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CEMENTITIOUS COMPOSITES MADE WITH FLY ASH – A CONTRIBUTION TO THE SUSTAINABLE CIVIL ENGINEERING

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Summary: *Very important and relevant problems of origin, as well as possible applications of fly ash in building industry, are discussed in this paper. The quantities of fly ash produced in power plants, both in Serbia and Europe, are analyzed. Considerable experimental research was conducted in the Laboratory for materials at the Faculty of Civil Engineering, University of Belgrade. The testing, performed on mortar, plain concrete and Self Compacting Concrete (SCC) specimens showed satisfactory results concerning possible application of fly ash as admixture for cementitious composites. Namely, in the first part of research – mortar was produced with fly ash (partially replacing cement from 0-30%) and in the second part – SCC was produced with fly ash partially substituting the aggregate (filler).*

Key words: *cementitious composites, cement, fly ash, mortar, concrete,SCC, superplasticizer, filler.*

1. INTRODUCTION

On a global scale there is an ongoing development of building industry, especially concrete structures, which in turn stimulates the production of cement for cementitious composites (concrete, mortar, etc.). As a result, cement production factories have to develop faster, and manage to keep up with the market pace. Cement is undoubtedly a basic material in building industry, because it is necessary for construction of residential, commercial and industrial structures, roads, tunnels and bridges, underground structures, etc.

In the last several years, substantial changes occurred, both in EU and in Serbia, regarding the types and strength classes of produced cement types. According to [1], more than 80% of the total production of cement in Serbia represents Portland composite cement. Also, in the period between 2000-2008, an increase in R-cements (cements with high initial strengths) was recorded.

The model of cement consumption in Serbia is typical for transition countries, where an individual way of building is still present, which means primary use of bag packed cement. In the future, development of concrete prefabrication and transported concrete application is expected, which will result in production of more different types of cement (with prescribed chemical and mineralogical compositions).

According to the investigations [2] from 2013, China was the biggest producer of concrete and cement in the world (Figure 1). Annual production is more than 2.2 billion tons of cement, which means 1.6 t per capita. There are more then 5000 factories of cement in China only, some of them equipped with old equipment and working with obsolete technology. However, this fact is not much of a barrier for China to be recognized as the biggest cement exporter in the world, for construction of infrastructural objects.

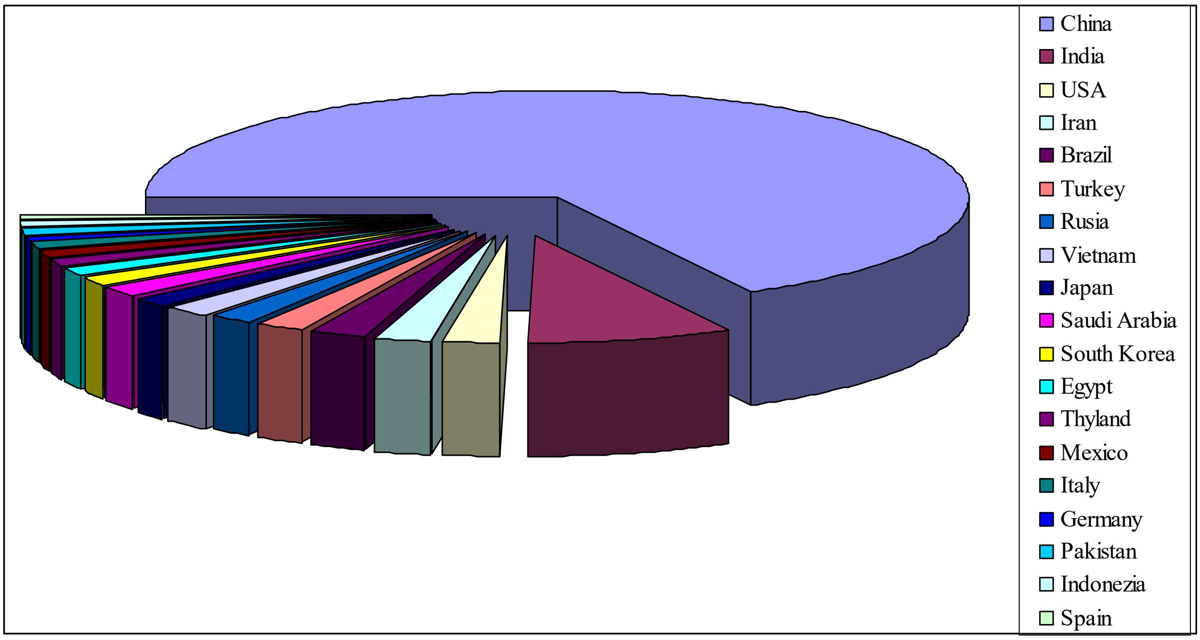

Figure 1: Worldwide cement production in 2013.

Nowdays, annual production of cement in the world exceeds 3.8 billion tons. This huge production is accompanied with environmental hazard. Namely, approximately 0.92 tons of CO_2 are produced in the process of cement production, for every ton of cement clinker produced. This CO_2 emission is mostly a result of de-carbonation of limestone and the use of carbon based fuels.

One of the ways of decreasing this evident threat is incorporated in production of cement. It is the use of pozzolanic materials, such as fly ash and slag. Through the use of such materials, the need for production of cement clinker (as a main component of cement) reduces, and with that reduces the need for energy consumption and CO_2 emission. This is only one of the possible applications of fly ash, which is regretfully used very poorly in Serbia, and represents a great potential. In this paper, several topics of interest will be discussed and illustrated, regarding fly ash and its use in building industry [18].

Usually the term "fly ash" is attributed to a fine-grained material that is deposited on the filters of the exit gases, as a result of the coal combustion in thermal power plants. The amount of this ash, which can be collected, varies widely: from about 1 ton per day, to several tons a minute, depending on a power plant [3]. Given the fact that fly ash has pozzolanic properties and relatively high fineness (its particle diameter is usually within limits of 1 - 150 microns), the powder material can be used as a mineral supplement in cement production, or as a partial replacement for Portland cement in cementitious composites. In this way, significant savings in the consumption of non-renewable resources (limestone, clay) can be achieved, as well as in the energy required for the production of cement. This also provides reduction of the CO_2 emissions (cement industry is responsible for the emission of at least 5-7% of the total carbon dioxide in atmosphere [6]). It should always be borne in mind that the possibilities of application of fly ash, in each particular case, depend on its origin and quality, i.e. the type of plant where fly ash was produced, types of filters, size of particles, quality of coal, types of combustion, the way fly ash was stored, etc. [7].

On the basis of the relevant assessment [4], it is estimated that there is more than 200 million tons of deposited ash and slag in Serbia today - as a result of decades of disposal of these by-products in the power plants. Additional 6 million tons of the waste is the amount added to the previous on an annual basis. Taking into account the fact that the Republic of Serbia obtains the largest portion of electricity from thermal power plants (about 72%, according to report [5]), as well as the fact that it plans to further increase power capacity based on coal combustion, it is clear that the problem of the fly ash utilization in the near future will be even more pronounced. Despite the constant control and monitoring, deposited fly ash and slag in Serbia occupy an area of about 1600 hectares, producing many environmental problems such as: pollution of air, soil, surface and groundwater, destruction of vegetation cover and disturbance of ecosystems, damage to human health, etc. These landfills are located mainly near thermal power plants (TPP Kolubara A, TENT A and B near Belgrade, Kostolac A and B, TE Morava Svilajnac). These are densely populated areas, close to the large rivers: Sava, Danube and Morava; the fact that makes this problem even more important. In addition, it is important to stress out that fly ash represents the most common waste material in our country, as well as secondary raw material that can be used primarily in the construction industry.

Numerous international experiences show that fly ash and slag can be used as cement admixtures, as a substitute for aggregates and fillers for concrete and asphalt, for the production of lightweight concrete masonry elements, gas concrete, bricks, etc. Also, geopolymers can be obtained through alkaline activation of the fly ash; geopolymers present a promising alternative to cement composites [5]. In this sense, application possibilities and synthesis of geopolymers [13] have been studied at the Faculty of Civil Engineering, University of Belgrade.

Regarding the situation in the Republic of Serbia, a package of legislation that deals with the issue of environmental protection came into force in 2004, as part of efforts to harmonize national with the EU legislation. In this regard, public company "Elektroprivreda Srbije" (EPS) is bound to harmonize the work of its plants with the provisions of the Law on Integrated Pollution Prevention and Control, by the end of 2015. This means that the EPS is bound to apply modern safety measures, such as plant fuel gas desulphurization, high efficiency electrostatic or facilities for waste treatment, both in the new objects and in the old objects to be reconstructed (revitalized).

The first attempts of application of fly ash in Serbia are dating back to the 70s of the last century, when this material was used for soil stabilization in combination with cement (a test section of the road route Lazarevac – Ibarski put, also in the street Beogradski Bataljon at Banovo Brdo, etc.). Later on, 15 km of roads were made around flyash dumps in "TENT A" and "TENT B" near Belgrade, and some local roads in the town of Obrenovac - during the 80's.

Currently, fly ash is utilized practically only in cement plants, and to a lesser amount, for brick manufacture. Only a few percents of the total annual production of fly ash are used this way [1]. Given the market potential, there is a strong possibility to significantly increase the degree of utilization of this resource. As an example, the following chart (Figure 2) can be used, which refers to the area of use of fly ash in the European Union in 2008 [8],[14].

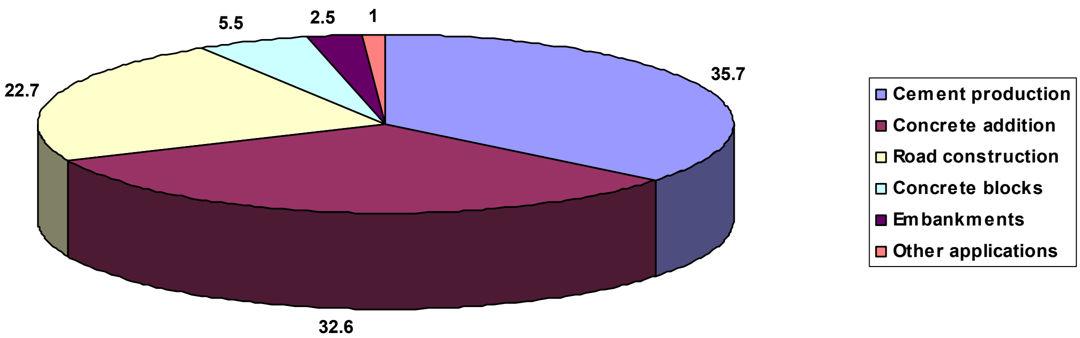

Figure 2: Use of fly ash in construction industry of the European Union (15 countries) in 2008 [%]

Regarding the legislation in Serbia, there is a set of laws, standards and other documents covering this topic of fly ash. For instance, there are standards about the use of fly ash in concrete, designated as SRPS EN 450-1 and SRPS EN 450-2, [16],[17].

2. EXPERIMENTAL INVESTIGATION

The main objective of the experimental research presented in this paper was to evaluate the possibilities of application of locally generated fly ash as a partial replacement for cement, in order to obtain cementitious composites (mortar and concrete) with acceptable physical and mechanical properties (such as density, compressive strength, tensile strength, flexural strength and modulus of elasticity). This research was carried out in the Laboratory for Materials, Institute for materials and structures, Faculty of Civil Engineering, University of Belgrade. Within the performed tests, percentage of fly ash as a partial replacement for cement was varied. In the Self Compacting Concrete mixtures, part of the filler was replaced with fly ash. Figure 3 shows this fine material precipitated from the stack gases of industrial furnaces burning solid fuels with predominantly spherical particles.

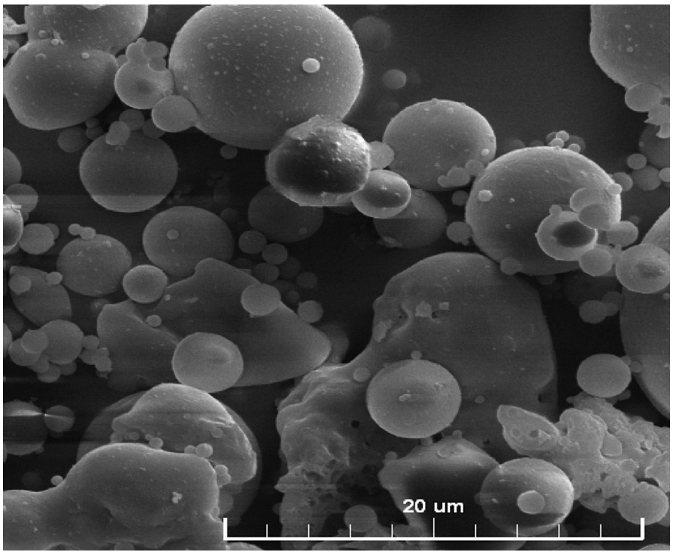

Figure 3: SEM microphotograph of fly ash [14]

2.1 Mortar

Cement designated as PC 20M (S-L) 42.5 R (producer: Lafarge, Beočin) was used for production of mortar mixtures, together with the fraction I (0/4 mm) of river aggregate "Moravac"; and fly ash originating from thermal power plant "Kolubara", Veliki Crljeni. Aggregate grain density amounted to 2.632 g/cm^3, and the bulk density of aggregate was 1.635 g/cm^3. Specific gravity of cement amounted to 2.945 g/cm^3, and fly ash 2.112 g/cm^3. The test results of particle size distribution of the fly ash are given in Table 1 [9] and Figure 4, and the results of its chemical composition in table 2 [12],[15].

Table 1: Fly ash sieve analysis

d (mm)	0,003	0,005	0,009	0,015	0,026	0,045	0,056	0,063
Y (%)	9,63	11,82	15,32	27,14	39,45	51,30	58,00	60,70
d (mm)	0,090	0,200	0,315	0,500	0,710	1,000	2,000	-
Y (%)	69,10	92,20	96,40	98,60	99,00	99,66	100,00	-

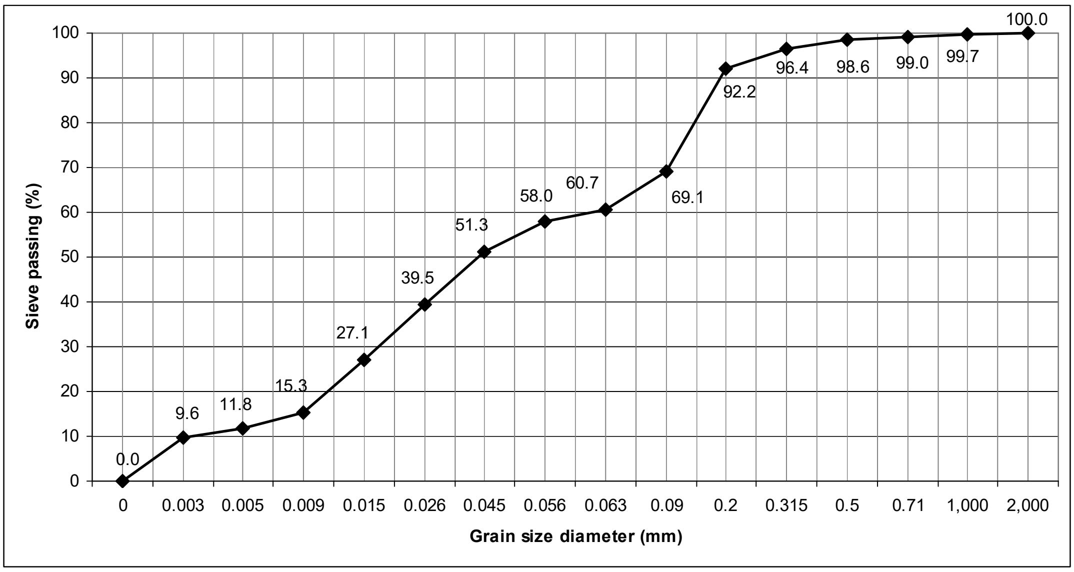

Figure 4: Passing percentages for fly ash

Table 2: Chemical composition of fly ash

Oxides content (%)											
SiO_2	Al_2O_3	Fe_2O_3	TiO_2	CaO	MgO	P_2O_5	SO_3	Na_2O_3	K_2O	MnO	CO_2
58.32	*18.88*	*6.75*	*0.57*	*8.71*	*2.3*	*0.025*	*1.29*	*0.5*	*1.16*	*0.026*	*0.11*
Microelements (mg/kg)											
Pb	Cd	Zn	Cu	Ni	Hg	As	Ba	Sb	Se		
24.4.	*0.2*	*56.6*	*36*	*135.1*	*0*	*182*	*86.2*	*1.1*	*1.4*		

Four series of mortars were made with the use of different amounts of fly ash as a partial replacement for cement (0% - R, 10% - F1, 20% - F2 and 30% - F3). The guiding principle for the design of these series was to maintain a constant total amount of binder component (cement + fly ash). The adopted quantities of component materials are shown in Table 3, together with the applied water-cement ratio (w/c) and the measured properties of the mortar in the fresh state (density and consistency – flow table method).

Table 3: Mortar mix composition and fresh state properties

Series	w/c	Water (g)	Cement (g)	Fly ash (g)	Sand (g)	Density (g/cm^3)	Flow diameter (mm)
R	0.57	256	450	0	1350	2.248	180
F1	0.70	284	405	45	1350	2.243	183
F2	0.85	306	360	90	1350	2.195	180
F3	1.00	315	315	135	1350	2.169	185

Density and tensile strength (f_z) of hardened mortar were tested at 28 days, while the compressive strength (f_p) was determined at the age of 28 and 90 days. Modulus of elasticity (E) and adhesion (f_{at}) were tested only at 28 days. Ultrasound pulse velocity was also measured, and it ranged from 3385 m/s to 3673 m/s. The test results of the aforementioned properties are shown in Table 4.

Table 4: Hardened mortar properties

	R	F1	F2	F3
Density (g/cm^3)	2.215	2.195	2.179	2.151
$f_{zs,28}$ (MPa)	7.17	7.29	6.13	6.54
$f_{p,28}$ (MPa)	38.5	36.1	30.7	27.3
$f_{p,90}$ (MPa)	41.2	39.4	39.1	34.8
$f_{at,28}$ (MPa)	2.79	2.85	2.58	2.39
E (GPa)	25.6	23.4	22.8	20.7
v (m/s)	3673	3587	3542	3385

For adhesion tests, specimens of mortar covered prefabricated concrete slabs (measuring 40x40x5cm) were carefully prepared in the subsequent manner: after the preparation of the surface of the concrete slab (steel brush roughing and cleaning, followed by wetting with water), a mortar was manually applied onto the substrate, in a layer with thickness of approximately 3 cm. After that, the mortar was cured for the following 28 days, by means of a wet jute cloth cover. Upon completion of the curing period, adhesion was studied on the basis of "Pull-off" method; using a "Controls" tensile testing machine, with loading velocity of 0.5 ± 0.1 kN/s.

Determining the modulus of elasticity of the mortar was performed at 28 days, on standard prisms (4x4x16cm), made of mortar (Fig. 5). During this test, the initial (lower) stress was 0.5 MPa, while the adopted value of the upper stress was $f_p/3$ (i.e. three times lower stress value than the compressive strength of mortar). Relevant values for modulus of elasticity (E) were calculated based on the differences in strain measured during the last (third) cycle of loading-unloading.

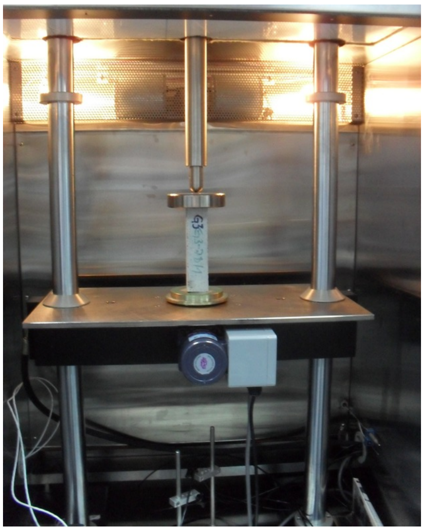

Figure 5: Modulus of elasticity investigation on mortars

Another laboratory investigation was made with ″pure″ cement designated as PC 42,5R and carboxilate based superplasticizer (0,56% of the binder mass). Compositions of these mortars are shown in Table 5, together with the measured properties of the fresh mortars (density and consistency – flow table method).

Table 5: Mix composition of mortars made with pure cement and their fresh state properties

Series	w/c	Water (g)	Cement (g)	Fly ash (g)	Sand (g)	Density (g/cm^3)	Flow diameter (cm)
E	0,56	252,5	450	/	1350	2,299	13,3
P1	0,562	227,6	405	45	1350	2,278	12,9
P2	0,71	254,4	360	90	1350	2,259	13,4
P3	0,8	268	315	135	1350	2,227	13,9

Hardened mortar properties are shown in Table 6. Investigations included 28-day density, flexural and compressive strength, adhesion (tested as previously explained) and ultrasound pulse velocity of mortar specimens.

Table 6: Hardened mortar properties (mortars made with ″pure″ cement)

	R	P1	P2	P3
Density (g/cm^3)	2,290	2,279	2,256	2,223
$f_{zs,28}$ (MPa)	8,09	8,50	7,50	6,67
$f_{p,28}$ (MPa)	57,6	58,6	48,4	42,5
$f_{at,28}$ (MPa)	2,03	2,32	1,39	1,31
v (m/s)	4255	4250	4092	3961

2.2 Plain concrete

Numerous studies of the impact of fly ash on the properties of fresh and hardened concrete have been conducted in the Laboratory for Materials at the Faculty of Civil Engineering in Belgrade over several years [10]. Results related to two typical concrete mixtures will be presented in this paper:

- Mixture "E"; - Standard mixture, prepared on the basis of the common component materials (cement, water and river aggregate), and
- Mixture "A"; - Concrete mixture prepared with 20% fly ash as a partial replacement for cement.

Two sets of boundary conditions were used simultaneously in the process of concrete mix design: requirements with respect to the consistency of fresh concrete and requirements related to the quality of the hardened concrete. As for the first group of the requirements, the mixture had to meet the requirement of liquid consistency (based on Abrams' method), with a maximum aggregate grain of 8 mm. Given the high value of fineness of ground fly ash (specific surface area more than 5000 cm^2/g), the need for a greater amount of water in the concrete mixture "A" occurred; which led to use of a carboxylate based superplasticizer in an amount of 1.6% (relative to the weight of cement). Pure Portland cement designated as PC 42.5 R "Lafarge" Beočin was used as a binder. River aggregate ″Moravac″ which was used consisted of 45% fraction I (0/4 mm) and 55% fraction II (4/8 mm) (grain size distribution curve is given on Fig. 6). In addition, the grain density of aggregate amounted to 2.682 g/cm^3 for the fraction I and 2.655 g/cm^3 for fraction II. Gravity of fly ash and cement amounted to 2.180 g/cm^3 and 3.00 g/cm^3, respectively. The complete composition of these concrete mixtures is shown in Table 5.

Table 7: Concrete mix design

Concrete	m_c (kg/m^3)	m_{ef} (kg/m^3)	m_{ad} (kg/m^3)	m_v (kg/m^3)	m_a (kg/m^3)	ω
"E"	350	-	-	217,5	1801	0,62
"A"	280	70	4,48	187,5	1842	0,67

The following symbols were used in Table 7: m_c - mass of cement, m_{ef} - mass of fly ash, m_{ad} – mass of additives, m_v - mass of water, m_a - mass of aggregates and ω - water-cement ratio.

The average values of density in the fresh state amounted to 2369 kg/m^3 for concrete ″E″, and 2384 kg/m^3 for concrete ″A″. Concerning consistency, concrete type ″E″ showed the average subsidence of 12.5 cm, while for concrete type ″A″ this property was 17.5 cm.

Tests on hardened concrete (compressive strength - f_p and tensile strength - f_s), were performed on 15 cm cube specimens, at the age of 7 and 28 days. The study included the determination of tensile strength (f_z), based on ″Pull-off″ method, ″ using the 20×20×5 cm plate-shaped samples. Examination of the modulus of elasticity (E) was performed on cylindrical specimens, with the base diameter of 15cm and height of 30cm, at the age of 28 days. The average values of the test results for hardened concrete are shown in Table 8.

Table 8: Hardened concrete properties

Concrete	$f_{p,7}$ (MPa)	$f_{p,28}$ (MPa)	$f_{z,28}$ (MPa)	$f_{zs,7}$ (MPa)	$f_{zs,28}$ (MPa)	E (GPa)
"E"	36,1	46,5	3,54	6,38	6,79	37,5
"A"	33,5	44,4	3,13	5,33	6,38	36,0

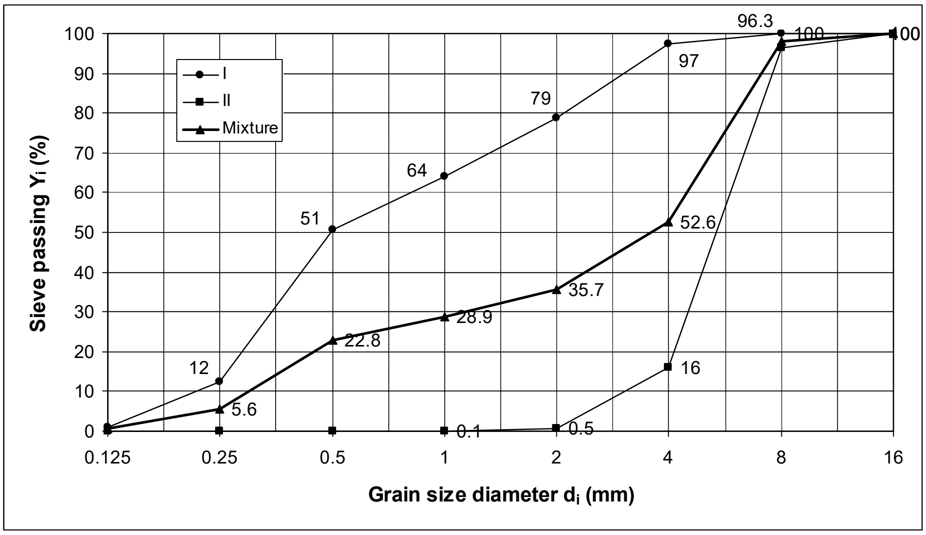

Figure 6: Passing percentages for the aggregate used in normal-vibrated concretes

2.3. Self Compacting Concrete

Self Compacting Concrete (SCC) is a modern composite material, which is becoming more accepted in the construction industry around the world - primarily for its good technological properties in the fresh state (consistency, workability, pumpability, etc.). The most commonly used definition of SCC says that it is a concrete with liquid consistency, a concrete that is capable of completely flowing through the most complex re-bar formations, filling complex sections of formwork and passing through narrow segments of formwork, without the occurrence of segregation and without the need for any mechanical compaction. The use of such concrete offers multiple benefits, both in engineering, but also in economic and environmental terms. These benefits are even more pronounced in the case when fly ash is used as a component for Self Compacting Concrete.

To verify the above-mentioned quotes, two SCC mixtures were made, one with the addition of fly ash and the other without fly ash [11] (Fig. 7). The aim was to obtain a class of SCC concrete SF2 (660 mm < Flow diameter < 750 mm).

The content of cement was constant in these series (″pure″ Portland cement designated as PC 42.5 R "Lafarge" Beočin was used), but the composition of the filler was different: in series designated as SCC1 only limestone powder (m_f) was used, and in series designated as SCC2 both fly ash (m_{ep}) and limestone powder (m_f) were used. The compositions of both types of SCC are given in Table 9.

Table 9: Composition of SCC

Concrete	m_c (kg/m^3)	m_{ep} (kg/m^3)	m_f (kg/m^3)	m_{ad} (kg/m^3)	m_v (kg/m^3)	m_a (kg/m^3)	ω
SCC1	260	-	304	10.92	190	1718	0,36
SCC2	260	104	200	10.92	190	1718	0,36

Densities of fresh concrete SCC1 and SCC2 amounted to average of 2337 kg/m^3 and 2310 kg/m^3, and the measured values of slump-flow were 780 mm and 730 mm, respectively.


Figure 7: Self Compacting Concrete prepared and tested in laboratory conditions

This concrete was poured in the molds without vibration and then cured in a customary manner under laboratory conditions. Testing of compressive strength (f_p) of SCC was performed on 15 cm cubes, at the age of 28 and 90 days. The investigation also included the adhesion strength test (f_z), carried out on 40×40×5cm plates using ″Pull-off″ apparatus, and splitting tensile test (f_{zc}) made on cylinders of diameter and height of 15 cm, at the age of 28 days [19]. The average values of the mechanical properties are given in the Table 10.

Table 10: Hardened SCC properties

Concrete	$f_{p,28}$ (MPa)	$f_{p,90}$ (MPa)	$f_{zc,28}$ (MPa)	$f_{z,28}$ (MPa)	$f_{zs,28}$ (MPa)
SCC1	30.1	37.2	1.92	2.82	5.08
SCC2	47.1	58.6	3.06	4.77	6.64

3. CONCLUSIONS

Based on the performed experimental studies, conclusion can be drawn that fly ash can be successfully applied for cementitious composites, such as mortar and concrete. Also, the replacement levels of cement with fly ash, as well as the properties of fly ash, depend on the physical and mechanical properties of the composite in design - both in fresh and in the hardened state. In order to eliminate the influence of the type of cement on the investigated properties of mortar and concrete made with the addition of fly ash, ″pure″ Portland cement should be used, instead of composite cements, whenever possible.

Generally, the increase in water content was necessary in order to achieve the same consistency of the composites with fly ash, which is in correlation with relatively large fineness of fly ash. This usually leads to a decrease in density of the composite, as well as to a reduction of the early strength, as compared to the reference (composites made without the addition of fly ash). This is fully in line with the results of other authors [3], [7]. On the other hand, due to its pozzolanic properties, fly ash contributes to the improvement of mechanical properties of mortar and concrete at ages greater than 28 days (see the values of strength at the ages of 90 days, shown in Tables 4 and 10).

Regarding the replacement of limestone filler (as the usual type of filler in Self Compacting Concrete) with fly ash as an alternative, conducted studies have shown that the addition of fly ash contributes significantly to the rise of the mechanical properties (compressive strength, tensile strength, flexural strength and adhesion - see Table 10).

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