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# Developing high-performance concrete using locally available materials

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## Abstract

In recent decades, the demands on concrete have increased beyond mere fulfilment of compressive strength requirements. Today, there is a demand for concrete with high resistance to aggressive environments so as to ensure the planned service life of structures. To provide for this high resistance, high-performance concretes are usually based on packing density achieved with a high proportion of fines provided by various supplementary cementitious materials. Silica fume and fly ash are some of the commonly used materials, while calcined clay and limestone are the emerging ones. The study presented in this paper aims to investigate the potential of different materials collected locally in Croatia for use as alternative supplementary cementitious materials in high-performance concrete. Initial experiments focused on the chloride migration resistance of four different combinations of mixed binders and a reference mix. Based on the results presented, the next phase will be to optimize high-performance concrete with calcined clays and fly ash.

*Key words: high-performance concrete, low* $CO_2$ *cementitious materials, calcined clays, non-steady state chloride diffusion, surface electrical resistivity*

# Razvoj betona visokih uporabnih svojstava na bazi lokalno dostupnih materijala

## Sažetak

Posljednjih desetljeća zahtjevi za beton povećali su se izvan ispunjenja tlačne čvrstoće. Danas postoji potražnja za betonom s visokom otpornošću u agresivnom okolišu kako bi se osigurao projektirani uporabni vijek građevina. Da bi se osigurala ova velika otpornost, betoni visokih uporabnih svojstava obično se temelje na gustoći pakiranja koja se postiže velikim udjelom sitnih čestica, što osiguravaju razni mineralni dodatci cementu. Silicijska prašina i leteći pepeo neki su od najčešće korištenih materijala, dok su kalcinirana glina i vapnenac materijali koji se sve češće koriste. Cilj ovog istraživanja je odrediti potencijal različitih lokalno dostupnih materijala kao alternativnih mineralnih dodataka u betonu visokih uporabih svojstava. Početna istraživanja usredotočena su na otpornost na prodor klorida četiri različite kombinacije miješanih veziva i referentne mješavine. Na temelju prikazanih rezultata, sljedeća faza bit će optimizacija betona visokih uporabnih svojstava s kalciniranom glinom i letećim pepelom.

*Ključne riječi: betoni visokih uporabnih svojstava, cementni materijali s manjom razinom* $CO_2$ *emisije, kalcinirane gline, difuzija klorida, električna otpornost*

# 1 Introduction

In the past few decades, a lot of special type of concretes have been produced to meet the unique requirements of construction industry. High-performance concrete (HPC) is one of the main variants among these, and the application-level of HPC is still growing and entering into broader application areas. A high-performance concrete always gives an optimized concrete with essential performance characteristics using given materials exposed to given condition, all provided with optimized cost and service life [1]. Development of HPC in the 1970s was mainly focused on reaching maximum compressive strength in the range of 80-120 MPa [1]. It is only later that the term HPC started to include high performance of fresh state properties and/or durability properties.

Proper material selection must be made for producing HPC of low porosity and fine pore structure. From the perspective of materials, alternative binders and chemical admixtures could be a proper choice for acquiring concrete of such required properties. In the current generation, supplementary cementitious materials (SCMs) are used as sustainable and economical alternatives for clinker at larger scale [2]. Silica fume (SF) and fly ash (FA) are two major SCMs that are used in HPC, providing reduced porosity in mortar matrix and improving the interfacial transition zone (ITZ) with aggregates. Additionally, secondary hydration products from pozzolanic materials also give significant contribution to the prolonged service life of concrete [3]. Nowadays, the availability of all these traditional SCMs is limited. Therefore, concrete industry needs alternative raw materials for the long-run. This issue can be overcome by the use of natural clay as SCM – which becomes reactive when calcined at temperatures between 600-800 $^0$C. In normal concrete, the usual replacement level of cement with calcined clay is around 30%, when the cost of calcination usually does not make this an economically viable option. Additional clinker substitution can be achieved with around 15% of limestone as ternary binder coupled with calcined clay, in what is known as limestone calcined clay cement -LC$^3$. The reasoning for such combination lies in the reaction between the aluminate component of the calcined clay with calcium carbonate (limestone) and calcium hydroxide to produce space filling carboaluminate hydrates [4]. Currently, these types of concrete have been widely used in countries like India, Brazil, Cuba, etc.

This paper aims to present preliminary data from initial tests on five mixes using locally available SCMs including fly ash, clays, and silica fume. The overall action plan is explained in Figure 1, with steps already taken and those planned in the forthcoming period. Based on the presented results, combinations of SCMs will be optimized, with the aim of developing a high-performance concrete that can be used in extreme environmental conditions.

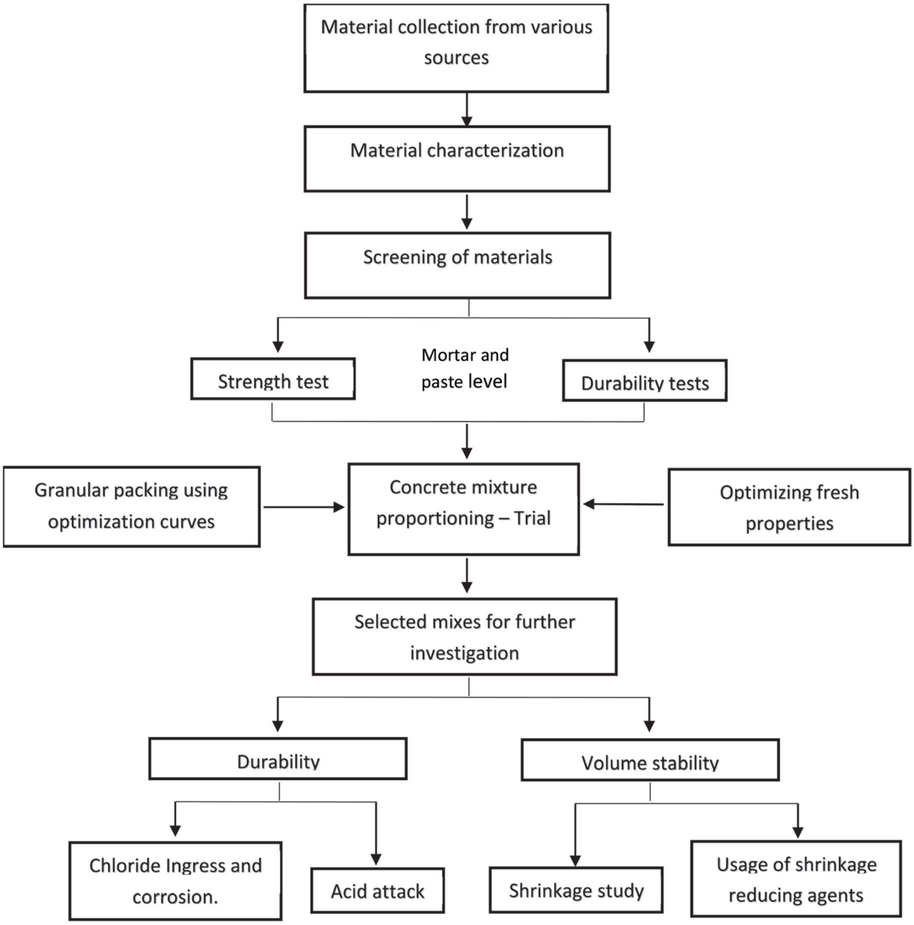

**Figure 1. Proposed methodology for producing high-performance concrete using locally available alternative binders**

# 2 General principles in HPC design

## 2.1 Mix proportioning

Generally, production of high-performance concrete is more complicated compared to conventional concrete. In most HPC designs, it is assumed that best engineering properties are gained when constituents are densely packed. Typical concretes are made with particles covering the size range from $10^{-3}$ m to $10^{-9}$ m. In HPC, all these solid particles should be arranged with minimum voids to get the composition of maximum density. It is the grading or packing of the whole range of particles, from coarse aggregate to fine aggregate, to cement grains, and to fine and ultra-fine cementitious materials, that determines the overall performance of a concrete mix [5]. It is assumed in the packing theory that the addition of finer particles to concrete matrix is beneficial to filling up the voids and reducing the space for water [6-8]. The particle packing density (ϕ) is defined as the solid volume of particles in a unit volume. Several methods are available for determining the particle packing density of composite mixtures. Particle packing of a mixture was initially achieved by means of optimization curves, Figure 2. In this method, a group of particles with different particle size distribution are combined in such a way that the total particle size distribution of the mixture matches an optimum curve.

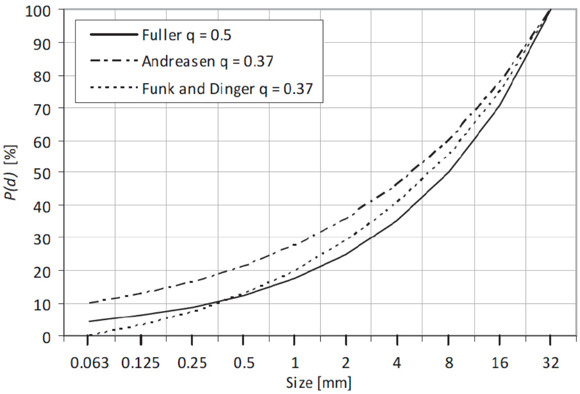

**Figure 2. Ideal packing curve according to various studies [9-11].**

Another method for determining the particle packing density is via particle packing models, Table 1. Several researchers reported that HPCs designed by the particle packing method exhibited excellent resistance against chemical attacks as well as a prolonged service life. For instance, Fennis et al. produced concrete using the particle packing method with fly ash, quartz powder, and bottom ash. The results showed that it is possible to design ecological concrete in which 50% of the cement is saved by using the particle packing technology in concrete mixture optimization [12].

**Table 1. Various particle packing models used for particle packing density determination**

| | | |
|---|---|---|
| The Furnas model | • Valid for two groups of mono-sized particles<br>• Most of the current models based on this model | [13] |
| The Toufar and modified Toufar model | • Determination of packing density of binary mixes with diameter ratio between 0.22 and 1.0.<br>• Assumed that each of the fine particles is placed between precisely four coarse particles | [14] |
| The Dewar model | • A smaller particle will fill the voids between larger particles<br>• Entire particle structure influenced by particle interference. | [15] |
| Linear Packing Density Model | • Furnas model improved by extending the two components to multi-components with geometrical interaction between the particles. | [7]<br>[16] |
| Compressible Packing Model | • Introduced virtual compactness factor, and is related to compaction energy.<br>• Larrard et al. included this factor in binary granular mixes and evaluated particle packing density. | [7] |
| The Schwanda model | • The model calculates the maximum void ratio, which corresponds to the minimum voids content, and then determines the maximum packing density. | [17] |

## 2.2 Workability

HPC contains a significant number of finer particles like fly ash and silica fume to minimize the void content in the mixture. Generally, workability of a concrete mix is dependant on the number of fine particles included in the mix. Studies from various researchers have proven that a very dense composition with too many fine particles produces a low-workability concrete [18]. To maintain the workability at a low water/binder ratio, the use of high-water reducer type superplasticizer (SP) is highly recommended in HPC. Nithya et al. report that a significantly higher dosage of SP is required for limestone-calcined clay cement compared to fly ash and ordinary Portland cement system. They also report that the use of the Poly Carboxylic Ether (PCE) based SP could provide more workability in LC$^3$ based concrete compared to ordinary concrete. The higher SP dosage in clay-containing concrete is attributed to the exchange of cations in the clay with the organic materials present in the admixture to neutralize electrical charges on the clay particle surface [19], causing lower dispersion of SP in the mixture. It is evident that the selection of SP type and dosage will play a key role in their performance in HPC design [20]. Therefore, for an optimized HPC, the compatibility of different superplasticizers with alternative binders needs to be studied.

## 2.3 Performance in harsh environments

Chloride diffusion is one of main factors that influence concrete durability and contribute to the reduction of service life of concrete structures. Better resistance to chloride diffusion of HPC compared to classical concrete can be expected due to high density and tightly packed structure. Tammi et al. [21] report that high-performance concrete with GGBFS exhibits chloride diffusivity below the chloride threshold of 0.40% at a depth of 10 mm, while standard concrete reveals higher than the threshold diffusivity at the same depth. Concrete with palm fuel ash, rice husk ash, and fly ash, significantly improves resistance against chloride attack in mortar by enhancement in nucleation sites for C-S-H precipitation, reducing the $Ca(OH)_2$ and improving the permeability of mortar [22]. In the $LC^3$ system, the pore refinement would be happening at an early age, and is dependent on the original kaolinite content in clay. The rapid pore refinement in $LC^3$ system is advantageous to limiting chloride ingress [23]. The chloride diffusion coefficient obtained with an accelerated method shows that the diffusion coefficient for $LC^3$-50 is ten times lower than that of the plain Portland cement [24]

Concretes exposed to environmental conditions such as those used in sewage pipes, oil refineries, and food processing factories, demonstrate significant interaction with chemical acids, including sulphuric acid, and hydrochloric acid. Standard concrete, exposed to these highly corrosive acids, might suffer considerable damage. It is, therefore, clear that HPC could be used to ensure higher resistance to acid attack. According to literature, concrete with a combination of 10% silica fume and 60% fly ash as an OPC replacement produced maximum protection against acid attack, which is represented in the study by sulphuric and hydrochloric acid [25]. Said-Mansour et al. reported that mortar with 10%, 20% and 30% calcined kaolin showed better resistance against 1% hydrochloride acid and 2.5% sulphuric acid. The weight losses are around 30% in sulphuric acid and 35% in hydrochloric acid, whereas OPC exhibited 40% weight loss in sulphuric acid [26].

# 3 Experimental Program

## 3.1 Materials

Mortar with Portland cement CEM I 42.5R was used as control mix in this research. Siliceous fly ash from Tuzla, Bosnia and Herzegovina, and natural clays from different sources (Našice and Maruševec, Croatia), were used after calcination (three-minute grinding in disc mill, followed by calcination at 800°C for 1 hour) with 30% replacement level of CEM I. Five combinations were used in this study, with one reference and four alternative composite mixes. In composite mixes, a suitable amount of gypsum was added to ensure the ideal sequence of chemical reactions (calcium silicate reaction followed by aluminate reaction). Chemical composition and physical properties of each material are given in Table 2, while mix design details are given in Table 3. The 0-4 mm fine aggregate was used for mortar and the water binder ratio was taken to be 0.50. All mortar specimens were mixed in Hobart mixer as per the EN 196-1 [27]. After casting, specimens were demoulded after 24 hours and stored in a humidity chamber (RH maintained at 95%) until testing.

**Table 2. Chemical and physical properties of each material**

| Compound | CEM I | Silica fume | Fly ash | Calcined clay - A (CCA) | Calcined Clay - B (CCB) |
|---|---|---|---|---|---|
| $SiO_2$ | 19.32 | 92.02 | 53.28 | 63.70 | 62.41 |
| $Al_2O_3$ | 4.86 | 1.68 | 19.11 | 19.53 | 21.35 |
| $Fe_2O_3$ | 2.94 | 0.45 | 9.05 | 6.80 | 7.26 |
| CaO | 64.04 | 3.06 | 11.52 | 2.57 | 2.17 |
| MgO | 1.83 | 0.77 | 2.78 | 2.34 | 1.78 |
| $SO_3$ | 2.75 | 0.27 | 1.48 | 0.12 | 0.07 |
| Kaolinite | -- | -- | -- | 13.4 | 17.6 |
| Density [ g/cm$^3$] | 2900 | 2200 | 2400 | 2200 | 2500 |
| Specific surface area [m$^2$/g] | 19834 | 137135 | 11694 | 3193 | 2984 |

**Table 3. Mix details**

| Mix | CEM I, [kg/m$^3$] | Silica fume [kg/m$^3$] | Fly ash [kg/m$^3$] | Calcined clay – A, [kg/m$^3$] | Calcined clay – B, [kg/m$^3$] | Water [kg/m$^3$] | Fine aggregate [kg/m$^3$] |
|---|---|---|---|---|---|---|---|
| CEM I | 450 | -- | -- | -- | -- | 225 | 1350 |
| SF10 | 405 | 45 | -- | -- | -- | 225 | 1350 |
| FA30 | 315 | -- | 135 | -- | -- | 225 | 1350 |
| CCA30 | 315 | -- | -- | 135 | -- | 225 | 1350 |
| CCB30 | 315 | -- | -- | -- | 135 | 225 | 1350 |

## 3.2 Methods

Three mortar prism specimens measuring 160 mm × 40 mm × 40 mm were prepared for each mix for the evaluation of compressive strength after 2, 7, 28, and 56 days according to EN 196-1. Electrical resistivity of each specimen was measured by the Wenner 4-probe resistivity meter on the side of saturated cylinder specimen measuring 100 mm in diameter and 200 mm in height [28]. Three measurements were taken at four different locations on the curved surface of specimen at two different specimens of each mix. All measurements were taken on fully saturated specimens so that resistivity values are not affected by drying. The resistance to chloride ingress was assessed after 7, 28, and 56 days of curing on three cylinder specimens measuring 50 mm in thickness using NT BUILD 492 [29] test method. This test is used to evaluate the non-steady state chloride migration coefficient of cementitious materials. In this method, an external electrical potential was applied axially across the specimen throughout the test. After the end of the test, the specimen was split axially and silver nitrate solution was sprayed to the freshly split section. Later, the chloride penetration depth was visible as the white silver chloride penetration, and the depth was measured using digital Vernier callipers. Chloride migration coefficient was calculated based on this penetration depth, applied voltage, and test duration.

# 4 Test results and discussions

## 4.1 Compressive strength

Fresh properties of all mixes are given in Table 4. All these mixes showed good workability, and their flow table diameters ranged from 146 to 166.5 mm. The compressive strength evolution of five mixes in 56 days is shown in Figure 3, while relative strength of each mix with respect to CEM I is presented in Figure 4. Compared to the control specimen, the mixes with calcined clays, CCA30 and CCB30, achieved 70% and 87% strength, respectively, within 28 days. Such results indicate that calcined clays are highly reactive in early ages. Regardless of the lower binder content in CCB30 and FA30, both mixes showed good compressive strength compared to CEM I after 28 days. Also, CCB30 revealed slightly higher strength compared to SF10 after 56 days. Unexpectedly, the compressive strength of silica fume mix showed less strength than the control mix, as was revealed by micro structural analysis. Though there was no significant addition in compressive strength at early ages, FA30 specimens showed better strength-gain behaviour in extended curing by prolonged pozzolanic reaction [30]. CCA30 gave average strength compared to CEM I, and it could be recommended for the use of ordinary types of concrete.

**Table 4. Fresh properties of five mixes**

| Mixes | Flow diameter, mm | Temperature [° C] | Density [kg/m³] | Air content [%] |
|---|---|---|---|---|
| CEM I | 160 | 18.7 | 2362 | 4.1 |
| SF10 | 157 | 19.7 | 2336 | 2.2 |
| FA30 | 146 | 19.6 | 2270 | 2.7 |
| CCA30 | 166.5 | 19.6 | 2310 | 2.5 |
| CCB30 | 165 | 19.5 | 2325 | 2.4 |

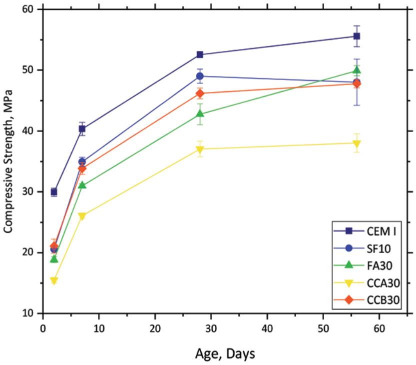

Figure 3. Compressive strength evolution of all mixes

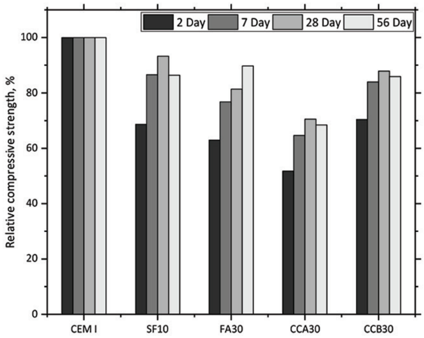

Figure 4. Strength of all mixes relative to CEM I mix

## 4.2 Surface Electrical Resistivity

The surface resistivity is employed as a quick indication of the resistivity of bulk concrete and is often used as a tool for determination of concrete quality and optimization of concrete mixes [31]. In this study, electrical resistivity of each mix was measured at 7, 28, and 56 days of curing, Figure 5. Results showed that the addition of silica fume gave enhanced resistance for the mixture, which is potentially due to denser microstructure of specimens, as caused by filler effect and additional pozzolanic reaction [32]. All other mixes showed better surface resistance values compared to the reference mix.

## 4.3 Resistance to chloride penetration

The non-steady state diffusion coefficient was measured at each mix according to NT Build- 492 at 7, 28, and 56 days. In the cases of calcined clay mixes, Figure 4 proves that migration coefficient values are comparable to CEM I values after 28 days and 56 days. As seen in compressive strength data, calcined clays specimens attained their maximum value within 28 days, whereas FA30 and SF10 mixes gained their maximum during extended curing. This is due to higher reactivity of calcined clays, and accelerated microstructure development at an early age of curing [33]. It is evident from Figure 6 that the mixes with silica fume and fly ash show reduced chloride ingress after each testing age. Though all mixes showed high migration coefficient values after 7 days, chloride ingress was reduced by 60% after 28 days, and by 70% at 56 days in SF10 mix, and by 64% and 66% in FA30 mix.

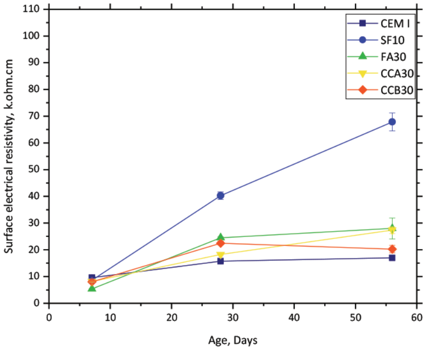

**Figure 5. Evolution of electrical surface resistivity of each mix**

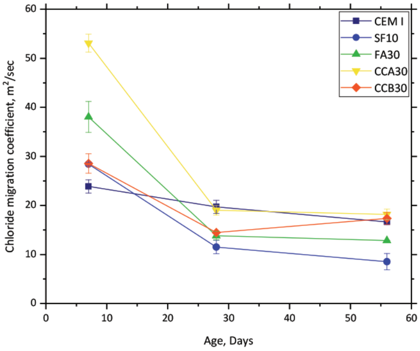

**Figure 6. Evolution of chloride migration coefficient by NT BUILD 492 method**

# 5 Conclusion

It is evident from literature that alternative SCMs such as fly ash and natural clays could be of interest for making high-performance concrete. Besides potential benefits in particle packing density, filling effect, and pozzolanic activity, they could prove advantageous to sustainable and economic aspects of HPC production.

As to availability of materials, calcined clays showed potential as alternative cementitious material in high-performance concrete. Mortar made with two different calcined clays with 30% replacement of cement showed results comparable to CEM I mortar in strength development and chloride migration coefficient after 28 days. The reactivity of calcined clays at early ages was higher compared to fly ash and silica fume. Other mixes showed enhanced properties in extended curing. The next phase of the project will involve mix optimisation for the high-performance concrete with calcined clays and fly ash.

## Acknowledgements

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