Effect of Surface Modifications of Recycled Concrete Aggregate on Concrete Properties

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Received: 25 November 2017; Accepted: 24 December 2017; Published: 26 December 2017

Abstract: The experiment aims to test the specific way of producing concrete with recycled concrete aggregate (RCA). To reduce its negative impact on the concrete properties, two different ways of treatment of the RCA with geopolymer slurry were applied—coating during the mixing using the specific mixing approach and coating prior to the mixing of concrete. As control samples, a mixture prepared by natural aggregate only and a mixture with RCA that was prepared by standard mixing with no coating process were tested as well. The results of density, total water absorption, and compressive strength in periods of 28, 90, 180, and 365 days of curing are presented and evaluated. Both methods of coating of the RCA with geopolymer slurry allow for the preparation of concrete with properties comparable to those of normal concrete (prepared by standard mixing with natural aggregate); thus, it seems to be a promising way to enhance the rate of RCA application. The positive effect of coating is clearly visible after a longer period of curing (180 days). When comparing the methods of RCA coating, coating directly during the mixing yields somewhat better results; it is also positive from the technological point of view, since the process is simpler in practice.

Keywords: coating; compressive strength; construction and demolition waste; geopolymer; interfacial transition zone; water absorption

1. Introduction

Waste not only represents a huge loss of resources when it is not used in the form of materials and energy, it can also endanger the environment if it is misapplied. The European Commission states that up to 80% of products are used only once and then discarded. Up to one-third of the total municipal waste in Europe will end up in landfills, which occupy space and can also cause air, water, and soil pollution. A quarter of European waste is incinerated, which can lead to the emission of hazardous pollutants if inadequately regulated [1,2].

On the other hand, recycling and waste recovery comprise a key tool for protecting the environment, ensuring the efficient use of natural resources, increasing employment in the waste management sector, and combating climate change. The European Union’s goal is therefore to reduce the amount of waste that ends up in landfills and to make production processes more efficient by lowering the use of primary resources and reducing the emitting substances [3,4].

In the Slovak Republic, 10.5 million tonnes of wastes were produced in 2015, consisting of 8,674,942 tonnes of industrial wastes and 1,888,456 tonnes of municipal wastes. About 20% of total waste was the construction and demolition waste (C&DW). The largest share of total waste (up to 76%) in Slovakia was landfilled, another 12% was combusted, and only 6% was recycled or composted. In the European Union, 28% of total waste was recycled and 16% was composted, on average [5,6].

Recycled aggregates are produced by crushing and sorting an existing structure or parts thereof. Recycled aggregate from crushed concrete—known as recycled concrete aggregate (RCA)—is used
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quite often. There have been attempts to use recycled material from crushed masonry. The properties of recycled aggregate are mainly influenced by its composition, affected by the quality of demolition, sorting, and recycling. Properties are divided into three groups—geometric, physical, and chemical properties. Geometric properties are determined by the sieving test for grain-size distribution, the fine particle fraction, and the softness modulus. Another important geometric property of recycled aggregate is the shape index, which, among other things, influences the design of the concrete recipe. Physical properties include crushing resistance, abrasion resistance, frost resistance, bulk density, water absorption, etc. The chemical properties are, for example, the content of alkali, soluble sulphides, and the total representation of different building materials in the recyclate. The more types of matter commonly found in C&DW, the worse will be the achieved properties of the RCA [7–10]. RCA made from waste concrete is a problematic material for concrete production, as it has a variable portion of original cement mortar adhering to the aggregate’s surface, resulting in higher water absorption. The quality is also affected by tiny cracks on the surface. Therefore, when using this RCA, a lot of water must be added, resulting in a decrease in strength properties. Moreover, the slump loss during transporting (workability) is observed to be higher than that of concrete made of natural aggregate [11,12].

In general, the factors determining the mechanical performance of concrete are the strength of the cement matrix, the strength of aggregates, and the strength of interface between the aggregate and the cement matrix and the stress acting on the interface of cement matrix/aggregates. This is a particularly weak area of concrete, which consists of pores around the hydrates and aggregate, and is characterized by a discontinuous porous area between the aggregate and cement paste [13–15].

While only one type of interfacial transition zone (ITZ) exists in normal concrete, lying between the virgin aggregate and the new cement mortar, three types of ITZ occur in RCA-based concrete—between the natural aggregate and the new mortar, inside the RCA between the old virgin aggregate and the old mortar, and between the old mortar and the new mortar [16,17].

It is clear that different qualities of various ITZ create an increased risk of unfavourable impact on concrete quality. According to [18–21], this suggests that the properties of recycled aggregate concrete can be enhanced by improving the new ITZ.

To improve the mechanical interface between the aggregate and the cement matrix, the coating methods are investigated. The surface of aggregate is coated with an inorganic material (fly ash, slag, etc.), thus improving the surface properties of RCA and ITZ and enhancing the mechanical properties of the resultant concrete [22–24]. There are two basic ways to coat the aggregate—coating in advance before mixing the concrete [25–28] and coating during the mixing using specific mixing methods. With the exception of standard single-stage mixing, in which the entire mixing process is done in one stage, several mixing approaches are under investigation, such as the two-stage mixing approach (double-mixing approach), in which the entire mixing process is done in two stages, and the three-stage mixing approach (triple-mixing approach), in which the entire mixing process is done in three stages. The principle of dividing the mixing process into two/three steps lies in the different timings of concrete’s components addition; the coating of aggregate occurs in the first stage in principle [29–32].

In this paper, an experiment is presented to test the specific way to produce the concrete with RCA. To reduce the negative impact of RCA on the concrete properties, two different ways of treatment by geopolymer slurry were applied. In total, four series of mixtures were tested in our study: the control mix containing 100% of natural aggregate in both fractions and was prepared by standard mixing, the mixture containing RCA as a full replacement for natural coarse aggregate (fraction 4/8 mm) and prepared by standard mixing, and same two aforementioned mixtures with surface treatment by coating of RCA during and prior to the mixing of concrete. Results in terms of density, total water absorption, and compressive strength in periods of 28, 90, 180, and 365 days of curing are presented and evaluated. The significance of the paper lies mainly in the specific materials that are used; (a) for coating the RCA, the geopolymer slurry was used—this decision was made on the basis of the knowledge of the geopolymer quality, for which excellent technical parameters are presented either in
bulk form [33] or in the form of artificial aggregate [34,35]. Usually, as visible in above given references, cementitious/pozzolanic materials are used for coating; (b) RCA is represented by specific kind of C&DW—concrete roof tiles remaining as waste in new construction. Moreover, the long-term testing was performed, which yields a comprehensive set of results.

2. Material and Methods

The experiment aims to test the specific way of producing concrete with RCA. To reduce the negative impact of RCA on the concrete properties, a specific approach was designed, involving the treatment of RCA with geopolymer slurry. This was applied in two different ways:

(a) coating of RCA during the mixing of concrete, and
(b) coating of RCA prior to the mixing of concrete.

As control samples, mixtures prepared by standard mixing were tested as well. Materials used are divided into materials for making the geopolymer slurry to coat the RCA and materials for subsequent mixing of the concrete.

For geopolymer slurry, the following materials were used:
- coal fly ash (FA) from the heating plant in Kosice, Slovakia,
- liquid glass (Na$_2$SiO$_3$),
- 8M NaOH solution, and
- water.

The chemical analysis of the fly ash is as follows: MgO (1.18%), Al$_2$O$_3$ (23.21%), SiO$_2$ (51.11%), CaO (2.58%), and Fe$_2$O$_3$ (6.4%). The calcium oxide content is less than 10%; hence, it can be classified as Class F, according to ASTM 618 standard.

The particle size is as follows: d(0.1) = 3.97 µm, d(0.5) = 20.44 µm, and d(0.9) = 84.73 µm.

Recycled concrete aggregate coming from concrete tiles was used (RCA). Residual tiles from construction site were crushed using laboratory crusher, following by sorting of 4/8 fraction. As a reference aggregate, the natural one (NA) was applied, too. Physical properties of aggregates are given in Table 1.

<table>
<thead>
<tr>
<th>Composition</th>
<th>Bulk Density (kg/m$^3$)</th>
<th>Void Ratio (%)</th>
<th>10 min Water Absorption Capacity (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>RCA uncoated</td>
<td>1370</td>
<td>30</td>
<td>9.9</td>
</tr>
<tr>
<td>RCA coated prior mixing</td>
<td>1320</td>
<td>32</td>
<td>8.2</td>
</tr>
<tr>
<td>NA 0/4</td>
<td>1830</td>
<td>30</td>
<td>1.2</td>
</tr>
<tr>
<td>NA 4/8</td>
<td>1580</td>
<td>39</td>
<td>1.4</td>
</tr>
</tbody>
</table>

The basic concrete mixture was designed to have following constant parameters:
- amount of cement per cubic meter (370 kg), and
- aggregate fraction ratio (0/4:4/8 = 60:40).

The amount of water and plasticizer was adjusted to keep the same consistency (slump S2). The real amounts are given in Table 2.

In total, four series of mixes were considered in our study. They differ in terms of mixing course-i.e., how the RCA is coated. B0 is the control mix containing 100% of natural aggregate in both fractions, prepared by standard mixing. B1 is concrete containing 100% of RCA as full replacement for natural coarse aggregate (fraction 4/8 mm), prepared by standard mixing. B2 and B3 samples are similar to B1, except that surface treatment of RCA by coating is applied in the latter. The principles of composition and mixing course for the individual samples are given in Table 2.
Table 2. Experimental mixtures details.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Agg. 0/4</th>
<th>Agg. 4/8</th>
<th>Mixing Details</th>
<th>Plasticizer (%)</th>
<th>Total Water (L)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>NA</td>
<td>NA</td>
<td>RCA</td>
<td>Standard Mixing</td>
<td>Coating RCA during Mixing</td>
</tr>
<tr>
<td>B0</td>
<td>●</td>
<td>●</td>
<td>-</td>
<td>-</td>
<td>0.5</td>
</tr>
<tr>
<td>B1</td>
<td>●</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>0.5</td>
</tr>
<tr>
<td>B2</td>
<td>●</td>
<td>-</td>
<td>-</td>
<td>0.5</td>
<td>210</td>
</tr>
<tr>
<td>B3</td>
<td>●</td>
<td>-</td>
<td>-</td>
<td>1.5</td>
<td>220</td>
</tr>
</tbody>
</table>

Note: “●” included; “-” not included in mixture composition.

To modify the surface properties of RCA, the geopolymer slurry based on FA was prepared. The composition is given in Table 3.

Table 3. Composition of geopolymer slurry.

<table>
<thead>
<tr>
<th>Component</th>
<th>Composition (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coal fly ash</td>
<td>75</td>
</tr>
<tr>
<td>Liquid glass</td>
<td>9</td>
</tr>
<tr>
<td>8 M NaOH solution</td>
<td>11</td>
</tr>
<tr>
<td>Water</td>
<td>5</td>
</tr>
</tbody>
</table>

For coating of individual batches of concrete samples, the slurry amount was calculated to make a layer thickness of 0.25 mm. It was then used in two specific ways.

I The coating of RCA during the mixing of concrete (sample B2). Here, the mixing procedure was divided into the following steps: the fraction 4/8 of RCA was coated by geopolymer slurry in the standard laboratory mixer. The coated RCA was left to rest for 30 min in the mixer and then the mixing continued by adding the cement to form a further layer on the wet aggregate. Finally, fine aggregate, water, and plasticizer were added.

II The coating of RCA prior to the mixing of concrete (sample B3): the RCA was coated in standard laboratory mixer and was subsequently cured for 28 days under standard laboratory conditions, as given for making the concrete specimens (95% RH and 20 ± 3 °C). Then, the concrete was mixed by standard course [36], using coated and hardened RCA and adding the cement, fine aggregate, water, and plasticizer.

The thickness of coating layer was chosen based on [28], who suggested 36–60 µm as typical thickness of interfacial transition zone (ITZ) in recycled aggregate concrete and the thickness 0.5 mm for covering whole ITZ. In this experiment, the 0.25 mm thickness was chosen as being between those values. Similarly, [37] found 0.25 mm thickness to perform well for coating the RCA since it gives optimal amount of paste to coat the grains, although it was for asphalt binder.

The volume of paste for coating the coarse aggregate was calculated using following equations: The volume of coating paste $V_{cp}$ [m$^3$]:

$$V_{cp} = F \times \delta$$  \hspace{1cm} (1)

where:
- $F$ is the surface area of aggregates (m$^2$), and
- $\delta$ is the thickness of layer (µm).

The surface area of aggregate $F$ [m$^2$]:

$$F = f \times \left(\rho_b / \rho_p\right) \sum(p_i / 0.1 \times d_i)$$  \hspace{1cm} (2)
where:
- $f$ is the coefficient characterising the surface quality of aggregate (12 is given for the poor surface quality),
- $\rho_b$ is the loose bulk density of aggregate (kg/m³),
- $\rho_p$ is the particle density (kg/m³),
- $p_i$ is the amount of aggregate having average grain size $d_i$ (%), and
- $d_i$ is the average grain size of aggregates fraction (mm).

For each of mixture type, 24 standard cubes of $100 \times 100 \times 100$ mm were prepared for testing individual properties (12 for density and compressive strength, while the same samples were used for measuring these properties, and 12 for total absorption). Samples were demoulded after 24 h and were then cured under standard moisture and temperature conditions (95% RH and $20 \pm 3 ^\circ$C) until the corresponding testing time. Density, according to [38], total water absorption using standard gravimetric method (samples were dried at 105 $^\circ$C, and, then immersed in water up to constant mass. Absorption capacity is determined based on the difference between soaked and dry mass of sample), and compressive strength, according to [39] (constant rate of loading $0.6 \pm 0.2$ MPa was applied gradually until the specimen’s failure, while satisfactory failures were achieved in each test) were tested after 28, 90, 180, and 365 days. The arithmetic means of the values obtained by measuring the three samples are presented below as test results.

### 3. Results and Discussion

The results of density are shown in Figure 1. The highest values were achieved by the reference sample B0, prepared with only natural aggregates, ranging from 2290 kg/m³ to 2340 kg/m³. The lower density values of B1–B3 samples are due to the substitution of natural aggregate by RCA. When comparing B1–B3, the lowest density was reached by sample B1, ranging from 2130 kg/m³ to 2190 kg/m³. This corresponds with the character of the materials surrounding the grains; in the case of B1, it is standard cement paste, while in the case of both the B2 and B3, it is the geopolymer.

![Figure 1. Density of samples after 28, 90, 180, and 365 days of curing.](image-url)

Figure 2 shows the results of total water absorption. The value for control sample B0 was about 3% during all the periods. Generally, the values of all samples show a decreasing tendency with time. All of the samples containing RCA (B1–B3) achieved higher values than those containing NA (B0). There is no significant difference between samples with coated RCA (B2 and B3) and sample prepared by standard mixing (B1) in the earlier periods (28 and 90 days). With progressive curing time, the water absorption of samples with coated RCA was significantly reduced and the value was closer to that of
the control sample. The rate of this reduction is evident by looking at the values in Table 4, where the difference between values for 28 and 365 days is given. Sample B1, containing RCA with no treatment, achieved the highest (worst) water absorption values for all testing times.

![Figure 2. Total water absorption of samples after 28, 90, 180 and 365 days of curing.](image)

The results of compressive strength are shown in Figure 3. The highest strength value was achieved by the control sample B0 after 28 days, while after 365 days, it was sample B2. In fact, the difference between the long-term values of control sample and both of the samples with RCA coated with geopolymer is negligible (B2: +3%; B3: −0.4%). In other words, the treatment of RCA by coating causes the concrete to have the same strength as the control containing the natural aggregate. Sample B1, containing RCA with no treatment, achieved the lowest compressive strength for all the testing times.

![Figure 3. Compressive strength of samples after 28, 90, 180 and 365 days of curing.](image)

The positive effect of geopolymer coated aggregate on the compressive strength of concrete is also presented in [40]; here, authors present the method of coating the RCA prior mixing, using both the geopolymer and cement slurry. As given here, geopolymer coated recycled aggregate concrete showed higher compressive strength compared to uncoated recycled aggregate concrete and also cement coated recycled aggregate concrete. The increase in compressive strength (21%) was achieved when geopolymer coated RCA was dried for 24 h at 85 °C prior the mixing of concrete. In our experiment,
the difference between B3 (coating of RCA prior mixing) and B1 (uncoated RCA) is +11.0% while no any thermal treatment was applied. As for 365-day values, the difference is even +37.0%.

The satisfactory type of failure was observed during the testing of all the samples (see examples given in Figure 4).

As for coating RCA during mixing, the results can be compared with that of presented for similar way of coating—wet RCA coated with powdery pozzolanic materials. Authors [41] have presented 37.0 MPa of the 28-day compressive strength for cement-coated RCA based concrete and 27.7 MPa for fly ash-coated RCA based concrete. In our experiment, sample B2 shows value 33.5 MPa; from this point of view, the geopolymer slurry gives worse results than that of cement, and better than that of fly ash. Similarly, in [29] the results of coating the RCA by powdery slag and fly ash has been presented. At 28 days of hardening, mixture based on slag-coated RCA achieved 53 MPa, while, when using standard mixing without coating process, it was only 40 MPa. In the case of fly ash-coated RCA, samples achieved 45 MPa, while when using standard mixing without coating process it was only 35 MPa. The positive difference between results achieved by coated RCA based concrete and concrete prepared by standard mixing was 32% and 29%, respectively. In our experiment, the difference between B2 (coating of RCA during mixing) and B1 (uncoated RCA) is 13.2% for 28-day values, and even 42.0% for 365-day values.

It is also evident that the time development of compressive strength all of samples increases, although the rate of increase is different. The difference between values in 28 and 365 days is given in the Table 4.

Table 4. Change in sample properties between 28 and 365 days.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Decrease in Water Absorption (%)</th>
<th>Increase in Compressive Strength (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>B0</td>
<td>10</td>
<td>42</td>
</tr>
<tr>
<td>B1</td>
<td>7</td>
<td>35</td>
</tr>
<tr>
<td>B2</td>
<td>61</td>
<td>69</td>
</tr>
<tr>
<td>B3</td>
<td>63</td>
<td>67</td>
</tr>
</tbody>
</table>

The change in sample properties between 28 and 365 days, given in Table 4, is expressed for water absorption and compressive strength—the properties directly defining the performance and durability character of concrete. As can be seen, samples B2 and B3 show significant rates of positive change. This confirms the positive effect of using the geopolymer material to coat the RCA, though it is a material that normally acquires positive properties over a long period of time. This is also presented in [42].
When comparing the two ways of coating the RCA, concrete with RCA coated during the mixing (sample B2) achieved somewhat better properties than that of concrete with RCA coated prior to the mixing (B3), in terms of both values and changes in values over time; however, the differences are not very significant.

To support the presented results, the relativity of total water absorption and compressive strength is presented (Figure 5), while all of the results regarding the samples and the testing time are included. The standard relation between these properties (negative linear relationship) is confirmed and the method of RCA application does not affect it; according to [43], \( R^2 = 0.6985 \) represents very strong correlation.

![Figure 5. Relation between the water absorption and the compressive strength of samples.](image)

4. Conclusions

To reduce the negative surface parameters of the RCA, it was modified by two methods of coating using the geopolymer slurry—coating prior to and during the mixing process. As control samples, a mixture prepared by natural aggregate only and a mixture containing RCA, but prepared by standard mixing with no coating process, were tested as well. Results of density, total water absorption, and compressive strength, in periods of 28, 90, 180, and 365 days of curing are presented and evaluated. The following conclusions can be formulated:

- Using the geopolymer slurry to coat the RCA results in higher density of concrete when compared with the same concrete prepared by standard mixing—6% increase in the case of B2 and 1.4% in the case of B3, when evaluating 28-day values.

- Both methods of coating the RCA with geopolymer slurry result in a positive effect on the water absorption of concrete, although it significantly occurs after a longer time. Decrease of water absorption is 3.3% for B2 and 9% for B3 sample when evaluating 28-day values, while it is 46% for B2 and 51% for B3 when evaluating 365-day values.

- Both of the methods of coating the RCA with geopolymer slurry result in a positive effect on the compressive strength of concrete; the early values are only a little lower than that of standard concrete with natural aggregate (13.0% for B2; 15.0% for B3), while the long-term values are fully comparable (even increase for B2 + 3.0%, 0.5% decrease for B3).

- Both methods of coating the RCA with geopolymer slurry allow for the preparation of concrete with properties that are comparable to those of concrete with natural aggregate prepared by standard mixing; thus, it seems to be a promising way to enhance the rate of RCA application,
When comparing the methods of RCA coating, directly applying the coating during the mixing yields somewhat better results, especially from long-term point of view. The density is higher by 4.0% and compressive strength by 3.4%.

Since the coating during mixing is technologically simpler and time-saving, it has a positive impact on total costs of coating process; thus, it seems to be better choice for practical purposes. However, after optimization and understanding all the possibilities, whether in one or the other way of coating, prior preparation of quality coated RCA ready for concrete production could also be a good way.

Acknowledgments: This work was supported by the Grant No. 1/0277/15 of the Slovak Grant Agency for Science and by the project NFP 26220220051 Development of progressive technologies for utilization of selected waste materials in road infrastructure (European Union Structural funds).

Author Contributions: Jozef Junak performed the experiment, analyzed data and wrote the paper; Alena Sicakova conceived and designed experiment, analyzed data and wrote the paper. Both authors read and approved the final manuscript.

Conflicts of Interest: The authors declare no conflict of interest.

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