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Effect of treatment types of recycled concrete aggregates on the properties of concrete

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Abstract: Reusing waste materials such as construction and demolition waste is an environmentally significant task. This article aims to improve the quality of recycled aggregates after the demolition of concrete structures. Two methods, mechanical cleaning and impregnation, were explored to enhance the quality of coarse aggregates. The findings indicate that mechanical cleaning is more effective than soaking in a sodium silicate solution. The compressive strength of concrete made with mechanically cleaned recycled aggregates was 2.3 times greater than that of concrete made with untreated rubble. Concrete with impregnated rubble had a compressive strength 1.1 times greater than that of concrete with untreated aggregates. Infrared spectroscopy was used to study the microstructure, revealing that the type of treatment does not influence the quantity of portlandite and ettringite in hydrated cement. However, the treatment of recycled aggregates alters the interaction in the gel portion of cement hydration products. Concrete with aggregates treated by sodium silicate has more aluminate components. Additionally, there is a shift of the bands assigned to Si-O stretching to higher wave numbers (from 995 cm–1 to 1088 cm–1), which can be attributed to the formation of calcium-silicate-hydrate gel with a lower calcium/silicon ratio.

Keywords: recycled aggregate, concrete, average density, compressive strength, FTIR spectroscopy

1. Introduction

The issue of reusing construction and demolition waste is becoming increasingly relevant due to the importance of sustainable development and environmental protection [1]. According to modern strategies for resource utilisation, there are two main methods for recycling demolished concrete. The first involves crushing it to a size of 10–40 mm to use as coarse aggregate in concrete. The second method involves obtaining fine aggregates smaller than 5 mm to use as fillers in concrete or composite materials. Numerous studies indicate a general trend towards a decrease in the strength of concrete made with recycled aggregates. For example, the results in [2] show that the higher the proportion of recycled rubble, the lower the compressive strength, flexural and tensile strength, elastic modulus, and toughness. Additionally, water absorption increases, while resistance to sulphate corrosion, chloride ion permeability, frost, acid, and carbonation decreases. A looser and more porous microstructure of the interfacial transition zone between old aggregates and new cement mortar leads to a reduction in bonding strength. This is why researchers are seeking ways to either remove the adhered cement layer or enhance it. Removal can be performed using various methods such as mechanical cleaning or thermal treatment. For mechanical removal of adhered cement paste, a concrete mixer drum or rotating drum with steel spheres was used in studies [3, 4]. The use of a mixer drum for 5 hours removes approximately 17% of the old cement mortar [3]. To increase the amount of removed paste, preliminary heating at 300°C, 600°C, and 900°C was carried out in study [4]. In this case, the amount of separated cement mortar reaches 96 wt.% after roasting at 900°C and subsequent mechanical treatment in a rotating drum for 15 minutes. A combination of microwave heating and impact crushing was used to obtain aggregates without cement paste in [5]. It was shown that the degree of aggregate liberation varies from 18% to 69%, depending on microwave power (kW) and exposure time (minutes). The old cement mortar becomes more brittle and weaker after heating, increasing the percentage of its removal. The disadvantages of these methods include the need for preliminary heating or grinding for many hours, which is economically unfeasible. In contrast, the present study examines processing at ambient temperature and grinding for 30 minutes, which distinguishes this study favourably from existing ones.

Even more research focuses on strengthening the adhered cement layer through impregnation or carbonation. Accelerated carbonation allows the residual old cement layer to achieve higher surface hardness under specific humidity conditions, high CO_2 concentration, and pressure. In the study [6], the influence of carbonated recycled concrete aggregate on the deformation behavior of concrete with recycled rubble was investigated. The recycled concrete aggregate was pre-soaked in lime water and then treated with accelerated carbonation for 24 hours at a pressure of 0.30 MPa. Four replacement ratios (0%, 30%, 70%, 100%) of coarse aggregate in the concrete were used. The results showed that as the substitution ratio increased, the tensile curve of concrete with non-carbonated recycled rubble gradually smoothed out, indicating a reduction in brittleness. Furthermore, its maximum stress, elastic modulus, and peak strain consistently decreased, while the ultimate strain consistently increased. In contrast, concrete with carbonated aggregates exhibited a similar tensile curve, but its maximum stress, elastic modulus, and peak strain increased, while the ultimate strain decreased. In the study [7], it was shown that the longer the carbonation time, the more complete the carbonation reaction and the better the properties of the aggregate. The degree of carbonation ranged from 17.65% to 40.6% when the exposure time varied from 0.5 hours to 72 hours. The efficiency of carbonation increases slowly as the CO_2 concentration changes from 40% to 60%. In addition to CO_2 concentration, pressure is also an important factor affecting carbonation efficiency. It was found that CO_2 absorption significantly increases under pressures of 0–0.01 MPa and slightly increases in the range of 0.01–0.5 MPa. The compressive strength of concrete with carbonated aggregates increases by approximately 9.6% compared to concrete with untreated aggregates. Furthermore, the compressive strength of concrete with carbonated aggregates is only about 3.49% lower than that of concrete with natural aggregates. However, the necessity for complex equipment to achieve the required pressure complicates the technological process of preparing aggregates in this case. The simplicity of the method presented here will be an advantage for manufacturers.

Strengthening the residual layer of cement mortar can be achieved through impregnation with various solutions. Treatment with polymer emulsions is considered one of the effective methods to reduce the porosity of recycled aggregate. Polymers such as polyvinyl alcohol, alkylalkoxysilanes (silanes), and polydiorganosiloxanes (siloxanes) can be used for this purpose [8]. The aggregate is soaked in the solution for a certain time, for example, a 5-minute soaking in silane or siloxane. After drying, the aggregates are used to make concrete. When a polymer emulsion is applied to the aggregates, holes and cracks, especially in the adhered cement mortar and interfacial transition zone (ITZ), can be filled with polymer molecules, and the aggregate surfaces can be sealed. Sodium silicate is another method to reduce the porosity of recycled aggregate. As is known, $Ca(OH)_2$ tends to concentrate in the ITZ, where porosity is higher than in the cement matrix. Similar to pozzolans, sodium silicate solution can react with $Ca(OH)_2$ in the ITZ to form a calcium-silicate-hydrate (C-S-H) gel. The C-S-H gel not only fills the pores of the ITZ but also enhances its load-bearing capacity, which, in turn, improves the load-bearing capacity of recycled aggregate concrete. To improve the properties of concrete with recycled aggregates, both single and complex impregnation of recycled coarse aggregates with a 5% sodium silicate solution by mass, an 8% silane suspension, and a 10% polyvinyl alcohol solution were carried out in the study [9]. The results showed that the slump, compressive strength, flexural strength, and splitting tensile strength of recycled aggregate concrete prepared with silane and sodium silicate impregnation increased by 9.8%, 26.53%, 14.72%, and 21.70%, respectively, compared to untreated concrete. Microstructure analysis showed that complex treatment by impregnation with silane and sodium silicate is most effective in filling pores and cracks on the surface of recycled rubble. In this case, the interfacial transition zone of recycled aggregate concrete becomes more compact, and the structure more stable [9]. Nevertheless, the microstructural analysis does not reveal the basis of strengthening the interfacial transition zone. The novelty of the present study is the use of infrared spectroscopy to investigate interfacial interconnections.

Modification of recycled crushed stone can be achieved by adding powder fillers such as silica fume, fly ash, nanosilica, basalt powder, granulated blast furnace slag, Portland cement, and metakaolin. As shown in [10], the 28-day compressive strength of concrete, which includes aggregates modified with silica fume, fly ash, and nanosilica, increased by 55.2%, 39.4%, and 17.6%, respectively, compared to concrete with unmodified rubble. In another study [11], the surface of recycled aggregates was treated with two strengthening mortars prepared with sulphoaluminate cement and the addition of basalt powder. The test results indicated that the quality of the treated surface improved, as evidenced by a reduction in the crushing value (by 23%) and water absorption (by 19%), while the mechanical properties and chloride and sulphate resistance of the concrete improved by 33% and 10%, respectively. As found in [12], the 28-day compressive strength of concrete with aggregates treated with Portland cement, metakaolin, and nanosilica increased by 25%, water absorption decreased by 7%, capillary rise reduced by 53%, and the chloride penetration coefficient decreased by 67%. Accordingly, concrete with recycled aggregates treated with metakaolin and nanosilica exhibits greater durability than concrete with untreated aggregates. However, the use of microfillers requires additional equipment for manufacturing and adjustments to the concrete mixture calculations. The use of conventional concrete manufacturing methods is one of the advantages of the present study.

Summarizing the results of the aforementioned studies, it can be concluded that in all cases, concrete with modified recycled aggregates demonstrates better properties compared to concrete with untreated crushed stone. However, the properties are still inferior to those of concrete with natural aggregates. To understand the reasons for this phenomenon, most researchers investigate the microstructure formation of concrete with different aggregates. For example, in article [13], the influence of fine recycled concrete aggregates on the structure of calcium silicate hydrates was investigated. The authors studied the properties of concrete with fine recycled aggregate and concrete with natural sand using analytical methods such as X-ray diffraction and scanning electron microscopy, as well as micro-indentation and nano-indentation. It was found that concrete with fine recycled aggregate contains a higher proportion of loosely packed, low-density calcium silicate hydrates compared to concrete with natural sand. Additionally, the nanostructure of C-S-H gel changes due to the presence of recycled fine aggregates. Calcium silicate hydrates in concrete with fine recycled aggregate demonstrate a higher C-S-H gel porosity, constituting 56.2% of the total porosity [13]. To enhance the mechanical properties of concrete with fine recycled aggregates, it is crucial to minimize the porosity of the C-S-H gel and maximize the proportion of densely packed, high-density calcium silicate hydrates. The objective of the investigation in study [14] was the utilisation of construction and demolition waste-based materials in the creation of geopolymer binders. Recent studies have shown that geopolymer binders are sustainable and eco-friendly cementitious materials. They are appealing because of their good cost-performance ratio, low-energy production process, and use of secondary raw materials instead of new cement. Geopolymer binders substitute Portland cement and create cement-free concrete, which is why the behaviour of these materials is similar to that of the substances studied. Microstructural analyses have shown that geopolymer pastes have higher compressive strengths and more homogeneous microstructures. The major reaction products of these geopolymer binders were mainly sodium aluminosilicate hydrate gels with zeolite-like structures. Additionally, some calcium aluminosilicate hydrate gels were present due to the use of materials with a high CaO content. Most researchers have observed that concrete with treated recycled aggregates has a denser mortar microstructure compared to standard concrete. The incorporation of such aggregates has a significant effect on the hydration products of building mortars, their amount, and packing density. However, the investigation of possible interactions at the micro level has not received sufficient attention in the literature. This paper aims to fill this gap by identifying possible interactions using infrared spectroscopy.

2. Materials and methods

The destroyed reinforced concrete railway sleepers were used as the material for this research. The parts of the sleepers consisted of continuous aggregates of several grains of crushed stone, connected with cement-sand mortar from the remains of old concrete. After removing the reinforcement, the sleeper parts were first loaded into a jaw crusher with a set distance of 20 mm between the working surfaces. The resulting material was then reloaded into a drum mill and ground for 30 minutes. After that, the mixture was discharged onto a standard set of sieves to determine the particle size distribution according to [15]. Following the crushing process, some of the crushed stone was placed in a sodium silicate solution and soaked for 1 hour, then dried at 40°C for 1 hour. Four series of concrete samples were produced for the study. All concrete mixtures contained 260 kg/m³ of CEM I 42.5R cement with a water-cement (w/c) ratio of 0.4. Fine aggregate fractions of 0-1 mm and coarse aggregate fractions of 5-10 mm and 10-20 mm were used. The sand consumption was 550 kg/m³, and the rubble consumption was 1400 kg/m³. For series 1 samples, untreated recycled coarse aggregates were used; for series 2, recycled coarse aggregates impregnated with sodium silicate were used; for series 3, recycled coarse aggregates after mechanical cleaning in a drum mill were used; and for series 4, natural rubble was used. Ten samples of each series were prepared. After curing under normal conditions for 7 and 28 days, the average density and compressive strength were determined for each series of concrete cube samples according to [16, 17]. For Fourier Transform Infrared Spectroscopy (FTIR), cement mortar samples were taken directly from the aggregate surface by scratching. The samples were dried to a constant weight at a temperature of 70°C and crushed until completely passing through a sieve with a cell size of 0.014 mm. Then, 3 mg of the cement powder was pressed into tablets with the addition of KBr under a pressure of 8–8.5 MPa. The IR spectra were obtained using a Bruker Alpha FTIR spectrometer in the wave number range of 400-4000 cm^{-1} with a spectral resolution of 4 cm^{-1}. The background spectrum was obtained using pure KBr pellets, and the sample spectra were adjusted with a linear baseline.

3. Results and discussion

3.1. Particle size distribution of recycled aggregates

After crushing the sleeper residues, the particles were sieved through a 40 mm sieve, and any particles remaining on the sieve were discarded. The particle size distribution of the recycled coarse and fine aggregate mixture was then determined using sieve analysis (Fig. 1).

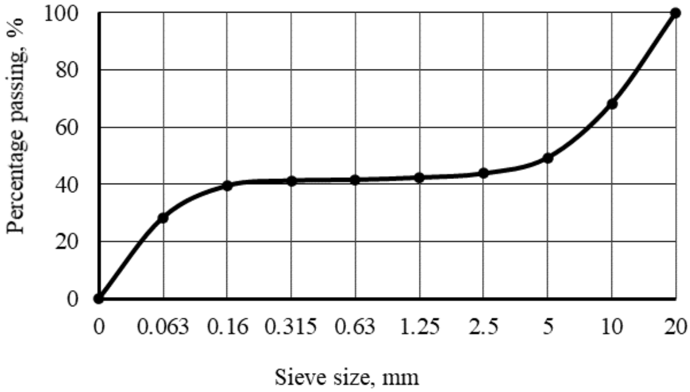

Fig. 1. Particle size distribution. *Source:* own study

As shown, about 50% of the recycled coarse aggregate passed through a 20 mm sieve, and about 50% passed through a 5 mm sieve. Based on Fig. 1, the quality of the fine aggregate does not meet the requirements of regulatory documents for fine aggregates used in concrete production.

3.2. Average density and compressive strength

The results for the determination of average density and compressive strength are shown in Fig. 2. As indicated, all concrete samples demonstrate a consistent trend in the development of average density and compressive strength, with these parameters increasing over the curing time. However, the addition of untreated recycled rubble (Series 1) significantly affects the concrete's compressive strength and average density. The results show that concrete made with untreated recycled coarse aggregate has lower compressive strength (10.4 and 19.2 MPa) compared to the control concrete with natural aggregate (24 and 32.2 MPa) at both test periods. After 7 days of curing, the concrete strength with recycled rubble impregnated with sodium silicate (Series 2) was 10% higher than that of concrete with untreated coarse aggregate. The compressive strength of concrete with mechanically cleaned recycled rubble (Series 3) was 2.3 times greater than that of Series 1 (concrete with untreated recycled crushed stone). After 28 days, the compressive strength of concrete with sodium silicate-treated and mechanically cleaned recycled coarse aggregate was 68% and 98%, respectively, of that of the control concrete with natural aggregate (Series 4).

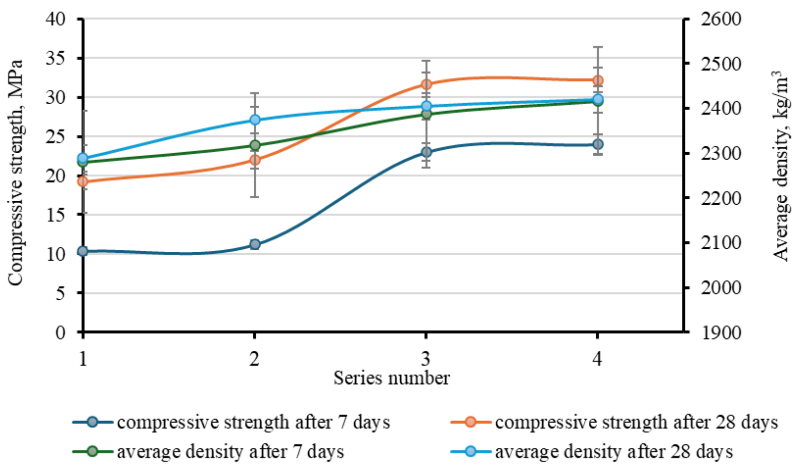

Fig. 2. Average density and compressive strength of the sample series. *Source:* own study

3.3. Analysis by infrared spectroscopy

The IR spectra obtained for all series of samples have a typical shape characteristic of hardened Portland cement (Fig. 3). As indicated in [18], a broad band with a maximum around 3440 cm^{-1} confirms the presence of water bound in the hydrate phases, while a medium-intensity band around 1645 cm^{-1} indicates the presence of capillary water in the samples. The main characteristic peaks of C-S-H are located in the range of 1100–900 cm^{-1}. The SO_4^{2-} ions cause sharp peaks at 1088 and 1100 cm^{-1}, characteristic of calcium monosulfoaluminate (ettringite). A sharp band at 3640 cm^{-1} indicates the presence of $Ca(OH)_2$ [18].

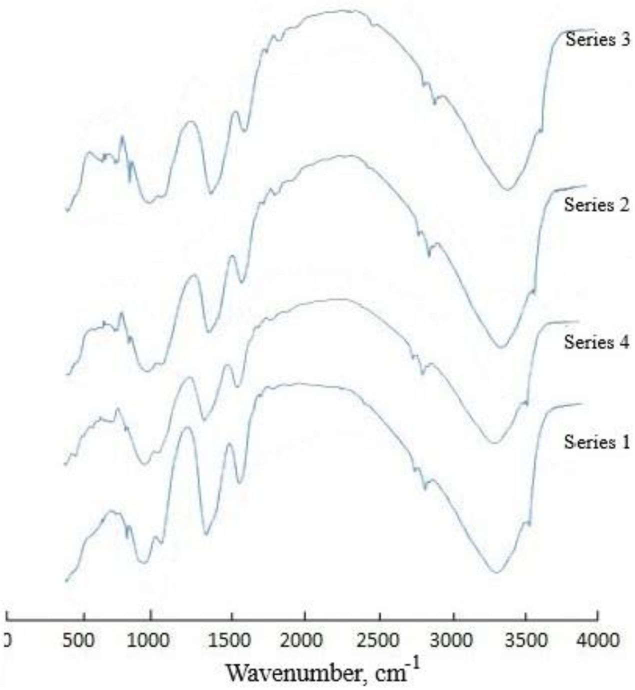

Fig. 3. IR spectra of the sample series. *Source:* own study

The analysis of the spectra (Fig. 3) led to the following conclusions. The type of aggregate treatment does not affect the amount of portlandite and ettringite in hydrated cement, as the placement of the peaks at 3640 cm^{-1} and 1100 cm^{-1} and their intensity are the same for all samples. However, the treatment method of the aggregates significantly influences the formation of the gel phase of hydrated calcium silicates and aluminates. The shape of the band characteristic of Si-O vibrations in the C-S-H phase widens, and its maximum shifts from 995 cm^{-1} for concrete with untreated rubble (Series 1) to 1000 cm^{-1} for concrete with natural and treated aggregates (Series 2, 3, 4). This indicates that the treatment modifies the calcium silicate hydrate structure of the material, affecting the Ca/Si ratio in the C-S-H phase and the degree of polymerization of the silicate links. This effect may result from a change in the concentration of Ca^{2+} and the Ca/Si ratio in the pore solution due to the presence of sodium silicate solution, potentially affecting the hydration products of the cement. The shape of the band characteristic of C-S-H and other silicate phases (900–1200 cm^{-1}) also indicates a different microstructure of the phases forming in concretes with different types of aggregates.

The intense band with a peak around 1420 cm^{-1} and sharp bands at 875 and 712 cm^{-1} indicate the presence of carbonates. These are most pronounced in concrete samples with untreated and mechanically cleaned crushed stone. For concrete with natural and impregnated rubble (Series 2 and 4), the intensity of these peaks is approximately the same. The peaks at 535 cm^{-1} and 650 cm^{-1} suggest the formation of a larger number of carbo-aluminates, sulpho-aluminates, hydrogarnets, and alumino-silicates, which explains the higher strength of concrete with treated aggregates compared to untreated rubble. However, hydrated calcium aluminate, silica aluminate, and portlandite have lower strength due to their lamellar structure compared to the stronger calcium silicate hydrate phase. This could explain the lower strength of concrete with impregnated or cleaned crushed stone compared to concrete with natural aggregates. The greater water retention capacity of porous residues of old cement mortar is confirmed by the larger amount of capillary water in the sample of concrete with untreated recycled aggregate (Series 1, peak at 1650 cm^{-1}).

4. Conclusions

The use of recycled aggregates requires enhancement of their quality. Mechanical cleaning and impregnation were investigated as methods for improving coarse aggregates. The results show that mechanical cleaning is more effective compared to soaking with sodium silicate solution. The compressive strength of concrete with mechanically cleaned recycled rubble was 2.3 times greater than that of concrete with untreated crushed stone. The compressive strength of concrete with impregnated aggregates was 1.1 times greater than that of concrete with untreated crushed stone. The study of microstructure using FTIR spectroscopy shows that the type of treatment does not affect the amount of portlandite and ettringite in hydrated cement. However, the treatment of recycled aggregates alters the interaction in the gel phase of cement hydration products. There are more aluminate components in concrete with aggregates treated by sodium silicate. Additionally, there is a shift of the bands assigned to Si-O stretching to higher wave numbers (from 995 cm^{-1} to 1088 cm^{-1}), which can be explained by the formation of C-S-H gel with a lower calcium/silicon ratio. Consequently, this could lead to higher concrete strength at a later age.

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