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Effect of water-to-cement ratio and soaking time of waste glass powder on the behaviour of green concrete

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ABSTRACT

Sustainability in the construction is becoming a major challanges due to the depletion of natural resources and the avialibility of landfill capacity. Additionally, there is an urgent need to improve the properties of concrete and produce environmentally friendly green concrete. From these points, the effect of varying the water-to-cement (w/c) ratios and the soaking time of glass powder (GP) on the activation of the pozzolanic reactivity and the mechanical properties of green concrete were examined. Two w/c ratios (0.6 and 0.7), two glass contents (5% and 30%), and six GP soaking times werer used in this study. The new mixing method (NMM) was used, in which the GP is partially dissolved in water and more Na^{+} ions than Ca^{+2} are formed in solution since Na^{+} ions have higher mobility than Ca^{2+} ions. The results indicate that greater normalized compressive strength (CS) is achieved under a w/c ratio of 0.7 for 5% and 30% GP content with soaking time of 6 h. This increase is correlated with the formation of more nucleation sites due to the growth of calcium silicate hydrate rather than portlandite. The relative activation index calculation showed that the 30% GP mixes have higher values than 5% GP mixes.

1. Introduction

Sustinable use of natural resources and effective waste utilization in green concrete is becoming increasingly necessary as natural resources become more limited. Further, the exponentially demand of individual habitaion has led to increased demand for new construction. Fabricating new bottles, flasks, or other tools from glass waste is infeasible due to the amount of energy required (collection, transportation and melting). Therefore, researchers have aimed to reuse crushed waste glass or glass powder (GP) as an aggregate in concrete or as a replacement for cement. The amorphous structure and chemical composition of GP and some types of ash are similar to those of cement, and thus induce a similar pozzolanic effect in concrete. The pozzolanic reaction involves the consumption of the weaker portlandite (CH) to produce calcium silicate hydrate (C-S-H), which has a higher CS and density [1–4].

The traditional or conventional mixing method (TMM), in which GP and water are directly mixed into the concrete mixture, has been used extensively to investigate the effect of using GP in concrete [5]. In the new mixing method (NMM), the GP is presoaked in water to partially dissolve it before it is mixed into the concrete mixture, thus activating the pozzolanic reaction earlier in the life (at 90 days for concrete made with TMM) of the concrete via hydrolysis of the GP and producing stronger concrete than the control [6,7].

The mechanical properties of fresh and hardened concrete using GP depend on the chemical composition, the diameter of the GP particles, and the curing conditions [8,9]. The mixing method used also contributes to the mechanical properties; the NMM allows for the partial dissolution of GP and inhance the compressive strength of the concrete [6,10,11].

The packing filling effect (PFE) and the pozzolanic reactivity enhance the mechanical behavior of concrete made with GP [12]. The shape of the GP particles impacts the PFE; an angular shape of the particles decreases the PFE, whereas round particles increase the PFE by increasing the density of the interfacial transition zone (ITZ) between the cement paste and the aggregates [13]. This increase in the PFE reduces the porosity of the concrete, thus improving its CS [14].

The consumption of CH by the pozzolanic reaction increases the concentration of secondary C-S-H and thus also reduces the porosity and increases the CS of the concrete [15–17]. The pozzolanic reactivity has been demonstrated to be responsible for the increase in concrete's CS after 90 days [4,18].

GP can be dissolved in mixtures with pH values between 12 and 13,

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Table 1
Cement type II (CEM II) and the glass powder (GP) composition.

Composition by Mass %	GP	CEM II
CaO	18.55	66.69
SiO_2	64.94	18.84
Al_2O_3	1.81	6.3
Fe_2O_3	1.97	3.72
SO_3	0	2.66
P_2O_5	0	0.70
MgO	3,12	0.61
K_2O	0.44	0.5
Na_2O	9.16	0

Table 2
Physical properties of aggregates.

Properties	Unit
Bulk Gs (SSD)	2,6 g/cm^3
Bulk Gs (Dry)	2,5 g/cm^3
Bulk Gs (Apparent)	2,7 g/cm^3
Absorption	3,6 %
Abrasion	21,5 %
U. Weight	1,4 g/cm^3

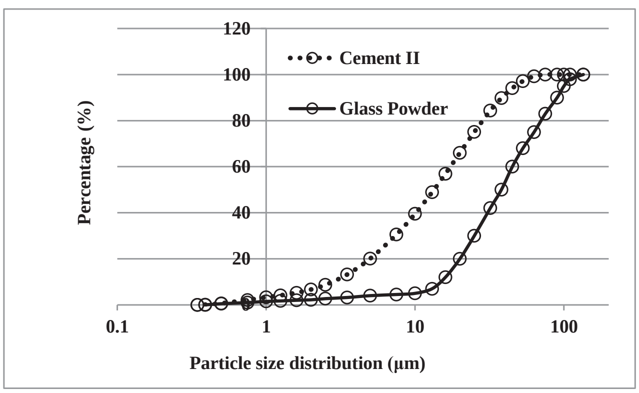

Fig. 1. Particle Size Distribution of cement and glass powder.

which is the pH range of concrete [19,20]. Fine GP has a high dissolution potential in the concrete pore solution at an early age (at 2 days) [21]. The dissolution of fine GP accelerates the binding between the aggregates and the cement paste via the densification of the ITZ [22].

As the GP is dissolved in water with pH between 12 and 13, the produced Na^+ ions have better mobility than the Ca^{2+} ions. The high surface area of the GP particles increases the exchange between ions [23].

As the GP soaks in water, the Na^+ concentration drops. Hydrolysis attack (rupture of chemical bond by water) leads to the formation of a silanol group on the silica's surface, which dissolves the silica and then reacts with OH^-ions present in the solution, according to the following equation.

$$SiOH + OH^- \rightarrow SiO + H_2O \tag{1}$$

The reaction between SiO and $H_3SiO_4^-$or $H_2SiO_4^-$depends upon the pH of the mixture according to the following two equations.

$$Si(OH)_4 \leftrightarrow H_3SiO_4^- + H^+ \text{ when pH} = 9.8 \tag{2}$$

$$H_3SiO_4 \leftrightarrow H_2SiO_4^- + H^+ \text{ when pH} = 11.8 \tag{3}$$

The Na^+ ions then react with H_2SiO_4 and $H_3SiO_4^-$ to form sites on the surface og GP. Higher dissolution of Na^+ ions occurs when the SiO_2: Na_2O ratio is equal to 5% [13].

After setting for 60 days, concrete mixes prepared with finer GP more had outer C-S-H observed near the GP particles. A reaction rim has been observed around the GP where the area between grains are well-filled [24].

Higher-density concrete has lower water absorption and porosity, which increases its performance due to the densification of the ITZ in destructive environments [25].

The water absorption of concrete has an opposite relationship with a high GP content and low water-to-cement (w/c) ratio due to the hydration degree if the concret [26,27]. Increasing the w/c ratio decreases the CS early in the life of the concrete due to the dilution effect of GP in the structure of concrete [28].

Prior researches have indicated that the PFE and the pozzolanic reaction improve the CS at early stage of the concrete particularly when the NMM is used. A lower leactched Ca^{2+} to Na^+ ratio in the concrte mixed water increases the CS [6,10,11].

Thecontribution of this work is to examinethe effect of varying the water-to-cement (w/c) ratios and the soaking time of glass powder (GP) on the activation of the pozzolanic reactivity and the mechanical properties of green concrete.

2. Experimental design

Cubic specimens (100 × 100 × 100 mm) with and without GP (≤75 µm in diameter) with two w/c ratios (0.7 and 0.6) and six dissolution times of the GP in water before mixing (0, 1, 2, 3, 6, and 12 h) were examined. Samples were tested via compressive tests at 2, 7, 14, 28, and 90 days after pouring to evaluate the mechanical behavior.

2.1. Chemical composition of cement and GP and particle size distribution

The same materials used in prior work were used; readers can refer to the work of Elaqra and Rustom [4]. Detail on the properties of the cement, GP and aggregates are provided in Table 1. The GP had a higher SiO_2 content (around 65%) whereas the cement had a higher CaO content (around 67%).Table 1

2.2. Physical properties and particle size distribution of aggregate

Fig. 1 shows the particle size distruction of cement and GP used in the study. The particle size distribution of the GP, cement, and aggregates was obtained using the standard test method for sieve analysis of fine and coarse aggregates (ASTM C136/C136M-14) [29]; readers are encouraged to refer to the work of Elaqra et al. [6,10,11] for particle size distribution results.

The cumulative particle size distribution of the aggregates shows a well-graded distribution located between the minimum and maximum limits authorized by the mix design as shown in Fig. 2 [6,10,11].

Physical properties of aggregates are given in table below.

2.3. Mixing methods and proportions

The NMM was employed, as detailed in ASTM C192/C192M-16a [30] for TMM and by Elaqra et al. [6,10,11], where GP was immersied in the water befor mixing in the concrete. Tap water was used in mixing. Control concrete mixes were prepared according to the w/c ratios equal to 0.4, 0.45, 0.5, 0.6, 0.66 and 0.7. Concrete blends using GP were made using 0%, 5%, and 30% GP, as detailed below in Table 3. The same table was used for mixes with w/c raio equal to 0.7, the amount of water is equal to 210 kg. where FA is the fine aggregate, MA is the medium aggregate and CA is the coarse aggregate.Table 3

2.4. Dissolution of glass powder in water before mixing with concrete

The glass powder was immersed in water and the amount of water was calculated according to the amount of water of each mixes. The flame atomic absorption spectrophotometry was used to determine the

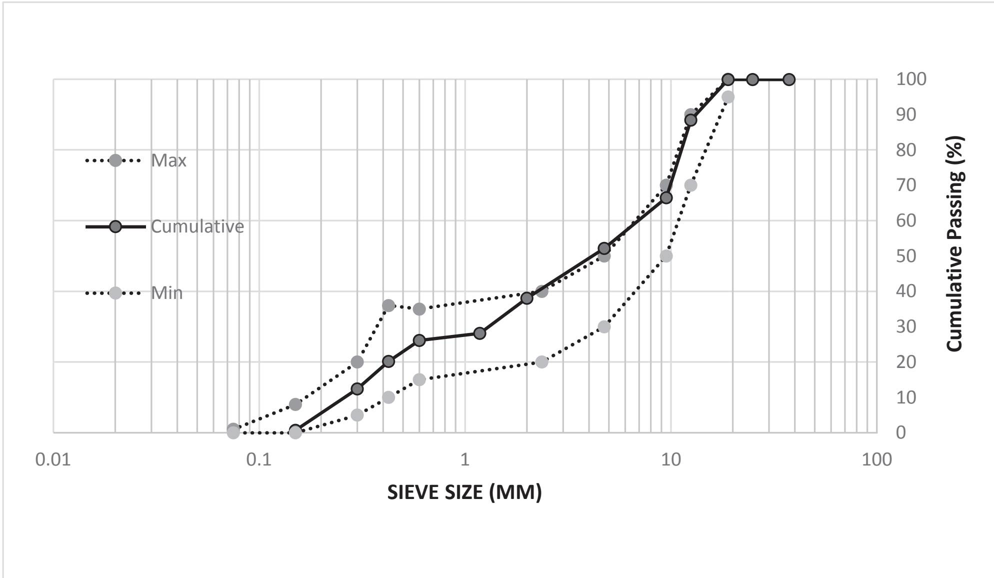

Fig. 2. Particles size distribution of aggregates according to ASTM C136 / C136M-14 [30].

Table 3
Concrete blends studied.

Mix	Cement (kg)	GP (kg)	Water (kg)	Sand (kg)	FA (kg)	MA (kg)	CA (kg)	W/C
0% GP	300	0	180	465.6	326	298	781	0.6
5% GP	285	15	180	465.6	326	298	781	0.6
30% GP	210	90	180	465.6	326	298	781	0.6

Table 4
RAI for mixes at 28 days.

w/c	GP = 5%	GP = 30%
w/c = 0.6	0,73	0,49
w/c = 0.7	1,13	0,65
w/c	GP = 5%	GP = 30%
w/c = 0.6	0,73	0,49
w/c = 0.7	1,13	0,65

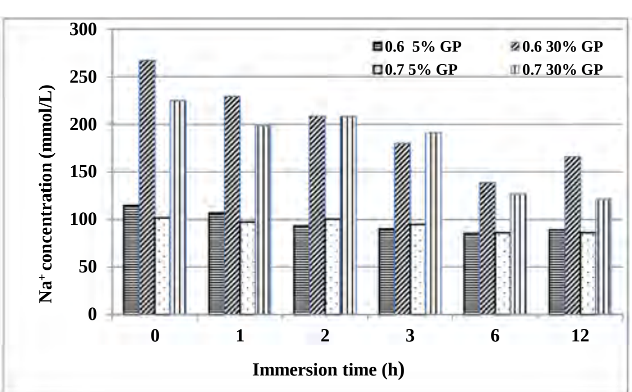

Fig. 3. Na^+ ion concentration in water under varying soaking times, w/c ratios (0.6 and 0.7) and GP content (5% and 30%).

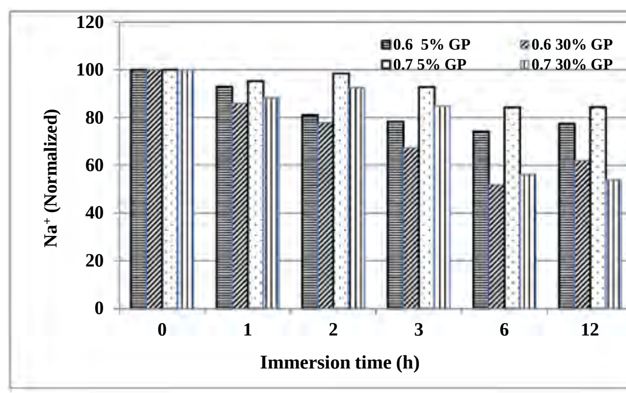

Fig. 4. Normalized Na^+ ion concentration in water under varying soaking times, w/c ratios (0.6 and 0.7) and GP content (5% and 30%).

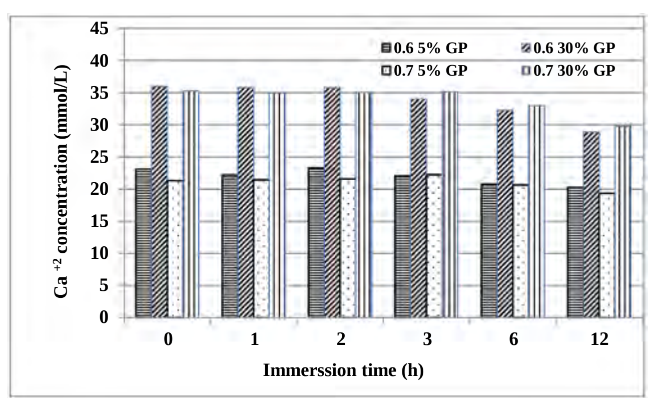

Fig. 5. Concentration of Ca^{+2} ions in water under varying soaking times, w/c ratios (0.6 and 0.7) and GP content (5% and 30%).

percentage of free ions according to ASTM D4691-17 [31]. The value of the amount of free ions is recalculated after deduction of the amount of free ions on the water before immersion.

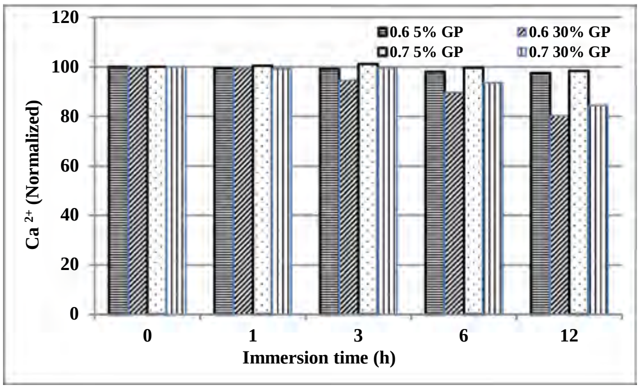

Fig. 6. Normalized Ca^{+2} ion concentration in water under varying soaking times, w/c ratios (0.6 and 0.7) and GP content (5% and 30%).

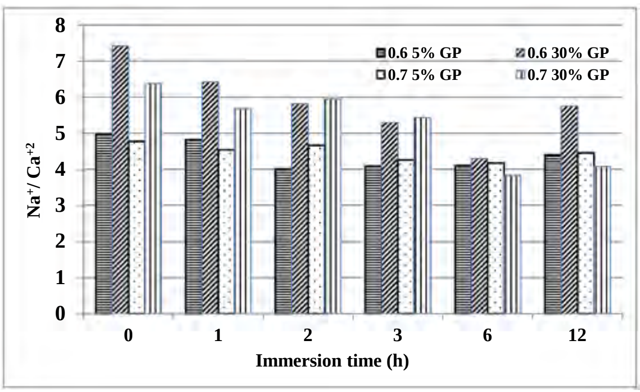

Fig. 7. Na^{+}/Ca^{+2} ion ratio with various soaking times, w/c ratios (0.6 and 0.7) and GP content (5% and 30%).

2.5. Mechanical properties

The density (ρ) of the concrete samples were obtained according to ASTM C642-13 [32] by dividing the measured mass (M) by the volume (V) just before the compressive tests.

Compression tests were then performed according to ASTM C39/C39M-18 [33] on cube-shaped samples with a side length of 100 mm 2, 7, 14, 28, and 90 days after pouring. Four samples from each mix were tested.

2.6. Relative activation index (RAI)

To examine the change in pozzolanic activity due to the replacement of cement, the relative activation index proposed by Pu [34] was calculated via the following equation.

$$RAI = \frac{100 * CompressivestrengthofGPmix}{Compressivestrengthofcontrolmix * (100 - GPcontent)} - 1 \quad (4)$$

A negative RAI indicates no pozzolanic activity from the substitution of cement with GP and a decrease in the CS. An RAI of zero indicates no change in the pozzolanic reaction process or the CS. A positive RAI indicates an increased pozzolanic reactivity and thus an increase in CS.

3. Results and discussion

3.1. Dissolution of the glass powder in the water

The resulting concentrations of Na^{+} ions for each soaking time, w/c ratio (0.6 and 0.7), and GP content (5% and 30%) are shown in Fig. 3. In each case, the ion concentration was the highest immediately after mixing (i.e., 0 h), after which the concentration decreased; the lowest concentration in all cases was found after 6 or 12 h of soaking. Additionally, the mixtures containing 5% GP had lower concentrations of Na^{+} ions than did the 30% GP mixtures. This come from the abundant amount of GP in the water for 30% (around 90 kg per M^3) GP rather than 5% GP (around 15 kg per M^3). The dissolution of Na^{+} ions from the GP is governed by the following reaction.

$$\equiv Si - O^{-}NaO^{+} + H^{+} \rightarrow \equiv Si - O^{-}H^{+} + Na^{+} \quad (5)$$

To better understand the global behavior due to the change in w/c ratio and GP, the normalized concentration of Na^{+} ions as a function of w/c ratio, GP content, and soaking time is shown in Fig. 4 The normalized concentration was calculate according to the amount of ions in the 0% GP mix (without GP). The lowest concentration of Na^{+} ions was found after 6 h of soaking timz. Mixes with higher water content (i.e., w/c = 0.7) had a higher loss of ions than mixes with lower w/cratio, which is likely caused by the dilution effect. The decrease of the amount of dissolved ions is probably due to the reaction between SiO_2 and Na^{+} to form nucleation sites favoring the formation of more CSH.

The resulting Ca^{+2} ion concentrations for each soaking time, w/c

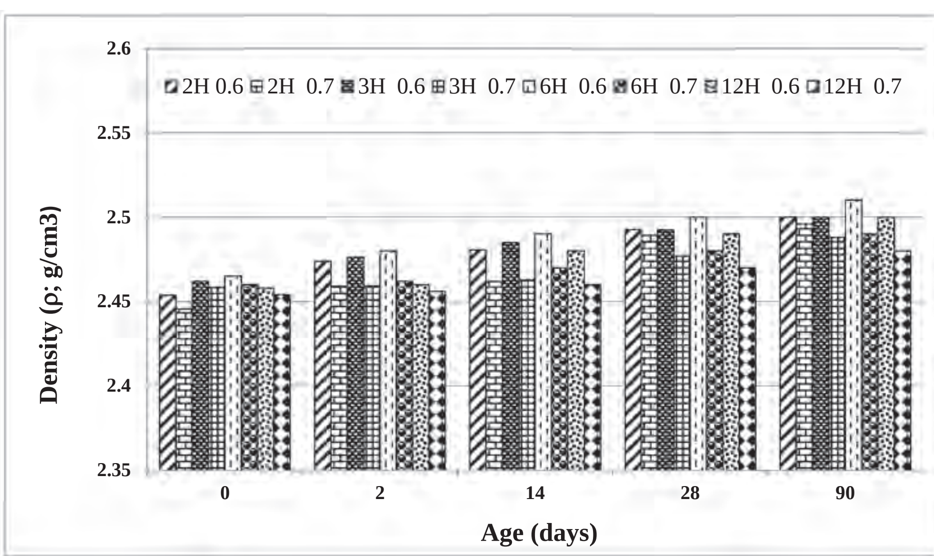

Fig. 8. Change in density with age of the 5% GP concrete with various GP/water pre-mixing times (2H, 3H, 6H and 12H) and w/c ratios (0.6 and 0.7).

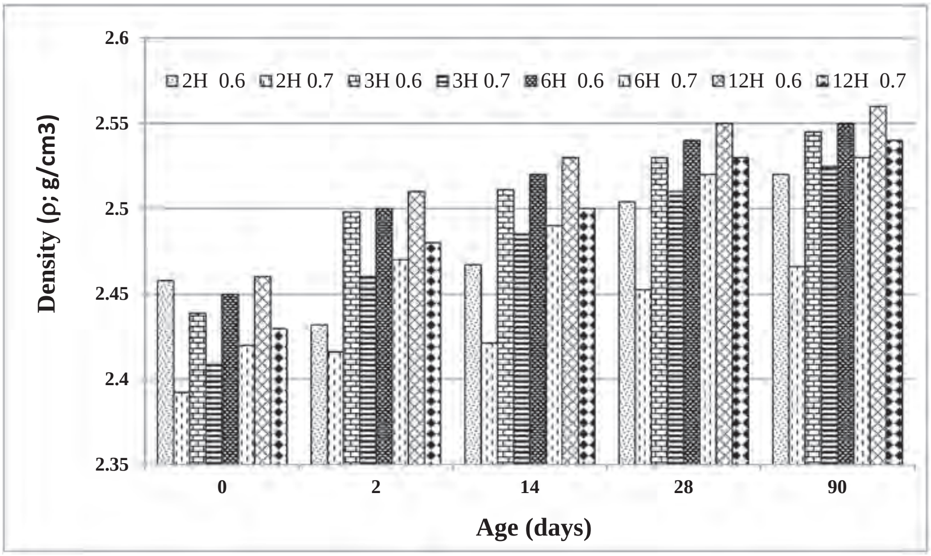

Fig. 9. Change in density with age of the 30% GP concrete with various GP/water pre-soaking times (2H, 3H, 6H and 12H) and w/c ratios (0.6, and 0.7).

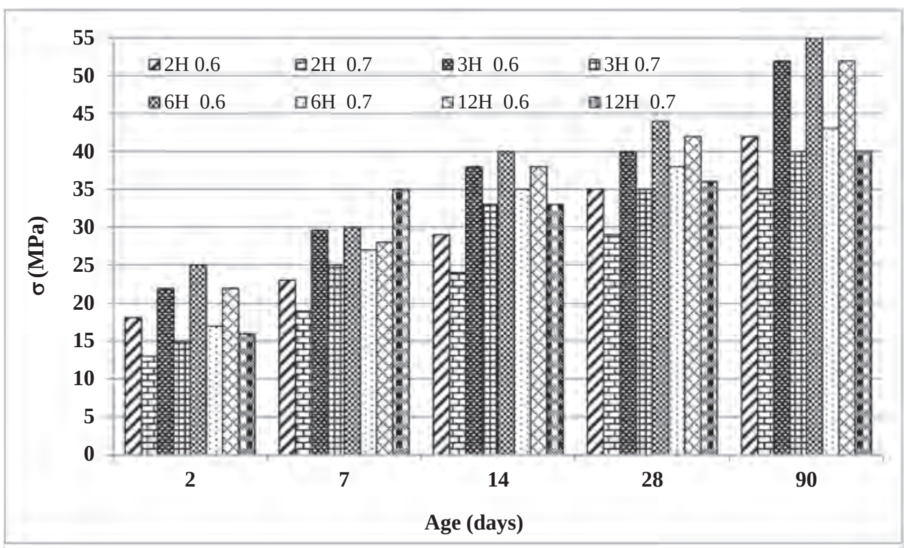

Fig. 10. Change in CS with age for concrete samples prepared with 5% GP with various w/c ratio (0.6, and 0.7) and pre-soaking time of the GP in water (2H, 3H, 6H and 12H).

ratio (0.6 and 0.7), and GP content (5% and 30%) are shown in Fig. 5. The concentration of Ca^{+2} ions remained steady between 0 and 3 h before decreasing; in all cases, the lowest concentration was found after 12 h. The concentration of Ca^{+2} ions was lower than that of the Na^{+} ions owing to their lower mobility. The water to cement ratio has insignificant decrease on the amount of ions release in the water, this might be caused by the lowermobility of Ca^{+2} ions.

In order to better evaluate the effect of water to cement ratio and the GP content, the use of normalized value is the best to show the changes in the measurement of each mixes.

The normalized concentration of Ca^{+2} ions as a function of w/c ratio, GP content, and soaking time is shown in Fig. 6. The lowest concentration of ions was found after 12 h of soaking, at this soaking time something happened on dissolution of Ca^{2+} ions from the GP.

The resulting ratio of Na^{+} to Ca^{+2} ions (Na^{+}/Ca^{+2}) under each GP content (5% and 30%), w/c ratio (0.6 and 0.7), and soaking time studies are shown in Fig. 7. The lowest ratio was found at 6 h of soaking time, thus indicating the presence of more nucleation sites at this time due to the fixation of ions on the surface of GP particles.

3.2. Fresh concrete behavior

3.2.1. Density of concrete mixes

The change in density of concrete made with 5% GP with time under various w/c ratios and pre-mixing times of the GP and water is shown in Fig. 8. The density increased with age and increased with the soaking time until reaching a plateau after 6 h of soaking time. The increase of the density with the soaking time may have been caused by the creation of denser hydrate products and a denser ITZ. The increase in nucleation sites allows for the formation of secondary C-S-H via the pozzolanic reaction. The highest density was obtained in mixes that underwent of 6 h of pre-mixing and lower water to cement ratio;while the lowest Na^{+}/Ca^{+2} ion ratio was obtained, as detailed above.

Similar results were seen in the change in density of concrete made with 30% GP under various w/c ratios and pre-mixing times of the GP and water, as shown in Fig. 9; the density increased with soaking time and age. The maximum density was obtained in mixes that underwent 6 and 12 h of pre-mixing and lower water to cement ratio, where the lowest Na^{+}/Ca^{+2} ion ratio was obtained, as detailed above.

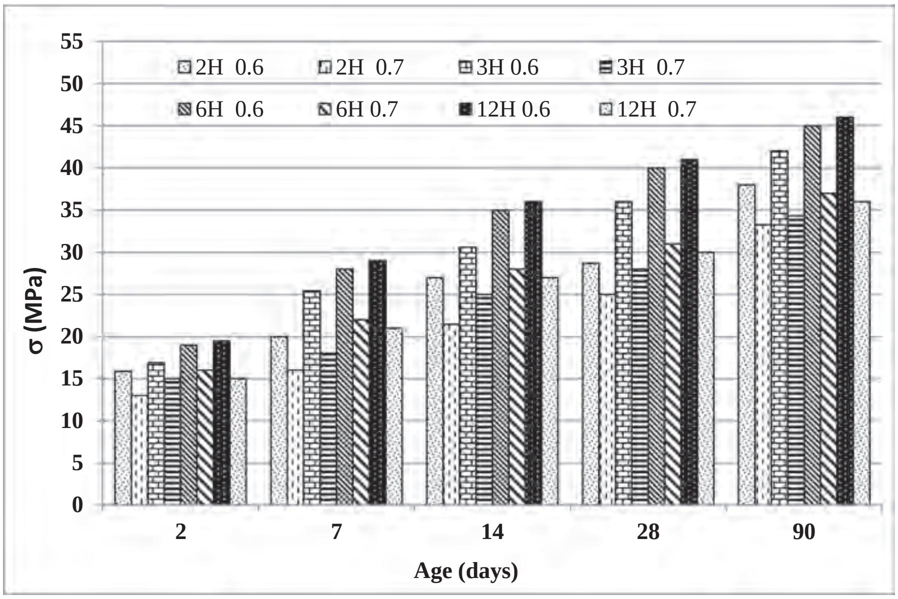

Fig. 11. Change in CS with age for concrete samples prepared with 30% GP with various w/c ratios (0.6 and 0.7) and pre-soaking time of the GP in water (2H, 3H, 6H and 12H).

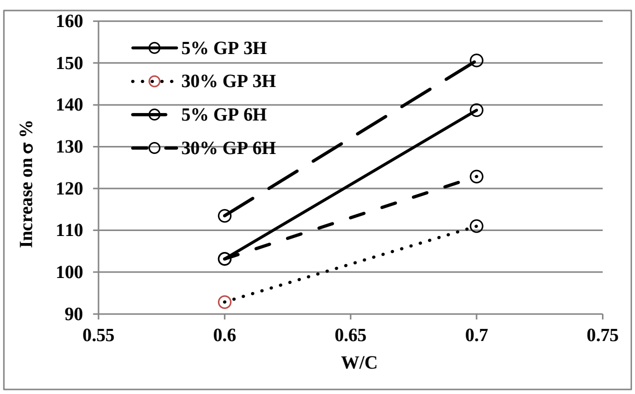

Fig. 12. The increase in CS with the w/c ratio and GP content at 28 days for 5% and 30% GP mixes.

3.3. *Mechanical behavior*

3.3.1. *Compressive strength (CS)*

The change in CS with age for concrete prepared with 5% GP and various w/c ratios (0.6 and 0.7) and soaking times (2H, 3H, 6H and 12H) is shown in Fig. 10. The CS increased as the soaking time increased, due to the formation of more nucleation sites. This result is in satisfactory relation with the density where the maximum densities were obtained for the same mixes with higher CS and with the lower ratio between Na^{+}/Ca^{+2} where the creation of nucleation sites was favorized to the CSH growth.

Similar results were obtained for the change in CS with soaking time for concrete prepared with 30% GP and various w/c ratios, as shown in Fig. 11; however, the CS was lower than for the samples prepared with 5% GP.

In order to better understand the effect of water to cement ratio on the activiation of the pozzolanic reaction, the CS at 28 days of concrete made with 5% and 30% with w/c ratio equal to 0.6 and 0.7 and 3H and

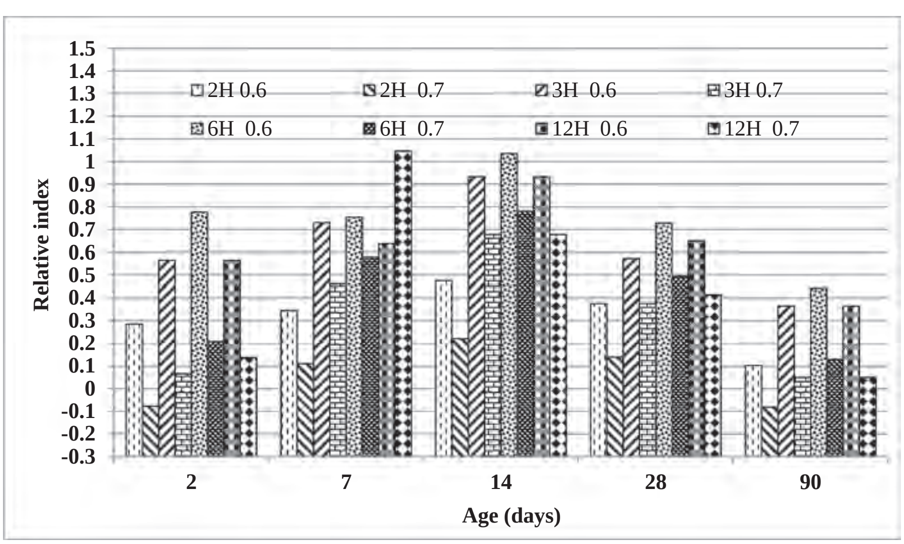

Fig. 13. Relative activation index (RAI) with the age, pre-soaking time of the GP in water (2H, 3H, 6H and 12H), and w/c ratio (0.6 and 0.7) for concrete made with 5% GP.

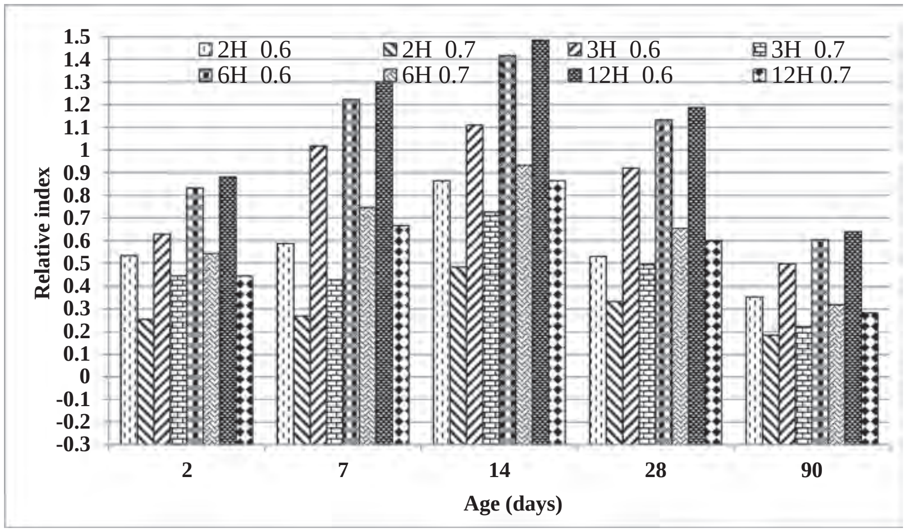

Fig. 14. RAI with the age, soaking time of the GP in water, and w/c ratio for concrete made with 30% GP.

6H pre-soaking time of GP in water is compared with the CS of the control at the same age.

The following equation was used in order to cacluate each point.

$$CSincrease = 100*\frac{CSwithGP - CScontrol}{CScontrol}$$

The change in CS with the w/c ratio and GP content after 28 days is shown in Fig. 12, calculated as a function of the CS of the reference concrete at w/c ratio of 0.6 and 0.7 (i.e., without GP). The CS increased more for both levels of GP content prepared with the higher w/c ratio than it did for that prepared with the lower w/c ratio. As noted, the increased w/c ratio increases the formation of nucleation sites by allowing Ca^{+2} ions to be fixed on the Si present on the surface of the GP particles, which then contributes to the formation of C-S-H in the cement paste and a higher-density ITZ.

3.4. Relative activation index (RAI)

The calculated change in RAI with age with various w/c ratios and soaking times of the GP in water for mixes prepared with 5% GP, shown in Fig. 13. The results demonstrate that the maximum value was obtained when the w/c ratio was 0.6 and the GP was soaked for 6 h. Pre-soaking the GP for 6 h allows for the formation of more ions and thus more nucleation sites for the growth of CSH. The presence of these ions increases the pozzolanic reaction.

Similarly, the calculated change in RAI with age with various w/c ratios and soaking times of the GP in water for mixes prepared with 30% GP replacement is shown in Fig. 14. The results demonstrate the maximum sample had a soaking time of 6 h and a w/c ratio of 0.6 or 0.7. After 2 days, the mixes prepared with 30% GP showed a higher RAI than for mixes prepared with 5% GP, likely owing to the formation of more ions due to the presence of more GP.

For both 5% GP and 30% GP, RAI calculation show an increase with curing age of samples until 14 days then it has an invers trend. This can comes from the hydration process of the cement, the cement used is CEMII, which has higher amount of C_2S than for CEMI. Where C_2S participate in the hydration process larer than at early age, which is the opposite for C_3S [35]. The participation of GP in the filling effect can represents raison for the increase of the RAI at early ages.

Table bellow presents the effect of w/c ratio as an activation factor for the pozzolanic reaction, the table shows that the mixes prepare with higher w/c ratio have higher RAI at same conditions. This results prove the results obtained in Fig. 13.

4. Conclusion

Using new mixing method by soaking the glass powder (GP) in water before mixing had increased the density,durability, andcompressive strength of the green concrete. Utilizing the new mixing method increased the compressive strength and relative activation index for green concrete mixes prepared with 5% and 30% GP. The mixes prepared with 5% GP had a higher compressive strength than those containing 30% GP. The maximum compressive strength was formed after allowing the GP to soak in water for 6 h, due to the increased formation of ions that participate in the hydration process. Soaking the GP in water for 6 h before mixing increased the pozzolanic reaction.

For the first few days, the compressive strength of the concrete increases due to the packing filling effect and pozzolanic reaction; later, the increase is due to the pozzolanic reaction. Both effects lead to a denser ITZ.

The use of a higher w/c ratio increased the compressive strength and relative activation index, due to the presence of more ions in the water for the first few days of hardening. In addition to the formation of more nucleation sites. Thus, increasing the w/c ratio in case of using the new mixing method represents a determinant key on the activation of the pozzolanic reaction of the GP and the increase in its compressive strength. Thus, the green concrete made with w/c of 0.7 at 5% GP had compressive strength of 36 MPa compared with 25 MPa given by control mix which represnt an increase of around 38%.

5. Data availability

All data, models, and code generated or used during the study appear in the submitted article.

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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