Effect of Rubber Treatment on Compressive Strength and Modulus of Elasticity of Self-Compacting Rubberized Concrete

I. Miličević, M. Hadzima Nyarko, R. Bušić, J. Simonović Radosavljević, M. Prokopijević, K. Vojisavljević

Abstract-This paper investigates the effects of different treatment methods of rubber aggregates for self-compacting concrete (SCC) on compressive strength and modulus of elasticity. SCC mixtures with 10% replacement of fine aggregate with crumb rubber by total aggregate volume and with different aggregate treatment methods were investigated. The rubber aggregate was treated in three different methods: dry process, water-soaking, and NaOH treatment plus water soaking. Properties of SCC in a fresh and hardened state were tested and evaluated. Scanning electron microscope (SEM) analysis of three different SCC patches were made and discussed. It was observed that applying the proposed NaOH plus water soaking method resulted in the improvement of fresh and hardened concrete properties. It resulted in a more uniform distribution of rubber particles in the cement matrix, a better bond between rubber particles and the cement matrix, and higher compressive strength of SCC rubberized concrete.

Keywords—Compressive strength, modulus of elasticity, NaOH treatment, rubber aggregate, self-compacting rubberized concrete, scanning electron microscope analysis.

I. INTRODUCTION

TODAY, SCC is implemented in all types of buildings and all types of elements due to its many advantages over ordinary concrete. It reduces construction time and noise at the construction site, no compaction is required, it is easy to build in, and it has a satisfactory early strength, which makes it economical. On the other hand, the number of waste car tires in the world is growing, creating a serious environmental problem because the decomposition of waste tires takes a very long time, even longer than half a century. The negative impact of waste rubber on the environment, due to its nondegradability, could be partially mitigated by its recycling. One way to recycle waste rubber is to add it to concrete in the form of rubber chips or crumb rubber.

Waste tire rubber can adversely affect the mechanical properties (compressive and flexural strength) of SCC and its workability due to inadequate connections between the cement paste and rubber. The main reason for this is that the cement paste is hydrophilic, while the rubber surface is hydrophobic. Therefore, surface treatment methods that could improve the adhesion between cement paste and crushed rubber have recently been investigated.

The influence of rubber treatment as a concrete or mortar admixture on the compressive strength and/or other properties has been experimentally investigated by various authors. In recent years, the surface treatment process has been applied to improve the adhesion between the cement paste and the crushed rubber.

Authors [1] tested the compressive strength of mortar with treated and untreated rubber in a volume fraction of 12%. The rubber was treated with NaOH saturated aqueous solution. The tests showed that the compressive strength decreased by 40% and 47% compared to the control sample. They conclude that the use of treated rubber in mortars did not present any significant improvement of the tested properties.

Authors [2] also treated rubber as a concrete admixture to evaluate the mechanical properties of concrete. The rubber was also treated with NaOH. The results of their tests showed that the mechanical properties improved; however, the improvement in the bond between the cement paste and the rubber pieces was barely noticeable.

Authors [3] treated recycled rubber with NaOH and added silicate dust to the concrete mixture in order to improve the mechanical properties of the rubberized concrete. They tested the 28-days compressive strength of three types of concrete mixtures (reference, with rubber, and with modified (treated) rubber). Based on the results of three samples for each concrete mixture type, the 28-days compressive strength of concrete with conventional rubber reduced by 67% on average compared with the reference concrete, while the 28-days compressive strength of concrete with the reference with the modified (treated) rubber reduced by 61% on average compared with the reference concrete, while the 28-days compressive strength of concrete with the modified (treated) rubber reduced by only 14%.

Another treatment, a so-called pre-coating process in which the rubber is coated with limestone powder was proposed by [4]. By evaluating the mechanical properties and durability of concrete, these authors found not only a slight increase in strength but also an improvement in the bond between cement paste and crushed rubber.

In order to determine the compressive strength and energy absorption capacity, a surface treatment method in which crushed rubber was coated with a chemically active agent and further treated with a silane coupling agent was implemented in [5]. The test results showed that higher values (between 10% and 20%) of compressive strength of concrete with coated crushed rubber were obtained compared with the control mixture.

Authors [6] performed surface treatment in two stages. In

I. M., M. H. N. and R. B. are with the Faculty of Civil Engineering and Architecture Osijek, Josip Juraj Strossmayer University of Osijek, Osijek, Croatia (phone: +385912240757; e-mail: ivana.milicevic@gfos.hr).

J. S. R., M. P. and D.D. are with the Institute for Multidisciplinary Research, University of Belgrade, Serbia (e-mail: jasna@imsi.rs).

the first, the rubber particles were treated with a silane coupling agent, and in the second the silane-treated particles were further coated with a layer of cement. The two-staged surface treatment was more effective than the silane coupling agent only. After the two-staged surface treatment, the compressive strength of the modified rubberized concrete was higher than the strength of the control/reference mixture, i.e., could be increased by up to 110%.

Authors [7] modified rubberized concrete using a combination coupling agent and carboxylated styrenebutadiene rubber latex to develop chemical bonds between rubbers and cement paste. The compressive strength and flexural strength of concrete with treated rubber were improved by 4%, i.e., 13% compared with the reference mixture. Moreover, the surface modification of rubber improved both the interfacial adhesive behavior of rubber to cement hydrates as well as the microstructure of concrete.

Authors [8] proposed a surface treatment in which the crushed rubber was treated by an oxidation and sulfonation process to allow groups of strong polarity to appear on the rubber surface. With this treatment, higher values of compressive strength of concrete with treated rubber were achieved compared to the control mixture with untreated rubber. Moreover, the adhesion between cement and rubber was improved.

Authors [9] provided research in order to improve the adhesion between crumb rubber and cement mortar by surface modification using organoclay composites. Obtained results showed that the bond between crumb rubber particles and cement matrix materials was improved due to the existence of hydrophilic groups on the crumb rubber surface.

II. MATERIAL CHARACTERISTICS AND PREPARATION

A. Materials

Considering disadvantages associated with the level of the aggregate replacement with the rubber, recycled tire rubber can be surface modified through chemical or physical processes to improve the interfacial transition zone and enhance the bond between the rubber and cementitious matrix.

Overall, the rubber-cement matrix bond can be enhanced by removing impurities, additives, and organic materials from the surface of the rubber aggregate by water-soaking, washing, filtering, and air drying rubber aggregates, and by the chemical treatment for surface modification, e.g., soaking in solutions like NaOH.

The rubber aggregate in this research was treated in three different methods: dry process (D), water-soaking (W), and NaOH treatment plus water soaking (T). The rubber was mixed with water for about 5 minutes and after 24 hours, water was drained, and rubber aggregates were dried at room temperature. After soaking rubber aggregates in 1 N NaOH for 20 min and washing with water, rubber aggregates were left in water for 24 h and after that dried at room temperature.

CEM I 42.5R type Portland cement was used in the study, from a cement factory in Našice, Croatia, which conforms to EN 197-1:2012 standard [10]. The density of cement was 3.17 g/cm³. Tap water from the local water supply that complies with HRN EN 1008 standard was used [11].

Chemical admixtures, superplasticizer Sika® Viscocrete® 20 Gold and viscosity modifying admixture Rheomatrix® 100 were used to achieve desirable SCC properties and flowability and viscosity classes.

Dolomite powder from a local quarry with a density of 2.97 was used as filler with 66 kg per m³. Dolomite aggregate fractions of 0-4 mm, 4-8 mm, and 8-16 mm and sand were used for the coarse and fine aggregates. In all mixtures, 10% of fine crumb rubber was used as a replacement for fine aggregate. SCC compositions are shown in Table I.

TABLE I									
SCC MIX DESIGN AND TREATEMENT									
Mixture	Cement W/C		SP	VMA	FA and CA	CR	NaOH	WC	
	(kg)	w/C	(%)	(%)	(kg)	(kg)	NaOH	w 5	
SCC-D	450	0.4	1.3	0.2	1578	66	-	-	
SCC-W	450	0.4	1.3	0.2	1578	66	-	WS	
SCC-T	450	0.4	1.3	0.2	1578	66	1N	WS	

B. Mixtures Design and Methodology

A total of three self-compacting mixtures, with 10% of TP as a replacement for fine aggregate, were tested in a fresh and hardened state. SCC mixtures compositions and treatment are shown in Table I.

Tests on fresh and hardened SCC were performed according to relevant European Standards. The slump flow test was measured according to HRN EN 12350-8 [12]. Flowability and viscosity of SCC were measured and classified through the slump flow test. Specimens were demolded 24 h after the casting and placed in a water tank for 4 weeks.

The mechanical properties tests were carried out after the specimens had been moist-cured for 28 days, and the next 28 days specimens were cast in room temperature $(22 \pm 2 \text{ °C})$ and relative humidity of 60%. Two mechanical properties were tested, compressive strength test on cylinders 150×300 mm, and modulus of elasticity test on cylinders 150×300 mm. Compressive strength was measured according to HRN EN 12390-3 [13] and modulus of elasticity was measured according to HRN EN 12390-13 [14].

Microstructures of the round cut rubberized cement samples with plane-parallel surfaces were analyzed using a SEM (Tescan VEGA TS 5130MM). Prior to investigation, the samples were sputter-coated with a thin layer of gold and micrographs were acquired in a backscattered electron mode at an accelerating voltage of 20 kV to identify regions of interest based on the difference in composition, i.e., atomic number over a sample (rubber-cement regions).

III. RESULTS OF EXPERIMENTAL TESTING

A. Properties of SCC in the Fresh State

Results of testing the fresh SCC are given in Table II. From the results, it can be seen that the NaOH treatment plus water soaking causing a reduction in flowability and an increase in viscosity. Water and dry treatment mixtures behave similarly, and they are better in comparison with SCC-T. Despite the presented results, slump flow values of all SCC mixtures are in accordance with EFNARC Guidelines [15].

TADLEII

TEST RESULTS OF FRESH SCC PROPERTIES								
	Viscosity				Flowability			
Minter	T500			Slump Flow				
witxture -	(s)	Class		d (mm)	Class	;		
SCC-D	1.9	<2	VS1	750	660-750	SF2		
SCC-W	1.9	<2	VS1	770	760-850	SF3		
SCC-T	2.4	>2	VS2	730	660-750	SF2		

B. Properties of SCC in the Hardened State

The test results of the SCC properties in the hardened state are given in Table III and Figs. 1-3. The unit weight of SCC cylinders and prisms was measured after 28 days. The results given in Table III show that the dry unit weight decreases within SCC treated with water and NaOH plus water. The differences between the dry unit weight of the mixtures are between 1.6% and 2.3% and it can be considered as negligible.

TABLE III Test Results of Hardened SCC Properties

TEST RESOLTS OF TRADERED SECTROFERTIES								
Mix	Dry Unit Weight (kg/m ³)	$f_{ck,cyl}\left(MPa\right)$			E (GPa)			
	Mean	Mean	st dev.	CV %	Mean	st dev.	CV %	
SCC-D	2250.4	34.8	0.2	3%	32.92	0.3	11%	
SCC-W	2214.3	35.2	0.1	1%	33.13	0.3	11%	
SCC-T	2199.5	37.9	0.2	3%	34.45	0.4	14%	

Generally, the use of tire rubber also increases the voids on the mixes and affects the mechanical properties. As expected, the mean value of three measurements of 28-day compressive strength was the lowest, i.e., 34.8 MPa, when the dry treatment was used. The negative impact of dry treatment on the 28-day compressive strength can be described with a poor rubber granule–cement paste bond and with a low rubber modulus of elasticity compared to the natural aggregates. The bondage of cement matrix with recycled rubber aggregates is weaker than it develops with normal aggregates, because of the inhibition in hydration of cement. Methods like watersoaking and NaOH plus water treatment (see Table III) are employed to enhance compressive strength. NaOH plus water treatment improves the compressive strength by up to 8% in our case.

Values of the 28-day modulus of elasticity were in a direct link with the compressive strength values. The lower results of modulus of elasticity indicate a higher capability to absorb strain after tire rubber addition with previous water-soaking and NaOH plus water treatment.

Figs. 1-3 display the SEM rubber-cement matrix surface morphology, in which a significant effect of the different treatment on the roughness of the rubber surface can be observed. Fig. 3 shows that the rubber surface with NaOH plus water treatment is rougher, with no gaps between rubber and cement matrix, and these attributes lead to a better rubbercement matrix interface while Figs. 1 and 2 revealed that the rubber crumbs treated without treatment (dry) or with watersoaking had a smoother surface with larger cracks and a weak rubber-cement matrix interface. A smooth surface inhibits the no cement paste coating on the rubber, and consequently the bonding is weaker, leading to lower compressive strength and modulus of elasticity. A smoother surface typically has a higher contact angle with poor wettability properties; therefore, the rubber cannot be well wetted and did not have a sufficient physical bonding system to strengthen the adhesion. As a consequence, the rubber will experience poor interfacial adhesion resulting in lower mechanical properties.



Fig. 1 SEM of SCC-D surface morphology



Fig. 2 SEM of SCC-W surface morphology





IV. CONCLUSION

The rubber aggregate was treated in three different methods: dry process, water-soaking, and NaOH treatment plus water soaking, to provide results of their influence to adhesion between the cement matrix and the rubber in SCC composites. The treatment aims to modify the rubber surface by mechanically etching the surface and to provide the rubber with a rougher surface, and remove a passive hydrophilic layer which prevents a good cement matrix adhesion to the rubber. Properties of fresh SCC, compressive strength, modulus of elasticity, and a microscopic surface texture study were analyzed to evaluate the effects of the treatments.

The results show that for all the tests, the treatment of the tire rubber surface with NaOH and water does not present significant change in self-compacting rubberized mixes. Although the flow test showed that the use of tire rubber decreases the mix workability, the difference between the specimens with different rubber treatment was not big. The lower results of modulus of elasticity indicate a higher capability to absorb strain after tire rubber addition with previous water-soaking and NaOH plus water treatment. The decrease of the mechanical strength of the specimens with the residue is attributed to the tire rubber capability to support fewer loads than the natural aggregate and also to the lack of adherence between the cement paste and the tire rubber.

Although this research presents a better performance of SCC after the rubber treatment with NaOH aqueous solution, the use of treated rubber in SCC does not present significant improvement of the studied properties, and this should be additionally investigated on larger numbers of mixes and with other different treatment methods.

ACKNOWLEDGMENT

This paper was supported by Croatian Science Foundation under project UIP-2017–05-7113 Development of Reinforced Concrete Elements and Systems with Waste Tire Powder and Bilateral scientific research project entitled "Microstructural and mechanical characteristics of concrete with recycled materials" 2019 – 2021.

REFERENCES

- Marques, A.C., Akasaki, J.L., Trigo, A.P.M., Marques, M.L.: Influence of the surface treatment of tire rubber residues added in mortars, The Ibracon Structures and Materials Journal, 1 (2008) 2, pp. 113-120.
- [2] Mohammadi, I., Khabbaz, H., Vessalas, K.: Enhancing mechanical performance of rubberised concrete pavements with sodium hydroxide treatment, Material and Structures, 49 (2016) 3, pp. 813-827.
- [3] Onuaguluchi, O., Panesar, D.K.: Hardened properties of concrete mixtures containing pre-coated crumb rubber and silica fume, Journal of Cleaner Production, 82 (2014), pp. 125-131.
- [4] Dong, Q.; Huang, N.; Shu, X. Rubber modified concrete improved by chemically active coating and silane coupling agent, Construction and Building Materials, 48 (2013), pp. 116-123.
- [5] Pelisser, F., Zavarise, N., Longo, T.A., Bernardin, A.M.: Concrete made with recycled tire rubber: effect of alkaline activation and silica fume addition, Journal of Cleaner Production, 19 (2011), pp. 757-763.
- [6] Li, G., Wang, Z., Leung, C.K.Y., Tang, S., Pan, J., Huang, W., Chen, E.: Properties of rubberized concrete modified by using silane coupling agent and carboxylated sbr., Journal of Cleaner Production, 112 (2016), pp. 797-807.
- [7] Huang, B., Shu, X., Cao, J.: A two-staged surface treatment to improve properties of rubber modified cement composites. Construction and Building Materials, 40 (2013), pp. 270-274, https://doi.org/10.1016/j.conbuildmat.2012.11.014
- [8] He, L.; Ma, Y.; Liu, Q.; Mu, Y.: Surface modification of crumb rubber and its influence on the mechanical properties of rubber-cement concrete, Construction and Building Materials, 120 (2016), pp. 403-407
- [9] Nagarajan, C., Shanumugasundaram, P., Anmeeganathan, S.R. (2019). Properties of high strength concrete containing surface-modified crumb rubber, GRAĐEVINAR, 71 (7), 579-588, https://doi.org/10.14256/JCE.2312.2018
- [10] European Committee for Standardization (CEN), EN 197-1:2012 Cement—Part 1: Composition, Specifications and Conformity Criteria for Common Cements, Brussels, Belgium, CEN, 2012.
- [11] European Committee for Standardization (CEN), EN 1008:2002 Mixing Water for Concrete—Specification for Sampling, Testing and Assessing the Suitability of Water, Including Water Recovered from Processes in the Concrete Industry, as Mixing Water for Concrete, Brussels, Belgium, CEN, 2002.
- [12] European Committee for Standardization (CEN), EN 12350-8:2010: Testing Fresh Concrete—Part 8: Self-Compacting Concrete—Slump Flow Test, Brussels, Belgium, CEN, 2010.
- [13] European Committee for Standardization (CEN), EN 12390-3:2009: Testing Hardened Concrete—Part 3: Compressive Strength of Test Specimens, Brussels, Belgium, CEN, 2009.
- [14] European Committee for Standardization (CEN), EN 12390-13:2013: Testing Hardened Concrete—Part 13: Determination of Secant Modulus of Elasticity in Compression, Brussels, Belgium, CEN, 2013.
- [15] The European Guidelines for Self-Compacting Concrete. Available online:
 - https://www.theconcreteinitiative.eu/images/ECP_Documents/European GuidelinesSelfCompactingConcrete.pdf (accessed on 5 September 2020).