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INFLUENCE OF ORGANIC ACID TREATMENT ON RECYCLED CONCRETE AGGREGATE

Abstract: As a result of increasing global environmental awareness and sense of responsibility for future generation lives, utilization of demolition waste as potential aggregate for structural concrete has been proposed. Reducing production and transport costs of natural aggregate, as well as solving the problem of removal and disposal of large quantities of demolished concrete waste, represent significant issues in contemporary civil engineering. The main subject of the study presented in this paper is the analysis of the possibility of recycled concrete aggregate (RCA) treatment using organic acid, in order to improve the properties of recycled aggregate.

Key words: Recycled concrete aggregate, acid treatment, physical and mechanical properties

UTICAJ ORGANSKIH KISELINA NA AGREGAT OD RECIKLIRANOG BETONA

Rezime: Kao rezultat povećane globalne ekološke svesti i odgovornosti za dobrobit budućih generacija, predložena je upotreba građevinskog otpada u vidu potencijalnog agregata za spravljanje betona. Smanjenje troškova proizvodnje i transporta prirodnog agregata, kao i rešavanje problema odlaganja velikih količina betonskog šuta, predstavljaju veoma značajne teme u savremenom građevinarstvu. Osnovni cilj istraživanja koje je prikazano u ovom radu, odnosi se na analizu mogućnosti tretiranja agregata od recikliranog betona (RCA) primenom organskih kiselina, sa namerom da se poboljšaju svojstva recikliranog agregata.

Ključne reči: Agregat od recikliranog betona, tretman kiselinama, fizičko-mehanička svojstva

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1. INTRODUCTION

Recycled concrete aggregate (RCA) differs from natural aggregate as the former contains hardened cement mortar. This cement mortar, adhered on recycled concrete aggregate, has higher porosity and water absorption and lower strength than natural aggregate. It has negative effects on the mechanical properties and durability of fresh and hardened concrete made with recycled concrete aggregate. Therefore, the wider application of recycled concrete aggregate should be enabled if we could enhance the quality of the adhered cement mortar. Removal and strengthening of the adhered mortar are the two main methods for enhancing the properties of recycled concrete aggregate.

Basic methods for removal of adhered mortar are: mechanical grinding (by ball-milling or heating) and pre-soaking in acid. Strengthening of adhered mortar can be done by: polimer emulsion, pozzolanic solution, using lime as a filler, calcium carbonate biodeposition or carbonation [1]. Pre-soaking in acid is a method proposed by many authors, usually consisting of using hydrochloric acid (HCl), sulfuric acid (H_2SO_4) or phosphoric acid (H_3PO_4).

In the research [2] the HCl and sodium sulfate (Na_2SO_4) were used; about 14% higher concrete compressive strength was achieved by using treated RCA in comparison with untreated RCA. In [3] HCl, HNO_3, H_2SO_4 and HCl with silica fume (SF) solutions were used. The mass loss was found to be 1.5%, 2.5%, and 5.6% for HCl, HNO_3 and H_2SO_4 treatment respectively. The crushing value for HCl, HNO_3 and H_2SO_4 treated aggregates was improved by 7%, 3% and 2%, respectively (compared to RCA). Similarly, the impact resistance was improved by 9%, 10% and 7% and the abrasion value by 19%, 24% and 34%, respectively. The higher resistance to crushing, impact and abrasion (19%, 23% and 29% respectively) was observed in HCl and SF impregnated aggregates. Acid treatment was found to effectively improve the quality of recycled fine aggregates as the acid-treated aggregates were characterized by improved density, absorption ratio and solid volume percentage compared to the original aggregates and the recycled aggregates treated with natural water [4]. The substantial quality improvement of RCA as a result of using sulfuric acid solution as the washing fluid is thought to result from the continuous elution of $Ca(OH)_2$ from cement paste, which in turn weakened the strength of cement paste [5]. The treatment process causes a decrease in water absorption and provides more dense and connected cement mortar compositions [6,7]. The RCA was soaked in an acidic solution composed of HCl (37%) and acetic acid ($C_2H_4O_2$) (99.7%) a low concentration of 0.1 M for 24 h at room temperature around 20°C. The acid treatment at low concentration is an effective technique to enhance the quality of RCA depending on the acid type, due to its corrosive influence on the attached mortar. However, it is concluded that using weak acid is more efficient than the strong acid, in order to decrease the influence of acid attacks on the RCA surface [8,9,10]. There is a linear correlation between the amount of mortar loss with the increase of the molarity of acid. The immersion time of RCA with acid did not have significant influence on the amount of mortar loss. A microstructure study by SEM on RCA surface indicates that the surface of treated RCA has cleaner crumb and becomes free of loose particles. However, increasing the acid molarity to 0.8 M in RCA treatment results in more brittle and fragile particles on the mortar surface because the acid tends to have corrosive effects on the adhered mortar [11].

Although removal of adhered mortar causes lower water absorption and higher specific gravity of RCA (as concluded by all authors), the chloride and sulphate contents are typically increased. Also, such processes increase the concrete production cost, and there is a problem of disposal of very aggressive and environmentally dangerous acids. Furthermore, the personnel employed in the process must be specially trained and equipped. In general, the authors are not addressing the potentially hazardous influence of strong acids on RCA and RAC (Recycled aggregate concrete) and the technological aspects are typically disregarded (in situ treatment procedures for large aggregate quantities, problem of used acid disposal, possible multiple application of the same acid solution, etc.).

There is a limited research on organic acid treatment of RCA, although it could lead to overcoming of the above stated disadvantages of strong inorganic acids application. In [12] the RCA is first soaked in acetic acid solution, in which the acetic acid reacts with cement hydration products attached to the surface of the aggregate. This reaction weakens the attached mortar, making it possible to remove it later from the RCA by mechanical rubbing. The treated RCA has lower water absorption and less cement mortar attached. When used as aggregate for the production of new concrete (RAC), this RCA can enhance the 28-day compressive concrete strength up to 25%.

2. EXPERIMENTAL RESEARCH

RCA used in this study was obtained by crushing concrete slabs that previously served as the sub-structure for tram rails in Vojvode Stepe street in Belgrade. During the service life, this concrete was covered with an asphalt layer and thus protected from potentially harmful environmental effects. At the time of removing and recycling, the concrete was more than 30 years old. After taking and testing the core specimens, it was determined that this concrete fulfilled requirements for compressive strength class C35/45, according to EN 206. After that, the concrete was crushed to a maximum aggregate size of 22.4 mm. The resulting RCA consisted of 98% of old concrete, 1.2% of asphalt, and 0.8% of brick particles [13].

Experimental research obtained in the laboratory on the RCA specimens was carried out to examine the reduction of the old (residual) cement mortar content. Before the research, aggregate was dried to a constant mass, and then sieved, i.e. separated in fractions 0/4, 4/8, 8/16 and 16/22.4mm. Only two coarse aggregate fractions (8/16 and 16/22.4) were the subject of further research, considering that fine RCA is less often used for making new concrete.

In the first part of the research on aggregate grains size 16/22.4, the degree of removal of residual cement mortar was determined, after these grains had been exposed to the treatment of immersion into the solution of acetic and citric acid at a concentration of 0.1 mol/dm, in the period of 1, 5, 7 and 9 days. Microscopic images of treated grains show that in the same period, at the same concentrations of acid solutions, citric acid in a higher degree manages to degrade the residual mortar. Therefore, the residual cement mortar falls off to a greater extent and in this way physical and mechanical properties of RCA are improved. Also, images show that there are no significant differences in the surface structure of the aggregate grains treated 5, 7 and 9 days compared to the grains soaked in acid solution for 24 hours.

In the second part of the research, the required quantity of the recycled aggregate size 8/16mm and 16/22.4mm was soaked. On these RCA fractions, after they had been treated with acid, oven dried particle density and water absorption were tested, then particle size distribution by dry sieving was determined, as well as frost resistance.

Change of citric acid solution PH value in time were also tested (Table 1). The research has shown that significantly more cement material removes from RCA grains during the first 24 hours than in the next few days. Also, from an economic point of view, it is more cost-effective for the treatment of aggregates to be as short as possible, bearing in mind the occupancy of premises in plants in which such treatment would be carried out, as well as the possibility of repeated use of citric acid, considering the small change in its PH value after 1, 5, 7 and 9 days.

Table 1 – Change of citric acid solution pH value in time

Time, t (days)	pH value
0	2.30
1	2.39
5	2.50
7	2.52
9	2.53

As already stated, in the second part of the research, the most important physical properties of RCA treated in citric acid were determined and then these properties were compared to the characteristics of the untreated aggregate. Table 2 shows the obtained results of these tests (for fraction 16/22.4).

Table 2 – Physical properties of untreated (A) and treated (A_T) RCA

Parameter	A	A_T
ρ_a (kg/m^3)	2603	2626
ρ_{rd} (kg/m3)	2433	2440
ρ_{ssd} (kg/m3)	2498	2511
WA_{24} (%)	3.30	3.08
Δm_l (%)	-	1,25

where:

ρ_a - apparent particle density

ρ_{rd} – oven-dried particle density

ρ_{ssd} – saturated and surface dried particle density

WA_{24} - water absorption of the aggregate after one-day immersion

Δm_l - aggregate mass loss after treatment with acid

Concerning RCA physical properties, obtained results show that, after the citric acid treatment, water absorption was reduced by about 7% (from 3.30% to 3.08%). At the same time, there was a slight increase in density compared to untreated aggregate: e.g. p increased by 0.30%. Thereby, the treated aggregate mass loss was 1.25%.

On a specimen of RCA fraction size 8/16mm, before and after treatment with citric acid, it was determined particle size distribution in accordance with the standard EN 933-1 (Figure 1)

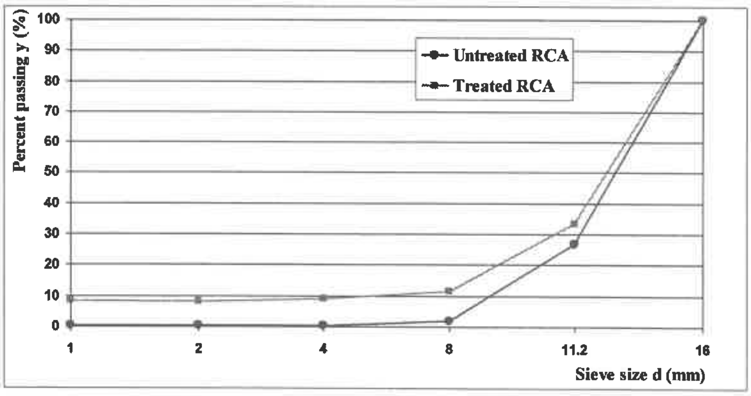

Figure 1 - Sieve analysis of aggregates

Particle size distribution of a test specimen after treatment with citric acid shows that there is no significant change in the amount of grain larger than 4mm, while the content of fine particles is significantly increased. This suggests that there has been a decomposition and a fall of one part of the cement mortar.

RCA frost resistance was determined by testing two aggregate specimens (fraction 10/14) using magnesium sulfate (SRPS EN 1367-2). Recycled aggregate specimens were marked with labels RCA-1 and RCA-2, and the specimens treated with citric acid for 24 hours, with labels RCA-T1 and RCA-T2. Aggregate categorization was done in accordance with the standards EN 12620 and EN 13043.Test results of this research are shown in Table 3.

Table 3 - Frost resistance testing results

Aggregate type	m_1 (g)	m_2 (g)	MS (%)	Average MS (%)	Category
RCA-1	422.58	377.93	10.6	10.0	MS_{18}
RCA-2	422.24	382.37	9.4		
RCA-T1	421.65	413.36	2.0	2.2	MS_{18}
RCA-T2	421.53	411.72	2.3		

where:

MS – aggregate frost resistance after 5 treatment cycles (mass %)

m_1 – initial specimen mass (g)

m_2 – final mass of the aggregate remaining on the sieve size

As shown in Table 3, frost resistance results of RCA treated with citric acid for 24 hours, are significantly better (for about 80%) than the results of the untreated RCA.

3. CONCLUSIONS

To remove the residual cement mortar from RCA, most researchers primarily choose strong inorganic acids, such as HCl and H_2SO_4, resulting in an increased levels of harmful salts (chlorides and sulphates) in the RCA, which is not acceptable concerning the durability of concrete and reinforced concrete structures.Also, there is a problem of disposal of the used acid as well as the need for specially trained and equipped personnel. Use of weak organic acids (such as citric or acetic acid) eliminates the aforementioned problems to a great extent. The research described in this paper shows that citric acid is more effective than acetic acid, i.e. for the same concentration of acid solutions, citric acid in a higher degree succeeds in degrading the residual cement mortar. By varying the duration of aggregate treatment in the solution of these acids it has been shown that this parameter has no significant effect on the quality of the RCA, i.e. that after 24 hours satisfactory results were obtained. Considering the small change in PH value of citric acid solution after 1, 5, 7 and 9 days it has been concluded that this solution can be repeatedly used.

Based on all of the above, it can be concluded that RCA obtained by crushing concrete of relatively high quality (class of strength C35/45) is generally not necessary to be treated with organic acids in order to improve its physical and mechanical properties. However, treatment of RCA obtained from concrete of somewhat lower quality (e.g. class C25/30), which is most often used in practice, would certainly significantly improve its physical and mechanical properties, compared to the untreated RCA.

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