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Original Article

Unprocessed calcareous fly ash as a partial cement replacement in self-compacting concrete

Adnan MUJKANOVIĆ*, **Dženana BEČIRHODŽIĆ**, **Ilhan BUŠATLIĆ**, **Marina JOVANOVIĆ**

Faculty of Metallurgy and Technology, Univesity of Zenica, Travnicka 1, Zenica, Bosnia and Herzegovina

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ABSTRACT

The effect of partial replacement of cement by unprocessed calcareous fly ash on fresh and hardened properties of self-compacting concrete was investigated. Concrete mixes containing 27.5%, 35%, 42.5%, 50%, 57.5% and 65% of fly ash based on the total weight of binder (cement + fly ash) were tested. All the mixes were designed to have the same volume proportions of paste, fine aggregate and coarse aggregate. Properties investigated were slump flow, J-ring, V-funnel, L-box and sieve segregation of fresh concrete mixes and compressive and flexural strength, dynamic modulus of elasticity, water absorption, volume of permeable voids and density of hardened concrete. The results showed that increase in fly ash content from 27.5% to 50% leads to improvement in workability without significant effect on segregation resistance and hardened concrete properties. Further increase in fly ash content results in worsening of both fresh and hardened concrete properties.

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INTRODUCTION

The construction industry is one of the least sustainable industries in the world, so there is an urgent need to apply available technologies to reduce its environmental impact. The south-east Europe region has substantial reserves of waste streams from various industries causing ecological and economic burden to the production industries [1]. Several industrial waste materials, such as fly ash, silica fume, or ground granulated blast furnace slag, have the potential to be used as a substitute for cement. Reducing the amount of cement allows for reducing carbon dioxide emissions and thus significantly improves the environmental performance of concrete [2, 3]. Fly ash, a residue produced during coal combustion in thermal powerplants, is one of the most common mineral admixtures for both cement and concrete. According to EN 197-1, there are two types of fly ash: siliceous and calcareous [4]. Siliceous fly ash has pozzolanic properties, and calcareous has both pozzolanic and hydraulic properties [5]. Whereas other national standards, for example, the American and Canadian ones, allow the

***Corresponding author.**
*E-mail address: adnan.mujkanovic@mtf.unze.ba

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use of calcareous fly ash in concrete [6], according to the European Standards (EN 450 [7] and EN 206 [8]), its use as an admixture to concrete is not permitted. This can be attributed to significant variability of its chemical composition and physical properties, and high content of free calcium and sulfur compounds [9, 10]. Also, the effectiveness of chemical admixtures in concrete may be lower when calcareous fly ash is used [11]. For these reasons, the potential of calcareous fly ash has been unexploited although its production amounts to more than 50% of the total fly ash output in Europe. High amounts of produced calcareous fly ash force the contraction sector for their utilization. Spain and Greece developed their own national standardization regulations for the use of HCFA in the composition of concrete. Research projects which aimed at assessing the applicability of calcareous fly ash in cement and concrete were carried out in Serbia and Poland [12, 13].

Self-compacting concrete (SCC) is considered to be one of the most successful innovations in the construction industry in recent decades. Fresh SCC mix flows under its weight; it fills the formwork and achieves full compaction without vibration. Compared to ordinary concrete, SCC has improved workability and segregation resistance as well as mechanical properties and durability [14]. It is particularly suitable for congested reinforced concrete structures with difficult casting conditions. SCC allows for rapid concrete placement with significantly reduced labor requirements. The same ingredients are used in SCC and ordinary concrete but their contents are different. In SCC, the content of coarse aggregate, as well as maximal nominal aggregate size, is decreased and the fines content is increased [15, 16]. Chemical and mineral admixtures are usually added in larger quantities compared to ordinary concrete. High range water reduction admixture (HRWRA) is typically used and a viscosity modifying admixture (VMA) may be used to reduce mix sensitivity to small variations of properties and content of SCC components [17, 18]. Compared to ordinary concrete, SCC has a greater share of cement which causes an increase in energy consumption and emissions at the production stage. For that reason, use substitution of cement with alternative binders, such as silica fume, fly ash, should be considered [19, 20]. Siliceous fly ash is widely accepted as a component of SCC, whereas use of calcareous fly is less common.

In the study carried out by Anastasiou and Papayianni unprocessed calcareous fly ash was used as 30% and 50% by mass of the total binder without changing the water to binder ratio. It was found that fly ash reduces workability of fresh concrete, while the mechanical properties of the test mixtures were comparable to the reference mix [21]. Ponikiewski and Gołaszewski tested SCC mixes containing 10, 20, and 30% ground calcareous fly ash and reported the negative influence of fly ash on rheological properties and workability of SCC [22]. Gołaszewski et al. produced SCC containing different types of cement with calcareous fly ash as the cement component (CEM II and CEM IV). It was found that by using cement containing calcareous fly ash it is possible to obtain SCC of acceptable properties. However, the flowability of fresh concrete and mechanical strengthof hardened concrete decreased with the increase of fly ash content in cement [23]. Miera et al. observed that replacing cement with as low as 20% calcareous fly ash reduces the heat of hydration [24]. Naik et al showed that the use of high-volumes of calcareous fly ash in SCC reduces the requirements for HRWRAand VMA and that economical SCC with 28-day strengths up to 62 MPa can be obtained using high-volumes of fly ash [25].

RESEARCH SIGNIFICANCE

Whereas siliceous fly ash use has been studied extensively, the data on calcareous fly ash utilization in SCC mixes are still limited. Many researchers stated the negative influence of calcareous fly ash on workability of fresh concrete which considerably reduces the attractiveness of this fly ash use in self-compacting concrete technology. However, the different physical properties and chemical reactivity of calcareous fly ashes would affect the performance of SCC in different manners. The properties of calcareous fly ashes are very diverse, mainly due to the diverse lignite used as fuel in different power plants. Thus, further research is needed, which assumes the influence of the physicochemical properties of calcareous fly ash. This paper provides the results of a comprehensive study about the calcareous fly ash effect on the most important fresh properties and mechanical properties of SCC. The fly ash used in the study has been utilized successfully for decades as a component of Portland cements (CEM II and CEM IV) produced in a local cement plant. In raw form, it has relatively low reactive lime content, high specific surface area, and spherical shape of particles. This raises the opportunity forits utilization in SCC without previous processing.

EXPERIMENTAL

Materials

The Portland cement used in this study complied with EN 197-1 and is labeled as CEM I 52.5N. Unprocessed calcareous fly ash obtained in "Kakanj" thermal power plant was used as a partial cement replacement. To determine particle shape and texture of fly ash, scanning Electron Microscope (SEM) MIRA3 TESCAN was used (Fig. 1). Limestone filler was used to obtain a given volume ratio of paste and aggregate in all the mixes. Chemical composition and physical properties of OPC, fly ash, and filler are given in Table 1. Locally available limestone aggregate was used. The saturated surface dry specific gravity of aggregate fractions 0-4 mm, 4-8 mm, and 8-16 mm were 2.68 Mg/m^3, 2.70 Mg/m^3, and 2.70 Mg/m^3, respectively. The water absorptions of

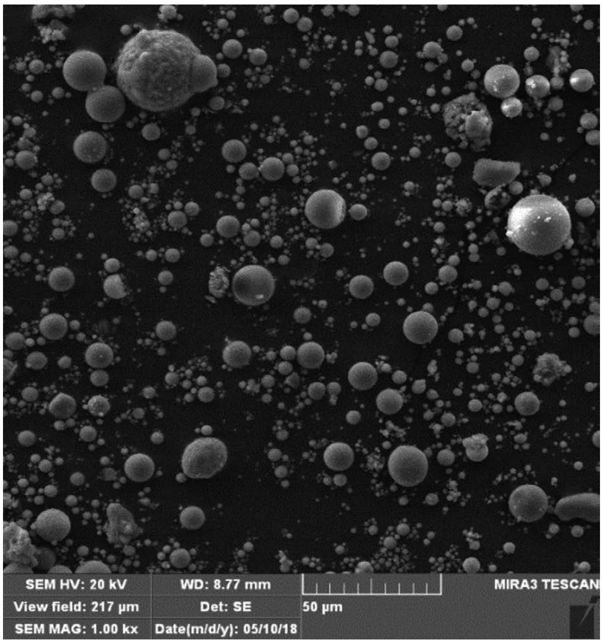

Figure 1. SEM image of fly ash.

Table 1. Chemical composition and physical properties of OPC, fly ash and filler

Component	OPC	Fly ash	Filler
L.O.I. (%)	-	0.14	41.62
SiO_2 (%)	20.71	44.85 (37.411)	0.20
Al_2O_3 (%)	5.69	20.20	0.15
Fe_2O_3 (%)	3.03	8.58	0.30
CaO (%)	65.12	16.83 (14.862)	47.34
MgO (%)	1.29	2.62	8.47
Other oxides (%)	3.96	5.61	1.92
Blaine SSA (cm^2/g)	4130	2530	2260
Specific weight (g/cm^3)	3.10	2.65	2.71
Particle size distribution			
<45 μm (%)	87.9	62.2	53.9
<90 μm (%)	98.1	84.9	85.2
<125 μm (%)	99.3	89.9	95.7

Reactive SiO_2; [2]Reactive CaO.

the aggregate fractions were 0.23%, 0.21%, and 0.21%, respectively. Particle size distribution of aggregate is shown in Figure 2. HRWRA based on polycarboxylate was used to increase flowability and VMA was used to ensure the required stability of SCC mixes.

Mix proportions

Concrete mixes were designed according to UCL (University College London) method which assumes that:

- paste consists of water and all particles less than 0.125 mm (cement, fly ash, filler, and aggregate particles less than 0.125 mm),
- fine aggregate consists of particles having a diameter between 0.125 mm and 4.0 mm, and
- coarse aggregate consists of particles greater than 4.0 mm.

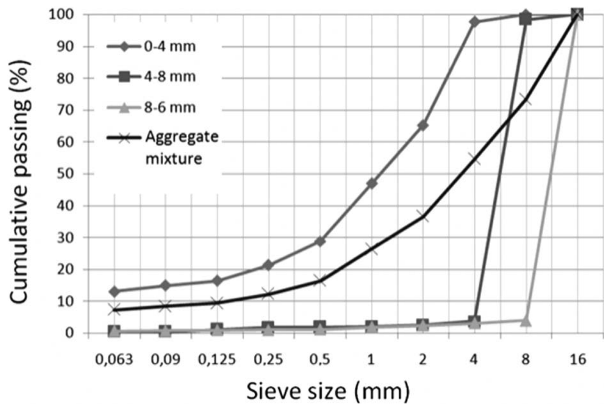

Figure 2. Particle size distribution of aggregate.

The detailed explanation of SCC mix design was explained in [16].

The target workability classes were:

- slump flow SF3 (760 to 850 mm),
- viscosity VF1 (V-funnel time ≤8s),
- passing ability PL2 (L-box height ratio H2/H1 ≥0.8 with 2 rebars) and
- segregation resistance SR2 (sieve resistance ≤15%) [16].

The following parameters were kept constant in the study:

- the total volume of paste (38.5 vol. % of concrete),
- the total volume of fine aggregate (31.5 vol.% of concrete),
- the total volume of coarse aggregate (30.0 vol.% of concrete),
- the total weight of binder (400 kg/m^3) and
- the type and amounts of admixtures (HRWRA based on polycarboxylate: 4.5 kg/m^3 and VMA: 1.7 kg/m^3)

Six concrete mixes were made with fly ash as 27.5%, 35.0%, 42.5%, 50.0%, 57.5% and 65.0% by weight of total binder (cement + fly ash) and the mix proportions are given in Table 2. An increase in fly ash content increases the total paste volume because fly ash has lower specific gravity than cement. For that reason, filler was added in each mix to maintain constant volume proportions of paste and aggregate. The required filler amount was calculated based on fly ash, cement, and filler specific gravities and content of particles less than 0.125 mm in the aggregate fractions.

SCC mixes preparation and testing

After the required quantities of materials were weighed, cement and fly ash were premixed in a dry state. Then coarse and fine aggregates were added to the mixer to ob-

Table 2. Mix proportions of SCC mixes

Components (kg/m^3)	Mixes					
	SCC1	SCC2	SCC3	SCC4	SCC5	SCC6
Cement	290.0	260.0	230.0	200.0	170.0	140.0
Fly ash	110.0	140.0	170.0	200.0	230.0	260.0
Filler	74.0	70.0	66.0	62.0	58.0	54.0
Water	176.2	176.2	176.2	176.2	176.2	176.2
Aggregate 0-4 mm	941.5	941.5	941.5	941.5	941.5	941.5
Aggregate 4-8 mm	316.6	316.6	316.6	316.6	316.6	316.6
Aggregate 8-16 mm	475.3	475.3	475.3	475.3	475.3	475.3
HRWRA	4.5	4.5	4.5	4.5	4.5	4.5
VMA	1.7	1.7	1.7	1.7	1.7	1.7

tain a homogeneous dry mix, followed by adding 2/3 of the total water amount. Finally, the remaining water with HRWRA and VMA was poured into the mixer.

Following tests were carried out on fresh concrete mixes: slump flow (EN 12350-8) [27], J-ring (EN 12350-12) [28], V-funnel (BAS EN 12350-9) [29], L-box (EN 12350-10) [30], segregation resistance (EN 12350-11) [31], density (EN 12350-6) [32] and air content (EN 12350-7) [33]. Specimens were then cast in steel molds and were not subjected to any compaction other than their self-weights. The cubes of size 150 mm were cast for determination of compressive strength and 100×100×400 mm beams for flexural strength. The specimens were removed from molds after 24 h and cured in water till testing. Compressive strength (EN 12390-3) [34] and ultrasonic pulse velocity (EN 12504-4) [35] were determined at 1, 2, 14, 28, 56 and 90 days. Ultrasonic pulse velocity (UPV) measurements were carried out on samples used in flexural tests by direct transmission through the longest side of a beam (400 mm).

Dynamic modulus of elasticity (E_{din}) was calculated according to:

$$E_{din} = \frac{v^2 \rho (1+\eta)(1-2\eta)}{(1-\eta)} \; [Pa] \qquad (1)$$

where: v – ultrasonic pulse velocity (m/s), ρ – concrete density (kg/m^3) and η – Poisson ratio (η = 0.2).

Flexural strength (EN 12390-5) [36], density and water absorption (ASTM C 642) [37] tests were performed at 90 days. All test measurements were taken as the average of three readings. A visual assessment of segregation in hardened concrete was carried out on vertical cross-sections of cubes.

RESULTS AND DISCUSSION

Properties of fresh SCC

The results of fresh concrete tests are summarized in Table 3. The results show that all concrete mixes fall within target slump flow class SF3 (with tolerances according

Table 3. Properties of fresh concrete

Properties	Mixes					
	SCC1	SCC2	SCC3	SCC4	SCC5	SCC6
Slump flow						
Diameter (mm)	745	755	780	790	770	750
T_{500} (s)	1.7	1.6	1.0	1.0	1.2	1,4
V–funnel (s)	7.4	6.7	5.2	4.5	5.6	5.7
L–box (H_2/H_1)	0.84	0.86	0.88	0.90	0.86	0.88
Segregation (%)	4.82	5.11	6.57	8.84	11.04	12.63
J–ring						
Diameter (mm)	740	750	780	790	770	745
T_{500} (s)	1.9	1.8	1.0	1.0	1.2	1,6
Height difference (cm)	1.2	1.0	0.8	0.6	1.0	1,0
Temperature (°C)	18	20	18	18	19	20
Apparent density (Mg/m^3)	2.49	2.52	2.54	2.56	2.55	2.53
Air content (%)	2.3	2.1	2.0	1 .8	1.9	2.1

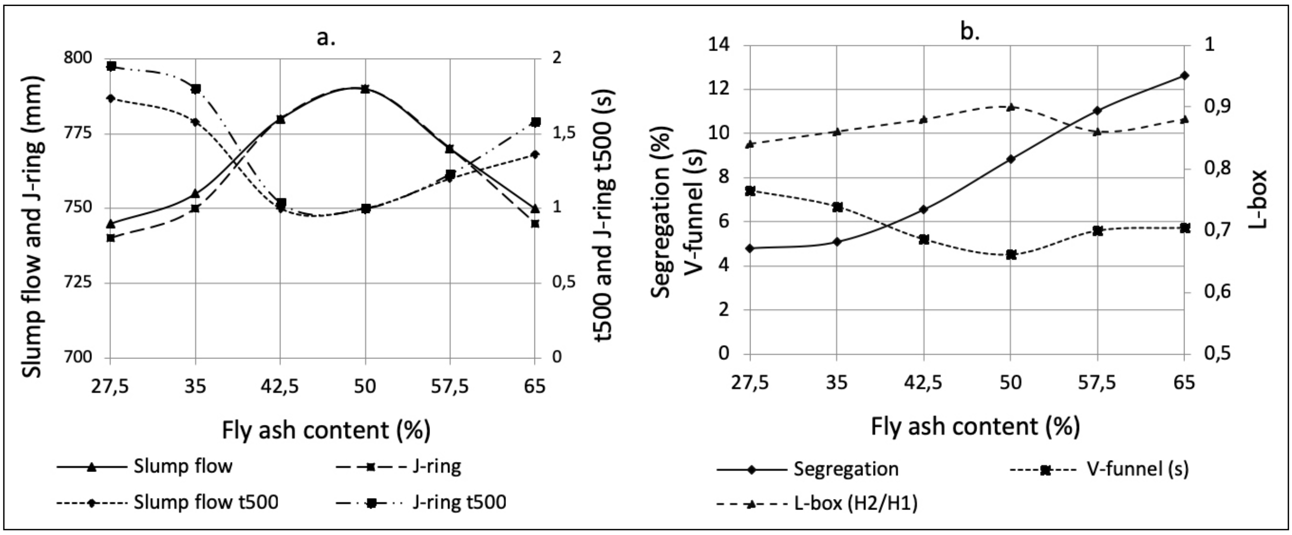

Figure 3. Properties of fresh SCC mixes.

to EN 201) as well as target viscosity class VF1, passing ability class PL2, and segregation resistance class SR2. Figure 3a and Figure 3b show that all workability properties tested have a similar dependence on fly ash content, with exception of segregation resistance. Filling ability (determined by slump flow and V-funnel) and passing ability (determined by J-ring and L-box) increase with fly ash content increase up to 50% (SCC4). Further increase in fly ash content results in decreasing filling ability and passing ability of the mixes. Figure 2b shows that segregation resistance continuously decreases with the increase of fly ash percentage from 27.5% to 65%. The results presented in Table 3 also show that density and air content were not significantly influenced by fly ash content, although minimal air content and maximal density were recorded in a mix containing 50% of fly ash.

The explanation of this complex effect of fly ash on fresh concrete properties probably lays in the facts that:

- Fly ash has lower SSA than cement (Table 1) and since it was not ground, the particles retained their rounded form and smooth surface (Fig. 1).
- Fly ash has lower specific gravity than cement. Therefore, the replacement of cement with fly ash by weight results in increasing the total volume of the binder. (This is mitigated to some extent since the filler content in the mixes decreases as fly ash content increase; Table 2).

The first assumption means that higher fly ash contents yield better self-compacting properties, whereas the second one leads to the opposite conclusion (greater volume → greater total SSA → more water needed). These two effects oppose each other and, depending on fly ash content in the mix, one of them becomes dominant.

Properties of hardened SCC

Results obtained by testing of hardened concrete are shown in Table 4. Water absorption and volume of permeable voids gradually decreases with fly ash content increasing, reaches its minimum at 50% fly ash (SCC4), and increases with further increase in fly ash content. Both bulk density after immersion and bulk density after immersion and boiling, as well as apparent density, reach their maximum at the fly ash content of 50%. Flexural strength at 90 days slightly decreases as fly ash content increases.

The compressive strength development of concretes with different fly ash contents is presented in Figure 4. Compressive strength at 1, 2, 14, and 28 days decreases as fly ash content increases. Early strengths (1 and 2 days) of mixes containing 57.5% and 65% fly ash are particularly low. However, their strength loss decreases at later ages. The mix SCC6 (65% fly ash) reaches only 5% of the 1-day strength of the mix SCC1 (27.5% fly ash), whereas it has nearly 60% of the 28 strength and 69% of the 90-day of the mix SCC1. After 56 days strength of mixes SCC2 (35% fly ash) and SCC3 (42.5% fly ash) exceeds the strength of mix SCC1 (27.5% fly ash).

Figure 5 shows the rate of dynamic modulus of elasticity development. The effect of concrete age on modulus is similar to the effect of age on its compressive strength. However, modulus grows more rapidly in the beginning, whereas in more advanced ages this growth is reduced. Also, modulus reaches a level of constancy at an earlier age than compressive strength.

Figure 6 provides vertical cross-sectional photos of the hardened concrete samples. Segregation was observed in the mixes containing 57.5% and 65% fly ash (SCC5 and SCC6) and, to some extent, in the mix containing 50% fly ash (SCC4). This complies with the fresh concrete segregation results given in Table 3. No mortar layer at the top of the cut plane and no variance in size and the percent area of coarse aggregate distribution from top to bottom are observed in the mixes SCC1, SCC2, and SCC3. Surface of the mix SCC4 shows no mortar layer at the top of the cut plane

Table 4. Properties of hardened concrete

Properties	Mix					
	SCC1	SCC2	SCC3	SCC4	SCC5	SCC6
Compressive strength (MPa)						
1 day	16.2	14.0	11.5	8.6	3.2	0.9
2 days	25.0	23.5	21.2	19.1	10.9	4.0
14 days	51.3	49.0	45.5	36.8	29.5	25.0
28 days	57.4	56.4	53.2	47.3	41.6	33.9
56 days	61.9	67.6	66.7	56.7	54.2	44.7
90 days	67.2	71.0	74.2	65.7	57.4	46.4
Flexural strength (MPa)						
90 days	9.76	9.23	8.89	8.71	8.54	6.97
UPV (m/s)						
1 day	4020.1	3910.0	3816.7	3565.0	3311.3	2375.3
2 days	4362.0	4333.7	4259.9	4232.8	4149.4	3937.0
14 days	4801.9	4767.6	4689.3	4662.0	4651.2	4587.2
28 days	4819.3	4807.7	4784.7	4728.1	4716.9	4694.8
56 days	4938.3	4914.0	4895.9	4878.0	4860.3	4842.6
90 days	4993.8	4987.5	4975.1	4962.8	4944.4	4895.9
Dynamic elasticity modulus (GPa)						
1 day	34.9	32.9	31.7	28.0	23.9	12.3
2 days	41.0	40.4	39.5	39.4	37.5	33.9
14 days	49.7	48.9	47.9	47.8	47.2	46.0
28 days	50.1	49.7	49.9	49.2	48.5	48.2
56 days	52.6	51.9	52.2	52.4	51.5	51.3
90 days	53.8	53.5	53.9	54.2	53.3	52.4
Water absorption after immersion (%)						
90 days	3.88	3.82	3.67	3.54	3.82	4.10
Water absorption after immersion and boiling(%)						
90 days	3.92	3.98	3.81	3.75	3.93	4.23
Dry bulk density (Mg/m^3)						
90 days	2.30	2.31	2.34	2.36	2.33	2.33
Bulk density after immersion (Mg/mv)						
90 days	2.40	2.39	2.42	2.44	2.42	2.40
Bulk density after immersion and boiling (Mg/m^3)						
90 days	2.40	2.40	2.42	2.45	2.42	2.43
Apparent density (Mg/m^3)						
90 days	2.54	2.54	2.56	2.59	2.57	2.59
Volume of permeable voids pore space (%)						
90 days	9,37	9,31	8,89	8,85	9,17	10,00

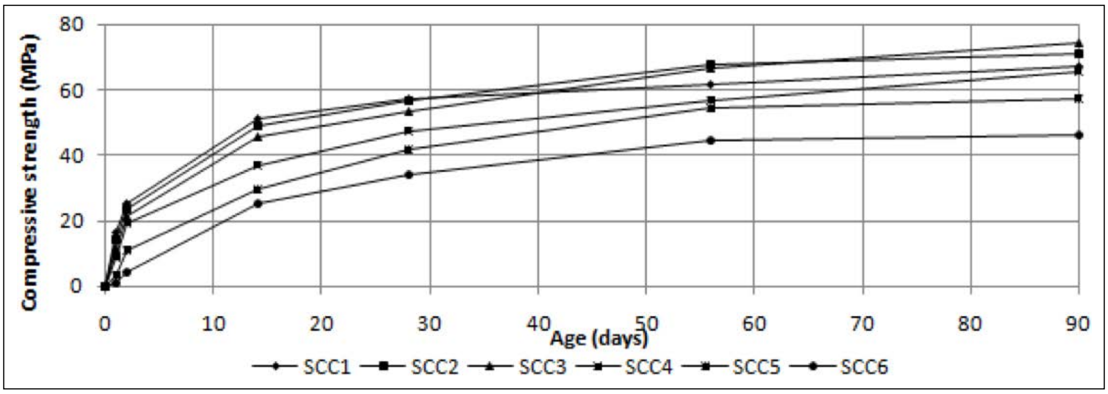

Figure 4. Development of compressive strength of SCC.

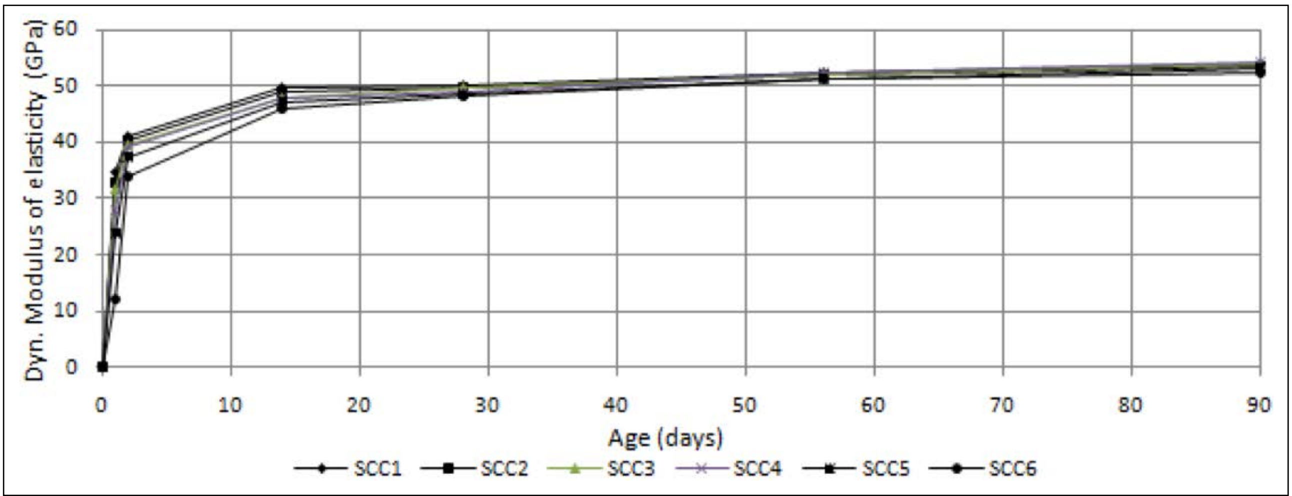

Figure 5. Development of dynamic modulus of elasticity of SCC.

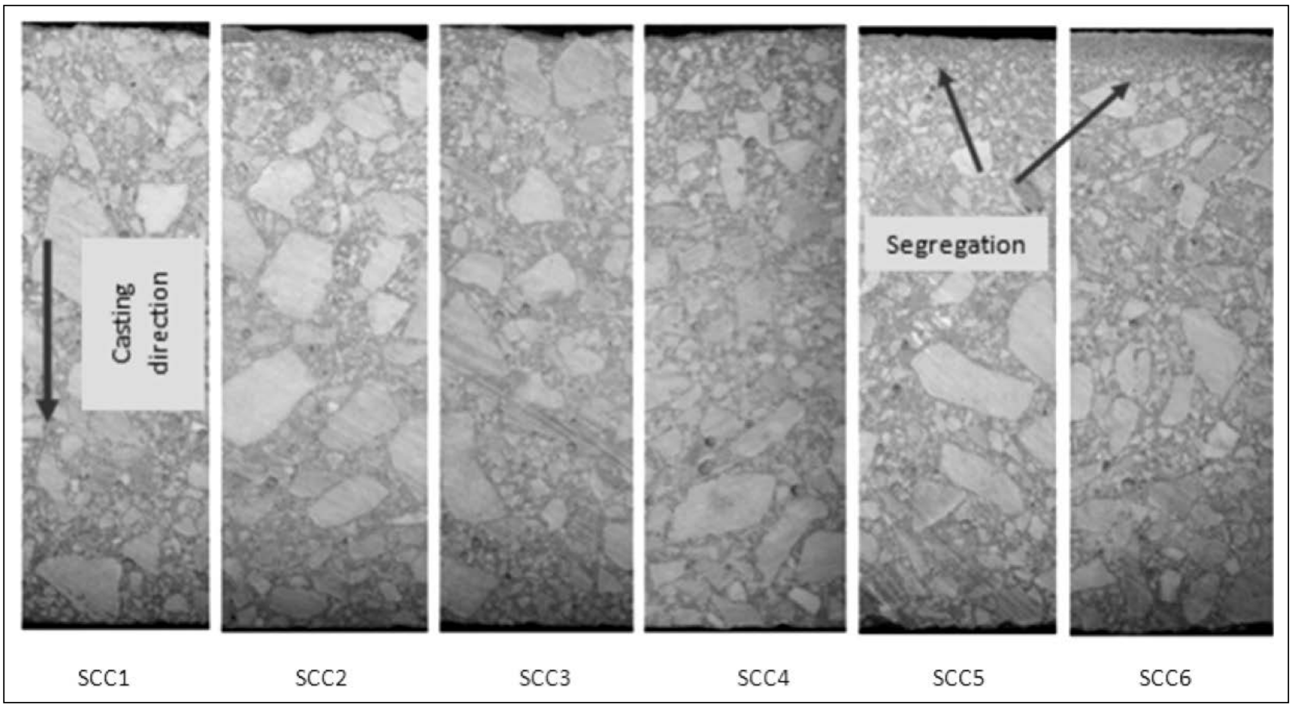

Figure 6. Vertical cross section of hardened SCC.

but a slight variance in size and coarse aggregate distribution. A slight mortar layer at the top of the cut plane and distinct variance in size and coarse aggregate distribution from top to bottom is seen in the mixes SCC4 and SCC5

CONCLUSION

Based on the obtained results, the following main conclusions can be drawn:

- Fresh concrete properties are significantly influenced by fly ash content. However, regardless of their fly ash content, all mixes remained in given slump flow, viscosity, passing ability, and segregation resistance classes.
- Flowability, passing ability, and filling ability increase with increasing fly ash content from 27.5% to 50%. Further increasing in fly ash content leads to worsening of concrete workability. Segregation resistance decreases with an increase in fly ash content.
- Compressive strengths of all mixes at 1, 2, 14, and 28 days decrease as fly ash content increases. Although the early strengths of concrete containing higher volumes of fly ash (57.5% and 65%) were very low, all concretes developed compressive strength in the range 33.9–57.4 MPa, at 28 days, and 46.4–67.4 MPa, at 90 days. Concrete containing 27.5% fly ash has the highest compressive strength at 28 days, while at 56- and

90-days mixes with 35% and 42.5% develop higher strengths.

- Compared to compressive strength, dynamic modulus of elasticity grows more rapidly in the beginning, whereas in more advanced ages this growth is reduced.
- Flexural strength at 90 days slightly decreases as fly ash content increases.
- Water absorption decreases and density increases as fly ash content increase from 27.5% to 50%. Mixes containing 57.5% and 65% fly ash have higher water absorption and lower density.

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Authorship Contributions:

Concept: A.M., I.B., M.J.; Design: A.M., Dž.B.; Supervision: A.M., I.B., M.J.; Data collection and/or processing: A.M., Dž.B.; Analysis and/or interpretation: A.M., Dž.B., I.B., M.J.; Literature search: A.M., Dž.B.; Writing: A.M. Dž.B.; Critical review: I.B., M.J.

Conflict of Interest:

The authors declare that they have no conflict of interest.

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