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Recycling of fly ash-slag Geopolymer binder in mortar mixes

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ABSTRACT
Fly ash-slag based Geopolymer cement (GPC) has demonstrated mechanical properties and environmental advantages that make it one of predominant sustainable alternatives to Portland cement (PC). Despite the fact that numerous environmental analyses about geopolymers are being published, their environmental impact after the end of service-life has barely been explored. Given that construction-waste management is a major sustainability issue, the present study is investigating the potential of recycling fly ash-slag GPC as a fine aggregate in mortar mixes. The major physical properties of the fine recycled aggregates (FRA) were tested and compared to those of PC FRA and natural sand of similar fineness. The effect of incorporating FRA in low (25%) and high (50%) percentage in PC or GPC matrix mortars was investigated. The 28day compressive and flexural strength of mortars were tested. Also the 28day water absorption and flow of mixes incorporating GPC FRA were recorded. GPC FRA exhibited properties similar to those of PC FRA and poorer than those of natural sand. The results of compressive and flexural strength proved that FRA addition had a negligible effect in all cases. The influence of the high water absorption of GPC FRA, relatively to that of natural sand, was prominent on the workability of fresh mixes and possibly affected the water absorption of mortar prisms. The effect of GPC FRA proved to be similar to that of PC FRA on compressive strength, while none of the tested mortar properties appeared to be jeopardised by the incorporation of the GPC FRA in the mix.

1. INTRODUCTION

Geopolymers were popularised by Joseph Davidovits in the 1970's and their production relies on minimally processed natural materials or industrial by-products. There are various categories of geopolymer binders (depending on the raw materials), but from a terminological point of view, geopolymer cement is a binding system that hardens at ambient temperature. Given that this relies on addition of calcium (usually through ground granulated blast furnace slag - GGBS), the most appropriate type for high volume construction applications is fly ash-slag based geopolymer (Davidovits, 2015). With the adoption of that type of geopolymer cement (GPC), extreme high temperature kilns with large expenditure of fuel and CO_2 emissions by the decomposition of calcium carbonate are avoided. Therefore a reduction of carbon emissions at the range of 40-80% is considered possible (Davidovits, 2013). But before proceeding to its wider adoption, the engineering community has to ensure tangible environmental benefits.
An increasing number of life cycle assessments and environmental evaluations of GPCs are being conducted. Most of them are focusing on cradle-to-gate issues relating to the availability of materials or the environmental impact of the activating solutions (Habert, et al., 2011) (Fawer, 1999) (Heath, et al., 2014). Hardly any research concerning the stage after the end of service-life has been conducted. Disposal of construction and demolition wastes is an issue that already troubles the construction industry as far as Portland cement (PC) concrete is concerned. Therefore, it has been deemed appropriate to investigate disposal or reuse scenarios for GPC since the potential of its wider adoption is becoming higher. The present study is investigating the potential of recycling fly ash-slag based GPC by utilising it as recycled aggregate in mortar mixes.
To do so, fine recycled aggregates (FRA) were produced in the laboratory and their basic physical properties such as water absorption (WA) and particle density were investigated. The potential of recycling GPC in mortar mixes with PC and fly ash-slag based GPC matrices was investigated. The effect of GPC FRA on the 28day flexural and compressive strength of mortars was tested and the results were compared to those obtained by similarly produced mortars with natural sand and PC FRA. Additionally, the flow and water absorption of mortars incorporating GPC FRA were recorded.

2. Materials and Experimental methodology

The GGBS used for the GPC precursor was provided by the Hanson Heidelberg cement group from the Port Talbot works. The fly ash (FA) was CEMEX 450-S (BS EN 450 - 1 Fineness Category S; LOI Category B). The activating solution was produced by mixing sodium hydroxide pellets (NaOH, 98-100.5%) and sodium silicate solution ($Na_2O(SiO_2)x \cdot xH_2O$, Na_2O, ~10.6%,SiO_2, ~26.5%) with distilled water. The chemicals were both supplied by Sigma-Aldrich and the mixing of the solution took place 24 hours prior to casting. For the PC elements, General Purpose Portland fly ash

cement/ Sulfacrete EN 197-1 –CEMII/BV 32,5R supplied by Tarmac Cement & Lime Ltd was used. The natural aggregate incorporated in the mortars was standard sand complying with BS EN 196-1 specifications. The same materials were used for the fabrication of both the recycled aggregates and the mortars. Although the PC used had fly ash addition, the specimens will be referred to as PC for convenience.
The FRA were produced in the laboratory by casting 100mm cubes of pure paste to establish a worst case. For the GPC binder the proportion of FA/GGBS was 1/1, water/GPC solids 0.4, the Na_2O/SiO_2 ratio 1 and the percentage of Na_2O in the activating solution was 5% by mass precursor. The water/GPC solids ratio was used and it indicates that the products deriving from the dissolution of the compounds of the activating solution were taken into consideration for the calculation of the free water proportion for the mix. For the PC paste the water/cement ratio was 0.4. The cubes were demoulded one day after casting and then stored in sealed plastic bags at 20±1℃ for a period of 28 days before crushing. The resulting FRA was crushed and sieved to achieve fineness and particle distribution similar to that of standard natural sand.

Table 1 Amount of materials used for the production of recycled aggregate mortars in kg/m^3

Matrix type	PC	PC	PC	GPC	GPC	GPC
RA	**0%**	**25%**	**50%**	**0%**	**25%**	**50%**
PC	960	960	960	-	-	-
FA	-	-	-	480	480	480
GGBS	-	-	-	480	480	480
NA	2160	1620	1080	2160	1620	1080
RA	-	540	1080	-	540	1080
NaOH	-	-	-	19	19	19
Silicate	-	-	-	53	53	53
Water	451	451	451	463	463	463

The replacement percentages of natural sand in the final mortars were 25% and 50% while the reference mixes (0% replacement) incorporated standard sand only. For the GPC mortars water/GPC solids ratio was 0.47 and Na_2O was 3% by mass of precursor, while all the other parameters were the same as for the original GPC binder. The water/cement ratio for the PC mortars was 0.47. For all mortar mixes the proportions were 2.25 parts of sand to 1 part of binder per mass. No presaturation of aggregates or water compensation took place. Mortar prisms of 40x40x160mm were fabricated and demoulded 24h after casting. Then they were kept at 20±1°C in moist conditions for 28 days. The amount materials used for the casting of the final mortars is presented in **Error! Reference source not found.**~~Table 1~~.
The recycled aggregates were subjected to water absorption and particle density testing according to BS EN 1097-6:2013 "*Test for mechanical and physical properties of aggregates*". The final mortars were tested according to BS EN 196-1:2005 "*Methods of testing cement"* and BS 1881-122:2011 *"Testing concrete"*. The notation used for the mixes follows the rationale Matrix type-Replacement percentage.

3. Results and discussion

The results of the FRA testing are presented in

Table 2~~Table 2~~. The apparent particle density of GPC FRA is about 20% lower than that of natural sand but still at the same order of magnitude. For oven-dried and saturated surface dried condition the difference increases at about 48% and 33% accordingly. When compared to the results obtained from the testing of similarly produced PC FRA, it appears that GPC FRA are less influenced by oven drying and water immersion.

Table 2 Particle density and 24hour water absorption of natural sand, GPC and PC FRA

Aggregate type	Natural Sand	GPC FRA	PC FRA
Particle density (kg/m^3):			
Apparent	2631	2041	2247
Oven-dried	2202	1151	895
Saturated-Surface dried	2367	1588	1497

The obtained results do not present significant divergence from those reported in literature regarding PC recycled aggregates of similar fineness. The usual ranges are 1,970-2,140 kg/m^3 for oven dried and 2,190-2,320kg/m^3 for saturated surface dried density (Dhir, et al., 1999) (Wai , et al., 2012) (Hansen, 1986) (Silva, et al., 2014b). Given the nature of the original material (binder without any aggregate) such values are considered reasonable.
The most striking observation is that GPC FRA exhibited water absorption (WA) 5 times that of natural sand. The general trends reported in literature are much lower. Specifically, PC recycled aggregates are reported to demonstrate WA 3–6 times higher than that of natural aggregates, while FRA present values at the range of 8-12% (Akash Rao, 2007) (Hansen, 1986). Given that the tested PC FRA exhibited an exceptionally high WA as well, it can be assumed that this significant increase is due to the nature of the original material and the fineness to which the FRA were crushed. When evaluating the obtained results comparatively though, it can be stated that GPC FRA had a better performance than PC FRA.
The remarkably high WA of GPC FRA was correlated with the results of the WA of the mortar prisms and the flow of fresh mixes. As it can be observed by the values demonstrated in
Table 3~~Table 3~~, the WA of GPC FRA had a prominent effect on the workability of mortars.
Specifically, a gradual reduction of flow was observed with increasing replacement levels for both matrix types. With incorporation of 25% GPC FRA in the mortar, a 13% reduction of flow occurred but the mixes maintained reasonable workability. With increased replacement level though, flow demonstrated a reduction of about 50% for both matrix types and the mixes were hard to cast.

Table 3 Flow and 28day water absorption of mortar mixes incorporating GPC FRA. The relation to the reference mixes with 0% GPC FRA is stated as reduction % and increase %.

	Flow	Reduction	WA	Increase
PC-0%	144%	-	4.9%	-
PC-25%	125%	13%	6.7%	37%
PC-50%	68%	53%	10.1%	108%
GPC-0%	145%	-	6.8%	-
GPC-25%	126%	13%	9.8%	45%
GPC-50%	77%	47%	10.6%	56%

These results are in accordance with the information reported in literature. Specifically, it is mentioned in most studies that WA of recycled aggregates influences directly the workability of the resulting mixes, as the hardened mortar tends to absorb the available water during mixing, resulting to less free water in the mix (Akash Rao, 2007) (Zhao, et al., 2015) (Evangelista & de Brito, 2007) (Pepe, et al., 2014) (Silva, et al., 2014a). Additionally, aggregate porosity and WA have been correlated with the water absorption of the resulting mortar (Zega & di Maio , 2011) (Evangelista & de Brito, 2010). This remark is confirmed by the results in

Table 3Table 3. PC mortars appeared to be more prone to the influence of GPC FRA addition, given that, for double replacement percentage, the WA doubled. For the GPC mortars the replacement level did not seem to have a remarkable impact as both cases showed increases of about 50%.

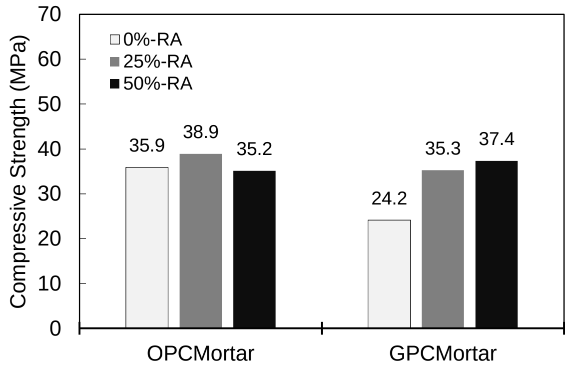

Figure 1 Results of 28day compressive strength testing of PC and GPC mortars incorporating GPC FRA

In fact, the high WA of FRA in combination with the hardened paste could lead to an overall modification of the nominal w/c ratio and the resulting compressive strength of the mix. This could be related to the obtained results for compressive strength presented in Figure 1Figure 1. In most cases the addition of recycled aggregates is reported to lead in decrease of mortar and concrete strength (Gerardu & Hendricks, 1985) (Zhao, et al., 2015) (Hansen, 1986).

It was observed that, regardless the replacement level, the addition of GPC FRA had a minor effect on the compressive strength of PC mixes. A slight increase was observed for 25% GPC FRA, while for 50% the strength is slightly lower than that of the reference mix. For high replacement level, the favourable effect was probably counterbalanced by the dryness of the mix. In the case of GPC mortars, a clear enhancement of strength is observed by the GPC FRA addition. Regardless the FRA level, a strength increase at the range of 50% took place. Apart from the justification given above for the case of PC mortars, an interpretation of the effect of GPC FRA addition could be based on the fact they derived from a binder with alkalinity higher than that of the final mortar. It is possible that alkalis from the FRA leached into the new mix resulting to the observed strength increase.

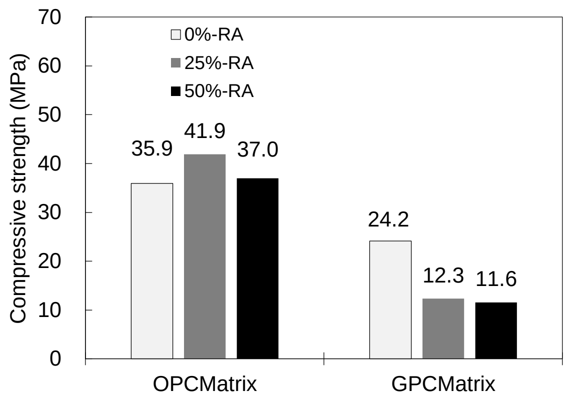

Figure 2 Results of 28day compressive strength testing of PC and GPC mortars incorporating PC FRA

The results obtained by corresponding mixes incorporating PC FRA are presented in Figure 2Figure 2. It is observed that in the case of PC mortar the replacement of natural sand had a similar effect, irrespective of the FRA type. For GPC mortars though, it is prominent that PC FRA had the opposite effect of GPC FRA. Summarizing the compressive strength results it could be stated that GPC FRA had a similar or enhancing effect compared to PC FRA.

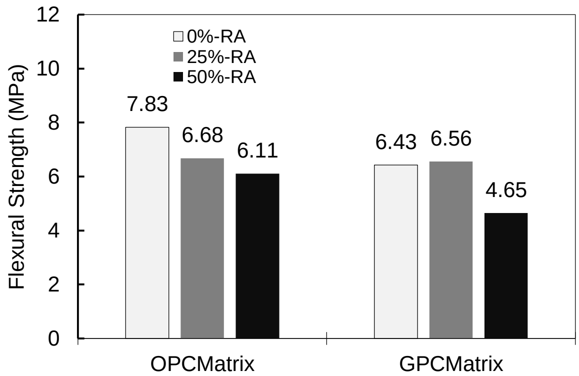

Figure 3 Results of 28day flexural strength testing of PC and GPC mortars incorporating GPC FRA

A similar trend was recorded for the results of the 28day flexural strength of mortars (Figure 3Figure 3).

4. Conclusions

The overall conclusion of the present research is that no indication that could be considered as preventive for the use of GPCs as recycled aggregates in mortar mixes was observed. GPC FRA demonstrated suitable physical properties for many applications. Although they were inferior to those of natural sand they were still within acceptable limits for recycled aggregates. Their high water absorption, which was attributed to their fineness and the nature of the original material, influenced most of the final mortar properties in both direct and indirect ways The physical properties of similarly produced PC FRA proved to be inferior, signifying that, despite the values obtained in the present case, the use GPC FRA is viable. The workability of final mixes was influenced in a way acceptable for the case of recycled aggregate mortars. With adaptation of the FRA percentage and appropriate mix design this effect could be easily overcome. The 28day water absorption of PC mixes appeared to increase proportionally to the replacement level, while in GPC mixes the increase of FRA percentage did not result to significant fluctuations. Increase of mortars' water absorption is generally accepted to occur with FRA use. In both matrices though the increase for low replacement level was not significantly high. The compressive strength of both mortar times exhibited slight enhancement with 25% GPC FRA addition. For high replacement level the effect was less favorable but still resulted to values almost equal to those of the reference mixes. The same trend was observed for flexural strength.

Overall, the effect of GPC FRA on both PC and GPC mortars was similar, while when compared to PC FRA they did not appear to be inferior in any aspect. For low replacement levels, all the investigated properties presented acceptable deviations from the reference mix. Considering that in most real life cases the percentage of recycled aggregates in mixes does not exceed 30-35%, with adaptation of the mix design the above results indicate that their use will be feasible.

The present study was limited in the investigation of specific physical and mechanical properties on mortars alone. Therefore the obtained results should be expanded in order to take into consideration the intrinsic structure and chemistry of geopolymer cements. Chemical aspects and microstructural parameters should be included into the investigation, while the potential of recycling GPC mortars and concretes with various matrix types has to be evaluated.

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