters, sulfur polymer concrete, partition blocks, plasterboard panels, ferrocement panels and wallboard. Roads, storage facilities, and parking lots can benefit from a layer of PG combined with fine and coarse aggregate. Valorization of PG also supports circular economy initiatives in the building sector; although it is currently considered waste, this material may serve as a

resource soon. This can potentially lessen the industry's harmful effects on the environment.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Dr. G. Murali completed his post-doctoral research in fracture mechanics at Peter the Great St Petersburg Polytechnic University. He is also listed in ‘The Stanford University’s list of world’s top 2% scientists- Oct-2022. He has published more than 139 research articles in an international journal. His research area includes Impact resistance of fibrous concrete, Preplaced aggregate fibrous concrete, Functionally graded fibrous concrete, Layered fibrous concrete, Slurry infiltrated fibrous concrete and Weibull Distribution, Recycled aggregate concrete, Geopolymer concrete, Ultra High-Performance Concrete, Topology optimization, Strengthening of structural members using FRP, Rubberized concrete and Fracture toughness of the composite.



Dr. Marc Azab is working as an Assistant professor, American University of the Middle East, Kuwait. His research area includes fracture mechanics, Sustainability, Construction Materials.