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Waste Glass Powder as Cement Replacement in Concrete

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Received 25 August 2014, accepted 2 November 2014 doi:10.3151/jact.12.468

Abstract

The pozzolanic reactivity of waste glass powder was experimentally studied at cement replacement levels of 0, 15, 30, 45 and 60% by weight. Results revealed that the concrete compressive strength was not decreased by the cement substitution after 28 days because of the pozzolanic reaction between glass powders and cement hydration products, if the replacement is below 30%. Also, the resistance to chloride ion and water penetration continuously increases with increasing glass powder content up to 60% cement replacement. At 60% replacement level, the electrical resistivity and water penetration depth were reduced by 95% and 80%, respectively, while the compressive strength was maintained as 85%. These improvements in durability properties are due to the refined microstructures, particularly at the interfacial transition zone. Pore size distribution was measured to confirm the refinement in the capillary pores, which partially block the pathways for water and chloride ions. This study also demonstrates that high performance concrete (improved strength and impermeability against chloride and water) could be achieved by using glass powder as 15% additive, which contributes to the pozzolanic reaction instead of being inert fines for compact packing.

1. Introduction

Due to the awareness on the need for environmental protection, turning solid wastes or by-products into ingredients of concrete has drawn increasing attention. Apart from material and energy conservation, reuse of some solid wastes could result in better performances of concrete in several areas. For example, use of waste glass as fine aggregates provides concrete with higher strength and impermeability against chloride and water) could be achieved by using glass powder as 15% additive, which contributes to the pozzolanic reaction instead of being inert fines for compact packing.

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rarely been reported (Shi et al. 2005). They found that curing temperature have a more obvious accelerating effect on the pozzolanic reactivity of glass powder than on that of fly ash. Both the hydration and pozzolanic reaction greatly govern the structure formation and hence all properties like strength development and durability.

As reviewed above, the properties of concrete with glass powder as cement replacement in higher proportion remains an unexplored area. Therefore, this research aims to study the pozzolanic reaction of GP up to 60% cement replacement and its influence on the microstructure of cement paste, which has not been investigated before. The hydration rate and rheological properties of paste were also characterized. The chemical composition and calcium hydroxide content of the hydrated products were analyzed at different ages. Also, the microstructure development of cement paste was determined by measuring the pore size distribution. At the same time, the compressive strength and resistance to chloride and water penetration were investigated for concrete with the same cement replacement levels as the paste. The influence of pozzolanic reaction on the transition zone between aggregate particles and cement paste is discussed. Based on the results, the optimum cement replacement content by glass powder is suggested.

2. Experimental program

2.1 Materials

The crushing process of recycled waste glass can be found in the authors’ previous work (Tan and Du 2013; Du and Tan 2014a, 2014b). Waste bear bottles (soda lime glass) were collected from a local recycler in Singapore. To finely grind the sand-sized particles, a ball miller was used. The size distributions of ground GP and cement used in this study are shown in Fig. 1. Both cement and GP show the same median particle size of around 10 µm. The chemical compositions of GP and OPC are displayed in Table 1. The surface appearance of GP and OPC are compared in Fig. 2. GP is amorphous and its X-ray diffraction (XRD) pattern is compared with OPC in Fig. 3. GP has a negligible water absorption capacity of 0.07%. Coarse aggregate with a maximum size of 10 mm was used in concrete. The fineness modulus of natural sand was 2.80.

2.2 Specimens

The microstructure of cement paste with various contents of GP was investigated. The water to cement (w/c) ratio for the paste is 0.485 by weight. The mix proportion for concrete is listed in Table 2. Cement was replaced by GP at 0, 15, 30, 45 and 60% by weight. An additional mix (OPC+15GP) was cast in which the GP was used as an additive, instead of replacing cement at a content of 15%. Compared to the use of cement replacement, the addition of GP in this mix leads to a lower water-to-binder (w/b) ratio of 0.42. This could help examine the utilization of GP as additive in high performance cementitious composites. For each paste mix, three 50 mm cube specimens were prepared. For each concrete mix, nine 100 mm cubes and five Φ100×200 mm cylinders were cast for compressive strength and durability tests, respectively. Paste and concrete were mixed in Hobart and pan mixers respectively. All the specimens were compacted on a vibration table and covered by plastic sheets to prevent moisture loss. After demolding on the next day, the paste specimens were cured in saturated lime water and concrete samples were cured in water until the test age. The curing method for paste is following ASTM C 109 (2005) to prevent calcium leaching and curing method for concrete specimens is following ASTM C 39 (2005).

![Fig. 1 Particle size distribution of OPC and glass powder.](image-url)
2.3 Test methods

The rheological properties of cement paste containing GP were determined by a coaxial-cylinder viscometer named RotoVisco 1. After mixing the cement paste in the Hobart mixer for about 3 minutes, a sample weighing about 150 g was taken out and placed in the outer cylinder of the viscometer, followed by inserting the inner cylinder immediately. The sensor then started to rotate at different shear rate while the torque was recorded. Isothermal calorimetry was performed on triplicate paste samples by using an eight-channel microcalorimeter (TAM AIR). Paste samples were taken from the center of 50 mm cube specimens for the purpose of mercury intrusion porosimetry (MIP), XRD and thermogravimetric analysis (TGA). For MIP test, paste samples (around 1 cm in cubic shape) were dried in dessicator at 50°C for 1 day before testing, at the age of 7 and 91 days. Micromeritics AutoPore III with a maximum mercury pressure of 412.5 MPa was used to measure the pore size distribution. For XRD and TGA analyses, paste samples were cured in 105°C oven for 24 hours and then finely ground to be less than 75 µm. Shimadzu XRD-6000 diffractometer was employed to qualitatively determine the influence of GP on the chemical composition of the hydrated production. The XRD scan was between 10° and 60° with a speed of 0.5°/min. TGA was carried out using LINSEIS L81-II. The powder sample was heated from ambient temperature up to 950°C at a rate of 10°C/min while the weight loss was recorded during the TGA test. The content of CH could be obtained from two different intervals in the weight loss curve, corresponding to the decomposition of Portlandite and calcite, respectively.
For concrete specimens, compressive strength and rapid chloride penetration test (RCPT) were carried out according to BS EN 12390-3 (2009) and ASTM C 1202 (2005), respectively. RCPT is commonly criticized for its drawbacks including temperature rise due to the applied high voltage of 60 V (McGrath and Hooton 1999). Thus, a reduced voltage of 20 V was applied for 24 hours, to minimize the side effects of RCPT. At 28 days, water penetration depth into concrete was determined using a water pressure of about 0.75 MPa for 7 days, in accordance to BS EN 12390-8 (2009). The side and top surfaces were both coated with impermeable epoxy while the bottom surface was exposed to the water pressure. After removed from the test rig, the concrete cylinders were axially split and the average depth water front measured.

3. Test results and discussion

3.1 Rheological properties

The shear stress ($\tau$) was recorded with shear rate ($\dot{\gamma}$) decreasing from 50 to 0.5 s$^{-1}$ during the descending branch of the test loop. Test results are shown in Fig. 4(a). Yield stress ($\tau_0$) and plastic viscosity ($\mu$) of fresh cement paste are linearly related using Bingham model (Mindess et al. 2003).

\[
\tau = \tau_0 + \mu \dot{\gamma}
\]  

(1)

The values of $\tau_0$ and $\mu$ for pastes with various GP contents are displayed in Fig. 4(b). Both the yield stress ($\tau_0$) and viscosity ($\mu$) decrease with higher cement replacement level. Yield stress indicates the minimal strength to make the mixture flowable. The reduced yield stress implies that the inter force between cement and glass particles is less than that between cement and cement particles. With increasing GP contents, the particle density of cement is diluted and hence lesser interaction between cement and water, leading to a smaller yield stress and plastic viscosity. This could also be due to the negligible water absorption and the smooth surface of GP (as shown in Fig. 2). Previous studies also indicate that the bond between cement paste and fine glass particles was decreased due to the surface smoothness of glass powder (Taha and Nounu 2009; Ali and Al-Tersawy 2012).

In this study, the cement has been replaced by weight instead of by volume. As the specific gravity of glass powder is lower than cement, the solid-to-water ratio by volume is higher for GP blended paste compared with pure cement paste. However, this adverse effect at a higher solid-to-water volume ratio is less pronounced compared to the dilution of cement and smooth glass surface, as mentioned earlier. The continuously decreased yield stress and plastic viscosity represent a better workability of paste mixture with higher content of GP. It is also noted that both the yield stress and viscosity increase substantially for the mix with 15% additional GP, attributed to the increased amount of solid-to-water ratio.

3.2 Heat of hydration

The heat evolution rate and cumulative amount of heat for pastes with varying contents of GP are shown in Fig. 5. It is clear that the maximum heat evolution rate and the total heat generated reduced continuously with higher OPC replacement level because of the dilution of cement and the slower rate of pozzolanic reaction of GP. The results in this study are consistent with previous findings (Dyer and Dhir 2001; Mirzahosseini and Riding 2014). The benefit of lower hydration heat is helpful in preventing the temperature cracking, especially for structure members with large thickness and mass concrete.

In the presence of GP, the time to reach the peak hydration rate is shortened, possibly because fine glass powders can accelerate the cement hydration via the adsorption of calcium ions from the liquid phase and play as nucleation and growth sites for C-S-H and other hydrates. At the same time, the high content of alkalis (Na$_2$O) in GP may act as catalyst in the formation of calcium silica hydrate at an early age (Jawed et al. 1978; Khmiri et al. 2013). The time corresponding to the peak rate reduces from 418 minutes for OPC paste to 377 minutes for paste with 60% glass powder paste.
3.3 XRD

The XRD patterns for paste with glass powder at age of 7, 28 and 91 days are shown in Fig. 6. The compositions of the hydration product for Portland cement paste are C-S-H and CH and their principal peaks do not change from 7 to 91 days. CH peaks tend to be weakened with increasing glass powder and longer curing time, especially for 60% glass powder paste at 91 days in which the CH peaks almost disappear. The XRD results directly reveal that CH is consumed to form the additional C-S-H in paste with glass powders.

At 91 days, peaks corresponding to CH can be obviously seen for OPC and 30GP paste, which means that CH remains even after 91 days of pozzolanic reaction with glass powder. However, it is not true for paste with 60% glass powder in which the CH seems to be insufficient for glass powder to be fully reacted.

3.4 Ca(OH)$_2$ content

The CH content was calculated from the TG and DTG curves, as illustrated in Fig. 7(a). Derivative curve was used to find the beginning and the end of the derivative peak and measure the weight loss correspondingly (Kocaba 2009). In the temperature range of about 425 and 550 °C, the amount of CH can be calculated by determining the water loss during the CH decomposition: Ca(OH)$_2$→CaO+H$_2$O. Also, the influence of carbonation on the CH content could be evaluated by the mass loss in the temperature range of 650 and 800 °C, where the calcite decomposes and release CO$_2$: CaCO$_3$→CaO+CO$_2$. Hence, the content of CH was calculated taking into account molecular weight of each component and were calculated as percentage of the dry weight at 105 °C.

As shown in Fig. 7(b), the CH content in the reference OPC paste remained at above 20% of the total weight after 28 days of curing, which is similar to previous findings (Jawed et al. 1978). With higher cement substitution level, the CH content drops, especially at later age such as 91 days. At the beginning, the reduction is caused by the dilution of Portland cement in the total cementitious material content. With longer curing and hydration time of 28 days, more CH is consumed in the pozzolanic reaction of glass powder. The initial percentage drop in CH content was higher for 30% GP paste than in 60% GP paste. After 28 days, the reduction in CH content slowed down for 30% glass powder paste and stabilized at about 11%, indicating the pozzolanic reaction was almost completed. In contrast, the CH content in 60% GP paste continued to drop and was less than 4% at 91 days. This depletion in CH reveals that 60% might be the maximum practical glass powder content since the CH content is insufficient for the further pozzolanic reaction of glass powder. Dyer and Dhir (2001) has reported that the amount of CH continuously decreased with increasing GP replacement up to 40%, at the age of 28 days, but no information for higher GP replacement.
3.5 Microstructure

The pore size distribution of cement paste containing glass powder at 7 and 91 days are shown in Fig. 8, from which the change in the microstructure of paste can be seen clearly particularly for paste with 30 and 60% glass powder. The pore structures become more refined, as indicated by the reduced threshold and critical pore diameter, as well as the higher fraction of the micro-pores. For paste with 60% glass powder, the threshold and critical pore diameter was reduced from 488 and 140 nm at 7 days to 139 and 74 nm at 91 days, respectively. Moreover, the volume fraction of capillary pores (10 µm – 50 nm) in the total porosity (10 µm – 3.7 nm) was lowered from 44.6% at 7 days to 30.7% at 91 days. According to Mindess et al. (2003), it is the capillary pores that govern the permeability and diffusivity of paste. Thus the MIP results reveal that the permeability of high volume glass powder paste is likely to be reduced. Pozzolanic reaction between glass powder and CH is a slower chemical process compared with OPC hydration. Since glass powder is inert in the initial stage, the effective w/c ratio is actually higher than that of the OPC paste. This results in a more porous microstructure for mix with glass powder as OPC replacement. However, at later age, most of the glass powders in 30GP and part of it in 60 GP mix have reacted with lime to form the secondary C-S-H. The pore structures are almost identical for each paste mix, regardless of glass powder content at 91 days.

3.6 Compressive strength of concrete

The compressive strengths of concrete with varying contents of GP, at different ages, are shown in Fig. 9. At 7 days, the strength generally decreases with GP content more than 15%. However, at 28 and 91 days, no reduction in strength was observed for concrete with 15 and 30% GP, due to the pozzolanic reaction between GP and cement hydration products. Instead, concrete with 15 and 30% GP possessed higher compressive strengths. This increase in later age strength was not observed for concrete in which more than 30% cement is replaced by GP. The pozzolanic reaction requires the hydration products, CH, whose amount is governed by the cement content. Therefore, there is an upper limit for cement replacement level, beyond which no further pozzolanic reaction of GP can occur. In that case, GP can only play
the role of inert filler without being activated. The results in this study indicate that GP will exhibit obvious pozzolanic reaction provided that the replacement level is lower than 30%. Mix of OPC+15GP exhibited much higher increase in compressive strength, that is 32%, 25% and 24% at 7, 28 and 91 days, respectively. It would be readily for the added fine GP to react as pozzolans instead of being inert fillers.

The pozzolanic reaction improves not only the pore structure in the bulk cement paste but also at the interfacial transition zone (ITZ) between coarse aggregates and cement paste. This ITZ governs the mechanical and transport properties of concrete since it is more porous compared to the bulk paste. CH content is also relatively higher at ITZ which favors the pozzolanic reaction of glass powder. The enhanced microstructure at ITZ has contributed to the compressive strength with up to 30% GP content.

3.7 RCPT result of concrete
The total charge passing the concrete with different OPC replacement levels are shown in Fig. 10, at 7, 28 and 91 days. Due to the low voltage applied in this study, the temperature rise during the 24 hours test was less than 3 °C. The RCPT results show that the total charge passed decreased significantly with increasing glass powder content, regardless of the test age. RCPT is essentially an indication of the electrical conductivity, instead of a direct measurement for chloride permeability. Therefore, the result depends on not only the pore structure but also the chemistry of the pore solution, such as ions (Ca²⁺, Na⁺, K⁺, OH⁻, etc) concentrations. However, it is the pore structure and not the pore chemistry that govern the transport of ions in cement composites. Therefore, it is also questionable to use RCPT to assess the permeability of concrete with supplementary cementitious materials such as silica fume and fly ash (Shi et al. 1998).

At 7 days, the total charge passed reduced almost linearly with decreasing OPC content. The primary reason could be the dilution of OPC in the concrete, as a result of which there are fewer ions in the cement paste. With longer curing time, all the concrete mixtures exhibited reduction in the total charge passed due to the further hydration of OPC. The reduction caused by glass powder became more distinct, especially for OPC replacement between 15 and 45%. This further reduction is attributed to the pozzolanic reaction between GP and cement paste, resulting in a more refined microstructure of secondary C-S-H especially at the ITZ as well as a lower concentration of ions. Glass powder has a similar effect on the RCPT result whether it adds to or replaces OPC as cementitious material, particularly in the long term. At an early age, concrete mix with 15% GP addition has a higher resistance possibly because of the hydration acceleration effect of GP, as discussed earlier.

3.8 Water penetration resistance of concrete
The water penetration depth into concrete with different contents of GP is shown in Fig. 11. Consistent with the RCPT results, the depth of water penetration continuously decreases with higher glass powder content. Relative to the plain concrete, the water penetration depth was reduced by 54, 65, 68 and 80% for cement replacement level of 15, 30, 45 and 60%, respectively. Concrete with 15% GP additive showed much lower water penetration depth compared to the plain concrete. The refined pore structure, particular the ITZ is the main reason for this reduced permeability. The reduction in the total charge passed in the RCPT tests at 28 days was 60, 81, 89 and 95% for 15, 30, 45 and 60% of glass powder content, respectively. Contrary to RCPT results, the resistance to water penetration can accurately reflect the pore structure. Hence, both accelerated and non-accelerated tests demonstrate the better resistance against chloride ions and water ingress, for concrete containing glass powder as cement replacement and additive.

3.9 Interfacial transition zone in concrete
The microstructures of paste at ITZ are compared for plain concrete and 30% GP concrete in Fig. 12. It is clear to find CH (plate crystals) and ettringite (needle shape) in the paste of the OPC concrete. Large pores
exist between different hydration phases. In contrast, a dense and more homogenous structure is found for concrete with 30% glass powder. The pozzolanic reaction over 91 days turned crystal CH into amorphous C-S-H. Large pores were much less noticeable. This change in the chemical compositions (more C-S-H formed) and pore size distribution (refined pore system) is the reason for the better mechanical and durability performances for concrete with glass powder as OPC substitution.

The pore size distribution of the microstructure was also investigated for concrete including the ITZ, as shown in Fig. 13. It is clear that with OPC partially replaced by glass powder, the pores are more refined, indicated by the downwards shift of the curve. From the figure, it can be seen that the critical pore diameter becomes smaller for glass powder concrete. Therefore, the effect of glass powder pozzolanic reaction on the concrete is distinct in comparison to that on paste. The reason is attributed to the existence of ITZ where the concentration of CH and water was higher and it was therefore easier to react with glass powder to form secondary C-S-H, as mentioned above.

4. Conclusions

This study reported experimental results on the use of recycled glass powder as supplementary cementitious material in paste and concrete. Unlike previous work, this study examined the effects of a higher glass powder dosage of up to 60%. Based on the newly obtained results, the following conclusions can be drawn:

1. The rate and total heat generated during hydration consistently decreased with higher GP content due to the dilution of cement in the mix. Owning to its negligible water absorption capability, higher glass powder content results in a smaller shear yield stress and plastic viscosity of the paste.

2. Calcium hydroxide content decreases with glass powder content and curing age, because of the reduced cement content in the initial stage and its consumption by glass powder pozzolanic reaction at the later stage. Calcium hydroxide was almost depleted at 91 days when more than 30% cement was substituted by glass powder.

3. An optimum cement replacement of 30% by glass powder was observed with respect to the development of compressive strength of concrete after 7 days.
However, with respect to the resistance to water and chloride penetration, 60% seems to be the optimum replacement. Compared to the reference concrete mix, the mix with 60% glass powder exhibited 75% strength at 7 days and 85% strength at 91 days.

4. Resistance to chloride ion and water penetration resistance were greatly improved by replacing cement with glass powder, due to the refined microstructure of paste, particular at the ITZ where calcium hydroxide concentration is higher and more readily available for pozzolanic reaction. With 60% cement replacement, the electrical resistance and water penetration depth of concrete decreases to 5% and 20% of the reference concrete.

5. When used as additional supplementary cementitious material at 15% level, glass powder can obviously reduce the porosity and the pore size distribution. Thus, large increases in compressive strength, resistance to water and chloride penetration were observed. This study only investigated finely ground glass powder as cement replacement or additive in concrete based on soda lime glass, which is the most common type of glass and accounts for 90% of manufactured glass. It is an open question as to whether the results could apply to other types of glass like lead glass and borosilicate glass. The pozzolanic reactivity depends on the chemical composition and properties of the glass. To understand the pozzolanic performance of a specific glass powder, it is recommended to (1) carry out chemical composition analysis to meet the minimum chemical requirement for pozzolans; (2) determine the strength activity index; and (3) investigate the different replacement levels on mechanical and durability performances on concrete.

**Acknowledgements**

The technical assistance of Ms. Li Wei and Mr. Ang Beng Oon from the Structural Engineering and Material Laboratory, National University of Singapore, in the conduct of TGA and XRD tests, is gratefully acknowledged.

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