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RECYCLING CERAMIC WASTE TO PRODUCE GREEN CONCRETE

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SUMMARY: Concrete is the most consumed construction material in the world. The cement production contributes around 5% of CO₂ emissions globally. Due to this substantial production, conventional resource supplies are in decline. This unsustainable supply has prompted a paradigm shift towards alternative raw materials to reduce the consumption of conventional concrete constituents and produce ‘green concrete’. One of the most effective ways is to replace cement with pozzolans derived from industry wastes, such as ceramic. Ceramics are the single largest contributor to construction and demolition waste globally, 54% in 2014 alone. There are approximately 68 million tonnes of ceramic waste sent to landfills annually in the UK. Current research into utilizing ceramics waste as a potential pozzolanic material in the production of ‘green concrete’ has had promising results. This study investigates the feasibility of using recycled ceramic waste in order to produce high performance concrete, while simultaneously reducing the consumption of cement. In this project, the mechanical performance of cement mortar samples, with a 15 % ceramic-cement replacement by weight was analysed and discussed. Further, the chemical properties of the concrete samples were determined using X-ray Powder Diffraction and the results were correlated with the mechanical strength tests. This study shows that ceramic waste of UK source can be a promising cement replacement to produce “green concrete”.

KEY WORDS: Cement, ceramic, hydration, XRD, compressive strength.

1 INTRODUCTION

A large amount of ceramic waste ends up in landfill causing severe worldwide environmental and economic burden. Around 100 million tons of construction and demolishing wastes are generated in the EU yearly, amongst which 54% are ceramic materials (i.e. tiles, walls, floors). In the UK, ceramic wastes consist of around 20% of construction materials and 12% of demolishing wastes for housing buildings [1, 2]. In developing countries like China, large amounts of ceramic powder waste are generated in cutting and polishing process for the furnishing industry. Around 10 million tonnes of ceramic waste powder are generated per year [3]. Due to dwindling natural resources, it is also a necessity to find a replacement for the cement content in concrete. The cement constituent within concrete is by far the most environmentally impactful material, as the process of mining raw materials (calcium carbonate, silica, alumina and iron ore), calcining to clinker form and then grinding to cement powder consumes considerable energy, requiring 850kcal/kg [4]. As well as being an energy intensive manufacturing process which emits high levels of CO₂, it also produces by-products in the form of mining waste. During the mining stage of cement production approximately 1.7 tonnes of material is extracted for every tonne of necessary minerals [5]. In order to address the high emissions and inefficient manufacturing process it is a necessity to reduce the consumption of cement within concrete production.
An innovative solution for the concrete mix is to replace a portion of cement with ceramic powder as a chemical reactive material. It has been stated in research that ceramics are in fact pozzolanic and provide the feedstocks required for secondary hydration to occur, forming a greater number of bonds in the ITZ [6]. Unlike most other pozzolanic materials being implemented today, ceramics are not a by-product of a manufacturing process (although a lot of ceramic waste is created during production and installation). As such, waste streams must be utilised in order for ceramics to be sensibly considered. Despite needing processing in order to attain adequate particle size, ceramics undergo calcination during manufacture and so already possess the required microstructure in order to be reactive. As such they are a potentially pozzolanic material in ample supply with no current reuse, and so should be investigated. This will provide comparable mechanical properties, hence suggesting that ceramics could be used as a suitable supplementary cementitious material (SCM) for Portland cement. The commonly used Portland cement concrete mainly consists of aggregates, cement and water. When the dry cement is mixed with water, calcium silicates (C₃S and C₂S) in cement react with water and produce calcium silicate hydrate (C-S-H) and hydrated lime (CH). This chemical reaction process is primary hydration. C-S-H is primarily responsible for providing concrete strength by binding with aggregates. More C-S-H can be generated in concrete if a portion of cement is replaced by constituents with active silicates (S) to facilitate secondary hydration with CH, as given below:

\[
\text{Hydration (mix cement with water): } \quad C_3S + 3H \rightarrow C-S-H + 2CH; \quad C_2S + 2H \rightarrow C-S-H + CH
\]

Secondary hydration (mix S containing constituents with CH from cement hydration): \[CH + S \rightarrow C-S-H\]

A typical example of secondary hydration is found though the use of metakaolin, which is the derivative from kaolin in heat treatment [7]. Hence, it is highly possible for ceramic powder to be chemically reactive in hydration processes. Heidari and Tavakoli [8] indicate that ceramic waste powders will participate in secondary hydration, and thus it should be added into concrete as cement substitute for enhancing performance. It is also suggested that 20% of cement can be replaced by ceramic powders.

In this study, we investigate the properties of a cement mortar mix design with a 15% ceramic-cement replacement by weight at different ages after casting, with the aim of correlating the mechanical strength with the mineralogy properties to evaluate the effectiveness of ceramic as a pozzolan for concrete.

2 EXPERIMENTAL WORK

2.1 Materials

Type 1 Portland cement was selected as the primary binding agent. This is the most commonly used cement in the UK, and as such has a substantial production (contributing large portions of CO₂ emissions). The ceramic source used in this study was a Johnson-Tiles glazed white ceramic wall tile, produced in Stoke, UK. This source was chosen as it is commonly used in the UK.

The ceramic tiles were initially broken using a hammer and then ground to a diameter of 1 – 2 mm using a jaw crusher. The ground pieces were then ground further to a fineness of less than 100 microns using an Orbital Mill. Details of the chemical composition and size distribution of raw materials are given in Table 1 and Figure 1.
Table 1: Chemical composition of the main components given by XRF (Top part) and crystalline composition by XRD-Rietveld (Bottom part) of the ceramic and the cement.

<table>
<thead>
<tr>
<th>Oxide Composition (%)</th>
<th>Cement</th>
<th>Ceramic</th>
</tr>
</thead>
<tbody>
<tr>
<td>SiO₂</td>
<td>18.87</td>
<td>68.92</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>4.28</td>
<td>19.78</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>2.89</td>
<td>0.89</td>
</tr>
<tr>
<td>MgO</td>
<td>2.12</td>
<td>0.31</td>
</tr>
<tr>
<td>CaO</td>
<td>64.78</td>
<td>7.03</td>
</tr>
<tr>
<td>Na₂O</td>
<td>0.08</td>
<td>0.33</td>
</tr>
<tr>
<td>K₂O</td>
<td>0.449</td>
<td>1.734</td>
</tr>
<tr>
<td>TiO₂</td>
<td>0.181</td>
<td>0.694</td>
</tr>
<tr>
<td>MnO</td>
<td>0.102</td>
<td>0.005</td>
</tr>
<tr>
<td>P₂O₅</td>
<td>0.112</td>
<td>0.173</td>
</tr>
<tr>
<td>LOI</td>
<td>5.93</td>
<td>0.15</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Main crystalline composition (Rietveld) (%)</th>
<th>Cement</th>
<th>Ceramic</th>
</tr>
</thead>
<tbody>
<tr>
<td>C₃S</td>
<td>85.7</td>
<td>3.1</td>
</tr>
<tr>
<td>C₂S</td>
<td>3.66</td>
<td>0.3</td>
</tr>
<tr>
<td>Quartz</td>
<td>0</td>
<td>47</td>
</tr>
</tbody>
</table>

2.2 Mixing, casting and curing

All constituents were carefully measured out (to the nearest gram) before a motorised hand mixer was used to homogeneously blend the mortar mixture in accordance with Eurocode 6: EN 1015-2 [9] before the mortar was poured into the moulds. The mortar was mixed using an Evolution Twister Mixer for a controlled mixing time of 2 minutes. The formulations investigated are given in Table 2. Mortars were cast at a cement/sand ratio of approximately 1:3 according EN 998-2 [10]. The water to binder ratio was empirically fixed at 0.581 to ensure constant and reasonable workability. The moulds selected were to Eurocode 6: EN 12390-1 [11] standard 40x40x160mm prims in 3-gang moulds. The specimens were then left to autoclave cure for 24 hours in the
moulds, before being demoulded. Once demoulded the specimens were placed in a heated, circulating curing tank. The first compressive strength tests were carried out when the samples were demoulded (at 24hrs), at which point all other replicates were moist cured until the required testing age (7, 29 and 56 days). The specimens were carefully removed from the curing tank and dried before testing began. The water is temperature controlled and maintained at a constant ambient room temperature, and the curing room is humidity controlled.

Table 2: Mix design

<table>
<thead>
<tr>
<th>Mix</th>
<th>Water [kg/m³]</th>
<th>Sand [kg/m³]</th>
<th>Cement [kg/m³]</th>
<th>Ceramic [kg/m³]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Control</td>
<td>292.969</td>
<td>1477.214</td>
<td>503.418</td>
<td>0</td>
</tr>
<tr>
<td>15% White</td>
<td>292.969</td>
<td>1477.214</td>
<td>428.059</td>
<td>75.521</td>
</tr>
</tbody>
</table>

2.3 Analysis and testing techniques

The compressive strengths of the specimens at different curing ages were determined using an Instron 4505 Universal Test Machine with a maximum capacity of 100 kN. The testing procedure and data interpretation follow the European Code 6: EN 1015-11 [12]. At least three samples were conducted for each mix. The specimen strength was calculated by using the measured loading plate dimension. Once the specimen had failed (when the maximum load was reached), the failure cracks were recorded photographically before the samples were broken by hand, such that the failure mode could be better observed (Fig. 2).

Figure 2: Failure mode of a 15% white ceramic at 29 days (after compressive testing)

The particle size analysis was performed by laser granulometry using a Beckmann Coulter LS100. The chemical and mineralogical composition of the samples were studied by XRF and XRD analysis respectively. XRF was performed on a Panalytical PW2404 wavelength-dispersive sequential X-ray spectrometer. For the XRD analysis, pastes with the same mix design as concretes were prepared without sand and cured under the same conditions. The samples were immersed in isopropanol to stop the hydration process after being crushed,
dried at 110°C for 2 h and stored in a desiccator. The samples were characterized by using Bruker D8 Advance with Sol-X Energy Dispersive detector with CuKα source (1.54Å) (Fig. 3). Samples were scanned on a rotating stage between 2 and 65 [°2θ] using a time per step of 5 s. Quantitative analysis was carried out by using TOPAS 3.0 Rietveld analysis software.

![Figure 3: Bruker D8 Advance and XRD tested sample.](image)

3 RESULTS

3.1 XRD analyses

![Image of XRD patterns](image)

Figure 4: XRD pattern of hydrated cement pastes Control (Right) and 15% White ceramic (Left) cured at 1 (d), 7 (c), 28 (b) and 56 (a) days. (1: C2S/C3S, 2: CH, 3: CSH, 4: Ettringite, 5: CaCO₃, 6: SiO₂).

Fig. 4 shows the XRD patterns of hydrated cement pastes control (without ceramic) and with 15%wt cured at 1, 7, 28 and 56 days. Using these patterns, it is possible to identify the main peaks characterising the hydration
process, such as SiO2, calcium silicates (C2S/C3S) and the hydrated products, CH, CSH, and Ettringite. The two samples present similar XRD patterns. The patterns show that C2S and C3S still exist up to 56 days. Their peak intensities decrease with the curing times (Fig. 5), as a result of their hydration. The decrease is similar for the control and ceramic specimens. We notice a stabilization of the consumption of C2S/C3S after 10 days of curing.

Figure 5: Variation of C2S/C3S with the curing time for the control specimen (black dots) and for 15%wt ceramic specimen (cross dots).

The presence of the hydration is also demonstrated by the formation of CH and CSH (Fig. 6). After 1 day of curing, we notice the presence of CH and CSH in the control and in the ceramic specimen.

Figure 6: Variation of CSH (A) and CH (B) with the curing time for the control specimen (black dots) and for the 15%wt ceramic specimen (cross dots).

Until 7 days, the two samples present the same evolution for CH and CSH. The content of CH and CSH is increasing with the curing time (Fig. 6), due to the primary hydration of silicate phases of cement with the liberation of CH and the formation of CSH. The hydration is very fast for both specimens. Lavat et al. [13] showed also that during the first 7 days, the samples were highly reactive. After 7 days, the behaviour differed noticeably between the two mixes. For the control specimen, the %CH content stabilizes whereas for the specimen with ceramics, it decreases with the curing time, indicating the presence of the secondary hydration. Further, with the ceramic, a pozzolanic reaction occurs between silicate and aluminates compounds and the CH produced by the first hydration of the cement. The variation of the CSH content indicates also the presence of the secondary hydration. Indeed, after 7 days, the sample with ceramic presents a higher content of CSH, which are produced during the secondary hydration. Others researchers reported also that the pozzolanic reaction takes place at longer curing times (over 7 days) [14-16].
3.2 Compressive strength

Compressive strength values of the control mortar and 15%wt ceramic mortar are given in Fig. 7. The behaviour of the two mixes is identical at 24 hours after casting. The compressive strength values are similar and are increasing with the curing time. The greatest growth in strength is experienced in the initial 7 days after casting, which is consistent with the XRD analysis. Further, a large increase in strength indicates occurrence of substantial hydration.

![Figure 7: Compressive strength of control mortar (black dots) and 15%wt ceramic mortar (cross dots) at 1, 7, 28 and 56 days.](image)

Between 28 and 56 days of curing, the compressive strength of the control sample stabilises whereas for the 15%wt ceramic replacement sample, it is still increasing. If this trend continues, it would exceed the strength of the control. Steiner [17] and Naceri [18] noticed the same trend, after 90 days of curing specimens with ceramic replacement levels of 10% exceeding the strength of ordinary Portland cement concrete. The increased rate of growth is an indication of secondary hydration. Actually, the secondary hydration process has been reported to be beneficial at long curing times [19].

4 CONCLUSIONS

The short term performance of 15%wt ceramic replacement cement was investigated, in terms of mechanical and physical-chemical properties. The presence of the first and secondary hydration has been proved using XRD analysis. The secondary hydration, corresponding to the pozzolanic reaction, occurs after 7 days. The compressive strength values of the cement with 15% ceramic replacement is similar to the control sample until 56 days. However, at longer curing times, the trend indicates that the sample with ceramic would exceed the strength of the ordinary Portland cement.

These results show the feasibility of using recycled ceramic waste as cement replacement. They demonstrate the potential of achieving structural grade concrete whilst simultaneously reducing the consumption of cement and providing a novel end-use of ceramic wastes.
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