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Quantification of Concrete Aggregate Liberation through Abrasion Comminution

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Abstract: Recycling concrete waste into structural concrete reduces the consumption of raw materials, decreases transport and production energy costs, and saves the use of limited landfill space. Since attached mortar is known for lowering the performance of recycled concrete aggregates (RCA) in concrete applications, current recycling involves the use of RCA as a road base material or in non-structural concrete with low strength requirements, therefore, the application in structural concrete is limited. In general, the applicability of RCA is improved by comminution through various crushing methods. Hereby, the parent particles are cleaved or shattered into a minimum of two particles with a comparable size and a number of much smaller particles. Through this technique, both mortar and aggregates fracture alike, resulting solely in a size reduction. However, to minimise undesirable effects, original natural aggregates (ONA) have to be cleared from attached mortar. Through the use of attrition and shear instead of pressure or impact, surface layers, edges and corners are removed thereby producing particles both slightly smaller and much smaller than the initial. In this study, ONA liberation through abrasion was investigated.

Keywords: recycled concrete aggregates; crushing; aggregate liberation; abrasion

1 Introduction

Deteriorated and obsolete constructions generate large amounts of demolition waste [1]. Reutilizing recycled concrete fines (RCF) and recycled concrete aggregates (RCA) into new concrete reduces the need for natural aggregates (NA) and also decreases the use of landfill areas, while simultaneously providing a high-quality recycling of construction and demolition waste. The main difference between NA and RCA is the attached (i.e. residual) mortar. This is the key factor and main concern causing impaired quality when applied to new concrete. Additionally, the fines generated by the production of RCA are considered to have a negative influence on the quality of the new concrete [2,3]. Their presence is therefore considered to be undesirable.

Several experimental concrete recycling techniques were developed recently, aiming to separate aggregates from hardened cement paste (HCP). These techniques include smart crushing [4-6], electrodynamic fragmentation [7], microwave fragmentation [8], and heating and rubbing [7,9]. This current work is part of a larger study into upscaling the Smart Crushing technique and is carried out on a modified jaw crusher with a typical capacity of up to 15 t/h.

preceding studies investigated the properties (PSD, density, oxide and mineralogical composition) of a number of recycled concrete fractions, obtained through both conventional jaw crushing and optimized crushing. Results showed that the optimized technique leads to RCA with a significantly lower amount of residual attached mortar [4-6]. Furthermore, the application of such RCA has shown no loss of mechanical properties. At short curing times, the mechanical properties of the recycled concrete sand mortars (RCS) is promising, achieving higher strengths than standard reference samples [5,6]. In contrast with regular crushing, the fines generated by smart crushing primarily consist of HCP and can be thermally activated to replace cement [5,6].

The mechanism at the root of shaping natural river gravel is abrasion [10]. Due to this, flaws in the material which could succumb to abrasion forces were tried long before the gravel is extracted, forming each particle to be resilient against these forces. Consequently, abrasion can be a suitable method to liberate the original natural aggregates (ONA) from RCA without fracturing it.

In general, the applicability of RA is improved by size reduction, making use of various crushing methods. Here, the main method of size reduction is the fracturing of particles under pressure or impact, whereby the parent particles are broken into a minimum of two particles with a comparable size and a number of much smaller particles. When this occurs, both mortar and aggregates fracture alike. Hence, the generated fractions have a composition close to the original concrete. With the intention of applying the recycled concrete aggregates into concrete, undesirable properties have to be minimised and can be done by liberating the ONA and clearing them from attached mortar. Thanks to this method, the properties of RCA and ONA would be similar, thus guarantee a similar applicability.

In this study, based on the concept of Smart Crushing (Figure 1), a jaw crusher-based crusher explicitly designed for

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Concrete recycling was used with the aim of producing RCA with a high degree of liberated DNA. The removal of HCP, from both fine and coarse RCA has been proven to be possible with the use of this technique \cite{5,6}. The produced RCA were found to have enhanced separation into the original constituents, namely gravel, sand, and HCP. The design of the prototype crusher used in the tests deviates from the drawing in Figure 1 by containing material between the jaws by means of a plate pivotally attached to the stationary crushing plate. The goal of this technique is to disperse the recycled concrete (RC) into its composing materials without damaging the components themselves. In order to achieve this, the occurring (indirect tensile) forces have to be intermediate between the failure strength of the HCP and the aggregates. Hereby, the aggregates stay intact while the HCP breaks under the applied stress.

Comminution through impaction is associated with high internal stresses, whereby particles are scattered; this type of loading is therefore avoided by keeping the eccentric speed of the jaws low. Additionally, direct compression or pinching, being the operational mechanism of a typical jaw crusher, also introduces yield stresses surpassing those of the aggregates and leads to subsequent cleavage of both HCP and aggregates. This type of loading is, therefore, to be avoided and is prevented by applying a jaw distance much larger than the maximum particle size. In this test setup, the shape of the crusher box coupled with the distinct movement of the single-toggle type jaw crusher (an eccentrically gyrating cycle) create a condition where pressure, shear, and attrition are present. When applying these conditions instead of pressure or impact to produce the RCA, surface layers, edges and corners are removed, producing particles both slightly smaller and much smaller than the initial. Herewith, the interlocking of angular pre-fractured concrete particles is beneficial for the abrasion process during the comminution. To aid this and maintain these conditions during the entire process, the removal of the finer fractions, created during the initial fracturing, was found to be beneficial for comminution through abrasion \cite{5}. The aim of this study is to develop a method for quantifying the level of aggregate liberation achieved without prior knowledge of the initial concrete constituents.

2 Experimental

2.1 Materials

The RC investigated in this study is released during the evacuation and renovation project of the AVF-cyclotron building at the campus of the Eindhoven University of Technology (TU/e) where it has been used in a structural application since 1967. As a first step in the recycling of this concrete, the rebar of the elements was removed by fracturing the concrete with a hydraulic jaw pulveriser. In preparation for a direct application of the material on the TU/e campus as a road base material, the pulverised material was crushed using an excavator-operated bucket jaw crusher before transferring the material to a flexible intermediate bulk container (FIBC). Then, prior to using the RC for the research, the pre packed material was exposed to outdoor weather conditions for several months. Prior to studying fracturing characteristics of the RC, the initial material was sieved with a Mogensen sizer (type 0554) equipped with effective mesh sizes of 16 mm, 10 mm, 4 mm, 1 mm and 150 μm. Only the >10 mm fractions were used as input material for this study.

Upon visual assessment of the concretes fractured surface, only non-angular course aggregates could be distinguished. This suggests that the origin of the course aggregates used in the production of this concrete can be found in natural gravel (e.g. deposited by rivers). In contrast to other countries, the use of this type of aggregates in concrete is common practice in the Netherlands and in all probability accordingly the case. However, the exact origin of the aggregates and the original recipe could not be retrieved.

| Table 1 Cement oxide composition (1982, supplied by manufacturer) |
|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|-------------------|
| CaO               | SiO2              | Al2O3             | Fe2O3             | K2O               | Na2O              | SO3               | MgO               | TiO2              |
| 62.6              | 21.4              | 5.6               | 3.0               | 0.8               | 0.2               | 3.0               | 1.7               | 0.3               |
| P2O5              | Cl                | LOI               |
| 0.1               | 0.0               | 0.9               |

It is assumed that the concrete is produced with a cement supplied by the market leader of that time, especially considering the year of construction. Compositional data of the cement of this producer only went back to 1982.
Nevertheless, the 1982 cement is produced in the same kilns under similar conditions with raw materials originating from the same quarries. From this, it is concluded that the composition of the 1967 cement closely resembles that of the 1982 cement given in Table 1.

2.2 Methods

The capacity of the crusher was regulated by the size of the outlet opening of the crusher box. In this study, the minimum distance between the containment plate and the moving jaw (hereinafter called aperture) was set at 30 mm.

The sampling of both the initial and the processed material was done in accordance with the DIN EN 932-1 standard [11] from respectively 1 m³ FIBCs and a conveyor. Contrary to a typical continuous process, the experiments were carried out as a batch process. Hereby, the phase of filling and emptying the crushers cavity deviates and could potentially influence the properties of the produced materials. To exclude these influences, samples were only taken after an initial purge with the material and prior to discharging the crusher’s cavity.

To avoid agglomeration when sieving the material, all materials investigated were pre-dried to an oven-dry state at 105 °C until constant mass was reached [12]. Particle size analysis was performed in accordance with EN 933-1 and EN 933-2 [13,14] whereby additional sieves were added to increase the detail of the study (45 mm, 22.4 mm, 11.2 mm, 5.6 mm, 2.8 mm, 1.4 mm, 710 µm, 355 µm, 180 µm, and 91 µm added). Consecutively, the material was split into corresponding fractions for further analysis.

The elements in the materials (computed as oxides) were determined by energy-dispersive X-ray fluorescence (EDXRF) using a PANalytical Epsilon 3 instrument (a 9 W / 50 kV rhodium X-ray tube, a silicon drift detector) equipped with Omnian software 1.0.E (PANalytical). The samples for EDXRF were 40-mm fused beads prepared from pre-dried powdered samples, 0.95 g, with a 67.00% Li₂BrK₂O₃ − 33.00% LiBO₂ flux, 9.50 g; and 0.32 ml of a 4 M LiBr solution as a non-wetting agent using a LeNeO fluxer (Claisse) at 1065 °C with Pt-Au crucibles.

SEM analyses were performed on a flat grounded sample provided with a gold sputter coated film by using a Phenom ProX desktop scanning electron microscope (SEM) equipped with a CeB6 thermionic electron source. Micrographs are recorded at 15 kV with a spot size of 4 by using a 4-quadrant back scattering electron detector.

The loss on ignition (LOI) of the materials was determined according to EN 196-2:2005 [15], by measuring the mass loss of samples at 950 °C to a constant mass. The additional thermal analysis using a Netzsch STA 449 F1 Jupiter was performed by thermogravimetric analysis (TGA) to determine the mass loss for specific temperature intervals and by differential scanning calorimetry (DSC) to determine the quartz content of the various size fractions. At about 573 °C, the endothermal crystalline phase changes from α-quartz to β-quartz without loss of mass [10]. The thermal analysis was performed in Pt crucibles in inert atmosphere (N₂) up to a maximum temperature of 950°C. The peak area on the DSC curve of both the heating and the cooling step of 20 °C / minute was analysed and correlated to measurements of samples containing a known amount of α-quartz, quantifying the quartz content of the measured samples. In contrast to prior studies, pure quartz is used for the calibration since the quartz content of the original aggregates used is unknown [5,6,17,18].

3 Results and discussion

During the abrasion comminution process, a perceptible amount of heat is generated in comparison with typical size reduction methods (e.g. jaw or impact crushing). This can be related to the number of fractures needed to produce the comparably large amount of finer material. Specifically, in a process with a limited throughput, in addition to the comminution, liberated aggregates remain present in the process and continue to rub over one another generating heat through friction. Generally, additional heat generated in comminution is considered as a waste of energy since it serves no purpose. In this process, the moisture level of the initial concrete may vary; the present water can cause the agglomeration of the fines, potentially complicating further extraction processes through clogging or smearing. Thus, the heat generated while particles are being both fractured and continuously mixed can be utilised for the (pre)drying of the material. In addition to dry screening and potential refeeding of the larger size fractions, this enables further dry removal of fines through air separation and subsequent storage in silos prior to further treatments.

3.1 Comminution

The smart crushing technique (in laboratory-scale tests on purposely designed concrete) was found to be the most effective with three passings through the crusher, with the finer material sieved out in between passings [5]. Since the characteristics of the materials are altered when the finer fraction is sieved out prior to further processing, in this study, only a single passing through the crusher is tested. Figure 2 shows the PSDs for the initial material and the resulting PSD of a single crusher passing at an aperture of 30 mm.

Overall, in Figure 2, it can be seen that all particles are substantially influenced by the abrasion comminution process whereby both PSDs have an amount of material in common between 5.6 mm and 22.4 mm only. The formed particles show an even distribution over the newly formed size range, only 0.2% being larger than 31.5 mm, and, in turn, making a refeed for further size reduction unnecessary.
3.2 Composition

Figures 3 and 4 show respectively the oxide compositions determined by XRF of the comminuted RC for the main (Al₂O₃, SiO₂, CaO, and Fe₂O₃) and minor oxides (MgO, SO₃, K₂O, TiO₂, and MnO) for the oxides in common with the cement oxide composition (Table 1). Corresponding LOI is given in Figure 7.

CaO shows a relatively constant decrease in content with an increase in particle size until it reaches a near stable value of 6.5% on average in particles larger than 5.6 mm. SiO₂ shows an opposite trend with a declining increase until it approaches a limit in particles larger than 5.6 mm with a maximum value of 84.6%. In the Netherlands, siliceous river aggregates are commonly used, therefore, these trends can indicate relatively clean aggregates and a concentration of cement stone in the fines. Even though visual inspection of the aggregates supports this theory, the absence of non-siliceous aggregates or siliceous fillers cannot be assured, making the quantification of the quality of liberation based on SiO₂ or CaO content uncertain. Other oxides might also not be used as cement specific tracer since Al₂O₃ and MgO can occur in natural aggregates, Fe₂O₃ content can be altered by contamination from either rebar or processing, and the deviations between the different samples for K₂O, TiO₂, and MnO can be within the measurement error of the equipment used. SO₃, however, can be used as a cement specific tracer oxide since the initial RCA is clean (from e.g. gypsum; CaSO₄) and it is an element unlikely to be found in natural aggregates in the Netherlands. Through the oxide composition of the unhydrated cement and the SO₃ content of the RCA sample, the unhydrated cement content is calculated (Figure 5).

The combination of the calculated cement content per fraction (Figure 5) and the PSD of the respective size fractions (Figure 2) results in a cement content of 14% by mass of hydrated concrete. Assuming that the density of the hardened concrete is close to 2400 kg/m³, the quantity of cement applied is estimated to be 345 kg/m³ which is in good correlation with the cement content used in a simple concrete mixture.

In order to assess the hydrated cement content, the chemically bound water and gel water present in the hydration products need to be accounted for. Brouwers states that for complete hydration under saturated conditions (allowing for unlimited water imbibition) or hardening at 80% RH (and imbibition) a w/c₀ of 0.32 and 0.27 are respectively needed. The calculated content of the unhydrated cement and the fully hydrated cement (for w/c = 0.32 and w/c = 0.27) are given in Figure 5. The rate of cement hydration is continuously decreasing over time and complete hydration is
Theoretically only possible for cement grains smaller than 50 \( \mu m \) \cite{23}. Therefore, complete hydration of all present cement is unlikely. This is confirmed by the SEM micrograph (Figure 6) taken from the mortar of a flat ground section of RCA, where the EDX analyses, performed at 3 different spots (1, 2, and 3; Table 2), indicate that the circular particles are agglomerated single cement grains.

![Fig. 5](image)

**Fig. 5** Calculated content of cement and completely hydrated HCP for saturated hydration (w/c = 0.32) and 80% RH (w/c = 0.27) according to \cite{19} vs. particle size.

![Fig. 6](image)

**Fig. 6** SEM-BSE micrographs of the mortar from a polished RCA, circular shapes (bright) are unhydrated cement grains (EDX analysis in Table 2).

<table>
<thead>
<tr>
<th>Spot</th>
<th>Mass fraction (At. %)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Ca</td>
</tr>
<tr>
<td>1</td>
<td>23.3</td>
</tr>
<tr>
<td>2</td>
<td>18.83</td>
</tr>
<tr>
<td>3</td>
<td>17.82</td>
</tr>
</tbody>
</table>

Table 2 SEM EDX elemental of spots indicated in Figure 6.

Hence, the cement used is not completely hydrated, whereby the HCP content of the analysed fractions of RCA is characterized by a lower boundary of non-hydrated cement and an upper boundary of completely hydrated cement under saturated conditions. To specify the amount of water present in the hardened cement paste (as both gel water and chemically bound water) thermal analysis is performed on the various fractions. Alarcon-Ruiz et al. \cite{22} state the occurring thermal decomposition reactions of cement paste and concrete as:

- 30–105°C: removal of evaporable water and partial removal of bound water. At 120°C all evaporable water is generally considered to be eliminated \cite{23},
- 110–170°C: decomposition of gypsum \cite{23,24} and ettringite \cite{23}, partial dehydration of carboaluminate hydrates \cite{26},
- 180–300°C: dehydration of C-S-H and carboaluminate hydrates \cite{23,27,28},
- 450–550°C: dehydroxylation of calcium hydroxide (CH) \cite{23,24} (Eq. 1):
  \[
  \text{Ca(OH)}_2(s) \rightarrow \text{CaO} (s) + \text{H}_2\text{O} (g) \quad (29)
  \]
  \[\text{(1)}\]
- 700–900°C: decarbonation of calcium carbonate (C\(\text{C}\)) \cite{23,30} (Eq. 2):
  \[
  \text{CaCO}_3(s) \rightarrow \text{CaO} (s) + \text{CO}_2 (g) \quad (31)
  \]
  \[\text{(2)}\]

Hereby, the most predominant mass losses occur for the decomposition of C-S-H, CH, and C\(\text{C}\) \cite{32}. Figure 7 displays the mass loss for 450-550°C (CH), 700-900°C (C\(\text{C}\)), and 105-950°C. Here, the magnitude of the dehydration of the other hydrates (e.g. C-S-H) can be seen. Additionally, the influence of both the CH dehydration and C\(\text{C}\) decomposition on the total LOI can be seen. Where both the total LOI and the LOI of CH (except for the fractions > 16 mm) show a decrease with increasing particle size, the LOI regarding C\(\text{C}\) shows to be nearly equal for particles between 355 \( \mu m \) and 16 mm. CH and C\(\text{C}\) mass loss together show to correlate well with the total LOI (Figure 8).
Fig. 7 LOI for Ca(OH)$_2$, CaCO$_3$, and total LOI (950°C) vs. particle size

Fig. 8 Correlation between LOI for Ca(OH)$_2$ and CaCO$_3$, vs. total LOI (950°C)

The presence of CH is related to the hydration of the alite (C$_3$S, Eq. 3) and belite (C$_2$S, Eq. 4) clinker phases (in absence of other clinker phases) into C-S-H and CH, according to the following equations:

\[
\begin{align*}
\text{C}_3\text{S} + (3-x+y)\text{H} & \rightarrow \text{C}_3\text{SH}_y + (3-x)\text{CH} \quad \text{(3)} \\
\text{C}_2\text{S} + (2-x+y)\text{H} & \rightarrow \text{C}_2\text{SH}_y + (2-x)\text{CH} \quad \text{(4)}
\end{align*}
\]

Here, the C/S ratio is subject to hydration conditions (e.g. particle size, and age) where x ranges from 1.5 to 2 [33]. The H/S ratio (y) is determined by the drying conditions (e.g. P-dried and saturated state). In the concretes lifespan, the CH is partially carbonated forming C\text{C} [29]:

\[
\text{Ca(OH)}_2(s) + \text{CO}_2(g) \rightarrow \text{CaCO}_3(s) + \text{H}_2\text{O}(l)
\]

(5)

Given that the samples are representative and under the assumption that the original concrete was made with aggregates and fillers absent of C\text{C} (e.g. limestone) both the CH content and the C\text{C} content (due to carbonation) are directly related to the cement content and should thereby, independent of the level of carbonation, have a linear correlation (see Figure 9).

Fig. 9 LOI of CH and C\text{C} vs. calculated cement content

Fig. 10 Loss of chemically bound water and gel water vs. calculated cement content

As expected, the LOI due to CH shows a linear correlation with the calculated cement content. However, the LOI attributed to C\text{C} appears to have two distinct sections: the fractions containing less than 20% cement, the particles larger than 355 µm, show a near constant and independent C\text{C} content with a LOI of approximately 2.15. Over 20%, particles smaller than 355 µm show a linear correlation with the cement content. It can be concluded that the carbonation of CH, intrinsically the cement, is the sole contributor of C\text{C}. If a C\text{C} filler was used in the production of the initial concrete, it would have been evenly distributed with the cement and after comminution, it would follow the distribution of the HCP. Additionally, SEM analysis of the mortar did not indicate the presence of a C\text{C} filler. In the case that some of the aggregates consist of C\text{C}, it is possible that, through abrasion comminution, these aggregates succumbed and got deposited in the finer fractions while the quartz aggregates stayed intact. This is consistent with the principle known from milling whereby C\text{C} (Mohs hardness H=3) grinds faster in the presence of quartz (H= 7) and quartz grinds slower in the presence of C\text{C} [34].

Overall considered, during the thermal analysis from 105 to 950°C, the material is decarbonated and all water is eliminated. Hence, exclusion of the mass loss due to decarbonation leaves the loss of chemically bound and gel water.
Figure 10 displays the linear correlation between the calculated cement content and the loss of water indicating a hydration w/c ratio of 0.145. Consequently, the HCP and aggregate content can be determined indicating the level of liberation in Figure 11. The α-quartz content determined through thermal analysis (DSC) for the different fractions is correlated with the calculated aggregate content and shown in Figure 12. With a slope of near 1, a good correlation between quartz content and aggregates is found whereby the influence of potentially present calcium carbonate is negligible.

![Composition of the RCA fractions divided into HCP and aggregate components](image1)

![α-quartz vs. aggregate content](image2)

### 4 Conclusions

In conclusion, it is shown that the investigated method is able to produce gravel and sand fractions which are clean of cement paste out of RC. To determine the quality of liberation, a method is developed making use of a cement tracer oxide. This is favourable when there is no knowledge of the composition of the original aggregates. In this case, the thermal analysis showed calcium hydroxide to correlate linearly with the cement content while the calcium carbonate showed to be disproportionately distributed. Hereby, it is concluded that a small extent of the original aggregates consist of calcium carbonate which ended up in the fines (through combined comminution with quartz). Further thermal analysis specified the RCAs non-evaporable water content through which the level of cement hydration and the HCP content were determined. SEM coupled with EDX analyses confirmed the absence of fine calcium carbonate in the cement matrix and acknowledged the presence of unhydrated agglomerated cement particles. Finally, DSC analysis indicated the aggregate content to have a good correlation with the α-quartz content whereby the influence of the present calcium carbonate is negligible.

To substantiate how the presence of finer particles in the input material influences the output material (e.g. by cushioning, energy absorption, or the prevention of interlocking), additional studies are required. Subsequently, this data can then be used to validate whether a relatively high throughput, together with the sequential removal of produced fines, and a refed of larger particles is beneficial for the output quality, quantity, and overall energy consumption compared to a single passing at a lower throughput.

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### References:
