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in cooperation with

CENTRE OF EXCELLENCE FOR INTEGRATIVE BIOETHICS

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POSSIBLE USE OF GROUND SULFUR AS PETROLEUM INDUSTRY BYPRODUCT IN SELF COMPACTING CONCRETE PRODUCTION

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ABSTRACT

The quantities of component materials used for production of concrete are high on the global scale, therefore alternative materials are experimented on, in order to replace the natural ones. The possibility of using ground waste sulfur from the oil refining (petroleum) industry as a filling component of self-compacting concrete is presented in the paper, based on common concrete laboratory tests. The physical and mechanical properties of self-compacting concrete decrease to some extend due to use of ground sulfur, but this concrete remains useful for some building applications.

***Keywords**: filler, self-compacting concrete, sulfur, fresh and hardened properties*

1. INTRODUCTION

Being a material produced in high quantities, concrete presents "number one" building material in the world. For the production of concrete huge quantities of water, aggregate and cement are used, resulting in a rising environmental concern. Therefore, the principles of sustainable development are being implemented in the building industry, presenting ever new aspects regarding materials and technologies.

One of the revolutionary changes in the field of concrete is Self-Compacting Concrete (SCC) [1]. This type of concrete is defined as a self-placeable concrete, which doesn't require any means of vibration (otherwise a must, for usual concrete types). It flows and fills the formwork by its own weight, under the influence of gravity, wrapping the re-bars, even densely spaced ones, and filling even the narrowest spaces in the formwork.

For production of SCC, in most cases, powder type material is needed, that, besides cement, provides resistance to the internal segregation of concrete. There are two types of such powder type materials, active (such as silica fume, fly ash, slag) and inert (limestone and dolomite powder, ground concrete and such).

This work presents a study of potential use of ground (pulverized) sulfur as filler for the production of SCC. Elemental sulfur is a major byproduct of metallurgy and oil refining industry, and its annual production was in 2015 estimated to 69.5 million tons [2] (figure 1).

Figure 1. Sulfur as a byproduct of petroleum industry

2. MATERIALS

Natural river aggregate originated from the river Danube was separated and washed, and used in production of SCC (figure 2). Self-Compacting Concrete is usually produced with the largest grain size restricted to less than 22.5 mm. Therefore, the fractions used for the synthesis of SCC included three standard fractions: I (0/4), II (4/8) and III (8/16).

Figure 2. River aggregate for production of concrete

Particle size distribution of used aggregate, tested according to the standard procedure [3], is shown in table 1, together with the particle size distribution of the mixture of aggregates used for this study.

Table 1. Particle size distribution shown as percentage passes in different sieves

	Sieve diameter	0.125	0.25	0.5	1	2	4	8	16	31.5	63
Pass (%)	I (0/4)	2.0	10.5	66.6	76.9	87.1	98.9	100	100	100	100
	II (4/8)	0	0	0	3.1	8.0	13.3	99.6	100	100	100
	III (8/16)	0	0	0	0	0	0.2	8.8	96.9	100	100
	Mixture	1	4	33	39	44	57	76	100	100	100

River aggregate can be successfully used for the production of Self-Compacting Concrete, as for the production of all of the commonly used concrete types. Nevertheless, the composition of the mixture of aggregate for the production of SCC can be tricky, in order to avoid segregation and provide proper workability and later strength.

Fines can present a particular problem when it comes to the production of SCC and therefore their content has to be determined for every fraction of aggregate used [4]. Also, the content of fines has to be periodically checked during the production of concrete, in order to notice any possible variation with time. The highest content of fines was recorded for the fine aggregate fraction I (0/4) and it amounted to less than 1.68 %.

Aggregate fractions proportions play an important role in the strength bearing system in the hardened concrete. In this work, mass ratio of fractions II and III was determined through the laboratory test that included the highest value of density for the mixture of these fractions in dry compacted state and was adopted to be 1:1. Content of fraction I in aggregate mixture was determined based on the pilot mixtures and it was 31.8 % of SCC total volume.

Portland cement *CEM I 42.5R, Lafarge, Serbia* was used in laboratory investigations. Density of cement was 890 kg/m^3 in loose, and 1440 kg/m^3 in compacted state, while gravity of cement was 3.04. Specific area of cement, measured by Blaine method, was 4240 cm^2/g. Limestone powder (*Granit Peščar, Ljig*) was used as filler. This material is characterized as carbonate filler with medium grain size of 250 μm. Density of the limestone powder was 780 kg/m^3, and gravity 2.72. Specific surface area of 3800 cm^2/g was measured by Blaine method.

Sulfur was obtained from the oil refining process by Claus`s procedure in the Oil Refinery Pančevo, and its purity was 99.9 %. Gravity of used sulfur was 2.05. Before the use in SCC, sulfur was ground to the size of particles below 0.09 mm with the aid of disintegrating mill (figure 3). The mineral filler obtained in such way had specific surface area measured by Blaine 2600 cm^2/g.

Glenium Sky 690 (*BASF Construction Chemicals, Italy*) was used as superplasticizer. This superplasticizer is a poly-carboxylate based, with gravity of 1.06, pH of 5 - 8, and with low amount of chlorides (chlorite ion content 0.01 %).

Figure 3. Disintegrating mill used for grounding of sulfur

3. MIXTURES AND METHODS

Five series of concrete were made, with 380 kg/m^3 cement and 220 kg/m^3 of filler. Fillers used in this study were limestone powder, and ground sulfur. Ground sulfur replaced the same mass amounts of limestone powder, i.e. in contents of 0 % (SCC1), 2 % (SCC2), 5 % (SCC3), 10 % (SCC4) and 20 % (SCC5). Water to cement ratio was constantly 0.482.

Fresh concrete mixtures were tested according to the standard methods for SCC, including slump-flow time and diameter measurement [5], V-funnel time [6], L-box test [7], and segregation ratio [8]. Entrained air content was measured according to [9], and fresh concrete density according to [10].

Results of investigations on hardened SCC with ground sulfur included compressive [11], flexural [12], and ultrasonic pulse velocity [13] measurements.

4. RESULTS AND DISCUSSION

4.1. Properties of fresh SCC

Based on the slump-flow test, flowability of SCC with added sulfur was higher than in referent SCC1 series, which was made without sulfur. It can be concluded that, with the slump-flow values ranging from 761 mm to 820 mm, all of the SCC series belong to the category of SF3 according to the relevant provisions [14]. Also, when it comes to the time of slump-flow up to the diameter of 500 mm, all series can be categorized as VS2/VF2 with t500 > 2 s and tv > 9 s (except in case of SCC2 and SCC3). Since differences among t500 values of all series are less than 1 s it turned out that content of ground sulfur has no significant influence on this property. According to the L-box test, all concretes achieved class PL2

(H2/H1 > 0.8). Segregation resistance of the SCC mixtures with ground sulfur turned out to decrease when compared to the reference SCC1.

The value of density of SCC decreases with the increase of ground sulfur content. This decline can be explained by lower density of ground sulfur in comparison to the substituted limestone powder. This decline is quite small and reaches 0.9 % for replacement of limestone powder with the ground sulfur up to 20 %.

As for the content of entrained air in these SCC mixtures, there was no significant change with the incorporation of ground sulfur. Entrained air contents ranged between 1.2 % and 1.9 %. Either way, the values of entrained air recorded on these SCC series were lower than expected for SCC generally, 2 % being the usual value of entrained air content for most SCC mixtures.

4.2. Properties of hardened SCC

Compressive strength was tested on three samples (cubes of 10 cm edge length) for each SCC series, at the ages of 3, 7, 14, and 28 days, while the average values are shown in the Figure 4. Results ranged from 54.6 MPa (for SCC5) to 62.2 MPa (for SCC1 as reference). All series achieved 75 – 85 % of final compressive strength (at the 28 days of age) already at the age of 3 days. Velocities of compressive strength increase for all tested SCC were similar, as it can be observed on the same figure (figure 4).

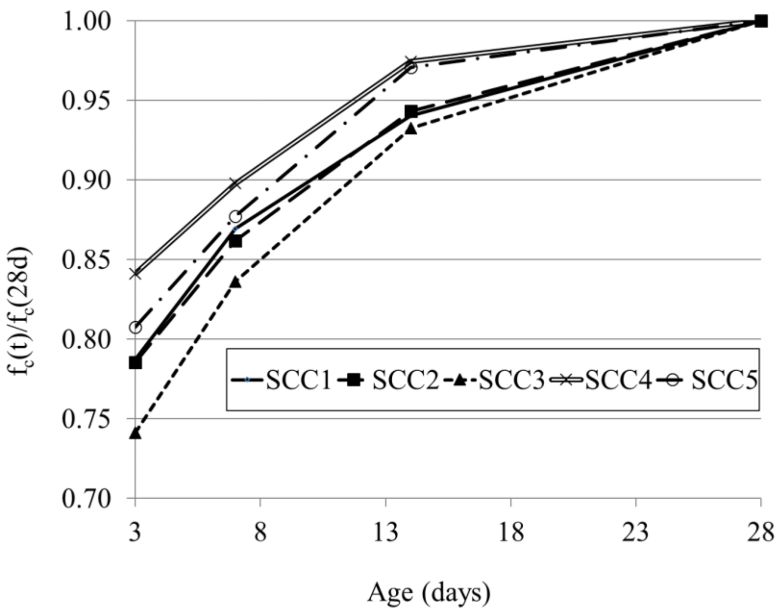

Figure 4. Compressive strength increase for all series

Flexural strength values were in the range of 6.9 (SCC5) to 10.5 MPa (SCC1), see figure 5. Bond strength values were within the limits of 5.8 MPa (SCC1) and 6.3 MPa (SCC4) for the Ø50 dolly, thus these concretes own sufficient bond strength for any structural application.

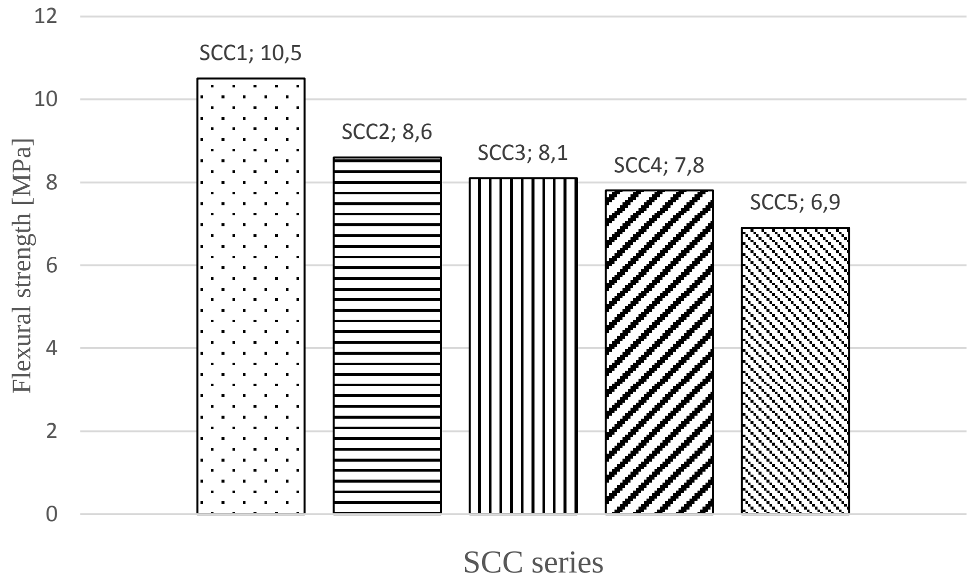

Figure 5. Flexural strength values

Overall analysis shows that ultrasonic pulse velocity is decreasing with the increase of sulfur content in the SCC while it is increasing with the age, see figure 6.

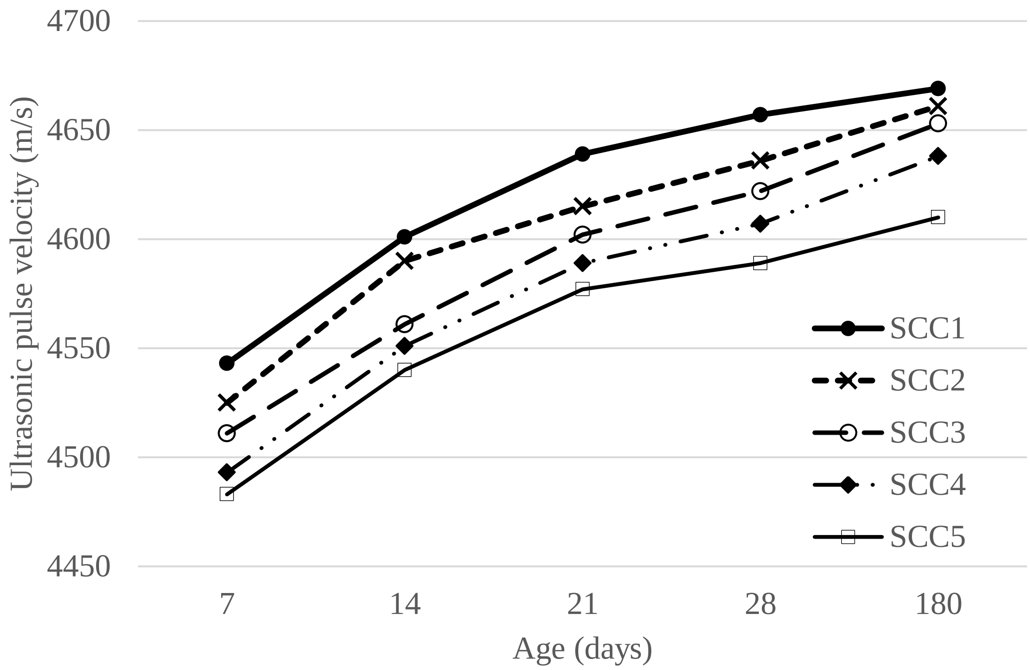

Figure 6. Change of ultrasonic pulse velocity with age

5. CONCLUSION

The results obtained in this study demonstrate the feasibility of ground sulfur use as filler for Self-Compacting Concrete. From the results and discussion, the important conclusions have been summarized as follows:

- that content of ground sulfur had no significant influence on flow ability, passing ability and segregation resistance. Also, there was no significant change in entrained air content,
- the increase in ground sulfur content resulted in a slight decrease of SCC density,
- compressive strength decreased up to 12 % with increase in ground sulfur content up to 20 % of filler. The reason is probably lower mechanical properties of sulfur itself (lower strength and hardness),
- when the curves slopes are analysed, velocities of compressive strength increase for all tested SCC were similar,
- flexural strength decrease was up to 34.3 %, with the highest content of ground sulfur (20 % of the total filer mass),
- ultrasonic pulse velocity through Self-Compacting Concrete with ground sulfur decreased for all ages with sulfur content almost linearly, up to 1.3 %.

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