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Sustainable Valorisation of Silane-Treated Waste Glass Powder in Concrete Pavement

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Abstract: This research presents new insights into the utilisation of waste glass powder in concrete pavements. Two different types of glass powder were used as a partial replacement for sand: 10% neat glass powder (untreated) and 10% silane-treated glass powder. The interfacial bonding properties, physical properties, and mechanical properties of concrete pavement were assessed at 7 and 28 days. Results exposed a reduction of 5% and 2% in the compressive and flexural strengths, respectively, and an increase of 15% in water absorption after the addition of neat glass powder to concrete after 7 days of curing. This is due to weak interfacial bonding between the glass powder and cementitious matrix. However, the incorporation of silane-coated glass powder led to an increase in the compressive and flexural strengths by more than 22% and 28%, respectively, and reduced the water absorption of concrete by 8%, due to the coupling functionality of silane. After 28 days of curing, the compressive strength of concrete increased by 15% and 22% after the addition of neat glass powder and silane-treated glass powder, respectively. In addition, water absorption dropped by 5% and 7% after the incorporation of neat glass powder and silane-treated glass powder.

Keywords: glass powder; sustainable development; concrete; silane; morphology; strength

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1. Introduction

The sustainable construction concept emerged two decades ago to ensure green development in the construction industry and to guarantee efficient use of resources in the industry [1–4]. However, the rise of sustainable construction involved many challenges throughout the design, management, implementation, operation, and maintenance phases of construction projects [1–6]. Moreover, moving toward sustainable construction, a significant challenge in material selection and valorisation was created, where the construction sector consumes more than 60% of natural resources as building materials [2,7–9]. Concrete is one of the construction materials that consumes more than 20 billion tonnes of raw material every year [10]. Aside from the high consumption of raw materials in the concrete production process, the manufacturing of its main constituent, cement, accounts for more than 80% of the total CO_2 emissions from concrete production [11–14]. Additionally, the production of coarse and fine aggregates is responsible for the other 20% of CO_2 emissions from concrete production [11,15]. As a result, researchers started to explore other alternatives, either by replacing cement and aggregate with environmentally friendly materials, or by replacing them with recycled materials [16–20].

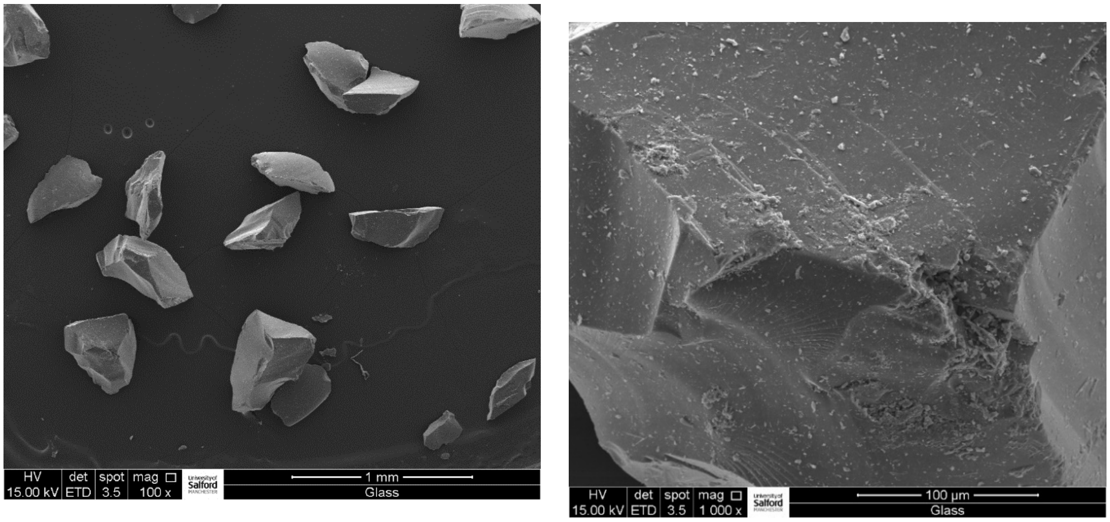
In recent years, more research, considering the influence of highway construction on the environment, started to emerge, which highlighted its vulnerability and the environmental risks that pavement construction imposes on the ecosystem after the deterioration of pavements [21]. Therefore, the sustainable construction of highways has recently arisen in order to downgrade its greenhouse gas emissions and reduce the dependence on virgin materials in its construction [22]. Many materials were used in concrete pavement as alternatives for cementitious binders, such as fly ash and geopolymers and recycled aggregates and rubber as alternatives for virgin aggregate [23–29]. Moreover, glass waste was one of the materials that was widely used as an alternative for both cement and aggregate in concrete [30–34]. In addition, glass waste was implemented as an eco-alternative to aggregates in asphalt mixtures in most of the flexible pavement layers, especially in the lower, coarser layers [35–37]. In addition, some research focused on the replacement of bitumen with glass powder in small ratios up to 10% [38,39]. However, not many studies discussed the incorporation of waste glass powder into concrete pavement in particular [40].

According to the United Nations environment program report of 2004, 7% of the solid wastes around the world, at that time, were composed of glass [41,42]. Moreover, all of these glass wastes go to a long-lasting landfill, impacting the environment [41]. Accordingly, recycling of glass waste became of high importance to researchers and decision-makers around the world to reduce its negative impact on the environment. Scenarios of different recycling techniques were suggested and followed, such as recovering and reusing glass waste and implementing it in ceramic production, container production, and the production of stoneware tiles [43–45]. Incorporating glass wastes into concrete started to gain importance, as well, to overcome this issue and, potentially, to enhance concrete's properties. The presence of more than 72% silicon and 11% calcium in the composition of glass wastes allows it to act as a pozzolan when incorporated within concrete mixtures and, at the same time, the potential to replace sand (sand contains 78% silicon) [33,34,41]. Furthermore, replacing virgin aggregate with glass wastes in concrete has shown promising results, despite its negative effect on some of the concrete's properties [42]. One of the main drawbacks of replacing sand with glass wastes was the propagation of cracks in the produced concrete due to the alkali–silica reaction [34,41,46]. Moreover, a reduction in the compressive strength of concrete was reported by many researchers after replacing sand with glass wastes [47–50]. Accordingly, the current work will discuss the possibility of treating glass powder with an environmentally friendly hydrophobic compound, 2-methoxyethoxy (vinyl) silane, before incorporating it into concrete as a replacement for sand. Silane materials are widely used hydrophobic compounds with the advantage of low cost and easy application methods as they can be applied by spraying, brushing, or soaking [51–53]. Therefore, the main objectives of this research are (1) to study the influence of replacing sand with treated glass powder on the performance of concrete, including mechanical and physical properties, and (2) to assess the interaction between glass powder, treated with silane, and the cement matrix.

2. Experimental Methodology

2.1. Materials

Coarse aggregate from crushed granite with 20 mm nominal maximum grain size, 2 mm fine sharp river sand, and CEM I Portland cement conforming to BS EN 197-1, were used in this research [54]. Glass wastes were obtained from Potters Ballotini Ltd. (Manchester, UK), which were ground into fine powder with a particle size of 0.2–0.5 mm (see Figure 1 for microstructure). Glass powder was coated with Vinyl tris(2-methoxyethoxy) silane prior to incorporation into concrete. The Vinyl tris(2-methoxyethoxy) silane material was provided by Sigma-Aldrich (London, UK), with no components considered to be persistent, bioaccumulative, or toxic. Vinyl tris(2-methoxyethoxy) silane is a vinyl-functional coupling agent with hydrophobic properties that improves the adhesion of inorganic substrates [55].



(**a**) (**b**)

Figure 1. Microstructure of (**a**) glass powder particles (100×) and (**b**) a magnified glass powder particle (1000×).

2.2. Mix Design and Formulation

In accordance with the British Standard BS 1881-125, a concrete mixture with water to cement ratio (*w*/*c*) of 0.40 was prepared, which is a commonly used mix in road construction [56,57]. The matrices used in this research consisted of a control mix, a mix with fine aggregate replaced with 10% glass powder, and a mix with fine aggregate replaced with 10% glass powder and treated with Vinyl tris(2-methoxyethoxy) silane. Due to the low *w*/*c* ratio, all concrete mixtures were compacted by using a mechanical vibrator to ensure full compaction of mixtures. Silane was applied to waste glass by spraying, following the manufacturer's guidelines. The mix design of the used concrete is shown in Table 1.

Table 1. Mix proportions of the used concrete.

Ingredient	Amount (kg/m^3)
Cement (CEM I/42.5 N; sulphates <3.5%, chlorides <0.10%, and initial setting time 40 min)	450
Water	180
Fine aggregate (sharp river sand with a uniform grain size distribution between 2 mm and 300 μm)	678
Coarse aggregate (crushed granite with sharp edges and maximum size of 20 mm)	1092

2.3. Test Specifications and Specimens

A total of 36 samples were produced: 18 cubes with the dimensions of 100 mm × 100 mm × 100 mm and 18 beams with the dimensions of 100 mm × 100 mm × 500 mm. Of these, 12 samples (6 cubes and 6 beams) were used as controls, 12 samples were produced with 10% glass powder, and 12 samples with 10% glass powder treated with silane. All samples were cured in a water tank at a temperature of 23 °C for 7 days and 28 days before testing.

After completing the curing periods, samples were dried in an oven at a temperature of 75 °C until a constant mass was achieved.

The experimental program was divided into six main sections: (1) workability, (2) microstructural analysis, (3) hydrophobicity, (4) water absorption, (5) compressive strength, and (6) flexural strength. Investigating physical properties such as workability, hydrophobicity, and water absorption is of great importance for concrete pavements. Workability testing will ensure the use of a proper mix design for pavements, where the *w*/*c* ratio should be kept to a minimum, as indicated by previous research [57]. Furthermore, due to the complications that water would bring to concrete pavements, especially when construction joints exist in pavements, it is useful to determine the effect of incorporating glass waste into pavement on its hydrophobicity and water absorption.

Scanning electron microscope (SEM) analysis was performed on all mixes to investigate the morphology of the used glass powder (treated and untreated) and its interfacial bonding with the cementitious matrix. Samples were coated with a gold film to make them conductive before placing them in the scanning electron microscope at 15 kV.

The workability of all mixes was determined by performing a slump test to assess the influence of replacing sand with glass powder [58]. The values were compared with control mixes.

The influence of using neat glass powder and glass powder treated with silane on the hydrophobicity of concrete and their ability to reduce water absorption was assessed by conducting a contact angle test. The contact angle (θ°) between a sessile water drop and the surface of concrete was measured by using a goniometer device [59,60].

The water absorption of all the used concrete mixtures was assessed after 7- and 28-day curing periods following the guidelines of the ASTM D 6489 [61]. The concrete cubes were weighted at the intervals of 24 h, 48 h, and 72 h after placing them in water, and their water absorption percentage was calculated by the following equation:

$$Percent\ Absorption\ (\%) = \frac{W_2 - W_1}{W_1} \times 100 \tag{1}$$

where,

W_1: dry weight of concrete samples (g);
W_2: weight of concrete samples after immersion in water (g).

Mechanical properties of concrete were assessed through compressive and flexural strength tests. The compressive strength test was performed on all concrete cubes after the curing periods of 7 days and 28 days ended, following the instructions of the British Standard BS EN 12390-5 [62]. Furthermore, the flexural strength of all mixes was determined by conducting the two-point loading method after 7 days and 28 days of curing, following the recommendations of the BS EN 12390-3 [63]. Figure 2 presents an illustration of the testing programme for this research.

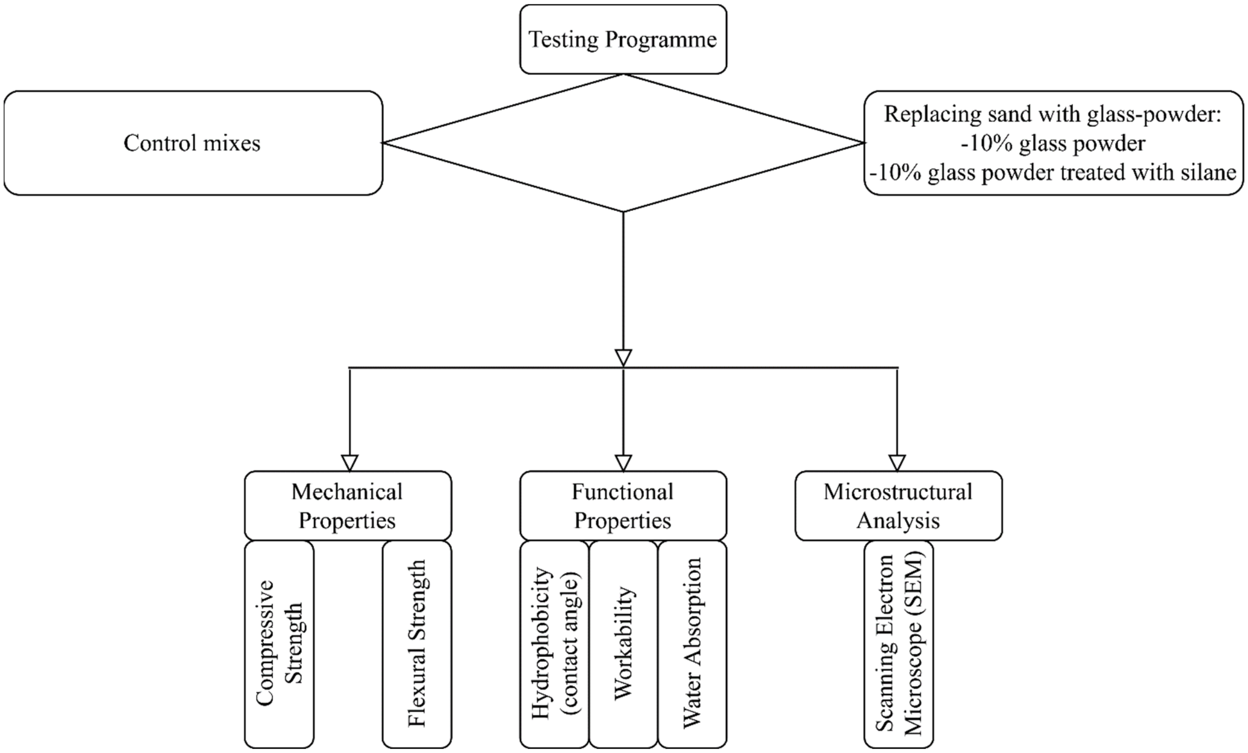


Figure 2. Testing programme chart.

3. Results and Discussion

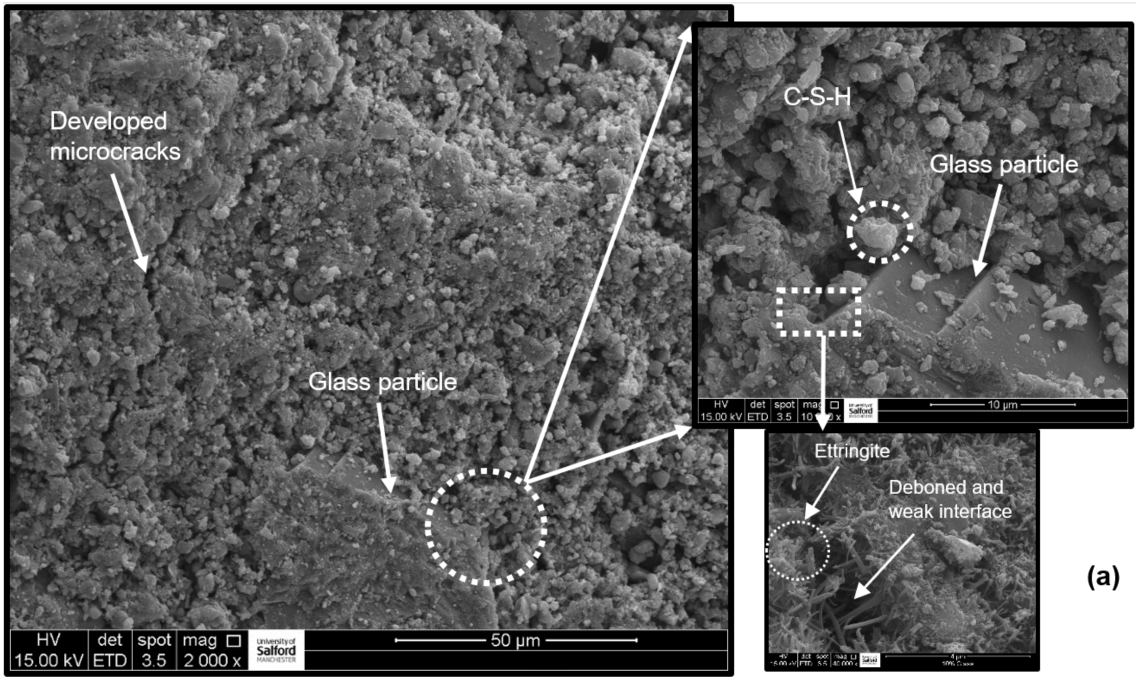
3.1. Microstructural Analysis and Interfacial Bonding Properties

The morphological analysis of untreated and silane-treated glass powder concrete specimens was conducted to elucidate the interaction system between hydrated cement and glass powder (treated and untreated) and to establish an aggregated understanding of their performance. Figure 3 illustrates the morphology of untreated and silane-treated glass powder concrete after analysing it under SEM.

As shown in Figure 3a, after the replacement of sand with neat glass powder (without treatment with silane), concrete showed a moderately high number of developed microcracks with an average width of 3 μm. This might refer to the reaction between the high content of silica in glass powder and calcium hydroxide in the hydrated cement, which leads to the formation of silica gel, which has a high affinity to water. Absorbing the silica gel to water eases its swelling and causes high pressure on the interface between the cementitious matrix and glass particles, resulting in the development of a large number of cracks [34,64,65]. Moreover, the surface of glass powder can be seen to have a rough texture (Figure 3a), which might reduce the filling effect that glass powder exerts inside the pores [66]. Accordingly, the overall strength and rigidity of the concrete matrix could be negatively affected due to the absence of support inside the pores. Additionally, it can be seen from Figure 3a that C-S-H is distributed with low quantities in the concrete mix, which might be caused by the hindering effect of glass powder on the hydration process of concrete, where the hydration of cement is faster in the absence of glass [66]. Ettringite (needle shape) is observed to form in large quantities after the inclusion of glass powder in the mix, which decomposed the bond between the cement paste and glass particles.

Interestingly, replacing sand with glass powder treated with silane improved the resistance of concrete to crack propagation (Figure 3b), as fewer cracks were observed compared with untreated glass powder concrete. This refers to the effect of silane in increasing the adhesion between glass particles and cement paste [67]. On the other hand, the surfaces of the glass particles were observed to have smooth surfaces due to the presence of silane as a coating on the particles (Figure 3b). This would increase the filling effect of glass powder within the pores and enhance its bonding with all concrete constituents through the developed silanol group (Si-OH) on the surface of glass, resulting in enhanced strength (Figure 3b).

With reference to Figure 3b, it is evident that a denser structure with large quantities of C-S-H was formed, especially at the interface between glass and the cementitious matrix, after the treatment of glass with silane. The presence of the hydroxyl group in the formed silanol (Si-OH) might have promoted the coupling effect of silane, which enhanced the linking between C-S-H matrices on one side and the linking between C-S-H matrices and other constituents of cement on the other side [68]. Accordingly, the presence of silane on the surface of glass might enhance the toughness of the inner structure of concrete. Moreover, ettringite was observed to form in smaller quantities when compared with untreated concrete, which might improve the strength of concrete.



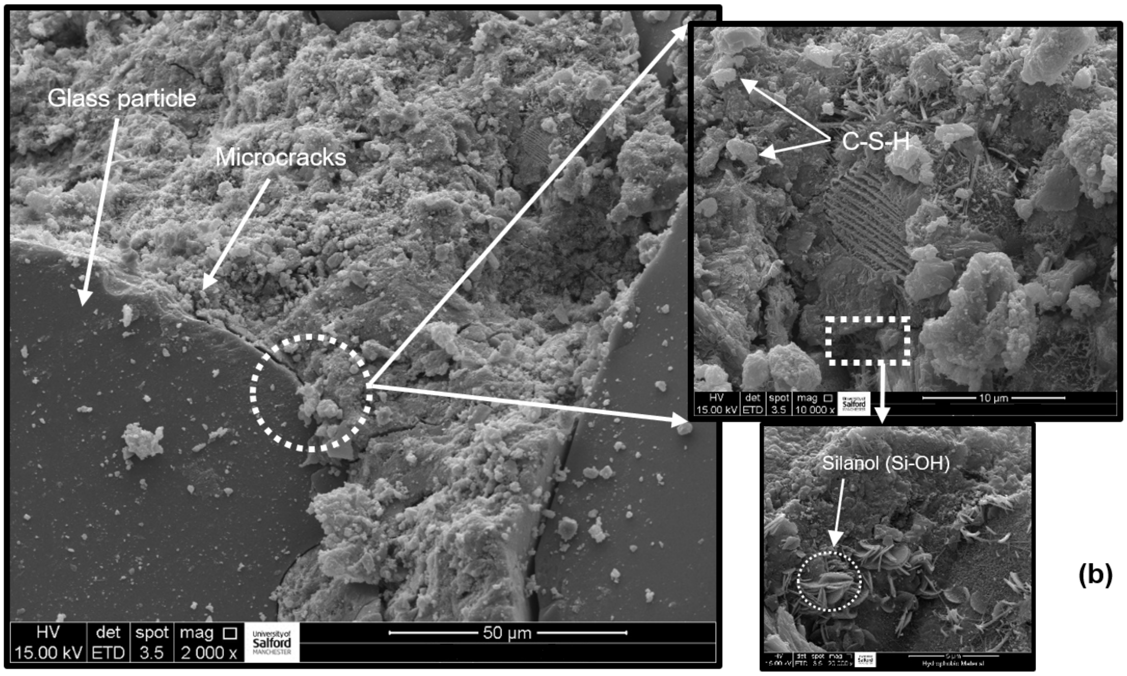


Figure 3. Microstructural analysis of (**a**) untreated glass powder concrete and (**b**) glass powder concrete treated with silane.

3.2. Functional Properties

3.2.1. Concrete Consistency

The slump test was used to differentiate the workability of the employed concrete mixes and explore the influence of neat and treated glass powder on the rheology of concrete. The slump test was performed by filling the standard cone of the BS EN 12350-2 with concrete, and the fall in the concrete's height was measured. Table 2 illustrates the slump values of all the mixes.

Table 2. Consistency of the used concrete mixes.

Concrete Mix	Slump (mm)
Control	29
Concrete with 10% glass powder	31
Concrete with 10% glass powder and treated with silane	33

Results from Table 2 demonstrate a clear increase in the slump values of both glass powder concrete and silane-treated glass powder concrete. However, the increase in silane-treated glass powder concrete was more than that in concrete with neat glass powder. The reduction in the slump of glass powder concrete may refer to (1) the hydrophobic nature of glass, which decreases its absorption to water, and (2) the high surface tension of glass powder, which repels water from its surface [69,70]. On the other hand, the high increase in the slump of silane-treated glass powder concrete is related to the presence of silanol composites on the surface of the glass, which increased its hydrophobicity and reduced its affinity to water. The reduction in the absorbed water from both neat glass powder and silane-treated glass powder would serve to increase its amount in the mix, which would increase the overall workability of concrete.

3.2.2. Hydrophobic Effect

The effect of glass powder and silane-treated glass powder on reducing the affinity of concrete to water and reducing its absorption to moisture was measured through the contact angle test (Figure 4). It is clear from Figure 4 that the addition of neat glass powder and silane-treated glass powder served to increase the contact angle of concrete by more than 42% and 69%, respectively, when compared with the control. The amorphous and smooth texture of the glass powder and the relatively dense structure of the formed concrete matrix, after the addition of glass, are capable of increasing the contact angle of concrete. Additionally, the size of the used glass particles is finer than sand, which would increase the compactness of the mix and reduce its porosity [34]. As a result, this would increase the hydrophobicity of glass powder concrete.

The presence of silane on the surface of the used glass powder increased the water contact angle to higher levels than untreated glass powder concrete and control mixes (Figure 4). Silane will react with calcium hydroxide in the mix and form a hydrophobic sheet of silanol (Si-OH) in the interface between glass particles and cement paste, as seen in Figure 3. The presence of silanol in the interfacial area between glass and the cementitious matrix would serve to increase the hydrophobicity of the mix. In addition, silane is expected to reduce the porosity of concrete by filling its pores and decreasing the affinity of concrete to water, which will increase its contact angle.

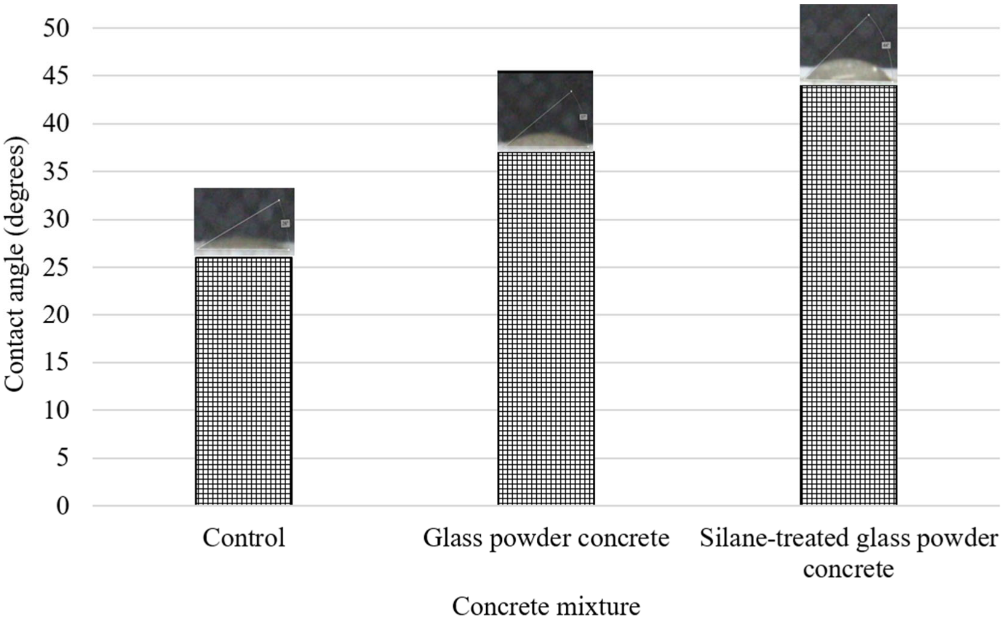


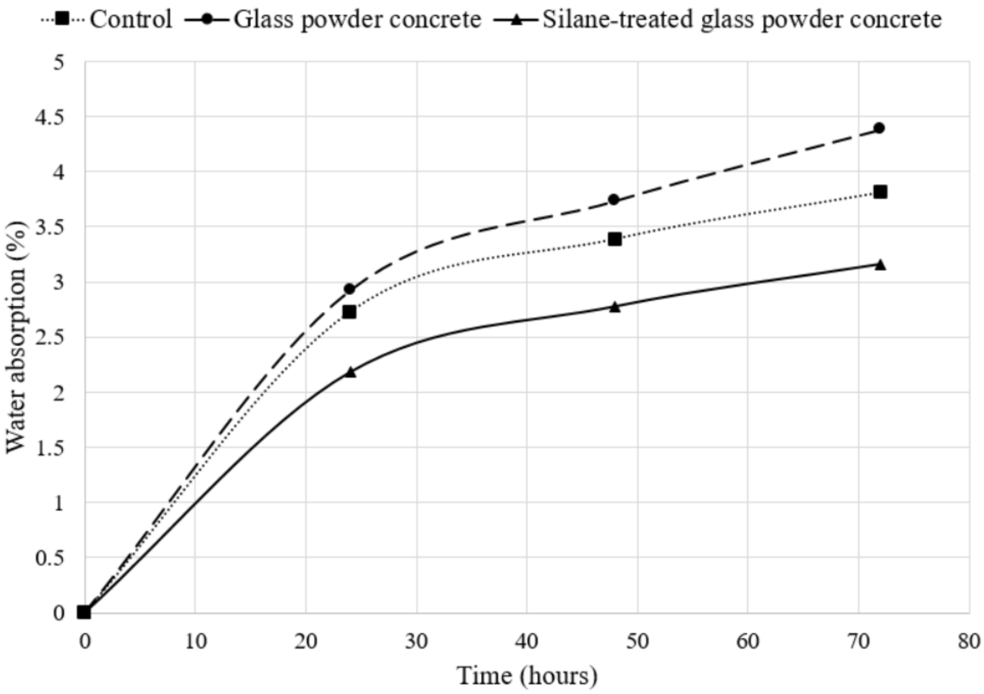
Figure 4. Hydrophobicity of the used concrete mixtures.

3.2.3. Water Absorption

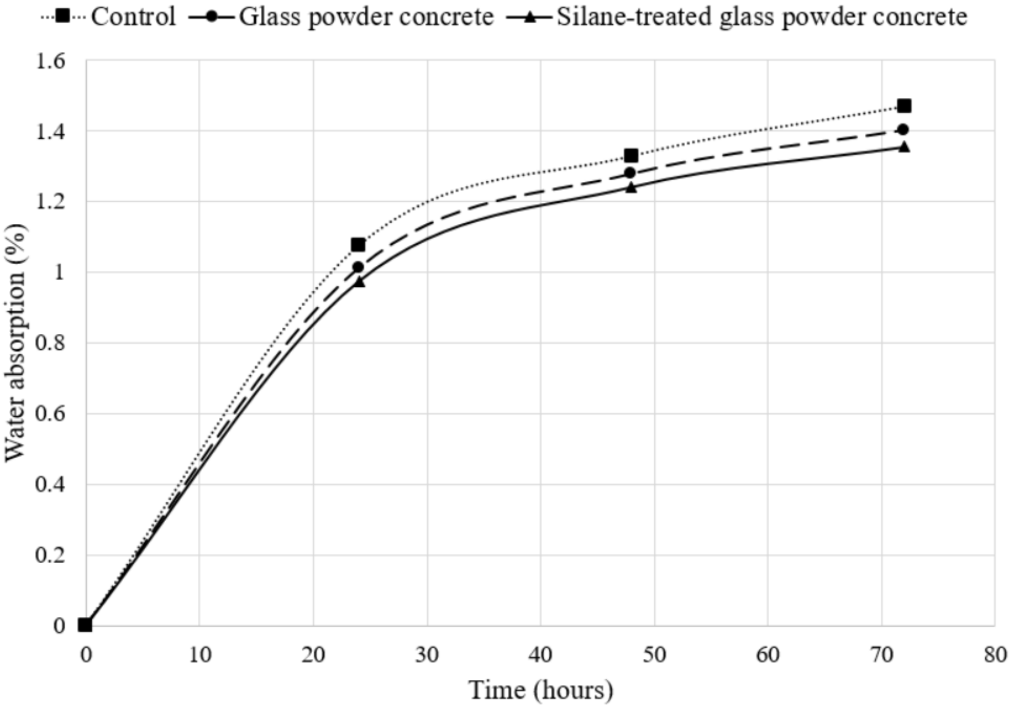
In order to examine the efficacy of glass powder and silane-treated glass powder in reducing the permeability of concrete, all concrete specimens were tested for water absorption at 7 and 28 days for a duration of 72 h. As shown in Figure 5a, replacing sand with neat glass powder increased the water absorption of concrete, at the age of 7 days, by more than 15% when compared with the control. This can be attributed to the presence of a high number of microcracks at the early stage of 7 days due to the incomplete hydration process [34,46]. In addition, the size of the pores of concrete at this early stage would be large and unfilled with glass powder, allowing for more water absorption [46]. On the other hand, an evident enhancement in the impermeability of concrete after the replacement of sand with silane-treated glass powder was noticed after 7 days (Figures 5a). The lower water absorption of silane-treated glass powder concrete, when compared with other mixes, may refer to the hydrophobic effect of the added silane that repels water out of the pores, reducing concrete's affinity to water, even in the presence of microcracks at this early stage.

In contrast, the influence of replacing sand with neat glass powder at 28 days was observed to be more effective in reducing the permeability of concrete when compared with the control (Figure 5b). At the age of 28 days, the degree of hydration would reach higher levels, which would serve to reduce the number of developed microcracks and increase the filling of pores with glass powder. In addition, despite the rough texture of glass powder observed in Figure 3a, the presence of glass powder in the pores would serve to reduce the absorption of water when compared with traditional concrete with sand. This could be attributed to the relatively smoother surface of glass compared to sand.

The presence of silane-treated glass powder within the mix was found to have the lowest water absorption rate between all mixes after 28 days (Figure 5b). This may refer to: (1) the presence of silane on the surface of glass increasing the cohesion between the glass particles and cementitious matrix, which would result in the formation of fewer microcracks in the matrix and reduce its porosity when compared with other mixtures (as illustrated in Section 3.1); and (2) the hydrophobic effect of silane would reduce the water absorption of concrete.



(**a**)



(**b**)

Figure 5. Water absorption rate of all concrete mixtures at the age of (**a**) 7 days and (**b**) 28 days.

3.3. Mechanical Properties

3.3.1. The Effect of Glass Powder on Compressive Strength

As seen in Figure 6, silane-treated glass powder concrete achieved the highest compressive strength levels between all mixtures, with an increase of 15% and 22% at 7 days and 28 days, respectively, when compared to the control. The presence of silane within the added glass powder promotes the linking between the glass particles and the cementitious matrix, which is reflected in the formation of a denser structure and more refined pores (as shown in Figure 3b). In addition, silane will create a thin film on the surfaces of glass particles that will serve to increase the bonding between the glass and cement matrix and increase the concrete toughness.

On the other hand, the addition of neat glass powder to concrete reduced the 7-day compressive strength by 5% and increases the 28-day strength by 15% when compared to the control (Figure 6). The reduction in the 7-day strength may be due to the development of microcracks at this early stage due to the incomplete hydration process. In addition, at the early stage of 7 days, fine glass powder particles perform as a catalyst for the hydration process and not as a pozzolanic material, which slows the strength development of concrete [34]. The increase in the 28-day compressive strength of the neat glass powder concrete (Figure 6), when compared to the control, is evident for the filling effect that glass particles play in the pore structure of concrete, which reduced the propagation of microcracks. In addition, the presence of high silica (SiO_2) content in glass powder will promote the pozzolanic reaction with alkalis in the cementitious matrix and increase the strength development of concrete. Despite this increase in compressive strength, the developed strength of neat glass powder concrete is still lower than that of silane-treated glass powder concrete. Based on the microstructural analysis in Figure 3, the number of developed microcracks is moderately higher in neat glass powder concrete than that in silane-treated glass concrete, which serves to reduce the strength of neat glass powder concrete.

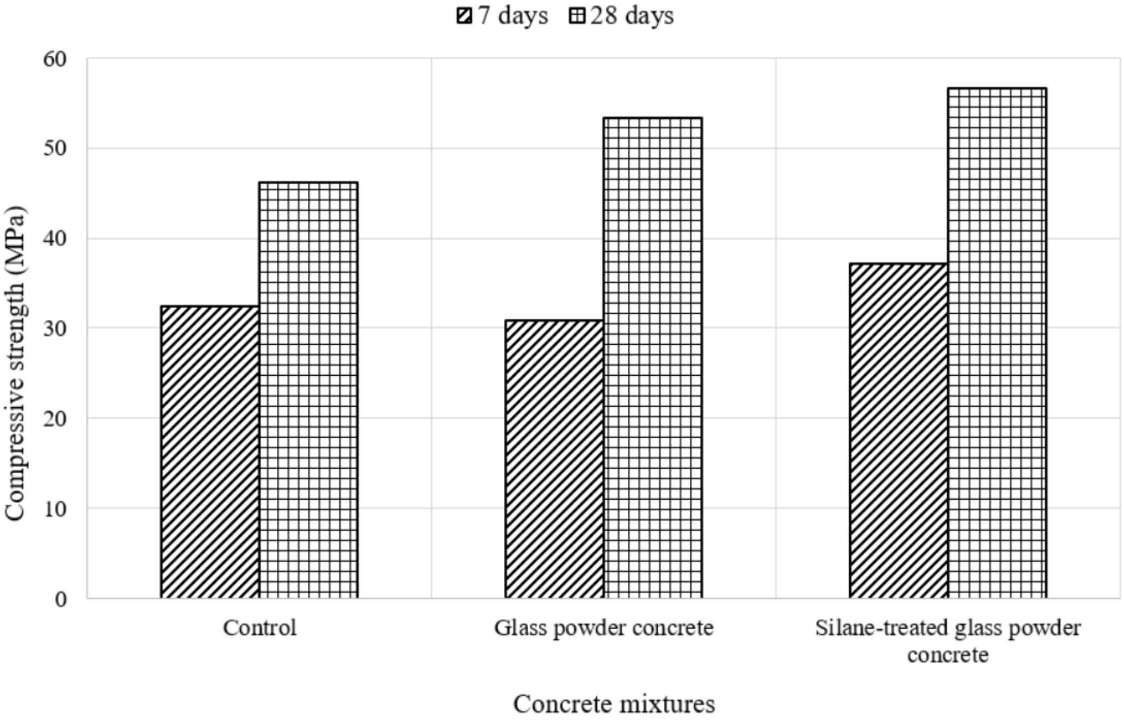


Figure 6. Compressive strength comparison of all concrete mixtures at the age of 7 days and 28 days.

3.3.2. The Effect of Glass Powder on Flexural Strength

The flexural strength of all tested concrete mixtures showed similar trends to those obtained by compressive strength. As depicted in Figure 7, replacing sand with neat glass powder reduced the 7-day flexural strength by 2% and increased the 28-day flexural strength by 10%, when compared to the control. However, the highest flexural strength of concrete was obtained after replacing sand with silane-treated glass powder, where an increase of 10% and 28% was observed at 7 days and 28 days, respectively. The addition of neat glass powder to concrete was noticed to have a negative effect at early stages (as shown in Section 3.1). In contrast, at later stages and with the progress of the hydration process, pores will start to be filled with glass powder, which will increase the strength of concrete. On the other hand, the presence of silane within the matrix will promote the linking between glass particles and hydrated cement through the developed silanol group (Si-OH) on the surface of the glass (Figure 3b).

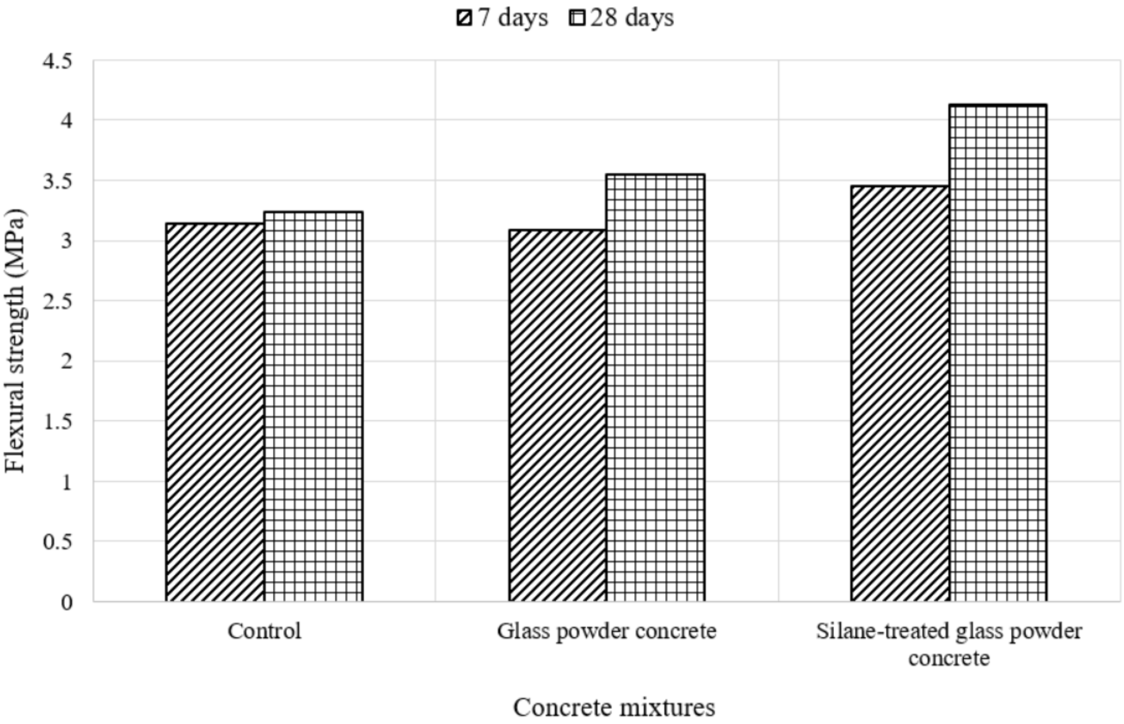


Figure 7. Flexural strength development of all the used concrete mixtures at the age of 7 days and 28 days.

4. Conclusions

In this study, the influence of replacing sand with glass powder on the properties of concrete pavement was investigated. Sand was replaced with 10% neat glass powder (untreated) and 10% silane-treated glass powder. Different properties of concrete were studied after the inclusion of treated and untreated glass powder, including interfacial bonding and mechanical and physical properties. The following conclusions can be drawn:

1. The microstructural investigation was able to show the changes in the morphology of concrete after the addition of treated and untreated glass powder, which explained the reasons behind the changes in the physical and mechanical properties of concrete.
2. Untreated glass powder can have a negative effect on concrete at the early age of 7 days and lead to increased water absorption and decreased compressive and flexural strengths compared to the control. In contrast, a decrease in water absorption and an increase in compressive and flexural strength were observed at later stages due to the progression of the hydration process and the filling effect of glass particles in the pores.
3. The silane coating of the added glass powder can enhance the compressive and flexural strengths and the impermeability of concrete at 7 days and 28 days. This is probably due to the coupling effect of silane, which enhances the linking between glass particles and the cementitious matrix.
4. The hydrophobicity of concrete was observed to increase after the inclusion of glass powder. However, higher levels of hydrophobicity were obtained after the treatment of glass powder with silane. Glass powder has an amorphous and smooth texture that increases the contact angle of concrete. Adding silane to glass powder will increase the contact angle to higher levels. This can be reflected in the enhanced impermeability of concrete.

Further work is underway to study the influence of replacing sand with higher ratios of silane-treated glass powder on the performance of concrete pavement.

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