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Effect of Hydrophobic Silica-Based Admixture on the Physical Performance and Durability of Waterproof Geopolymer Composite

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Abstract

Sustainability construction technology initiatives have a great concern for green construction materials that offer high durability properties comparable to conventional alternatives. Geopolymer concrete, synthesised from industrial by-product-based precursors such as fly ash and slag, presents an eco-friendly alternative to Portland cement, but requires improved water resistance for widespread adoption and high durability binders. This study systematically evaluates polydimethylsiloxane (PDMS) as a hydrophobic modifier (0-5% by binder weight) to enhance the performance of geopolymers. Comprehensive testing revealed PDMS's dual mechanism: surface modification (an increase in contact angle from 40.1° to 97.3° at a 4% dosage) and pore structure refinement (a 76% reduction in water absorption at a 5% dosage). Mercury intrusion porosimetry showed PDMS effectively seals micropores while creating strategically isolated macropores that maintain low permeability. The 5% dosage emerged as optimal, delivering balanced surface hydrophobicity (93.9° contact angle) with superior matrix densification (0.93% water absorption). These findings establish PDMS-modified geopolymer as a viable, durable construction material for moisture-prone environments, addressing both sustainability and performance requirements in modern and resilient infrastructure.

1. Introduction

The global construction industry is undergoing a critical transition towards sustainable practices to mitigate its significant environmental impacts. This industry is largely attributed to the production of Ordinary Portland Cement (OPC). The manufacturing of OPC requires an intensive energy process that is responsible for a substantial portion of global Carbon Dioxide (CO_2) emissions. Thus, research and development of blended cement is

exponentially increasing to reduce the high production of CO_2 [1]-[3]. Instead of blended cement, Geopolymer concrete (GPC) utilises industrial by-products such as FA and slag, which have emerged as a promising solution to reduce the carbon footprint and the beneficial reuse of waste materials [4]. It is formed through the alkaline activation of aluminosilicate-rich materials, such as FA, slag, metakaolin, or marble waste, resulting in a high compressive strength cementitious binder up to 20% higher compared to conventional binder [5].

Despite the environmental advantages and good mechanical properties of GPC, its potential application may be limited due to its poor durability. Its hydrophilicity and porous microstructure often lead to high water uptake and poor moisture resistance [6]. This susceptibility to water penetration is likely to promote premature deterioration through corrosion of the reinforcement, spalling, and other similar effects [7]. In light of the growing need for fast-strength development and durability of repair materials in the construction industry, a better understanding of the fundamentals of geopolymer synthesis is necessary to customise such cementitious materials for these purposes [8]. The enhancement of water resistance is the ultimate direction of research on the long-term performance of GPC.

GPC can be modified to enhance its hydrophobic properties, either externally or internally. This has sparked interest in creating Waterproof Geopolymer Composites (WGC) by integrating hydrophobic admixtures during the mixing process, resulting in enhanced waterproofing durability [9]. Researchers have investigated the hydrophobic modification of the GPC matrix to address the challenges posed by GPC. One effective strategy to enhance GPC resistance to water penetration and other severe environmental factors is by altering its surface characteristics [10]. Coating is a technique often utilised due to its affordability, minimal energy consumption, and effectiveness [11]. This encompasses surface coating that can be applied via spraying, painting, or sealing. It offers initial water resistance, but this can often be diminished by wear, flaking, or exposure to the elements [12]. Nonetheless, the durability of traditional surface coatings tends to be limited over time, particularly when exposed to adverse environmental factors and mechanical strain. To overcome these limitations, researchers have explored the incorporation of hydrophobic agents into coating formulation treatments to impart durable water-repellent properties. These agents function by altering the surface energy of the substrate, thereby minimising capillary water ingress and improving the resistance against moisture-related deterioration.

Hydrophobic agents are typically categorised based on their waterproofing mechanisms and chemical composition, including silicone-based compounds, fatty acids, metallic stearates, and polymer emulsions [13]. Another study by Qu et al. [14] incorporated biofilm in alkali-activated materials. The study demonstrated that biofilm enhances hydrophobicity and reduces capillary tension. Past studies support the potential of internal hydrophobic modification. For instance, silane-based treatments have been shown to reduce water sorptivity, but at the expense of reduced mechanical strength [15]. Other strategies include incorporating nanomaterials to improve durability in a saline environment [16]. In addition, studies have shown that modification of the liquid-to-solid ratio during the mixing process affects the microstructure of GPC, consequently influencing the wettability properties of the composite [17]. Using silicone-based compounds, such as PDMS, has been found to be the most effective way to make things more water-resistant [18]. This study investigates the effect of adding PDMS to GPC at varying concentrations on its physical and durability performance. The porous nature of GPC means that the structure of its pores is also very important for its performance. These pores are crucial for sorption and support GPC, which lasts longer [19]. This study also examines the pore structure of WGC to investigate how hydrophobized GPC affects its hydrophobicity. It is essential to understand this relationship because the presence and connectivity of pores significantly impact the movement of water and other substances through the matrix. By modifying the pore structure to make it hydrophobic, you can increase the resistance of GPC to moisture infiltration and the resulting damage.

One of the primary research objectives is to enhance the water resistance of GPC, enabling it to reach its full potential as a long-lasting and environmentally friendly building material. Different amounts of PDMS had different effects on the physical and durability performance of WGC. There is currently insufficient research on how hydrophobic admixtures function to enhance water resistance and prevent moisture from entering. Additionally, it is a concern that WGC may not last long when exposed to the environment, especially in areas where water is likely to come into contact with it. Previous research has demonstrated that hydrophobic modifications can enhance the waterproofing properties of GPC [20]. The higher WCA, which measures hydrophobicity, supports this finding. The water contact angle (WCA) is a method for quantifying the effectiveness of hydrophobic materials in repelling water. A surface with a WCA more than 150° and a sliding angle less than 5° is considered superhydrophobic [21]. This study helps us learn more about creating long-lasting, environmentally safe building materials that can be used in challenging environments. This study provided valuable insights into creating composites that are more water-resistant and long-lasting by determining the optimal dose of PDMS. The results can help GPC be used more widely in difficult situations.

Despite these encouraging improvements, significant problems still need to be addressed. The geopolymerization process can be affected by the incorrect amount of hydrophobic admixture or by uneven distribution of it. This could cause the material to not fully hydrate, which would make it weaker [18]. Additionally, practical issues such as the high cost of specific admixtures and the lack of standardised testing

Int. Journal of Integrated Engineering Vol. 17 No. 7 (2025) p. 209-219

techniques for WGC make it more challenging to make the technology commercially viable and widely used. Most importantly, there is a distinct gap in the research regarding the optimal dosage of an admixture. There is yet no systematic investigation to determine the optimal concentration of PDMS that balances the best waterproofing performance with the least amount of material needed. To address this knowledge gap, this study investigated the influence of varying PDMS concentrations on the properties of FA and slag-based geopolymer composites. This research involved analysing the chemical composition of selected aluminosilicate precursors to verify their suitability for geopolymer synthesis. Then, the effect of varying the concentration of PDMS was analyzed through physical and durability characteristics. Finally, the study employed MIP to characterise the changes in pore structure incorporated with PDMS. The scope of this investigation focused on geopolymer composites incorporated with Class F FA and slag, and then activated by sodium silicate and sodium hydroxide solution. This study exclusively utilised PDMS as a hydrophobic admixture, with the concentration varying from 0% to 5% by weight of the binder. The evaluation is specifically limited to WCA as a test for physical performance, and the capillary water absorption test as a measure of durability performance. The tests were conducted in accordance with ASTM D7344 and ASTM C1585, respectively. This research did not extend to analysing the mechanical properties of WGC.

2. Methods

2.1 Materials

The materials used in this research include aluminosilicate precursors, which were Class F Fly Ash (FA) and slag, alkaline activators (sodium silicate and sodium hydroxide), water, and hydrophobic admixtures, which were PDMS. The suppliers for FA and slag were Klang Energy Venture (KEV) Sdn. Bhd. and Desjaya Concrete Products Sdn Bhd, respectively. Meanwhile, the sodium silicate was sourced by South Pacific Chemical Industries Sdn. Bhd. (SPCI), Malaysia, and the sodium hydroxide was supplied by Formosa Plastic Corporation, Taiwan. The sodium silicate was in liquid form, while the sodium hydroxide was in pellet form. For the hydrophobic admixture used, which was PDMS, a silicone-based compound that was provided by Sigma Aldrich, USA.

2.2 Mix Proportions and Specimen Preparation

WGC specimens were prepared according to the mix proportions in Table 1. The alkaline activator solution was formulated by combining both sodium silicate solution and sodium hydroxide solution. Sodium hydroxide solution was prepared beforehand by dissolving NaOH pellets in distilled water. The molarity of NaOH was maintained at a constant value of 12 M. The ratio of sodium silicate solution to sodium hydroxide solution was kept constant at 2.5:1; meanwhile, the ratio of binder to alkali activator remained constant at 2:1, as optimised in previous work by Pasupathy et al. [22]. Six distinct mixes were prepared, including a control specimen with no addition of PDMS. Five modified specimens with PDMS concentrations ranging from 1% to 5% by weight of the total binder were prepared.

Table 1 *Mix design of control and modified WGC (in kg/m³)*

Mix design	PDMS (%)	FA (kg/m³)	Slag (kg/m³)	Na^2SiO^3 (kg/m³)	NaOH (kg/m³)	PDMS (kg/m³)	Water (kg/m³)
G-0	0	1270.0	1306.0	2224.0	768.0	0.0	386.0
G-1	1	1270.0	1306.0	2224.0	768.0	26.0	386.0
G-2	2	1270.0	1306.0	2224.0	768.0	52.0	386.0
G-3	3	1270.0	1306.0	2224.0	768.0	78.0	386.0
G-4	4	1270.0	1306.0	2224.0	768.0	104.0	386.0
G-5	5	1270.0	1306.0	2224.0	768.0	128.0	386.0

The preparation of WGC paste followed a standardised, multi-stage mixing procedure. Firstly, the dry precursors (FA and Slag) were mixed manually for 2 minutes. Then, a pre-mixed alkaline activator solution and water were added to the dry mix, and it was manually blended for an additional 2 minutes. The paste is then mixed using a mechanical mixer with the addition of a specified dosage of PDMS for the final 6 minutes at a medium speed to ensure uniform dispersion of the hydrophobic admixture.

After mixing, the fresh paste was cast into moulds of two different dimensions for a specific test, which were 50 mm × 50 mm × 20 mm and 100 mm × 100 mm × 50 mm. The moulds were ensured to be clean, and a thin layer of release agent was applied to prevent sticking. The paste was compacted in three separate layers and vibrated

for two minutes to eliminate any air pockets. The samples were cured at ambient temperature and demoulded once they had completely hardened. The tests were conducted at 3-day and 28-day intervals.

2.3 Test Methods

The chemical composition of the precursors, FA and slag, was determined using X-ray Fluorescence (XRF) analysis to evaluate their suitability as precursors in geopolymer synthesis. The XRF analysis is an analytical technique used for the elemental analysis of solid materials. Representative samples of Class F FA and slag were collected and prepared. Both samples were then submitted to SIRIM Berhad, Shah Alam, for XRF analysis.

The contact angle used in this paper was measured using the profile image analysis method, as specified in ASTM D7334. A droplet of liquid was dispensed vertically onto the sample surface using a syringe. The angles formed were recorded using a high-resolution camera and analysed using software.

A capillary water absorption test was executed in compliance with ASTM C1585. The prepared, modified, and control specimens were tested at the 28-day curing period. The procedure began by drying the specimens in a vacuum oven at a temperature of 50 ± 2 °C. The specimens were then dried for 2 days, after which no further changes in mass occurred. The specimens' periphery was sealed with epoxy resin, leaving the surface exposed for testing. Then, the weight of sealed specimens was measured and recorded as M0. After that, water was added to a water bath maintained at 20 °C, and the water level was maintained 2 ± 1 mm above the specimens' surface. The mass of specimens was measured as Mt at different times until the mass remained constant over time. The average of repeated experiments and the capillary water absorption were calculated using Eq. (1).

$$\text{Capillary water absorption} = \frac{M_t - M_0}{M_0} \times 100\% \quad (1)$$

MIP was employed to analyse the pore structure of hydrophobized geopolymer. MIP can be employed to characterise the distribution of pores, which offers a thorough comprehension of the pore structure modifications that result from hydrophobization. The physical and durability performance of the material is directly correlated with the results of the MIP analysis. The pore structure of WGC can be observed using MIP with the addition of hydrophobic admixtures. To assess the pore structure resulting from the influence of hydrophobic agents, the control specimen was compared to hydrophobized geopolymer samples. To determine the optimal dosage of PDMS and the impact of hydrophobic admixture on the enhancement of WGC properties, the results of all tests were analysed. The results emphasised the optimal blend design, which has improved the physical and durability properties of WGC. All the insights obtained can serve as a basis for further research on sustainable construction materials.

3. Results and Analysis

3.1 Chemical Composition of Precursors

The elemental composition of the raw precursors was quantitatively determined using XRF spectroscopy in powdered form. This characterisation step was crucial to verify the chemical suitability of the materials for their intended role as aluminosilicate sources in the synthesis of WGC. As illustrated in Table 2 and Table 3, the XRF-derived compositional profiles of Class F FA and slag reveal key oxide percentages that directly influence their reactivity and binding potential. These results confirm that both FA and slag possess the necessary chemical attributes to serve as effective precursors in formulating a high-strength, durable geopolymer matrix.

Table 2 presents the chemical composition of FA, confirming its viability as a primary precursor for geopolymerization. The material predominantly consists of SiO_2 (56.3 wt%) and Al_2O_3 (33.4 wt%), which are critical for forming the three-dimensional aluminosilicate framework that defines geopolymer binders. These oxides provide the necessary reactive silica and alumina, ensuring effective dissolution in alkaline solutions to initiate the geopolymerization process. The low CaO (2.65 wt%) content aligns with its classification as Class F FA, promoting the formation of N-A-S-H gel - the primary binding phase in low-calcium geopolymer systems. Minor constituents such as K_2O (1.54 wt%) and Na_2O (0.2 wt%) further enhance reactivity by increasing alkalinity and facilitating the dissolution of amorphous phases. Additionally, the low LOI (2.54 wt%) indicates minimal unburned carbon, which improves workability and reduces water demand. Collectively, these properties confirm that Class F FA meets the essential criteria for use as a precursor for geopolymer. However, Table 3 defines the chemical composition of the slag, which is predominantly composed of CaO (40.17 wt%), followed by SiO_2 (33.9 wt%) and Al_2O_3. The elevated CaO content is indicative of slag, facilitating its function as a supplemental cementitious ingredient in geopolymer systems. During alkaline activation, slag undergoes hydration reactions similar to those of OPC, resulting in the formation of a C-S-H gel. The presence of calcium and aluminosilicates promotes the production of C-A-S-H gel, enhancing mechanical strength and durability. The MgO content of 8.3

Int. Journal of Integrated Engineering Vol. 17 No. 7 (2025) p. 209-219

wt% and a low LOI of 1 wt% indicate few organic contaminants, hence assuring optimal reactivity. Incorporating slag into geopolymer blends improves early-age strength development and sulfate resistance, rendering it a viable supplementary precursor. When slag is added to geopolymer blends, it accelerates the development of early-age strength and increases sulfate resistance, making it a very valuable supplementary precursor.

Table 2 *Chemical composition of Class F FA*

Mineral	Composition (wt%)
Silicon Dioxide (SiO_2)	56.3
Aluminium Oxide (Al_2O_3)	33.4
Potassium Oxide (K_2O)	1.54
Iron (III) Oxide (Fe_2O_3)	1.59
Calcium Oxide (CaO)	2.65
Magnesium Oxide (MgO)	0.83
Sulphur Trioxide (SO_3)	0.23
Sodium Oxide (Na_2O)	0.2
Phosphorus Pentoxide (P_2O_5)	0.27
Titanium Dioxide (TiO_2)	0.45
Loss in ignition	2.54

Table 3 *Chemical composition of slag*

Mineral	Composition (wt%)
Calcium Oxide (CaO)	40.17
Silicon Dioxide (SiO_2)	33.9
Aluminium Oxide (Al_2O_3)	11.65
Magnesium Oxide (MgO)	8.3
Sulphur Trioxide (SO_3)	1.56
Iron Oxide (Fe_2O_3)	1.26
Sodium Oxide (Na_2O)	0.86
Manganese Oxide (MnO)	0.73
Potassium Dioxide (K_2O)	0.59
Loss in ignition	2.54

3.2 Effect of Varying Concentration of PDMS on WGC

For the purpose of this investigation, the qualities of WGC were examined by analysing its physical and durability properties in response to varying amounts of hydrophobic admixtures that were incorporated into the mixture.

3.2.1 Physical Performance of WGC

The physical performance of WGC is crucial for applications in environments exposed to moisture and chemically aggressive conditions. Hydrophobicity was evaluated via WCA measurements (ASTM D7334), where angles <90° indicate hydrophilicity and >90° hydrophobicity. Six samples, cured for 72 hours under ambient conditions, were tested at three intervals to capture dynamic wetting behaviour.

WCA measurements quantified surface wettability and hydrophobic performance of WGC specimens with 0-5% PDMS. Fig. 1 shows the highest WCA values recorded for samples G-0 to G-5 after 3 days of curing. Results in Fig. 2 revealed a clear trend of increasing WCA with PDMS content, transitioning from hydrophilic (40.1° for G-0) to hydrophobic (>90°) at ≥3% PDMS. The WCA peaked at 97.3° for G-4 (4% PDMS) - a 143% increase over control - before slightly decreasing to 93.9° for G-5. This nonlinear progression aligns with known polymer saturation behaviour, where excessive PDMS may cause agglomeration. The findings match previous reports [20] of threshold concentrations (typically 2-4% PDMS) for optimal hydrophobicity in similar systems.

While the maximum WCA (97.3°) didn't reach superhydrophobic levels (>120°), all PDMS-modified specimens (G-3 to G-5) maintained stable hydrophobicity (>90°), exhibiting significantly improved water repellency compared to conventional materials. The slight WCA reduction at 5% PDMS (93.9° vs 97.3° at 4%) suggests a saturation point, consistent with literature on diminishing returns at higher concentrations [20], [23].

Fig. 2 clearly shows the critical 2-3% PDMS transition zone, where surface properties shift from hydrophilic to hydrophobic, matching theoretical predictions for such modifiers. This performance enhancement, achieved through a simple modification, demonstrates WGC's potential for use in moisture-resistant applications in demanding environments.

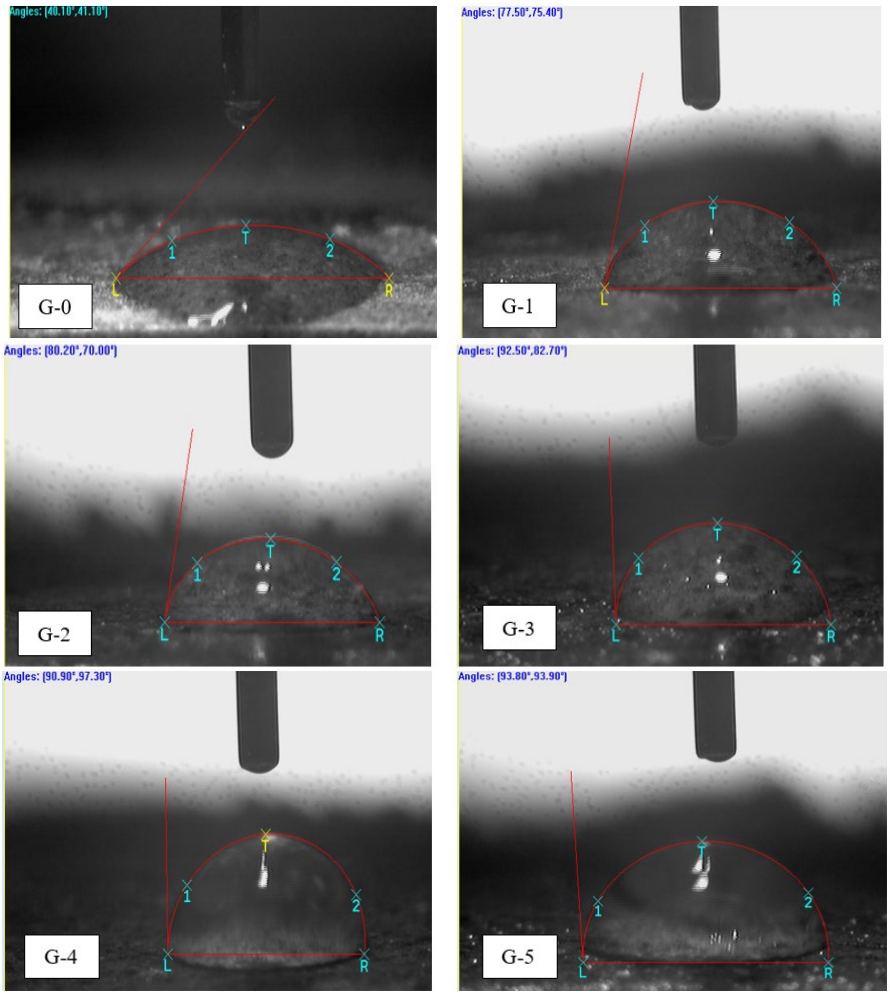


Fig. 1 *Measurements of the contact angle of WGC after 3 days of ambient curing*

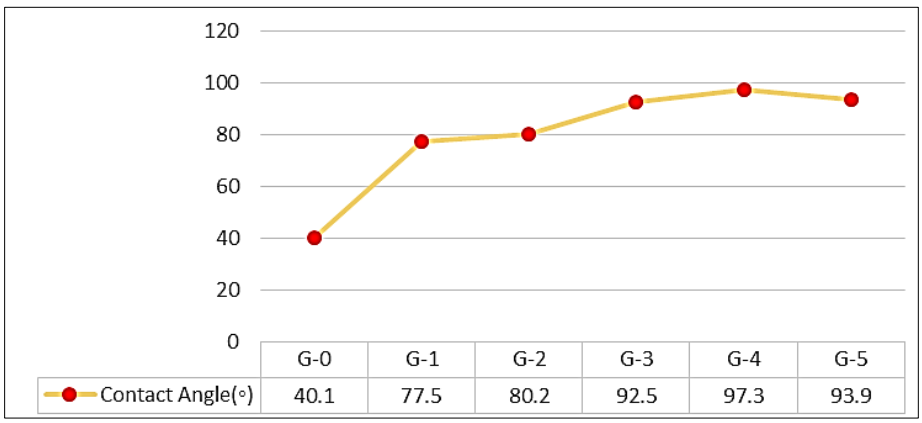


Fig. 2 *Contact angle of WGC after 3 days of ambient curing*

3.2.2 Durability Performance of WGC

The capillary water absorption test, conducted per ASTM C1585, measured WGC's resistance to water ingress through porous networks - a critical durability parameter influencing waterproof performance. Specimens were oven-dried after 28 days of curing, epoxy-sealed on all sides except one, and partially immersed (2±1 mm water

Int. Journal of Integrated Engineering Vol. 17 No. 7 (2025) p. 209-219

depth) while recording mass changes from 2 minutes to 7 days. Results revealed a strong inverse correlation between PDMS content and water absorption, with the control sample (0% PDMS) absorbing 3.91% water after 7 days, demonstrating the inherent porosity of the unmodified geopolymer. In contrast, PDMS-modified specimens showed a progressive improvement, culminating in a 76% reduction (from 0.93% absorption) for the 5% PDMS mix (G-5). Notably, absorption rates stabilised after day 4 across all samples, indicating pore saturation while still maintaining PDMS's superior water resistance.

The results established a distinct inverse relationship between PDMS concentration and water absorption capacity. The control sample (0% PDMS) exhibited maximum absorption (3.91% after 7 days), revealing the inherent porosity and moisture susceptibility of unmodified geopolymer matrices. As illustrated in Fig. 3, PDMS incorporation progressively reduced absorption: G-1 (1% PDMS) decreased to 3.43%, G-2 (2% PDMS) showed a more pronounced reduction to 2.25%, with G-5 (5% PDMS) achieving only 0.93% absorption - a remarkable 76% improvement over the control. This performance progression suggests PDMS effectively alters the pore structure and surface energy, with optimal effectiveness observed between 3-5% concentrations.

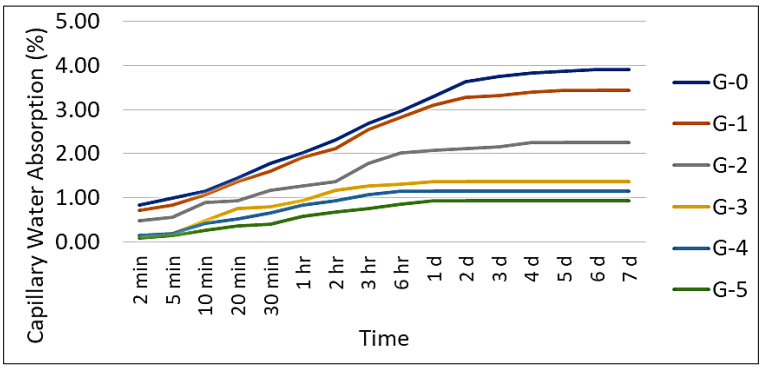


Fig. 3 *Capillary water absorption of WGC after 28 days of ambient curing versus time*

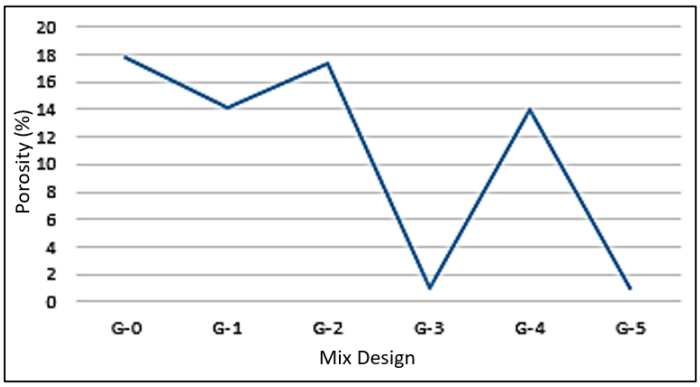
Between days 4-7, absorption rates stabilised across all samples, indicating capillary pore saturation. Notably, PDMS-modified specimens maintained significantly lower final absorption values despite this saturation, demonstrating persistent water resistance. The nonlinear reduction pattern with most substantial improvements occurring at 1-3% PDMS implies a percolation threshold for hydrophobic network formation. While G-5 (0.93%) showed only marginal improvement over G-4 (1.12%), both concentrations outperformed conventional materials, confirming the effectiveness of PDMS even at saturation. These findings corroborate previous work by Zhang et al. [23] and Ruan et al. [24] while providing new insights into the 1-3% PDMS range as the critical transition zone for optimal performance. The 76% absorption reduction demonstrates WGC's enhanced durability potential for moisture-exposed applications; however, further studies could explore the use of complementary additives to further enhance performance while maintaining cost efficiency.

3.3 Pore Structure Analysis

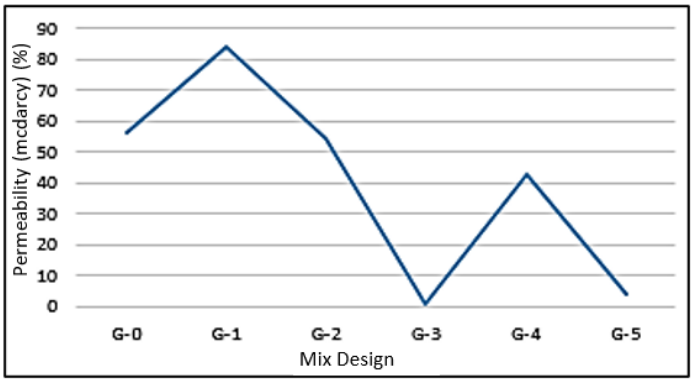
MIP analysis was employed to evaluate the critical pore structure parameters of both control and modified specimens, providing essential insights into their performance characteristics. As presented in Table 4 and Fig. 4, the MIP testing yielded comprehensive data on porosity, total intrusion volume, permeability, and average pore diameter. These parameters are particularly significant as they directly influence the material's durability and mechanical properties. The control specimen exhibited a relatively open pore structure, while PDMS-modified samples demonstrated notable reductions in total porosity and average pore diameter. This microstructural refinement correlates well with the observed improvements in hydrophobicity and water resistance reported in previous sections.

Table 4 *Results of MIP analysis*

Mix design	Porosity (%)	Total intrusion volume (mL/g)	Permeability (mcdarcy)	Average Pore Diameter (nm)
G-0	17.8	0.0969	55.89	237.3
G-1	14.1	0.0732	83.61	348.1
G-2	17.3	0.0908	54.03	204.4
G-3	1.0	0.0612	0.503	193.1
G-4	14	0.072	42.45	169.6
G-5	1	0.0551	3.69	289.6



(a)



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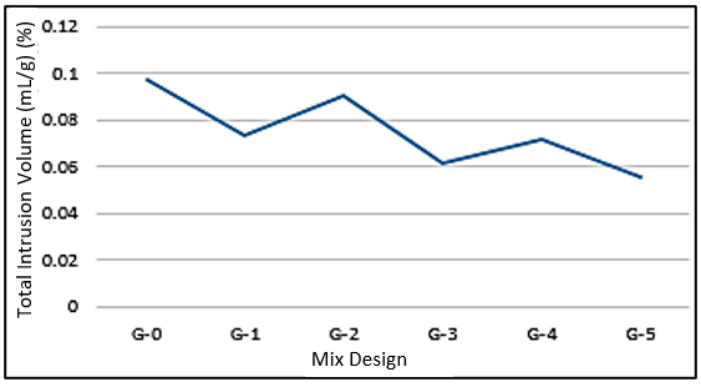

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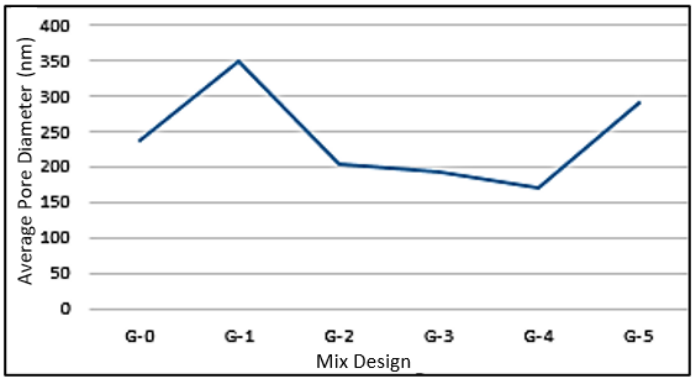

(d)

Fig. 4 *MIP analysis of geopolymer mix designs (G-0 to G-5) - (a) Porosity (%); (b) Total intrusion volume (mL/g); (c) Permeability (mdarcy); and (d) Average pore diameter (nm), illustrating the influence of mix design variation on pore structure and transport properties*

The G-1 specimen exhibited a notable anomaly, displaying the highest permeability and average pore diameter (see Table 4 and Fig. 4), despite a moderate reduction in porosity. This apparent contradiction aligns with previous findings by Qu et al. [20] regarding the modification of PDMS-induced pore networks. The observed "pore coarsening" phenomenon, where PDMS merges smaller pores into larger, interconnected channels, explains these results. While total void space decreased slightly in G-1, the formation of continuous transport pathways significantly increased permeability, demonstrating PDMS's active role in microstructural evolution even at low concentrations.

The results reveal a clear optimal PDMS dosage, with G-3 achieving peak performance (0.503 mD permeability, refined matrix). However, G-4 showed a sharp porosity rebound to 14%, indicating that it had exceeded a critical threshold where PDMS began to interfere with geopolymerization. This dosage sensitivity corroborates the warning about careful concentration control [24]. The G-5 mix presented further complexity - achieving an exceptional 1% porosity but unexpectedly large average pore diameter (289.6 nm). This represents PDMS's dual action: effectively filling small/medium pores while creating limited but larger voids that disproportionately influence diameter measurements. These findings collectively demonstrate PDMS's role as an active pore modifier whose effects transition from beneficial refinement to potential degradation across specific concentration thresholds.

3.4 Analysis of Optimum Dosage of PDMS

Through comprehensive evaluation of pore structure characteristics, surface hydrophobicity measurements, and water absorption performance, the optimal PDMS dosage for WGC enhancement has been determined to be 5%. While previous observations indicated that 4% PDMS (G-4) achieved the peak contact angle of 97.3°, a complete performance assessment incorporating internal pore structure refinement, total water absorption capacity, and permeability characteristics demonstrates superior overall improvement at 5% PDMS (G-5). The G-5 formulation demonstrated outstanding performance characteristics, including the lowest recorded porosity at just 1%, a minimal intrusion volume, and a markedly reduced capillary water absorption of 0.93%.

These results conclusively demonstrate G-5's superior capability in preventing water penetration. Although a marginal reduction in contact angle occurred (93.9° versus G-4's 97.3°), this slight decrease in surface hydrophobicity had a negligible impact on overall durability performance. Microstructural analysis revealed that the 5% PDMS formulation created an exceptionally dense internal matrix, resulting in the second-lowest permeability measurement among all tested mixes (surpassed only by G-3). This confirms that at optimal dosage, PDMS functions as an extremely effective pore-blocking agent, successfully sealing the majority of fine capillaries and substantially limiting fluid transport pathways within the geopolymer matrix.

The apparent contradiction of a higher average pore diameter in G-5 (289.6 nm) can be attributed to PDMS's unique pore modification mechanism at elevated concentrations. While creating a limited number of larger isolated pores, these voids demonstrate poor interconnectivity, as evidenced by the material's maintained impermeability and water resistance. Comparative analysis reveals that the G-5 formulation achieved a 76% reduction in water absorption compared to the control, exhibited only a 3.4% decrease in contact angle relative to G-4, and demonstrated superior pore refinement when compared to both G-3 and G-4 formulations. This performance profile establishes 5% PDMS as the optimal dosage, providing the most favourable balance between surface hydrophobicity and internal barrier properties. The formulation successfully addresses the critical durability requirements for WGC applications in moisture-exposed environments, demonstrating that minor surface property trade-offs are more than compensated by substantial improvements in bulk material performance. These findings provide valuable guidance for practical implementation, suggesting that maximum durability enhancement requires consideration of both surface and bulk material characteristics rather than optimisation of any single parameter.

4. Conclusion

The purpose of this study was to explore the influence of PDMS concentration variations on the physical and durability properties of WGC. The results of this investigation demonstrated that this silica-based hydrophobic admixture significantly improves both the water-repellent features and the microstructural refinement of the material. With contact angle measurements revealing a significant rise from 40.1° in the control specimen to values exceeding 90° in modified mixes, reaching a maximum of 97.3° at a 4% PDMS content, the experimental results demonstrate that PDMS incorporation efficiently transitions WGC from hydrophilic to hydrophobic behaviour. These findings were further confirmed by complementary capillary water absorption tests, which showed a gradual decrease in water uptake. These reductions culminated in the 5% PDMS formulation (G-5) achieving an impressive 0.93% absorption rate, representing a 76% improvement over the unmodified control specimen. To gain essential insights into the underlying modification mechanisms, microstructural analysis using MIP was employed. This study revealed that PDMS serves a dual role as both a pore-sealing agent and a network modifier. While the admixture effectively sealed small and medium pores, it occasionally created a limited number

of larger, isolated voids, particularly at higher concentrations. This phenomenon explains the apparent discrepancy in the 5% PDMS mix (G-5), which exhibited both the lowest porosity/intrusion volume, as well as a relatively high average pore diameter. Importantly, permeability measurements confirmed these larger voids remained poorly interconnected, maintaining the composite's overall impermeability despite their presence. The comprehensive dataset demonstrates that PDMS modification operates through multiple complementary mechanisms to enhance WGC performance. Through integrated analysis of surface hydrophobicity, water absorption resistance, and pore structure characteristics, this study identifies 5% PDMS as the optimal dosage for WGC enhancement. This concentration achieves an exceptional balance between surface water repellency (a 93.9° contact angle) and internal barrier properties (0.93% water absorption), without compromising the integrity of the matrix. The findings position PDMS-modified geopolymer as a promising sustainable material for demanding applications in moisture-prone environments, offering superior durability through simultaneous surface protection and bulk matrix refinement. The research provides both a fundamental understanding of modification mechanisms and practical guidance for material optimisation in field applications.

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Conflict of Interest

The authors declare that they have no conflict of interest regarding the publication of this paper.

Author Contribution

The authors confirm contribution to the paper as follows: ***Study conception and design****: Warid Wazien Ahmad Zailani, Nazirah Mohd Apandi;* ***Data collection:*** *Maisarah Nur Norain, Nur Hanan Hazwani Askaimi Abot, Zadariana Jamil;* ***Analysis and interpretation of results:*** *Warid Wazien Ahmad Zailani, Naeem Aziz Memon, Adeyemi Adesina;* ***Draft manuscript preparation:*** *Maisarah Nur Norain, Naeem Aziz Memon. All authors reviewed the results and approved the final version of the manuscript.*

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