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Combined use of waste glass powder and cullet in architectural mortar

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Abstract: The use of 100% waste glass cullet (WGC) as fine aggregates in architectural cement-based mortar had been proven to be feasible in previous works. This paper reports a further study on investigating the influence of using waste glass powder (WGP) as a supplementary cementitious material on the properties of glass-based architectural cement mortars. The experimental results showed a good linear relationship between the particle size of WGP and the flow values of the fresh mortar, revealing that the particle size of WGP played an important role in controlling the workability. For the hydration of white cement, the inclusion of WGP not only affected the second exothermic peak of hydration but also changed the third peak. In particular, the results indicated that the use of finer WGP had an advantage in increasing the flexural strength of the cement mortar when compared with the corresponding compressive strength, which was attributed to the morphological and pozzolanic effect of WGP. In addition, the very fine WGP could act as micro-fibers and micro-aggregates in filling the microstructure of the mortar. At 90 days of curing, the mortar prepared with finer WGP showed a distinct improvement in strength due to the improved interfacial transition zone and the pore-size refinement.

Keywords: Waste glass powders (WGP); Heat of hydration; Contribution rate to strength (CRS); Interfacial transition zone (ITZ); Pore structure

1 Introduction

Waste glass beverage bottles are a major solid waste type in Hong Kong. Although the public has paid more attention on municipal waste separate collection, the recycling rate of waste glass beverage bottles is still very low (less than 10%) [1]. Due to the low commercial value, waste glass beverage bottles are mostly landfilled in Hong Kong rather than collected for recycling. It is estimated that Hong Kong's landfills will be exhausted one by one by 2020 if waste levels continue to increase at current levels. Therefore, the need to recycle more glass

waste is crucial to Hong Kong.

Previously, there had been studies aiming at studying the feasibility of using waste glass cullet (WGC) to replace natural fine aggregates in concrete blocks or cement mortars. The conclusions showed that this glass recycling option was a cost-effective and attractive for construction material applications. The WGC was used as replacements of natural aggregates for the purpose of reducing the drying shrinkage [2][3] and water absorption [3] taking advantage of its impermeable properties, enhancing fresh properties leveraging its smooth surface [4], improving the durability based on its high resistance to abrasion and acid [5], strengthening compressive strength and elastic modulus after exposure to 800 ℃ on account of glass melting and filling the cracks and pores [6], reinforcing phtocatalytic activity because of its light transmittance properties [7].

Based on the past research of this group, several practical and potential applications have been developed such as using WGC in eco-glass concrete paving blocks [8], glass-based self-compacting concrete [9] and architectural mortars [10][11]. However, there is a potential detrimental effect if WGC is used in self-compacting concrete or architectural mortars, that is, the presence of high amorphous silica content in WGC can react with alkali in cement to cause expansion due to the Alkali-silica-reaction (ASR) in the cement matrix. In this regard, attempts have been made to use waste glass powder (WGP) as an ASR suppressor although it has a high alkali content [12-14]. Recently, some studies have also pointed out that the finer glass powder showed significantly improved ability to enhance the durability characteristics of concrete products [15][16]. Encouraging results were also obtained by Du and Tan [17-19] in concrete with high replacement levels (up to 60%) of glass powder. They found that as glass the powder content was increased up to 60% in concrete, the improved interfacial transition zone (ITZ) and refinement in pore structure were responsible for the significant reduction in the chloride diffusion, water penetration depth, sorptivity, conductivity, and migration coefficients. Therefore, it would be possible to manage waste glass environmental friendly through a combined use of WGC to fully replace aggregate and WGP to partially replace cement.

In order to understand the effects of particle size on the reactivity of WGP, different fineness of WGP were used in previous studies. Shao et al. used 75-150 μm, 38-75 μm, and 0-38 μm as partial cement replacements. The result showed that the smaller particle size of ground glass led to a higher reactivity of glass with lime [20]. A similar conclusion has been recently reported by Mirzahosseini and Riding [21]. They evaluated three narrow size ranges of glass powder, 63-75 μm, 25-38 μm, and 0-25 μm and found that surface area of finely ground glass was an important factor on glass reactivity.

In this study, different particle sizes of WGP were just ground by using a ball milling approach for specified periods (0.5h, 1h, 2h, and 4h, respectively), and then the WGP were used to partially replace cement at a level of 20% by weight. Moreover, in the light of the appearance of WGP is nearly white, so that the inclusion of WGP would not cause any adverse impacts on the aesthetic properties of the architectural mortar.

Furthermore, few studies have been done on the combined use of WGC and WGP in architectural mortars. Therefore, the aim of this work was to explore the feasibility to maximize the re-utilization of waste glass both as a fully replacement of fine aggregates and a partial replacement of cement in architectural mortars. Fly ash (FA) was also used in this study as a comparison with the WGP. A comprehensive experimental work including fresh behavior, hydration characteristics and mechanical properties was performed. In addition, the morphology of the ITZ and the porosity of the mortars containing WGP or FA were also investigated.

2 Materials and methods

2.1 Materials

2.1.1 Cement

A type of white ordinary Portland cement (WPC) (TAIHEIYO Cement Corp., Japan) was used in this study due to the aesthetic requirement of the architectural mortar. The chemical composition of the WPC is listed in Table 1.

Table 1

Chemical composition of WPC, WGP and FA (ms %).

	WPC	WGP	FA
SiO_2	21.36	73.5	45.70
Al_2O_3	5.27	0.73	19.55
Fe_2O_3	0.20	0.38	11.72
CaO	67.49	10.48	12.27
MgO	1.14	1.25	4.10
K_2O	0.077	0.69	1.71
Na_2O	0.048	12.74	1.36
TiO_2	0.14	0.087	1.09
SO_3	2.60	-	1.82

2.1.2 Waste Glass Cullet (WGC)

The WGC was sourced from post-consumer beverage bottles which were collected from a glass waste recycling facility in Hong Kong. Because most of the waste glass was contaminated with paper and other substances, the WGC was rinsed with tap water first and then oven-dried for a minimum of 24 h at 105 ℃ to reduce the moisture content. The gradation curve of WGC is presented in Fig. 1.

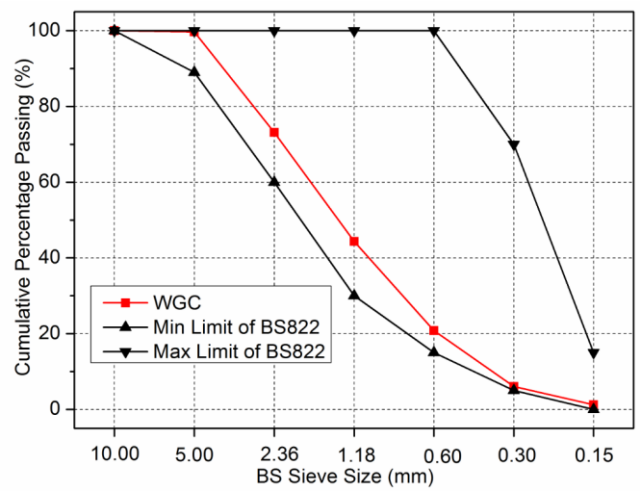


Fig. 1 Gradation curve of WGC

2.1.3 Waste Glass Powder (WGP) and Fly Ash (FA)

Different sizes of WGP were obtained after grinding the WGC with a laboratory ball mill for 0.5h, 1h, 2h and 4h, respectively. The ball mill used was a cylindrical barrel with internal diameter of 260 mm and length of 330 mm. The rotation speed of ball mill was 60 r/min and the ratio of ball to WGC was set to 6:1. Different sizes of steel ball with maximum diameter of 23 mm and minimum diameter of 11 mm were used to crush the WGC. The particle size distributions of the different WGP are shown in Fig. 2. FA was produced as a by-product during the generation of electricity from a local coal-fired power plant. The chemical compositions of WGP and FA as tested by X-ray Florescence are given in Table 1. Scanning electron microscopy (SEM) was employed to observe the morphologies of WGP (milled for 2h) and FA. The micrographs (Fig. 3) show distinctive differences between the WGP and the FA. The WGP showed a smooth surface texture, irregular shape with sharp edges and high aspect ratios, while the FA was made up of many spherical particles in micrometer range.

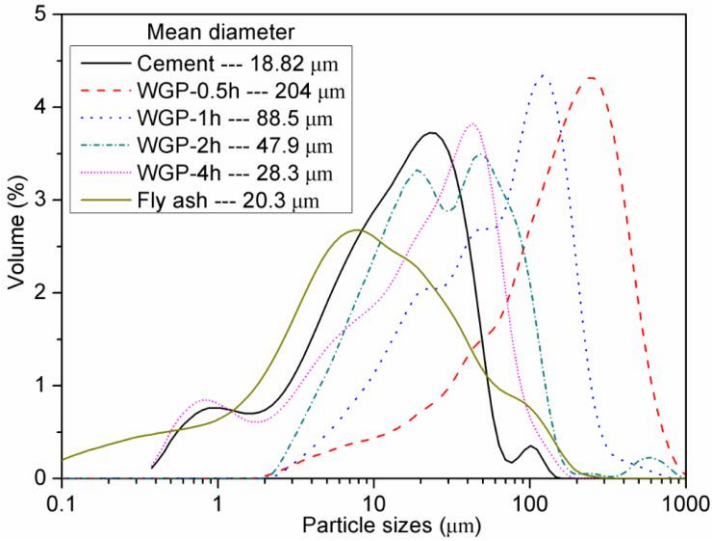


Fig. 2 Particle size distribution of WPC, WGP and FA

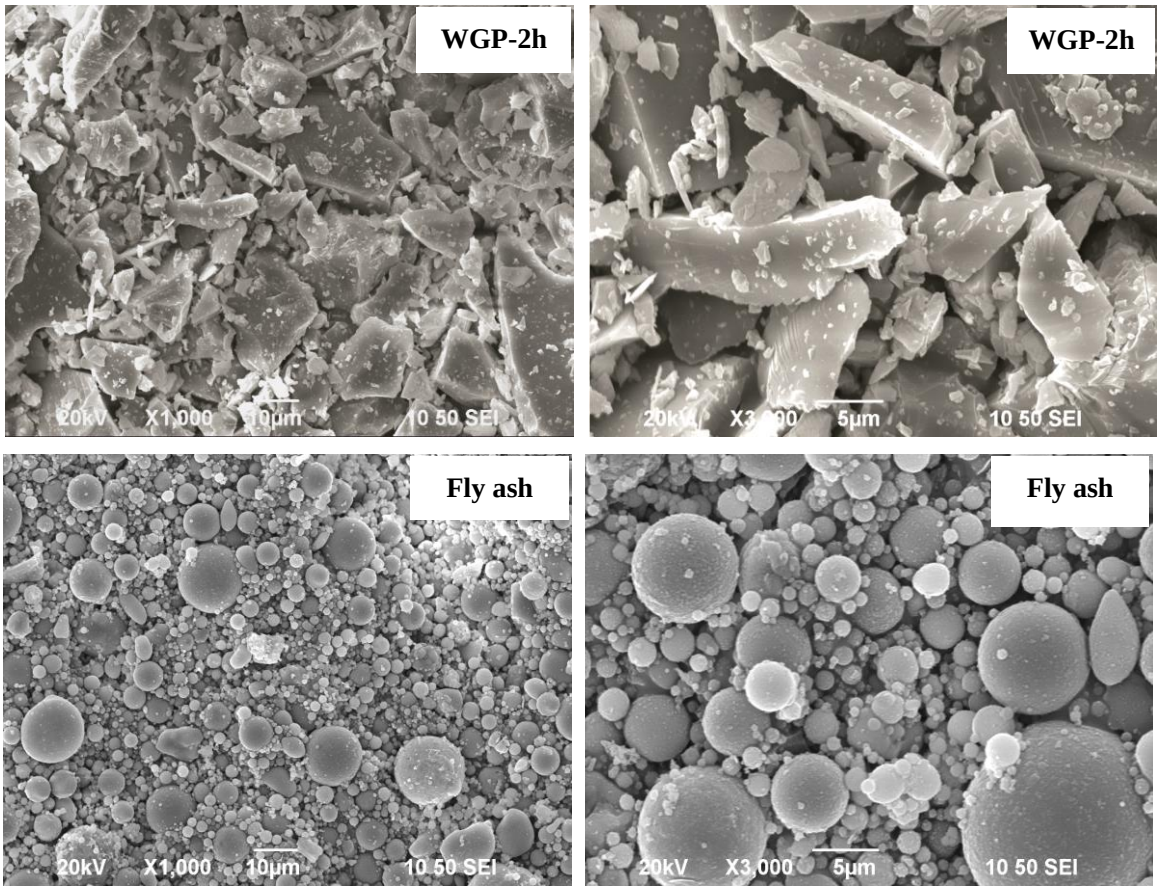


Fig. 3 Morphology of WGP and FA

2.2 Research Framework

The methods used were divided into four parts. The first part aimed to determine the effects of WGP with

different fineness on the early fresh behavior of the cement-glass mortars. The heat of hydration associated with the corresponding heat release rate were studied in the second part. The third part aimed to evaluate the influences of the WGP on the hardened properties of the cement-glass mortar, which focused on comparing the mechanical properties of the mortars prepared with different types of SCMs (WGP and FA). The last part was to study the microstructure of the cement-glass mortar (morphology of ITZ and pore structure). An overview of the research framework is illustrated in Fig. 4.

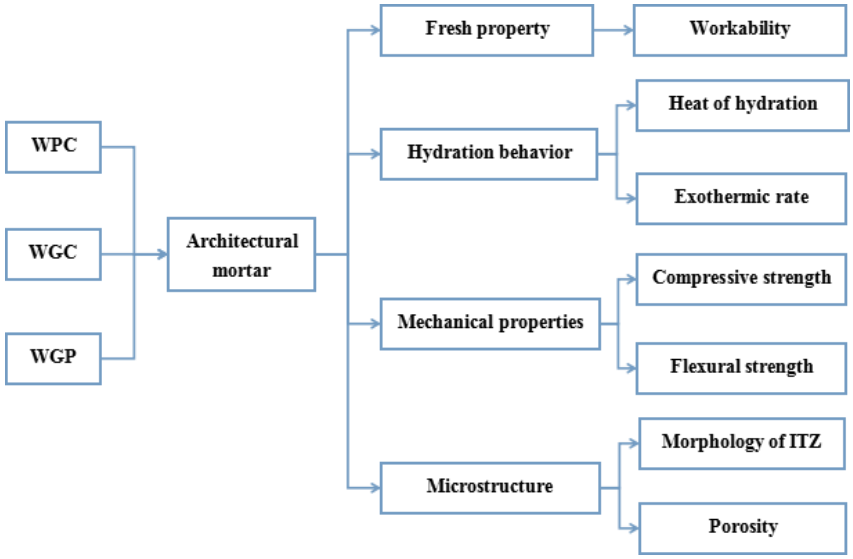


Fig. 4 Research framework

2.3 Test methods

2.3.1 Workability

The flowability of the mortar was determined according to BS EN1015 [22]. Table 2 shows the mix proportions of different mixtures prepared. The flow value was measured by means of a mini-slump flow cone with a 100 mm internal diameter on a 250 mm flow table disc. The mould was firstly filled with the fresh mortar, and then raised vertically to spread out the mortar on the disc by jolting the flow table 15 times at a constant frequency. Two perpendicular spread diameters of the mortar before and after jolting were measured and recorded.

Table 2

Mix proportions of cement-glass mortar mixtures

Mix	WPC (kg/m^3)	WGP (kg/m^3)	FA (kg/m^3)	WGC (kg/m^3)	Water (kg/m^3)	SP (kg/m^3)	*w*/*b*
Control	706	0	0	1412	283	4.2	0.4
WGP-0.5h	565	141	0				
WGP-1h	565	141	0				
WGP-2h	565	141	0				
WGP-4h	565	141	0				
FA	565	0	141				

Note: A superplasticizer (SP) ADVA-109 (W.R. Grace) without the presence of chloride was used to control the workability.

2.3.2 Heat of hydration

The heat hydration test was utilized to investigate the effect of WGP incorporation on the hydration of the cement. During the test, 20% of WGP was used to replace the cement by mass. The cement and WGP were firstly mixed in an insulated container thoroughly, then 60 grams of this mixture were mixed with 24 grams of water (*w*/*b* = 0.4) for 2 minutes in the container used for the heat of hydration test. Having completed the above steps, the container was sealed and then placed into the isothermal calorimeter (Calmetrix I-CAL). The instrument was set to a constant temperature of 20 ℃. After 48h, the measurement was stopped and the data were exported and analyzed.

2.3.3 Mechanical properties

The mortar mixes containing the WGP or FA (based on the total weight) replacements were prepared by adopting a *w*/*b* of 0.4 and aggregate-to-binder ratio of 2. The mix proportions of the mortars for the strength tests are given in Table 2. All the mixtures were mixed thoroughly before the fresh mortars were cast into steel prisms molds with the size of 40 mm × 40 mm × 160 mm. Each mold was put on a vibrating table for 15s for compaction. After 24h, these prisms were demolded and kept in a standard water curing tank until 7, 28 and 90 days of curing. Then, the specimens were removed from the tank and the three-point flexural strength test was carried out in conformity with ASTM C348 [23]. The equivalent compressive strength test was carried out after the completion of the flexural strength test according to ASTM C349 [24].

2.3.4 Microstructure tests

2.3.4.1 Preparation of samples

The samples for the porosity test and SEM morphology observation were collected after the strength test at 90 days. The fractured mortar specimens were further broken up into fragments and stored in sealed bottles which were filled with anhydrous ethanol to stop the cement hydration. After at least of a week of immersion, the small pieces of mortar were transferred to an oven at a constant temperature of 60 ℃ for another 3 days to remove the residual ethanol.

2.3.4.2 Morphology

Morphological investigation was carried out by SEM and energy dispersive X-ray spectroscopy (EDS) on the gold-coated fractured surface of specimens using a JEOL Model JSM-6490.

2.3.4.3 Porosity

As known, cementitious materials are porous and there are three main categories of pores including air voids (micrometres to millimetres), capillary pores (micrometres to nanometres) and gel pores (nanometer-sized) [25]. In this study, the pores in mortar detected included capillary pores, and pores in the ITZ, and air voids. The size distribution of the accessible pores in the mortar was examined by a mercury intrusion porosimeter (MIP, Micromeritics AutoPore IV 9500 Series) with a maximum mercury intrusion pressure of 207 MPa. The sample preparation for porosity test was described in Section 2.3.4.1. The porosity tested in this method was a wide range of pore sizes from 150 μm down to 7 nm. The mercury could only intrude the pores that were interconnected and accessible from the outside. The volume of mercury penetrating a sample was measured as a function of increasing pressure (p), which was related to the diameter (d) of the pores that had just been penetrated at that pressure by the equation: $p = 4\gamma \cos\theta/d$. Where γ is the surface tension of mercury and θ is advancing contact angle with the solid. This test used the usual assumption that the pores were cylindrical and θ was 140°.

3 Results and discussion

3.1 Workability

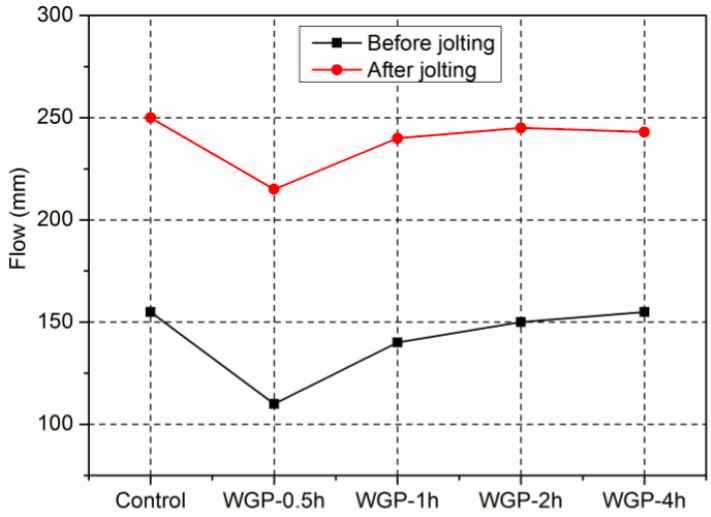


Fig. 5 Flow values of mortars with different WGP fineness

Fig. 5 presents the influence of WGP on the workability of freshly mixed mortars. The result shows a big drop in workability when the WGP-0.5h was incorporated into the mortar. This is mainly due to the larger particle size and irregular shape of the WGP. Similar results had been obtained by Park et al. [26] and Tan et al. [27], who pointed out that the sharper edge and more angular shape of glass particles reduced the fluidity of cement mortars or concrete. However, this result was contrary to other studies using glass cullet as aggregates which showed improved flowability due to the smooth surface and non-absorbent nature of glass cullet [28][29].

To explain this, two characteristics of WGP should be taken into account. One is the non-hydrophilic and smooth surface of WGP, and the other is the irregular and angular shape of WGP. The former might cause an increase in the effective water to cement (*w*/*c*) ratio, which would improve the fluidity of the fresh mortar. On the contrary, as a result of the sharp edge and high aspect ratio, the incorporation of WGP would hinder the movement of the mortar. Therefore, it is expected that if the workability is controlled by the former, the flow value will be increased. If the latter becomes predominant, the flow value would be reduced. In the case of WGP-0.5h, it was found that the average particle size was ten times larger than that of cement grains. Undoubtedly, the bigger and irregular particles would increase the flow resistance of the fresh mortar. In fact, it is of interest to notice that the flow value gradually increased with decreasing particle size of WGP, probably because the finer particles could reduce the friction due to the irregular shape.

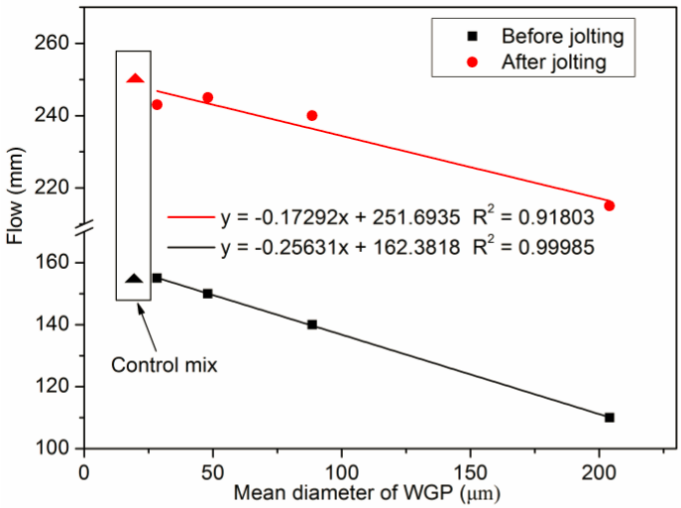
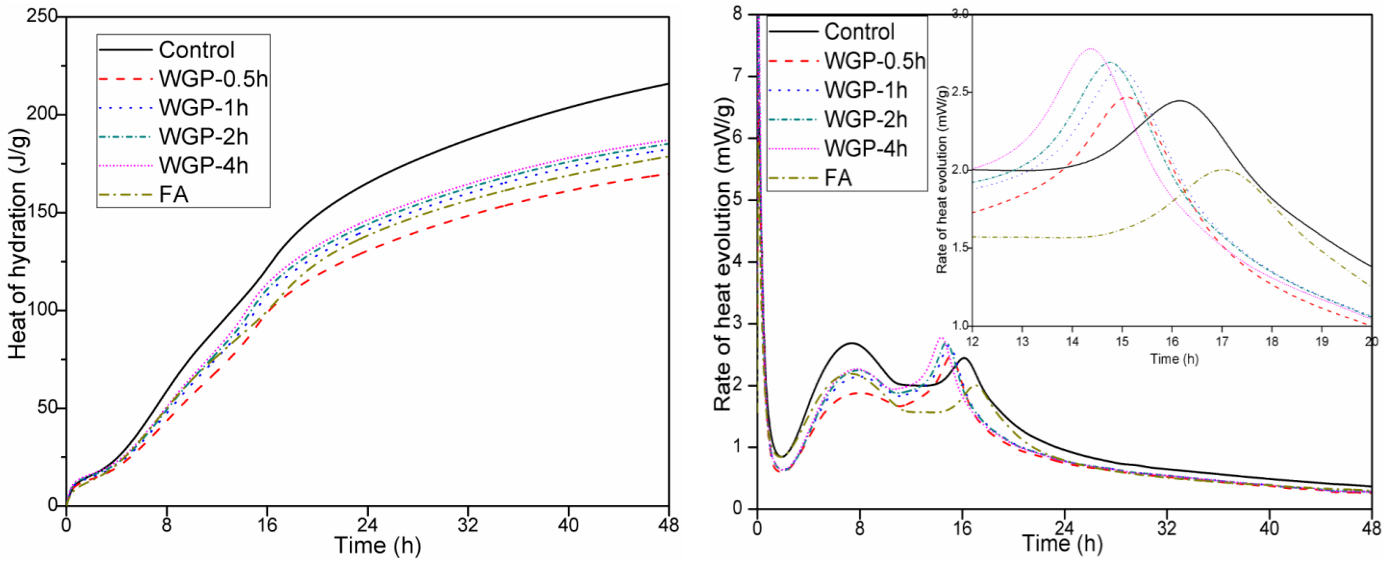


Fig. 6 Effect of particle sizes of the WGP on the flow of mortar

The correlation between the glass particle fineness and flow value of the mortar is illustrated in the Fig. 6. As shown in the Figure, a good linear relationship could be noticed between the two indexes regardless of whether jolting was applied. It can be noticed that if the mean diameter of WGP was reduced to less than 50 μm, the mortar had a similar workability to that of control mortar although the particle size of WGP was still larger than that of cement. This suggests that the non-absorbent surface of WGP could make up the loss of workability due to the irregular shape.

3.2 Heat of hydration

Fig. 7 shows the results of the isothermal calorimetry tests. As indicated in Fig. 7a, during the early stage, the plain cement paste gave out more heat than that of the WGP and FA blended pastes. Heat evolved tended to decrease with the partial replacement of cement by WGP and FA due to the dilution effect of WGP and FA with low reactivity at this early stage. It is clear that the heat output of the WGP-0.5h modified paste was the lowest among all the mixtures for its largest particle size and lowest surface area. However, it can be seen that the heat outputs of WGP-2h and WGP-4h pastes were only slightly higher than that of the WGP-1h blended paste. A possible explanation is the similar fineness of WGP-1h, WGP-2h and WGP-4h as compared with that of WGP-0.5h.



(a) Heat of hydration (b) Rate of heat evolution

Fig. 7 Heat evolution pastes with different fineness of WGP

It is widely accepted that there are two peaks in a typical isothermal conduction calorimeter curve of Type I Portland cement. However, it is interesting to note that in this study three peaks are present in the heat evolution rate curves (Fig. 7b). It should be noticed that the cement used in this study was a type of white cement. Several previous studies have shown the similar observation when the white cement was used [30][31]. Bye [32] stated that the heat evolution could produce a third peak as a shoulder on the broad second peak if a cement has a C_3A content of higher than the usual range of 8-10%. Based on the chemical composition of the white cement used and using the Bogue equations [33], the estimated C_3A content in the white cement is 13.63%, which is much higher than that of Type I Portland cement (9.31%) reported in a recent publication [34].

Based on earlier literatures [35][36], it is known that the hydration reaction of the C_3A phase is a two stage process. In stage 1, C_3A reacts rapidly with water and calcium sulfate to form AFt phase ($C_3A.3C\bar{S}.H_{32}$), which is usually referred to as the mineral ettringite (Eq. 1). The AFt phase has a distinctive needle-rod like morphology. It is widely accepted that the reaction of stage 1 makes a significant contribution to the initial exothermic heat evolution [37]. In stage 2, if there are surplus and unreacted C_3A and when the supply of sulfate ions runs out, the ettringite reacts with C_3A and water to form a AFm phase ($3C_3A.C\bar{S}.H_{12}$). This process is also known as the conversion from AFt to AFm (Eq. 2). Also, the richer C_3A content in the white cement may provide another approach for producing C_4AH_{13} due to the direct hydrolysis of C_3A (Eq. 3) [32][35]. Therefore, other than the C_3A reaction of stage one, the C_3A hydration of stage two was mainly responsible for the third peak in the isothermal conduction calorimetric curve.

Stage 1:

$$C_3A + 3C\bar{S}H_2 + 26H \rightarrow C_3A.3C\bar{S}.H_{32} \quad (1)$$

Stage 2:

$$C_3A.3C\bar{S}.H_{32} + 2C_3A + 4H \rightarrow 3C_3A.C\bar{S}.H_{12} \quad (2)$$

$$C_3A + 21H \rightarrow C_4AH_{13} + C_2AH_8 \quad (3)$$

Note: C = CaO; S = SiO_2; A = Al_2O_3; H = H_2O; $\bar{S}$ = SO_3.

It is particularly worth pointing out that the insert to Fig. 7b shows the time of appearance of the third peak was accelerated and its intensity was increased when the WGP was used to partially substitute cement. In spite of the replacement of cement by WGP, the C_3A to calcium sulfate ratio in the cement pastes was still constant. Moreover, as a result of the low reactivity of WGP in the early age and its non-absorbent nature, the major difference between the WGP blended pastes and the reference paste was the *w*/*c* ratio. As mentioned, an incorporation of WGP led to an increase in the effective *w*/*c* ratio, which means that more water was available for hydration. For the C_3A hydration at stage two, the excess water would probably accelerate the reaction of Eq.1 and Eq.2 based on chemical equilibrium, thus causing an enhancement of heat output.

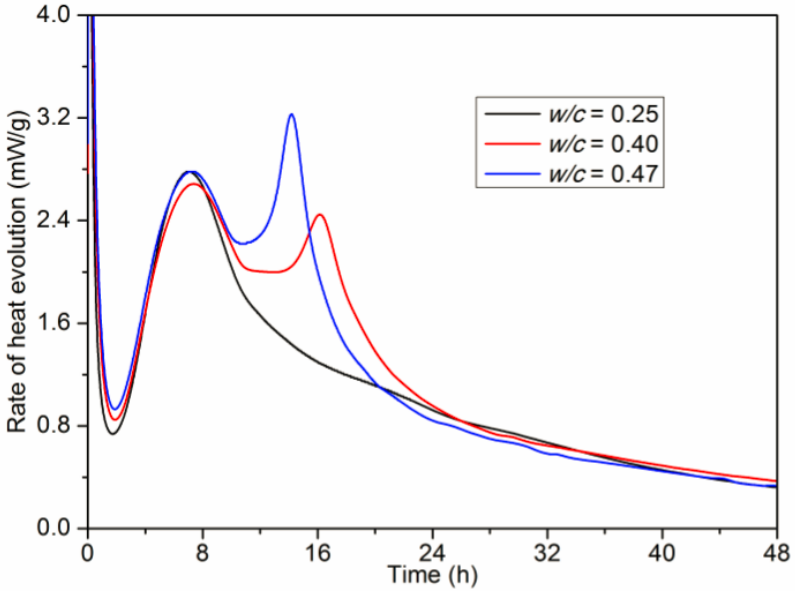


Fig. 8 Exothermic rate of WPC with different *w*/*c* ratios

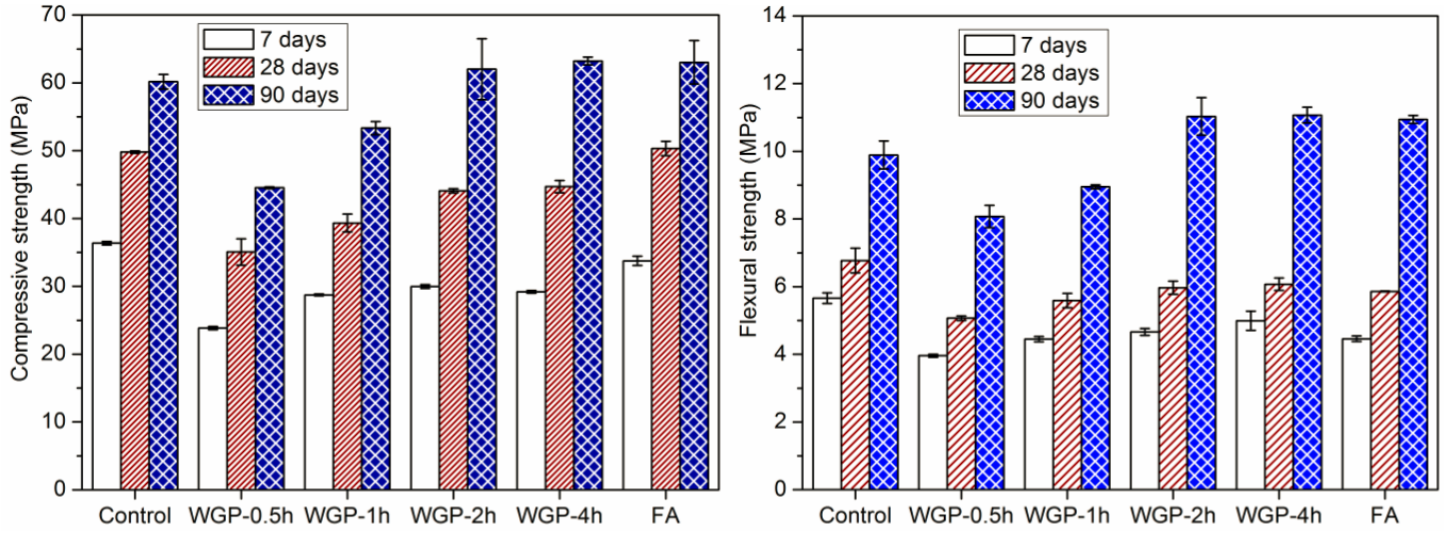
In order to verify this explanation, the heat evolution curves of the pure white cement pastes prepared with different *w*/*c* are obtained in Fig. 8. It can be clearly seen that the third peak became more distinctive with increasing *w*/*c* ratio (from 0.40 to 0.47). In contrast, the third peak disappeared when the *w*/*c* ratio was reduced from 0.40 to 0.25 due to the lack of water for C_3A hydration during stage two. The above results indicate that

the water content noticeably affected the hydration of C_3A, especially for the stage two, which provides strong evidence that the addition of WGP would facilitate the formation of AFm phase from AFt phase due to the richer amount of water in the system.

From the insert in Fig. 7b, it can be seen that the third peak became bigger with the increase in the fineness of WGP. According to the study of Nehdi et al. [38], the improved gradation of the binder imparted by the fine particles could possibly reduce the particles interlocking. Also, particles with small size could provide an internal lubricant effect by displacing water from the voids between the particles [39]. Therefore, it is believed that the inclusion of fine glass powder could promote the hydration of C_3A compared to the coarse glass powder. On the contrary, in the case of FA blended cement paste, it can be noted that the addition of FA retarded and reduced the third peak in the heat evolution curve. The reason may be caused by the finer FA particles were adsorbed on the surface of the cement particles due to the electrical charges [40], which may hamper the hydration of the C_3A phase. The retardation phenomena of C_3A phase as a result of FA or other pozzolanas were also reported by Wei et al. and Collepardi et al. [41][42].

3.3 Mechanical properties

The compressive and flexural strength of the mortar at 7, 28 and 90 days are presented in Fig. 9. It is easy to find out that the compressive strength of mortar decreased obviously by using WGP to replace cement. This can be contributed to the fact that WGP had a low pozzolanic activity at the early ages. The observation agreed with the result obtained by Tan et al. and Mirzahosseini et al. [27][43]. Except that the compressive strength of mortar in this study was found to be increased with an increase of WGP fineness. It seems that the particle size of WGP was one important factor responsible for the increased reactivity. This is consistent with the results of the conduction calorimetry tests.



(a) Compressive strength (b) Flexural strength

Fig. 9 Mechanical properties of mortars prepared with WGP or fly ash

It is worth to notice that, in the cases of WGP-2h and WGP-4h, there were remarkable increases in the compressive strength at 90 days, which even exceeded the strength of the control group by 3.1% and 5.1%, respectively. This strength level was also similar to that of the FA blended mortar, demonstrating that the finer WGP with larger specific surface areas facilitated the pozzolanic effect. This result was in agreement with the study of Afshinnia and Rangaraju [44], which showed that the long-term pozzolanic effect of the finer glass powder was likely to be sufficient to overcome the dilution effect. The early compressive strength of FA blended mortar was noticed to be relatively higher than that of WGP mortar. This is probably due to the relative smaller particle size of FA than that of WGP.

A similar trend was observed in the development of the flexural strength. The introduction of WGP or FA at 20% cement replacement level caused a significant decrease in the flexural strength. However, one point should be noticed was that the flexural strength of WGP-2h and WGP-4h blended mortars were the highest even higher than the FA blended samples, which was not consistent with the development of the corresponding compressive strength. This suggested that the finer WGP had an advantage in increasing the flexural strength of the mortar.

In fact, based on the above strength results, we cannot intuitively understand what the actual contributions of WGP and FA to the strength development were. For this purpose, it is necessary to determine the contribution to strength attributed to the reactivity of WGP or FA. In most publications, a strength activity index (Eq. 4) [44][45] and a strength efficiency factor [46] was often adopted. However, these methods still cannot directly express the contribution by the SCMs or other admixtures. Therefore, in this present study, a contribution rate to strength (CRS) concept was established to evaluate the pozzolanic activity of WGP or FA. The CRS can be calculated by

the following derivation process:

$$I_S = \frac{S_m}{S_c} \quad (4)$$

I_S stands for the strength activity index, which is defined as the ratio of S_m to S_c. In this study, S_m is the strength of mortar prepared with WGP or FA, while S_c is the strength of the control mortar.

$$R_I = \frac{S_m}{S_c * k} \quad (5)$$

R_I is defined as the relative strength index, k is the percent of cement to total amount of binder. Due to the cement was replaced by the WGP or FA at a 20% level, k is 0.8. The relative strength index of control mortar $R_{I(control)}$ is 1, which can be calculated by following formula ($S_m = S_c$, k = 1):

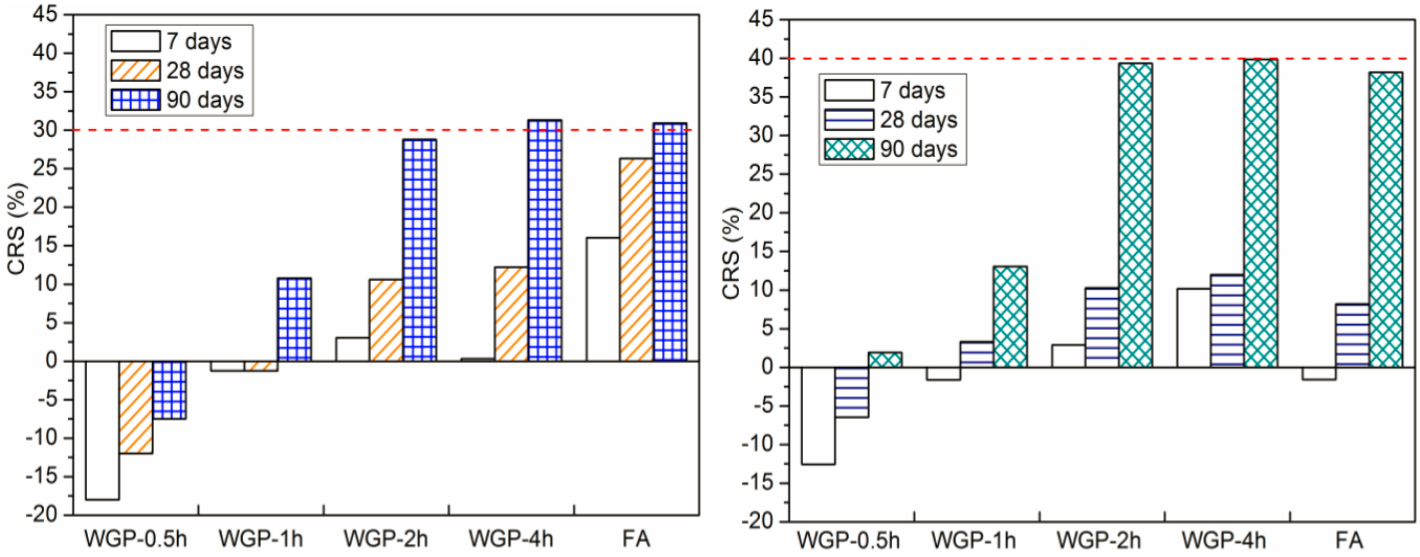
$$R_{I(control)} = \frac{S_m}{S_c * k} = \frac{S_c}{S_c * 1} = 1 \quad (6)$$

Consequently, the contribution rate of WGP or FA to strength of mortar can be derived as follows:

$$CRS = \left(R_I - R_{I(control)}\right) * 100\% = \left(\frac{S_m}{S_c * k} - 1\right) * 100\% \quad (7)$$

According to Eq. 7, it is easy to calculate CRS if we know the strength of samples containing the admixtures (S_m), the strength of control group (S_c) and the percentage of cement in the binder (k).

The CRS to the compressive strength and the corresponding flexural strength derived from WGP or FA are illustrated in Fig. 11. In theory, 20% by weight of cement in the mortar can offer 20% of strength. That is, if the CRS is higher than 20%, the 20% of WGP or FA replacement is considered to make a higher contribution to strength than the equal amount of cement. A positive CRS value, means the admixtures can make a positive contribution to the strength, and vice versa.



(a) Compressive strength (b) Flexural strength

Fig. 10 Contribution rate to strength of mortars with WGP or FA

It can be seen from Fig.10a that the WGP-0.5h had a negative effect on the compressive strength due to its low reactivity and the higher effective *w*/*c* in the mortar. However, the CRS gradually increased with the increasing of WGP fineness and the negative effect of finer WGP on the strength appeared to be insignificant. At the age of 90 days, the CRS was about 30%, which was even comparable to that of the FA. In the case of WGP blended mortars, it can be observed by comparing Fig.10a and Fig.10b that the contributions to flexural strength derived from WGP were higher than that to compressive strength, irrespective of the particle size of WGP. This is in particular significant at 90 days of curing. Also, it is interesting to notice that the incorporation of 20% finer WGP into the mortar contributed approximately 40% of the flexural strength grow (about two times higher than the theoretical value of 20%). Correspondingly, the contribution to compressive strength were only about 30%.

It can be seen that the WGP-2h or WGP-4h blended mortars had a higher contribution rate to the flexural strength than the FA blended mortar, particularly at 7 and 28 days, while the contributions of WGP to the compressive strength were much lower than that of FA at early ages. In general, the flexural strength was proportional to the compressive strength [26], but contrary results were recorded for the case of WGP and FA mortars. According to Poon et al. [47], even at 28 days of curing, there were still more than 80% FA playing the role as space filler or micro-aggregates, and the pozzolanic reaction between the FA and the calcium hydroxide (CH) only became important at a later age. Therefore, it was expected that the FA contributed relatively little to the flexural strength at the early age. In comparison, although the WGP had larger particle size and lower reactivity than the FA, WGP with irregular shapes and high aspect ratios (see Fig .3) possibly played the role as micro-fibers in the mortars, which may have significantly improved the flexural strength. This explanation is

consistent with the study of Dey and co-workers [48], who reported that the flexural strength of a cement mortar was enhanced by 40% when wollastonite micro-fibers with an average particle size of 55 μm were used as 15% of cement replacement after 28 days of curing. Furthermore, comparison with the increase of compressive strength, the micro-fibers were far more effective for flexural strength enhancement when cured for longer durations. At the later age, the morphologic effect of WGP would become more significant due to the secondary hydration reaction taking place at the surface of WGP particles. This might be another important factor in increasing the flexural strength by chemical bonding between the glass particle and the cement matrix. Therefore, the combination of morphologic and pozzolanic effects may explain why the WGP was beneficial to the flexural strength of the mortars.

3.4 Microstructure

3.4.1 SEM Morphology

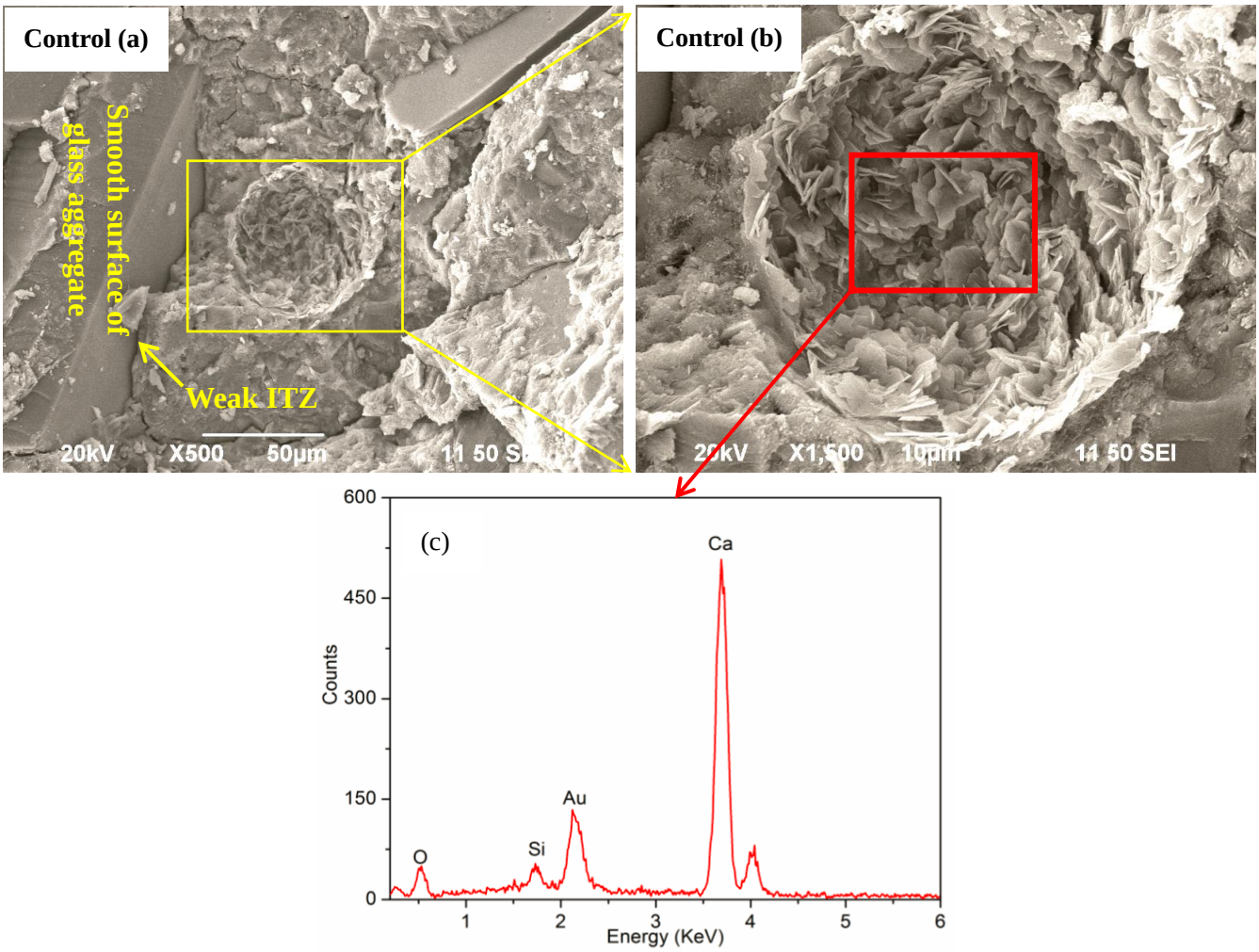


Fig. 11 Morphologies of control mortar and corresponding EDS

The SEM images and the corresponding EDS results of the reference mortar are shown in Fig. 11. From the

fracture surface (Fig. 11a), it can be seen a big void (about 50 μm) was present near the glass aggregate. This was because water was enriched around the glass aggregates (localized bleeding) due to the non-absorbent nature of the glass, along with the hydration between water and anhydrous cement, the sites originally occupied by the water would form many voids. It is envisioned that a more porous area was present between the cement matrix and the glass aggregates, (known as the ITZ), would possibly lead to a reduction in strength and durability of the mortar [33]. A weak ITZ can also be seen in Fig. 11a on account of the smooth surface of the glass aggregates. This is consistent with the previous research [3]. There were a lot of platelets present (as Fig. 11b illustrated). As shown in the EDS (Fig. 11c), calcium was the main elemental component, which demonstrated these coarsely crystalline products were CH. A similar phenomenon had also previously been identified [49]. It is possible that water in the voids could generally react with or dissolve the still unhydrated cement grains, resulting in the nucleation and crystallization of CH from the supersaturated solution. Furthermore, the available free spaces of bigger voids were favourable for the growth of CH crystals. As known, the crystalline CH is a weak link in the hardened cement paste because of its chemical instability and low specific surfaces [37]. Also, a higher concentration of CH would precipitate in the ITZ, whilst the higher porosity and probable higher connectivity of this porosity suggested that transport of chemical species should be faster in the ITZ [50]. Therefore, for the control mortar, one can speculate that the enrichment of CH crystals and the higher porosity in the vicinity of the ITZ may cause a decrease in strength and durability.

Fig. 12 shows the morphology of the mortars prepared with WGP-0.5h, WGP-4h and FA. As shown in Fig. 12a and Fig. 12b, it is apparent that porous structure and loose interfacial zone were present in the WGP-0.5h matrix and the ITZ, while relatively denser structures were exhibited in the case of the mortars prepared with WGP-4h and FA (see Fig. 12d and Fig. 12f, respectively). This explained the lower strength of the mortar prepared with WGP-0.5h compared to that of WGP-4h and the FA blended mortars. It seems to be due to the fact that, the degree of pozzolanic reaction between the CH and the WGP-0.5h was low due to the lower reactivity of coarser glass particles. When the cement was replaced by the finer WGP or FA, the secondary hydration products due to the pozzolanic reaction between the finer WGP or FA and the abundant calcium ions in the ITZ would fill the pores and compact the paste matrix, thus resulting in an increase of strength. Nevertheless, the SEM observations in Fig. 12c and Fig. 12e show that unreacted glass particles and FA particles were still available in the matrix even at 90 days, revealing that the micro WGP and FA particles could act the same role as micro-aggregates in filling the space of the mortar structure. In addition, it is noticed that the edges of glass and FA particles had been effectively activated by the alkaline hydration products, and the distinctive interfacial zones between the particles and the paste disappeared because of pozzolanic effect of WGP and FA. Therefore, it can be inferred that the strength of the mortar prepared with WGP or FA would be enhanced due to the continuous reaction between the WGP or FA particles and the remnant CH.

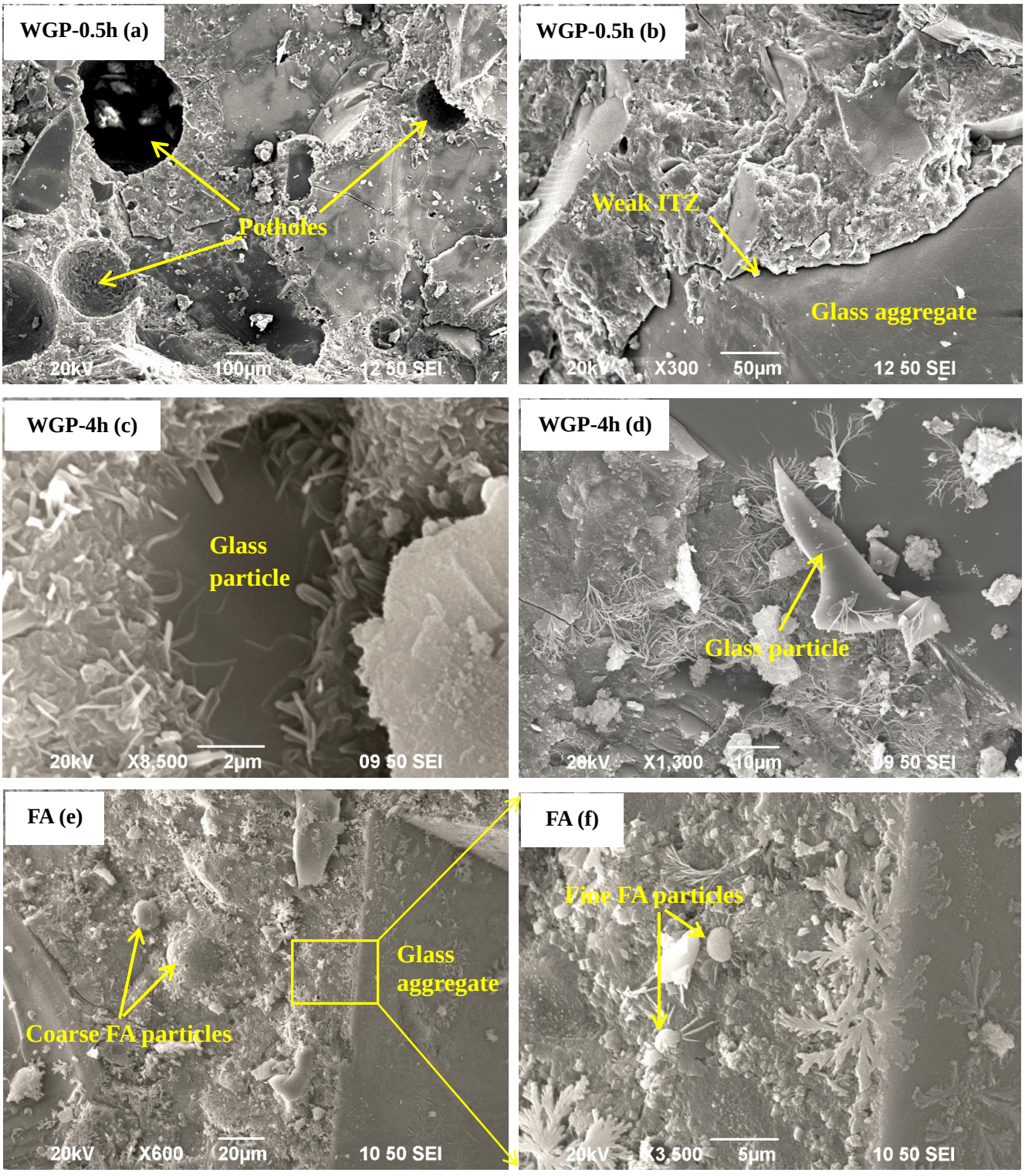


Fig. 12 Fracture morphology of mortars with WGP or FA

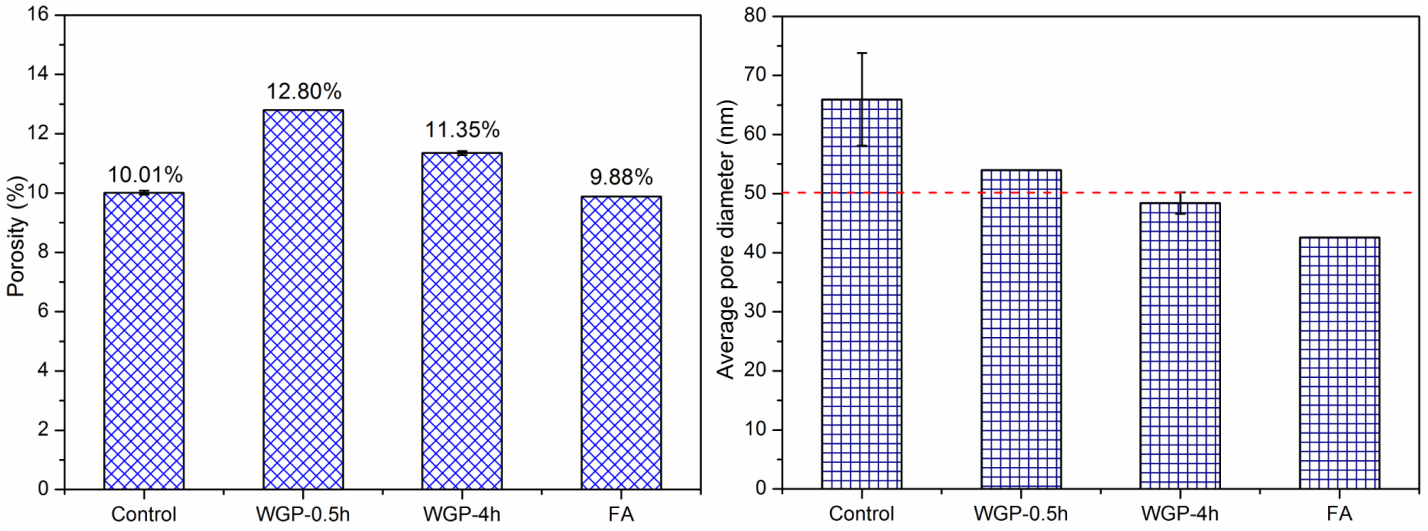
An interesting difference in morphology is noticed in Fig. 12d and Fig. 12f. The surface of the sample prepared with WGP-4h showed many fibrillar hydration products, while the FA sample showed many cluster

shape products. These products were mainly produced by the pozzolanic effect of WGP and FA. This is in agreement with the finding by Hu and Stroeven [51], who indicated that increasing the concentration of silicate ions or addition of mineral admixtures were effective ways to improve the microstructure of the ITZ. Due to the high amorphous silica content in the WGP, the formation of fibrous shaped products (Fig. 12d) in the ITZ were conducive to strengthening the bonding between the aggregates and the paste.

3.4.2 Pore structure

It is well-known that high porosity is highly detrimental to the mechanical properties of cement-based materials, and the pore size distribution also plays an important role in determining the strength and permeability of the composite materials [52]. The structure of the pores is considered at different levels, and the main interest is concentrated on the so-called micro-level (< 50 nm) and macro-level (> 50 nm) pores [33].

The pore structure of the mortars prepared with and without WGP and FA were determined by MIP, the results are summarized in Fig. 13. The experimental results (Fig. 13a) indicated that replacement of cement by WGPs increased the porosities of the mortars. This can be explained by the morphology of WGP that the sharper edge and more angular shape of glass particles led to a reduction in the fluidity of mortar and hence poorer compaction. In addition, the inclusion of WGP would bring about a higher effective *w*/*c*, it is therefore expected that the porosity of matrix would be increased with the increasing *w*/*c*. The results also showed that the porosity of WGP-4h blended mortar was smaller than that of WGP-0.5h (11.35% Vs 12.80%), which means that as the fineness of WGP was increased the porosity of the mortar decreased. This is probably because the mortar prepared with a finer WGP had a denser structure due to the filling and pozzolanic effects. The explanation is also consistent with those of the strength test results.



(a) Porosity (b) Average pore diameter

Fig. 13 Pore structure of mortars prepared with WGP or fly ash

However, it can be seen that the porosity of FA mortar is comparable to that of the control mortar. The observation is in agreement with the study conducted by Poon et al. [53], who found that using 15% and 25% FA to replace cement had little effect on the porosity of the mortar, whereas a more significant effect was on reducing the average pore diameter because of the improved interfacial microstructure between the paste and the aggregate. A similar trend of reduced pore size is found in Fig. 13b. Also, the addition of WGP-0.5h and WGP-4h reduced the average pore diameter although the total porosities of WGP mortars were higher than that of the plain mortar. This may be explained by, in the presence of WGP, the liberated CH was consumed to produce the additional C-S-H (pozzolanic effect), particularly for those plate-like crystals of CH present adjacent to the glass aggregates in the ITZ (as shown in Fig. 11 and Fig. 12). The formation of secondary hydration products could fill the large pores and microcracks with microporous products. Therefore, a positive contribution is expected from the effect of WGP addition on the pore size distribution of the mortar. As indicated in Fig. 13b, the average pore diameters of WGP-4h and FA mortar were less than 50 nm, which means that the incorporation of finer WGP or FA shifted the pore size distribution towards smaller sizes and more fine pores were present in the matrix compared to the control mortar. Therefore, the grain-size refinement in oriented CH crystals and pore-size refinement could be responsible for enhancing the strength of the mortars.

4 Conclusion

The effectiveness of WGP of various particle sizes on improving the fresh and mechanical properties of architectural mortars were evaluated. The study findings suggested that the improved ITZ and the pore-size refinement due to the inclusion of finer WGP (< 50 μm) could overcome the dilution effect and compensate for the drawbacks of the smooth surface of the glass aggregates. Therefore, the use of a combination of 100% WGC to replace natural aggregates and 20% WGP as a partial replacement of cement is feasible to produce high performance of architectural mortars. Based on the experimental results, the following conclusions can be drawn:

1. The effect of WGP on the workability of mortar depends on its surface characteristics including the smooth texture, non-absorbent nature, sharp edge shape with high aspect ratios. When the particle size of WGP was reduced to less than 50 μm, the workability of WGP blended mortar was similar to the plain mortar without addition of the WGP. Moreover, there was a good linear relationship between the particle size and the flow values, revealing that the particle size of WGP played an important role in controlling the workability.
2. The use of WGP as a cement replacement led to a reduction of hydration heat, not only reducing the second exothermic peak of hydration but also changing the third peak where the formation of AFm by secondary C_3A hydration with AFt.
3. The mechanical results indicated that the WGP had low pozzolanic activity at the early ages, whereas the strength of the mortar prepared with finer WGP showed a distinctive improvement at 90 days of curing, eventually surpassing that of control and FA mortar due to the finer pore structure and the denser ITZ.
4. A concept of contribution rate to strength (CRS) was proposed to evaluate the actual contributions of SCM to the strength development of cement based materials. Based on the CRS results, it was found that the incorporation of finer WGP in the mortar could develop an efficient advantage in increasing the flexural strength over the compressive strength, especially in the early age. The finer WGP blended mortar showed a higher contribution rate to the flexural strength than that of FA. This was probably due to the fact that the finer WGP could play the role as micro-fibers in enhancing the flexural strength.
5. Based on the experimental results, the WGP with particle size less than 50 μm was recommended to replace 20% cement in the production of architectural mortar with 100% glass aggregates.

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