Residual cementing property in recycled fines and coarse aggregates: Occurrence and quantification

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ABSTRACT

The adhered cement mortar in coarse aggregates and fines from demolished concrete has the potential to induce a residual cementing property upon recycling. A procedure for quantifying the contribution of recycled fines to the strength gain within the new mortar matrix is proposed. The strength gain is found to be more significant in recycled aggregates and fines from brick aggregate concrete than in those from stone aggregate concrete. Isothermal calorimetry measurements indicate the existence of large heat flows immediately after wetting and a prolonged initial reaction period during the hydration of recycled fines due to the presence of unreacted cement compound fractions and depleted amounts of gypsum. This phenomenon has been further confirmed, particularly in recycled fines from brick aggregate concrete, through electron microscopy observations of the formation of new gel structures due to rehydration. The chemical compositions determined using two independent methods indicated possible interactions between CH (the hydration product) and pozzolan (from brick) to induce strength gain in new mortars and new concrete obtained by recycling brick aggregate concrete. This explanation is consistent with the strength test results and the evolution of the heat events observed calorimetrically.

Authors’ keywords: Recycled brick aggregate concrete, Recycled stone aggregate concrete, Adhered mortar, Hydration products, Compressive strength, Testing method.

INTRODUCTION

Recycling stone and brick aggregate concretes by crushing and converting them into coarse aggregates (Figures 1 and 2) and fines (Figure 3) for use in new concreting is an established trend. Buck (1977), Frondistou-Yannas (1977), Hansen and Narud (1983), Hansen and

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Angelo (1986), Forster (1986), Topçu and Günçan (1995), Rashwan and AbouRizk (1997), Poon et al. (2004), Tam et al. (2005), Tam et al. (2007a,b), Casuccio et al. (2008) and Huda and Alam (2014) investigated the possibilities of recycling stone aggregate concrete. Historic use of clay brick aggregate concrete is common in Bangladesh and some parts of India (Akhtaruzzaman and Hasnat 1983). Mohammed et al. (2014) investigated the possibilities of using recycled products from brick aggregate concrete. The possibility of recycling construction wastes from ready-mix plants, precast concrete product plants, construction sites and brick fields into another generation of concrete is also being explored (Poon 1997, Poon and Chan 2007, Kou et al. 2012). In the recycling of demolished concrete, reliable achievement of the expected strength after recycling is a major concern. To address this concern, characterizing the mortar of old concrete (Figures 1-2) attached to the parent aggregates is the focus of the current study. The presence of adhered mortar significantly changes the Los Angeles abrasion (LAA) value, absorption capacity and unit weight of recycled aggregates compared with the parent aggregates (Islam et al. 2015). To determine the strength performance after recycling, two distinct points demand consideration: (i) whether the adhered mortar causes a weak plane to have an effect on aggregate strength and (ii) whether the recycled products, which contain adhered mortar, are inert or possess any cementitious properties to impart any benefit in strength properties after recycling.

![Fig. 1. Recycled aggregates of different sizes from stone aggregate concrete (RS). (a) 12.7 mm; (b) 19.0 mm; (c) 25.0 mm. Image by AFMS Amin.](image1)

![Fig. 2. Recycled aggregates of different sizes from brick aggregate concrete (RB). (a) 12.7 mm; (b) 19.0 mm; (c) 25.0 mm. Image by AFMS Amin.](image2)
Fig. 3. Micrographs of recycled fines in a loose state recorded using a scanning electron microscope. (a) Recycled fines from stone aggregate concrete (RFS) at 800x magnification; (b) Recycled fines from brick aggregate concrete (RFB) at 1000x magnification. Images by AFMS Amin.

In this context, two approaches are reported in the literature. In the first approach, Buck (1977), Poon et al. (2004), Tam et al. (2005), Tam et al. (2007a,b), and Casuccio et al. (2008) proposed different methods for removing weak fractions of old adhered mortar from their recycled materials for improving the surface morphology of recycled coarse aggregate with the aim of improving the strength performance. In the second approach, improving the viability of achieving desired strength properties upon recycling was attempted by relying on the force exerted on the old concrete by the crushing method (Florea and Brouwers 2013). The application of a larger force can better separate the weaker parts of the adhered mortars from the old concrete and produce stronger recycled aggregates and fines. The volume of finer fractions produced after crushing will be proportional to the applied pressure but inversely proportional to the strength of the parent concrete (Rashwan and AbouRizk 1997). The remaining fraction (larger aggregates) will be converted into recycled coarse aggregate. Recycled materials that are collected from a commercially operated construction and demolition waste recycling facility should also exhibit better surface uniformity of recycled products. Studies of Huda and Alam (2014) for recycled stone aggregate concrete and Mohammed et al. (2014) for recycled brick aggregate concrete reported promising outcomes by directly using the recycled aggregates without any further surface modifications. This notable difference in performance between the two approaches requires additional evaluation to establish whether the employed crushing process is capable of fracturing the partially hydrated cement grains and opening the unhydrated surfaces that are available for rehydration after recycling. To this end, a careful review of the literature convincingly highlights the existence of residual cementing properties in fines (generally with particle sizes of <2.4 mm) from recycled stone aggregate, particularly when used as soil stabilizers (Hansen and Angelo 1986, Arm 2001, Katz 2003, Poon et al. 2006, Vegas et al. 2011). The recycled fines were collected from a jaw crushe or a mechanical crushing plant.
Zakaria and Cabrera (1996) further reported that concrete with coarse aggregates from recycled brick aggregate concrete presents relatively higher strengths at later ages compared with 28-day strength due to the pozzolanic effect of finely ground burnt brick. Concrete from natural gravel aggregates does not exhibit this effect. Khatib (2005) noted the presence of residual cementing fractions in recycled fines from burnt clay bricks and stone aggregate concretes. Silva et al. (2008) attempted to reduce the cement content in mortars by exploiting the potential pozzolanic effect in recycled and very fine red clay ceramic wastes. Florea et al. (2014) reported the possibility of using recycled concrete fines obtained from crushed concrete to replace part of the cement in new mortar formulations.

However, there has been no attempt to explain the fundamental reasons for these significant observations, which were recorded over the past three decades by numerous independent research groups. A method for quantifying the additional strength gain in the recycled fines-sand-cement mortar matrix due to the residual cementing property may enable a designer to judiciously incorporate this property into the mix design as an added advantage.

FRACTURE PROCESS, PRESENCE OF UNHYDRATED CEMENTITIOUS PARTS AND REHYDRATION PHENOMENA

Tricalcium silicate ($C_3S$), dicalcium silicate ($C_2S$), tricalcium aluminate ($C_3A$), and tetracalcium aluminoferrite ($C_4AF$), which are the four primary constituents of hydraulic cement, remains substantially unhydrated in concrete for decades. Hydration that only occurs on the surface of a cement grain gradually proceeds inside the particle (Neville 1995, page 14) and subsequently liberates heat produced from exothermic reactions over time (Mehta and Monteiro 2006, Table 6.4). The reaction rate for $C_2S$ is the slowest, whereas that for $C_3S$ is moderate. The presence of an optimum gypsum content slows the reaction rates (Lerch 1946) for the faster-reacting compounds, including $C_3A$, $C_4AF$ and $C_3S$ (refer to Mehta and Monteiro 2006, Sections 6.3.3, 6.3.4), to provide the cement with a controlled setting time and to maintain the early strength-gain properties. Figure 4(a) schematically illustrates such a concrete matrix in which hydrated cement grains bind coarse and fine aggregates. However, the crushing of old concrete by fracturing the mortar (the hydration product) and the exposure of unreacted cement particle surfaces during the crushing of old concrete in a crusher can expose the unreacted surfaces (Figure 4b, see also, Wittmann 2002; Hu and Wittmann 1992; Florea and Brouwers 2012). The gypsum content in recycled coarse aggregates and fines with unreacted cementitious surfaces should remain at a depleted level because the amount of gypsum that is available in the cement of the parent concrete should have already been exhausted during the previous hydration process (Lerch 1946, Jansen et al. 2012; Dittrich et al. 2014). In this situation, the existence of a fast-
reacting compound, for example, C₃A in the presence of gypsum in a lesser amount than the optimum amount, should generate larger heat flows compared with a Portland cement for the ‘initial reaction period’ (Bullard et al. 2011; Kumar et al. 2012) of the first 30 minutes of wetting the recycled fines. No calorimetric study has described the fundamental hydration phenomenon in recycled fines for the ‘initial reaction period’, which would confirm the trace of unreacted cementitious compounds and provide direct physical evidence.

Fig. 4. Schematic representation of the fracture of concrete in a crusher and production of the recycled aggregate under a certain pressure. (a) Fracture line within the concrete matrix; (b) Fracture line (crack path) passing through a partially hydrated cement grain.

Furthermore, the evolution of the residual cementing effect in recycled aggregates and fines is expected to be related to very complex physicochemical reactions between the active mineralogical/chemical constituents and new cement (with additional gypsum) in the presence of water. The active constituents include pozzolans from old mortar, old brick aggregate concrete and Ca(OH)₂, i.e., CH, which is a hydration product present in old concrete. The possibility of a binding effect occurring analogous to the well-known lime- pozzolan action exhibited by ancient roman cement composed of clay brick dust and lime mortar (Baronio et al. 1997, Ubbriaco and Tasselli 1998, Franzini et al. 1999, Giergiczny 2004, Bakolas et al. 2008) cannot be excluded for recycled aggregates and fines from brick aggregate concrete. Finely ground brick dusts, composed mainly of silica and alumina found after burning clay minerals show pozzolanicity (Baronio and Binda 1997). Poon et al. (2004) reported that recycled concrete produced from high-performance concrete using a low w/c ratio and pozzolanic additives has a dense microstructure at the interfacial transition zone, which exhibited better strength performance after recycling. A later study by Poon et al. (2006) supported the roles of calcium silicate hydrate, i.e., C-S-H (product of the slow reaction between pozzolan and Ca(OH)₂), and C₂S in smaller sized grains in reducing the permeability and increasing the compressive strengths of recycled fines after
hydration. Tam et al. (2009) investigated physicochemical reactions in the formation of C-S-H through differential scanning calorimetry (DSC) conducted at high temperatures. The rehydration of recycled mortar after thermal treatment was studied by Shui et al. (2008). All of these observations suggest that the microstructure, composition of micrograins and interactions between active components are the origin of the residual cementing property in such aggregates and fines upon rehydration.

However, the results of a comparative study to differentiate the reactivity between RFB and RFS in terms of heat production per unit mass in the initial reaction period, decay features of heat generation in the depleted gypsum situation and the general chemical composition of the system may provide considerable evidence regarding the presence of reactive components and their possible role in strength gain over time. Synthesizing the results obtained from different independent observations is crucial for confirming the existence of the phenomenon that may provide added strength after recycling. Because no standard method for quantifying the contribution of the residual cementing property to strength gain is currently available, this beneficial property remains unaccounted for in any design.

OBJECTIVES AND METHODOLOGY

This study employs isothermal DSC and scanning electron microscopy (SEM) to physically examine and compare the residual cementing property in recycled fines from stone and brick aggregate concrete. The active chemical constituents in recycled fines were quantified using two standard methods to investigate plausible explanations for the occurrence of this significant property. To quantify the effect of the residual cementing property on the strength gain, compressive strengths attained at different ages with fresh recycled coarse aggregates are compared with those obtained from pre-hydrated recycled aggregates. A standard test method (ASTM C109/C109M-13) was modified and applied to quantify the additional strength gain in the recycled fines-sand-cement mortar matrix resulting from the residual cementing property.

MATERIALS

Akhtaruzzaman and Hasnat (1983), Mansur et al. (1999), Debieb and Kenai (2008), and Cachim (2009) summarized the engineering properties and extensive historical use of brick aggregate as an alternative to stone aggregate in concrete production, particularly in Bangladesh and in a part of India where natural aggregates are scarce. This study investigates the respective properties of recycled aggregates and fines from both stone and brick aggregate concretes.
Parent Materials
Concrete cylinders (diameter of 150 mm and height of 300 mm) produced from stone aggregate concrete and brick aggregate concrete, which were aged from 28 to 90 days, collected from different construction sites in Bangladesh and crushed in a testing machine to determine the compressive strength, were used as the parent concrete stocks to produce recycled aggregates and fines. The use of these concrete cylinder samples provides a representation of the construction materials utilized in Bangladesh. It also offers a unique opportunity to record the strength pattern of the parent concretes that were employed to produce the recycled aggregates (refer to Huda and Alam 2014). An independent statistical analysis conducted by Hoque (2009) reveals that the compressive strengths of the parent cylinders for brick aggregate concrete ranged from 10 to 45 MPa with an average strength of 25 MPa. The compressive strengths of 7% of the brick aggregate concrete cylinders were less than 12 MPa, and the compressive strengths of 5% of the cylinders were greater than 40 MPa. The compressive strengths of the parent cylinders produced from stone aggregate concrete ranged from 7 to 50 MPa, with an average strength of 25 MPa. The compressive strengths of 7% of the cylinders were less than 10 MPa, and the compressive strengths of 9% of the cylinders were greater than 40 MPa.

A jaw crusher with a 25-mm opening in the closed position was used to crush demolished concrete cylinders of stone aggregate and brick aggregate concretes to produce RS and RB, respectively. The crusher fines from the recycled stone aggregate (RFS) and recycled brick aggregate (RFB) were collected. Well-burnt clay bricks with a compressive strength of at least 25 MPa were crushed separately in the same jaw crusher. Brick dust (crusher fine) was collected from the crusher for use as a reference material. Table 1 lists the materials and their respective acronyms used throughout this paper. The material properties of RS and RB are summarized in Table 2 (see also Islam et al. 2015). The gradation curves are presented in Fig. 5 for RS, RB, RFS and RFB. A comparison of the material properties for different aggregate types indicates that RB typically has a lower density but higher absorption capacity than RS. RFS and RFB grains finer than <150 μm were selected to search for the origin of the residual cementing properties (Poon et al. 2006) due to small particle sizes and larger surface areas.

Choice of Control Materials
To explain the residual cementing effect through comprehensive measurements, aged recycled aggregates (pre-hydrated/aged for 90 days to complete hydration of the partially hydrated cementitious fractions) were adopted as control materials for the recycled aggregate concretes. Fine sand (FS, grain size of less than 150 μm) from riverbeds, brick dust (grain size of less than 75 μm) and cement were used as control materials (Controls 1-3, Table 1) to explain the effect in recycled fines. FS should have no cementitious fractions, whereas the Portland cement has a known strength gain property. Brick dust (Control 2)
only contains pozzolans (Zakaria and Cabrera 1996). In comparison, RFB contains CH, which is a hydration product with the potential to interact with pozzolans present in the brick of the parent concrete to produce C-S-H. Thus, Control 2 can provide a contrast with RFB to exemplify the role of CH in interacting with pozzolan.

Table 1. Materials and acronyms used in the study.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Acronyms</th>
<th>General terminology</th>
</tr>
</thead>
<tbody>
<tr>
<td>Recycled coarse aggregate from stone aggregate concrete</td>
<td>RS</td>
<td>Recycled coarse aggregates</td>
</tr>
<tr>
<td>Recycled coarse aggregate from brick aggregate concrete</td>
<td>RB</td>
<td></td>
</tr>
<tr>
<td>RB and RS after 90 days immersion in water</td>
<td></td>
<td>Control</td>
</tr>
<tr>
<td>Recycled fines (&lt; 75 μm in size) from stone aggregate concrete</td>
<td>RFS</td>
<td>Finer fraction of recycled aggregates</td>
</tr>
<tr>
<td>Recycled fines (&lt; 75 μm in size) from brick aggregate concrete</td>
<td>RFB</td>
<td></td>
</tr>
<tr>
<td>Cement of 197-1:2003, CEM II/A-M (S-V-L), 42.5 N class</td>
<td>Cement</td>
<td>Control 1</td>
</tr>
<tr>
<td>Brick dust (&lt; 75 μm in size) obtained from crushing of well burnt clay bricks</td>
<td>Brick dust</td>
<td>Control 2</td>
</tr>
<tr>
<td>Fine sand (&lt; 150 μm) from river bed</td>
<td>FS</td>
<td>Control 3</td>
</tr>
</tbody>
</table>

Table 2. Properties of recycled aggregates.

<table>
<thead>
<tr>
<th>Material properties</th>
<th>RS</th>
<th>RB</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bulk specific gravity (OD)</td>
<td>2.49</td>
<td>1.88</td>
</tr>
<tr>
<td>Bulk specific gravity (SSD)</td>
<td>2.57</td>
<td>2.10</td>
</tr>
<tr>
<td>Absorption capacity (%)</td>
<td>3.37</td>
<td>11.47</td>
</tr>
<tr>
<td>Unit weight (SSD) kg/m³</td>
<td>1311</td>
<td>1272</td>
</tr>
<tr>
<td>Unit weight (OD) kg/m³</td>
<td>1287</td>
<td>1138</td>
</tr>
<tr>
<td>Los Angeles Abrasion Value (%)</td>
<td>38</td>
<td>39</td>
</tr>
</tbody>
</table>

Fig. 5. Gradation of crusher fines and recycled aggregates as produced by a jaw crusher. The coarse aggregate (CA) and sand as the fine aggregate (FA) used in producing the concrete after adjustment in the gradation are shown with solid lines.
**Determination of Chemical Compositions**

A comparison of the heat that is produced immediately after the wetting of different fines (RFS and RFB) in water and measured via calorimetry with the corresponding values for cement (Control 1) and FS (Control 3) can provide some physical evidence regarding the existence of the residual cementing property in recycled fines. To determine the fundamental origin of this property, the chemical compositions of RFS and RFB should be examined. The compositions were determined using two independent methods. The chemical compositions of the samples were determined after digesting in aqua regia, and the extracts were analyzed by atomic absorption spectrophotometry (Shimadzu AAS6800). The ASTM C114-07 method was employed as the second method to analyze the chemical compositions of the four samples. The results are presented in Tables 3 and 4. The compositions of the three reference materials, i.e., cement, brick dust and FS, were measured for comparison with RFS and RFB.

**METHODS**

**Calorimetric Studies**

Heats of hydration can be measured using an isothermal calorimeter, where measurements are performed for at least 24 to 48 hours. In this approach, the total heat quantity is measured, whereas individual exothermic reaction events, which frequently occur immediately after the addition of water, cannot be distinctly isolated. Figures 4-6 in Lerch (1946) indicate the use of a special method different than isothermal calorimeter to measure the heat liberated in the first 30 minutes (see also Adams 1976). To trace the events, the heat that was generated immediately after the wetting of RFS, RFB, cement (Control 1), brick dust (Control 2) and FS (Control 3) was measured in this study using computer-controlled DSC (TA Instruments Model Q10, 159 Lukens DriveNew Castle, DE, USA) in a dry nitrogen environment at three isotherms: 303 K (30ºC), 318 K (45ºC) and 333 K (60ºC). The magnitudes of these three isotherms are selected as a natural choice because the majority of the previous fundamental studies concerning the evolution of heat due to hydration were conducted between 293 K and 353 K (Figure 6-12 in Mehta and Monteiro 2006; Table 1.7 in Neville 1995). Distilled water was added to the pre-weighed samples, which were placed in an encapsulated aluminum pan to hydrate the samples within the calorimeter. The computer was set to record the time history of the heat production in the calorimeter for 60 minutes. The time history of the heat generated in each test was recorded beginning immediately after closing the DSC pan following the addition of water. By increasing the isotherms, the reaction can be accelerated, i.e., to advance the reaction events over time. Increasing the isotherm can bring the samples to a higher energy level and cause earlier and faster hydration. Greater amounts of heats are produced due to the exothermic nature of these reactions. A specimen that instantly produces more heat at a lower isotherm after wetting should ideally possess a component with low activation energy. Measuring the heat flow time history of the material at a higher isotherm offers the opportunity to identify
components with higher activation energies to complete the reactions at an earlier time. These components shall have a delay to complete the reaction at room temperature.

Microscopy Studies
Scanning electron microscopy (SEM) images of RFS and RFB were recorded on a Phillips XL 30 SEM at 30 kV. Gold-coated specimens were attached to the mounting devices with double-sided adhesive tape.

Casting of Concrete and Mortars
The present study used the same mix design ratio of 1:1.4:2.5 (weight basis) for four mixes of recycled concrete using (i) freshly broken RB, (ii) freshly broken RS, (iii) RB after 90 days of immersion in water and (iv) RS after 90 days of immersion in water for casting concrete. River bed sand with a bulk specific gravity (saturated surface dry) of 2.58, a bulk specific gravity (oven dry) of 2.54, a bulk unit weight (saturated surface dry) of 1520 kg/m³ and a fineness modulus of 2.62 was used. The water/cement ratio was maintained at 0.5. After the green concrete was placed in the cylindrical molds, compaction was achieved using a vibrator. The cast cylinders were removed from their steel molds 24 h after casting and cured in lime water for 28 days. The compressive strength of the cast concrete was determined as per ASTM C39 (2014).

Contribution of recycled fines (RFS and RFB) in strength gain of sand-cement mortar has been assessed by modifying a standard test method (ASTM C109/C109M–13). The modified method proposed for the strength assessment is presented in later part of this paper.

PHYSICO-CHEMICAL EVIDENCE FOR THE OCCURRENCE
Observations from Chemical Compositions
Tables 3 and 4 present the amounts of Ca and Al detected in the RFB and RFS, which are distinctly greater than those in the FS for all the measurements. Furthermore, Table 4 shows that the amount of gypsum as indicated by the SO₃ content is considerably lower in RFB and RFS than that present in the cement (Control 1). The presence of low SO₃ content in RFB and RFS can be considered as the gypsum content that remains as residue after the end of the hydration process in the old concrete. SiO₂ contents in RFS and RFB are distinctly higher than Cement (Control 1) but lower than that present in FS. However, from this comparison the reactive silica content (Lothenbach et al. 2011) taking part in hydration cannot be ascertained. A calorimetric study of the hydration process of dry RFB and RFS immediately after wetting with water (occurring before the end of the first 60 minutes), also referred to as the initial reaction phase (Bullard et al. 2011), should be able to reveal the interaction of high Ca ,Si and Al contents in the presence of a gypsum content that is less than the optimum amount present in a fresh cement (Mehta and Monteiro 2006).
In the event of a more favorable chemical environment for interaction, the hydration products in old concrete that contains CH and Pozzolans in brick that are rich in alumina and silica have the potential to initiate the progressive formation of C-S-H products or to initiate a property that is analogous to a lime-Pozzolan action (Fu and Beaudoin 1996; Baronio and Binda 1997; Baronio et al. 1997; see also Section 9.3.6 in Taylor 1997).

Table 3. Chemical constituents of cement and fines. Total content (gm/kg) extracted with aqua regia.

<table>
<thead>
<tr>
<th>Chemical constituents</th>
<th>Cement (Control 1)</th>
<th>Brick dust (Control 2)</th>
<th>FS (Control 3)</th>
<th>RFS</th>
<th>RFB</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ca</td>
<td>179.5</td>
<td>1.4</td>
<td>0.3</td>
<td>3.5</td>
<td>46.6</td>
</tr>
<tr>
<td>Al</td>
<td>17.5</td>
<td>8.1</td>
<td>9.6</td>
<td>16.7</td>
<td>15.1</td>
</tr>
<tr>
<td>Fe</td>
<td>14.8</td>
<td>2.9</td>
<td>12.0</td>
<td>11.5</td>
<td>9.0</td>
</tr>
<tr>
<td>Mn</td>
<td>0.5</td>
<td>0.1</td>
<td>0.2</td>
<td>0.3</td>
<td>0.2</td>
</tr>
<tr>
<td>Na</td>
<td>0.4</td>
<td>0.5</td>
<td>0.2</td>
<td>0.5</td>
<td>0.9</td>
</tr>
<tr>
<td>K</td>
<td>2.3</td>
<td>0.2</td>
<td>0.8</td>
<td>1.6</td>
<td>2.0</td>
</tr>
</tbody>
</table>

Table 4. Chemical constituents of cement and fines (%) determined as per ASTM C114-07.

<table>
<thead>
<tr>
<th>Chemical constituents</th>
<th>Cement (Control 1)</th>
<th>FS (Control 3)</th>
<th>RFS</th>
<th>RFB</th>
</tr>
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<tbody>
<tr>
<td>CaO</td>
<td>50.3</td>
<td>4.0</td>
<td>18.8</td>
<td>12.2</td>
</tr>
<tr>
<td>Al₂O₃</td>
<td>10.0</td>
<td>16.0</td>
<td>10.2</td>
<td>16.0</td>
</tr>
<tr>
<td>Fe₂O₃</td>
<td>2.7</td>
<td>1.4</td>
<td>2.6</td>
<td>1.4</td>
</tr>
<tr>
<td>SiO₂</td>
<td>27.4</td>
<td>70.7</td>
<td>46.9</td>
<td>50.2</td>
</tr>
<tr>
<td>MgO</td>
<td>2.8</td>
<td>1.9</td>
<td>2.0</td>
<td>2.0</td>
</tr>
<tr>
<td>SO₃</td>
<td>2.0</td>
<td>0.13</td>
<td>0.75</td>
<td>0.37</td>
</tr>
<tr>
<td>Na₂O</td>
<td>0.11</td>
<td>0.06</td>
<td>0.10</td>
<td>0.19</td>
</tr>
<tr>
<td>K₂O</td>
<td>0.27</td>
<td>0.45</td>
<td>0.36</td>
<td>0.12</td>
</tr>
<tr>
<td>IR</td>
<td>16.6</td>
<td>61.2</td>
<td>92.3</td>
<td>48.6</td>
</tr>
<tr>
<td>L.O.I.</td>
<td>8.1</td>
<td>36.7</td>
<td>15.9</td>
<td>12.7</td>
</tr>
</tbody>
</table>

Calorimetric Measurements
The immediate heat flow (per unit mass) due to the exothermic processes initiated in the RFB, RFS, cement (Control 1), brick dust (Control 2) and FS (Control 3) after adding water at the three isotherms over the first 60 minutes is illustrated in Fig. 6. The measurements generally show the exothermic events that initiate after the addition of water to gradually decay within the first 60 min (Lerch 1946). This time span adequately covers the initial period (Period I, Jansen et al. 2012) of hydration but does not extend beyond the slow reaction/induction period (Period II) because no acceleration in heat flow is observed. The acceleration period (Period III) followed by the retardation period (Period IV, containing sulfate depletion peak) mark the other two major periods of cement hydration.
Figure 6(a) shows the typical heat flow vs. time plots for cement (Control 1) recorded for 60 minutes immediately after wetting with water at three isotherms (303 K, 318 K and 333 K). The initial heat flow is 0.27 W/g at 303 K and gradually increases to 2.6 W/g at 333 K. This result is attributed to the immediate dissolution of C₃A and sulfates (Kumar et al. 2012, Jansen et al. 2012, Dove et al. 2005). At higher temperature, the dissolubility and the reaction rate are both increased. A rapid reduction in heat production is isochronously observed after 6 min, particularly at 318 K and 333 K, which can be attributed to the activation of gypsum, which is responsible for the onset of ettringite formation. At 333 K, a temporal step-down plateau in heat production is observed between 8-16 min followed by a rapid decrease (see also Dittrich et al. 2014). This result may indicate the possible progress of ettringite production by C₃A after the onset.

A careful examination of the heat flow vs. time plots for RFB (Figure 6b) and RFS (Figure 6c) compared with the plots for cement [Control 1, Figure 6(a)] shows that the heat flow immediately after wetting at all isotherms is generally higher than the heat flow recorded for cement at the corresponding isotherm. Conversely, heat flow due to the immediate dissolution of C₃A and sulfates ceases gradually in RFB and RFS compared with cement (Figure 6a). The presence of a limited amount of gypsum (refer to Table 3) after the passage of the sulfate depletion peak (Period IV) in the previous hydration of the parent concrete (Jansen et al. 2012) can be attributed to this observation. Therefore, the higher amounts of heat flow in RFS and RFB and the delayed cessation of heat flow in the initial reaction period of hydration indicate the presence of chemically active aluminous compounds that originate from the crushing process of old concrete (Figure 4).

The formation of the temporal step-down plateau in the heat flow observed in RFB between 10-25 min at 333 K and 8-33 min at 318 K contrasts with RFS; this event was not distinctly observed in this study. To explain this difference, the corresponding heat flows from brick dust (Control 2), which is also rich in aluminous substances (Table 3) for the same three isotherms, are presented in Figure 6(d). The lower amount of heat of dissolution compared with RFB and the absence of any step-down plateau may suggest that brick is a secondary source of aluminous substances for initiating the largest heat flow (among all tested materials) at the onset of the hydration process in RFB in the presence of hydration products of old concrete.

The smallest amount of heat flow was recorded in the FS (Fig. 6e) at 303 K and 333 K. This finding reveals an absence of any significant components that can react with water and result in the cementing property exhibited by an exothermic event within the observed duration. Tables 3 and 4 show the absence of any Ca, which is the primary element offering a cementing effect for the range of materials examined in this paper. The difference in heat growth between Fig. 6a and Fig. 6e can therefore be attributed to the possible contribution
of partially hydrated cement particles and hydration products present in RFS and RFB at the initiation period (Period I). The weak existence of a step-down plateau at 333 K for FS cannot be explained in this study.

![Graphs showing heat flow vs time for different materials](image)

**Fig. 6.** Production of heat immediately after the addition of water in different fines. (a) Cement (Control 1), (b) RFB, (c) RFS, (d) Brick dust (Control 2), and (e) FS (Control 3).

**SEM Observations**
SEM observations conducted by the authors (Figure 7 in comparison to Figure 3) on recycled fines from stone and brick aggregate concrete in the presence of additional water clearly demonstrate the formation of a new gel structure on the surface of the fines. This new formation is markedly denser for recycled fines from brick aggregate concrete than for those from stone aggregate concrete.
Fig. 7. Reformation of hydration products in the space between the fine particles in a recycled fine matrix. (a) Recycled fines from stone aggregate concrete (RFS) at 1000x magnification; (b) Recycled fines from brick aggregate concrete (RFB) at 800x magnification; (c) RFB at 2000x magnification taken from (b) Section-X. Images by AFMS Amin.

QUANTIFICATION OF THE STRENGTH GAIN PHENOMENON

The physico-chemical evidence presented in the preceding section, particularly the DSC results, provided considerable evidence of the role of unreacted aluminous substances in the depleted gypsum environment in initiating the hydration process. The possible role of $C_A$ originating mostly from the crushing process (Figure 4) was discussed. This discussion, however, implies the co-existence of three other unreacted fractions of cementing compounds, e.g., $C_S$, $C_S$, and $C_AF$, and pozzolan to offer a residual cementing effect in recycled coarse aggregates and fines in the subsequent periods of rehydration (Periods III and IV, Jansen et al. 2012) after recycling. The existence of the residual cementing property in recycled aggregates should evidently manifest as an increase in the mechanical strength.
of the recycled aggregate concrete. The strength increase phenomenon for recycled fines should also ideally be observed in a recycled fines-sand-cement mortar system. This section assesses the compressive strength increase in recycled aggregate concrete and a recycled fines-sand-cement mortar system.

**Contribution of the Residual Cementing Property in Recycled Aggregates**

The compressive strengths attained by the two sets of recycled aggregate concrete are compared in Fig. 8. In the first group, RB and RS were cast in new concrete immediately after crushing (thus, the term ‘fresh’), whereas the RB and RS obtained after crushing in the second group were aged for 90 days in water to ensure that any partially hydrated cementitious fractions are exhausted by rehydration to enable an effective comparison, according to the premise illustrated in Figure 4.

The comparison between the compressive strength vs. age curves shows higher strengths in recycled aggregate concrete produced from fresh recycled aggregate. The difference is more prominent in recycled aggregate concrete produced using fresh RB, particularly at an early age (before 7 days). This result may be consistent with the DSC observations obtained from the RFB (Fig. 6b) if the contribution of fast-reacting cementitious components on strength gain is considered. The recycled aggregate concretes that were produced with aged (hydrated) recycled aggregates show a plateau in strength gain with time after 28 days, whereas the recycled aggregate concretes that were made with fresh RB and RS show an increasing trend, even beyond 28 days, due to the residual cementing property that possibly originated from pozzolans (Khatib 2005) and slow-reacting compounds of cement, for example, C₂S (Mehta and Monteiro 2006, Table 6.4). The results conform to the initial observations reported by Rashwan and AbouRizk (1997).

![Fig. 8. Strength vs. age for RB and RS for four different mixes using RB and RS at fresh and aged conditions.](image-url)
Contribution of Residual Cementing Property in Recycled Fines

A standard ASTM test method ASTM C109/C109M is in use to determine the compressive strengths of hydraulic cement mortars using 50 mm × 50 mm × 50 mm cubic specimens. In this method, ASTM-graded Ottawa sand (standard sand) in a specific proportion (Ottawa sand:cement = x:y, Fig. 9a) is mixed with cement (to be tested) to create a uniform sand-cement mortar mix. The mortar is compacted in molds following a standard procedure. Cubes cured in a controlled environment are tested at specific ages for strength assessment.

To assess the residual cementing property of RFB and RFS, the ASTM C109/C109M method is used in this work in a modified form. For this purpose, a fraction (r) of cement (y) is replaced by RFB, RFS (Fig. 9b) and FS (Fig. 9c) individually. The cubes were prepared following the ASTM-specified compaction method and cured in water at the prescribed temperature. The compressive strengths attained by the cubes in accordance with the mix proportions shown in Figures 9a, 9b and 9c at different ages of curing are compared in Figure 10. The DSC measurements, which determined the heat generated in an exothermic process initiated by adding water, demonstrated that FS (Fig. 6e) exhibits no substantial cementing property at room temperature, whereas RFB/RFS possesses the property to a considerable extent. The grain sizes of RFS and RFB are larger than the grain size of FS (Table 1). Therefore, the difference in strengths of the specimens in Fig. 9b (with recycled fines, RFS or RFB) and Fig. 9c (with FS) should logically quantify the contribution of recycled fines to the compressive strength gain due to the inherent cementing effect. The experimental trials with different r values should provide a trend in response to the addition of RFS/RFB contents. The novelty and technical simplicity of the approach derived by modifying the ASTM standard test method are valuable for assessing the strength contribution of recycled fines (RFS/RFB) in new mortar matrices. The modified procedure was implemented by casting two independent trial batches (Batch I and Batch II) with a total of 10 castings on the same material set. In Batch I, r had values of 0.20, 0.25, 0.40, 0.50, 0.60 and 0.75. In Batch II, r had values of 0.10, 0.25, 0.50 and 0.75.

![Fig. 9. Scheme for modifying ASTM specification; (b) matrix in which a fraction of cement is replaced with RFB or RFS; (c) matrix in which a fraction of cement is replaced with FS.](image-url)
Figures 10(a) and 10(b) present the compressive strength vs. age curves for the two batches for higher replacement levels ($r$ between 0.50 and 0.75), which are plotted at a higher magnification of strength values for clarity. The corresponding results for lower replacement levels ($r$ between 0.10 and 0.40) are plotted in Figures 10(c) and 10(d). The reference curve ($r = 0.00$) for the Ottawa sand-cement matrix per ASTM C109/C109M is also plotted to show the upper reference boundary for the mix proportion depicted in Figure 9(a). The trends of all the strength vs. age curves generally show a progressive increase in strength over time in all specimens. A comparison of the curves shows a characteristic reduction in strength after the replacement of cement by RFS, RFB or FS because none of these replacement materials have a cementing property that is equivalent to the cementing property of a Portland cement (Control 1). Increases in the replacement level $r$, however, appeared to consistently correspond to a reduction in strength compared with the Ottawa sand-cement mix. A more detailed comparison reveals that the strengths attained in the specimens with RFS (Figs. 10a-d) and RFB (Fig. 10b,d) as replacements are markedly higher than those of the specimens replaced with the same proportion of FS at a particular age. In all cases, however, RFB produced higher strengths than RFS at all replacement ratios. This observation is consistent with the physicochemical evidence discussed in the preceding section.

The net percentage increases in strength, i.e., $(\alpha - \beta)/\beta$, due to the residual cementing effect with the progressive replacement of cement with RFS and RFB are plotted in Fig. 11 for different mortar ages. Here, $\alpha$ is the measured compressive strength at a particular age for $r$ fraction of cement replaced with RFS or RFB (Table 1), and $\beta$ is the measured compressive strength at the same age [defined in Figures 10(a) and 10(b)] for the same $r$ fraction of cement replaced with FS (Table 1). Thus, the contribution of RFB or RFS to the strength gain at a replacement level ($r$) for a particular age can readily be determined by calculating the quantity $\alpha - \beta$. A detailed examination of the figure indicates that the increase in strength given by $(\alpha - \beta)/\beta$ generally increases with an increase in the replacement level, $r$. The increase in strength is significant in all cases, particularly for RFB during the early stages. This observation corresponds with the discussions in the earlier sections of this paper. The percentage strength increases for RFS appeared to be higher in subsequent stages due to the contribution of slow-reacting cementitious compounds. The general variation in the quality of the demolished parent concretes with respect to the cement content, cement type and age of parent concrete may have played a role in the absence of a better trend in Figure 11 for RFS in Batch I. This scatter is frequently encountered in concrete recycling investigations that employ actual demolished concrete from various sources as the parent material stock.
Fig. 10. Residual cementing effect in recycled fines observed via compressive strength comparisons using a method that is schematically presented in Figure 9. (a) Batch-I for between 0.50 and 0.75; (b) Batch-II for between 0.50 and 0.75; (c) Batch-I for between 0.00 and 0.40; (d) Batch-II for between 0.00 and 0.25. The strength increases for RFS and RFB at 28 days are shown in (a) and (b), respectively.
CONCLUSIONS

1. A modification to the ASTM method (ASTM C109/C109M) is proposed for measuring the compressive strength of hydraulic cement mortar to assess the contribution of the residual cementing property of recycled fines to the strength increase.

2. Recycled fines from brick aggregate concrete (RFB) manifested the strength increase more prominently than the recycle fines from stone aggregate concrete (RFS). The strength increase expressed as a percentage was determined to be more pronounced for RFB at early ages, whereas the strength increase for RFS was likely to exhibit an opposite trend.

3. The manifestation of the residual cementing property in recycled aggregate concrete was significant in concrete with fresh recycled brick as the coarse aggregate; however, the effect weakened when the recycled aggregate was aged in water prior to casting. The property was weaker in fresh recycled stone aggregate concrete.

4. The evolution of this residual cementing property in recycled aggregates and recycled fines obtained from jaw-crushed stone aggregate concrete and brick aggregate concrete were evident from coherent observations of the physicomechanical properties, chemical compositions and microstructures. The heat flow measurements and chemical compositions indicate the role of fast-reacting cementitious components, which are rich in aluminum in the presence of depleted amounts of gypsum, in the contribution to early strength-gain properties. The role of slow-reacting cementitious components, CH and their interactions with pozzolans have been attributed to weakly contributing additional strengths at later ages in concretes and mortars with recycled aggregates and fines. Measurements of the CH,
C-S-H, free lime and quantification of reactive silica components remain topics for future studies. A complete mineralogical study may further shed light on the precise identification of the sources of dissolution heat as observed in the present study.

5. The quantitative information provided in this paper on recycled aggregate, recycled fines and recycled aggregate concretes is specific to the concrete stocks that were sampled from different construction sites in Bangladesh. However, the authors believe that the methodology, discussions and evaluations provided in this paper from physical, mechanical and chemical perspectives are general in nature and suitable for application to other samples of recycled aggregates and recycled fines to qualitatively and quantitatively evaluate their respective contributions to strength gains.

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