CSA-BASED PORTLAND-FREE BINDERS TO MANUFACTURE SUSTAINABLE CONCRETES FOR JOINTLESS SLABS ON GROUND

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ABSTRACT

The paper is aimed to the evaluation of the influence of water/binder ratio, tartaric acid dosage and curing condition on rheological, elastic and physical properties of sustainable shrinkage-compensating concretes manufactured with calcium sulphoaluminate cement (CSA), anhydrite ($C\overline{S}$) and supplementary cementitious materials (SCMs)/lime (CH) compound in place of ordinary Portland cement (OPC). Results indicated that tartaric acid-based set-retarding admixture governs the behavior of concrete both in fresh and hardened state. In addition, according to Abram's model, results evidenced the water/binder ratio as a key factor in strength gain. Moreover, tartaric acid allows the production of shrinkage-compensating Portland-free concretes particularly indicated for slabs on ground. Finally, by replacing OPC with SCMs and lime, it is possible to obtain, both for CO_2-emissions and energy consumption, a reduction up to 60% at equal strength class respect to an OPC-based concrete.

KEYWORDS

Concrete slabs; Calcium sulphoaluminate cement; Portland cement; Supplementary cementitious materials; Sustainability.

INTRODUCTION

Nowadays, reinforced concrete slabs on grade are increasingly present both in infrastructures and in residential or industrial buildings. Unfortunately, in many cases these elements suffer from severe damage due to a poor design, a wrong materials selection and/or an inaccurate concrete casting and curing [1]. Drying shrinkage is one of the common causes of cracking and curling of concrete slabs-on-ground, also because these structures possess an high ratio between the surfaces exposed to the air and the concrete volume [2]. The high shrinkage typical of these slabs, in the presence of internal and external constraints (such as reinforcing bars, floor foundation or other structural elements), determines notable internal tensile stress [3,4]. Cracking can be avoided only if tensile stress induced by shrinkage, reduced by creep, is always lower than the tensile strength of concrete. The cracking risk limitation can be achieved through a proper mix design (reducing the cement factor and increasing both the maximum size of aggregates and the dosage of superplasticizer) [5–8], adequate placing and curing [9] and by using high stiffness natural or artificial aggregates [10,11].

Use of expansive or shrinkage-compensating concrete (EC), although more expensive than Portland cement-based mixtures, is valuable in concrete structures where a reduction in cracking is of crucial importance, such as in pavement slabs, bridge decks and liquid storage tanks. This technique is based on the early restrained expansion that occurs between the expansive agents and water [12–14]. Generally, EC are manufactured with expansive agents that lead to the formation of ettringite ($C_3A \cdot 3C\overline{S} \cdot H_{32}$) or calcium hydroxide (CH) according to the following reactions:

$$C_4A_3\overline{S} + 6C + 8CS + 96H \Rightarrow 3\, C_3A \cdot 3C\overline{S} \cdot H_{32} \qquad \text{(I)}$$

$$C + H \Rightarrow CH \qquad \text{(II)}$$

However, several authors [15,16] and standards [17,18] show that EC can be advantageously used in reinforced concrete slabs-on grade without control joints only if an adequate wet curing is ensured. In particular, depending on the nature of expansive agent, 2- or 7-day wet curing period is needed. Otherwise, use of expansive agents is totally unsuccessful.

Collepardi et al. [19] and Maltese et al. [20] showed that the combined addition of an ethylene glycol-based shrinkage-reducing admixture (SRA) with a CaO-based expansive agent seems to have beneficial effects on concrete shrinkage even in absence of wet curing. On the other hand, a wrong choice of the type and dosage of expansive agent can lead to an inadequate expansion and, therefore, to crack formation in concrete slabs [21].

Another effective method to produce EC involves the use of expansive binders, alternative to Portland cement, based on a controlled production of ettringite. Between these special binders, ternary mixtures based on calcium sulphoaluminate cements (CSA), Portland cement (OPC) and gypsum ($CSA:OPC:C\overline{S}$) are certainly the most widespread [22]. Recently, Coppola et al. [23] showed the possibility to manufacture environmentally friendly shrinkage-compensating mortars using CSA-based ternary mixtures in which OPC is totally replaced by supplementary cementitious materials (SCMs, such as fly ash and ground granulated blast furnace slag) and lime (CH). In particular, the experimental data showed the primary role of tartaric acid in the expansive behavior of Portland-free CSA-based mixtures. In fact, dosages between 0.4% and 1.2% of tartaric acid vs binder mass guarantee a quite stable behavior over time (free shrinkage lower than 500 μm/m after 270 days at 20°C and 60% R.H.), without affecting negatively mechanical performances. On the other hand, as opposed to OPC-based concretes, Portland-free CSA-based mortars evidenced higher compressive strength values when cured in dry environment respect to those measured on specimens stored under water. This behavior has strong consequences on job-site operations and could make unnecessary wet curing operations (often not done or carried out wrongly). Finally, from an environmental point of view, many authors have shown the beneficial effects deriving from the use of SCMs/lime [24–27] replacing Portland cement, reaching up to 60% reduction in CO_2 emissions and energy requirements to produce 1 cubic meter of concrete, at equal 28-day strength class.

The purpose of this paper is the evaluation of rheological, elastic and physical performances of shrinkage-compensating Portland-free concretes (for slabs on grade without control joints) manufactured with CSA:SCM:CH:C$\overline{S}$ and tartaric acid-based set-retarding admixture at different water/binder ratios.

MATERIALS AND METHODS

Materials

A commercial CSA clinker, ordinary Portland cement (OPC) type I 52.5 R (EN 197-1 compliant) and technical grade anhydrite (C$\overline{S}$) were used in this study to manufacture the reference shrinkage-compensating concretes (CSA:OPC:C$\overline{S}$ = 40:40:20). Ground granulated blast furnace slag (S: according to EN 15167-1), type V (according to EN 450-1 and EN 197-1) low calcium siliceous fly ash (FA) and hydrated lime (CH) CL90-S (according to EN 459-1) were employed to replace totally OPC in environmentally friendly mixtures (CSA:SCM:CH:C$\overline{S}$ = 40:35:5:20). The physical properties and the environmental parameters (Gross Energy Requirements: GER and Global Warming Potential: GWP) of binders were reported in Table 1. Furthermore, four different types of natural calcareous aggregates (maximum diameter equal to 32 mm) were combined to meet the Bolomey curve (Equation I, Figure 1 and Table 2).

$$P = \left[A - C + (100 - A)\sqrt{\frac{d}{D}}\right] \cdot \left(\frac{100}{100 - C}\right)$$

With P: percentage passing
A: empirical coefficient based on workability of concrete and shape of aggregates
C: ratio between cement factor and cement + aggregates mass
D: maximum size of aggregates

Tartaric acid-based set-retarding admixture was added up to 0.6% with respect to binder mass in order to control the expansive behavior and the workability loss over time. Finally, the mixing water was fixed equal to about 200 kg/m^3 to achieve the consistency class S4 (EN 12350-5) and the water/binder ratio was varied between 0.55 and 0.70. Composition of concretes are reported in Table 3.

Table 1 – Physical and environmental properties of binders (* Source: Ecoinvent 3.0 Databased)

	OPC	**CSA**	**$C\bar{S}$**	**CH**	**S**	**FA**
D$_{50}$ [µm]	5.19	8.18	2.93	3.00	5.48	11.1
Specific surface [cm^2/g]	3175	2722	4837	4678	3049	2283
Specific mass [kg/m^3]	3150	2650	2670	2120	2730	2010
GER [MJ/kg] *	5.50	2.70	1.30	4.50	0.31	0.10
GWP [kg CO$_2$/kg] *	$9.8 \cdot 10^{-1}$	$7.4 \cdot 10^{-1}$	$2.4 \cdot 10^{-1}$	$4.2 \cdot 10^{-1}$	$1.7 \cdot 10^{-2}$	$5.3 \cdot 10^{-3}$

Table 2 – Min/max size**,** water absorption and specific mass (EN 1097-6) of natural aggregates

	Fine sand S	**Fine gravel G1**	**Coarse gravel G2**	**Coarse gravel G3**
Diameter min/max [mm]	0 / 6	6 / 12	10 / 20	20 / 30
Water absorption [%]	1.69%	2.12%	1.62%	1.16%
Specific mass [kg/m^3]	2550	2660	2680	2650

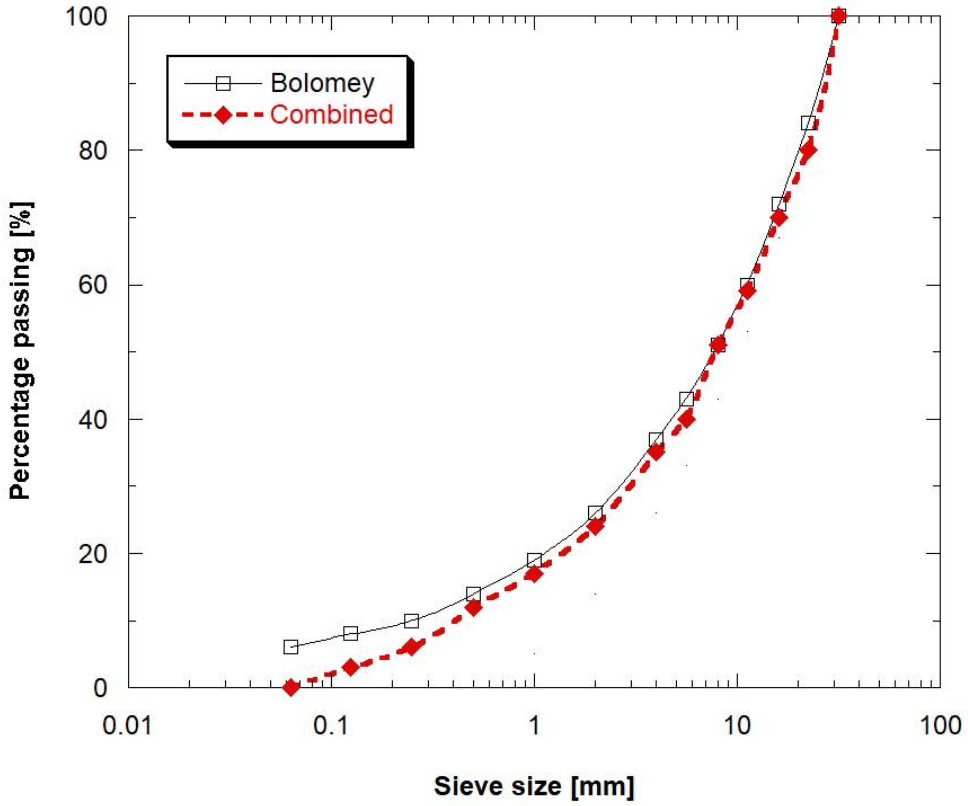

Figure 1 – Bolomey's and combined aggregate grading curves

Table 3 – Composition and fresh properties of concretes

Ingredients [kg/m^3]	**CSA**	**CEM I 52.5 R**	**$C\bar{S}$**	**CH**	**S**	**FA**	**Aggregates**	**Water**	**Tartaric acid**	**Specific mass in fresh state** [kg/m^3]	**Entr. air** [%]
RC 0.55-0.4	142	142	72				1788	196	2.20	2340	1.5%
RC 0.60-0.4	132	132	66				1818	197	2.22	2345	1.5%
RC 0.65-0.4	122	122	61				1845	199	2.23	2350	1.4%
RC 0.70-0.4	113	113	57				1857	199	2.23	2340	1.5%
S 0.55-0.4	142		71	18	123		1776	195	2.18	2325	1.3%
S 0.60-0.4	131		65	16	115		1806	196	2.20	2320	1.0%
S 0.65-0.4	121		60	15	105		1821	197	2.21	2330	1.1%
S 0.70-0.4	113		57	14	98		1845	198	2.22	2325	1.1%
FA 0.55-0.4	142		71	18		124	1780	195	2.19	2330	0.9%
FA 0.60-0.4	131		65	16		115	1802	196	2.20	2325	0.8%
FA 0.65-0.4	121		60	15		106	1825	197	2.21	2325	0.8%
FA 0.70-0.4	113		57	14		98	1849	198	2.22	2330	0.8%
RC 0.55-0.6	142	142	72				1788	196	3.35	2340	1.5%
S 0.55-0.6	143		72	18	125		1799	197	3.37	2320	1.4%
FA 0.55-0.6	142		71	18		124	1780	195	3.33	2330	0.9%

Tests on concretes

Fifteen concretes were manufactured according to EN 12390-2. At the end of the mixing procedure, workability was measured over time (at 0, 30, 60, 90, and 180 minutes from mixing) by means of Abram's cone according to EN 12350-5. In addition, specific mass and entrapped air were evaluated on fresh concretes according to EN 12350-6 and EN 12350-7 standards. Specimens were produced and cured (Table 4) both under water at 20°C (W) and in a climatic chamber at 20°C and R.H. 60% (D). Specific mass and compressive strength at 1, 7 and 28 days were also determined (EN 12390-3). In addition, only for mixture containing 0.6% of tartaric acid, free and restrained shrinkage/expansion were measured up to 56 days on specimens stored both under water at 20°C (W) and in dry environment (D: 20°C, R.H. 60%) according to EN 11307 and EN 8148, respectively. Finally, tensile strength on 28-day cured cylindrical specimens (according to EN 12390-6), elastic modulus (in accordance with method B, EN 12390-13) and water penetration under pressure (according to EN 12390-8) were measured.

Table 4 – Specimens manufactured for each concrete

Test	Ages	Curing conditions	Format specimens	Number of specimens	Note
Compressive strength	1-7-28 days	W - D	Cube 100 mm	18	3 specimens for each age and curing condition
Tensile strength	28 days	W – D	Cylinder h/d : 2 d : 100 mm	6	3 specimens for each curing condition
Elastic modulus	28 days	W – D	Cylinder h/d : 2 d : 150 mm	6	3 specimens for each curing condition
Water penetration	28 days	W – D	Cube 150 mm	6	3 specimens for each curing condition
Free shrinkage/expansion	up to 56 days	W – D	Beam 100x100x500 mm	6	3 specimens for each curing condition
Restrained shrinkage/expansion	up to 56 days	W – D	Beam 80x80x240 mm	6	3 specimens for each curing condition

W: curing under water at 20°C – D: curing in climatic chamber at 20°C and 60% R.H.

RESULTS AND DISCUSSIONS

Workability at the end of the mixing procedure remains almost constant independently of the water/binder ratio by using 0.4% of tartaric acid dosage with respect to binder mass (Figure 2-4). In particular, reference concretes (RC) and mixtures manufactured with slag (S) show an initial slump equal to 200 mm, reaching the consistency class S2 (100 mm slump) after about 60 minutes. On the contrary, FA-based concretes, at the same initial consistency class, evidenced a lower workability loss over time, achieving the consistency class S2 30 minutes later than the references (RC) and S concretes (S2 after 90 minutes from casting). According to Coppola et al. [23] the tartaric acid dosage strongly influences the slump of concretes. A general increase in the initial workability (more marked in FA-based mixtures than those containing OPC and S) and a reduction in workability loss over time are observed by using 0.6% tartaric acid with respect to binder mass. In detail, reference and S concretes (RC) reach the consistency class S2 after about 120 and 90 minutes, respectively. On the contrary, mixtures based on fly ash (FA) show an excellent maintenance of workability over time, reaching the S2 consistency class after about 180 minutes. In general, it is possible to conclude that, for practical uses, OPC- or S-based concretes require greater set-retarding admixture dosage (0.6% by binder mass) than that (0.4% by binder mass) needed for FA mixtures.

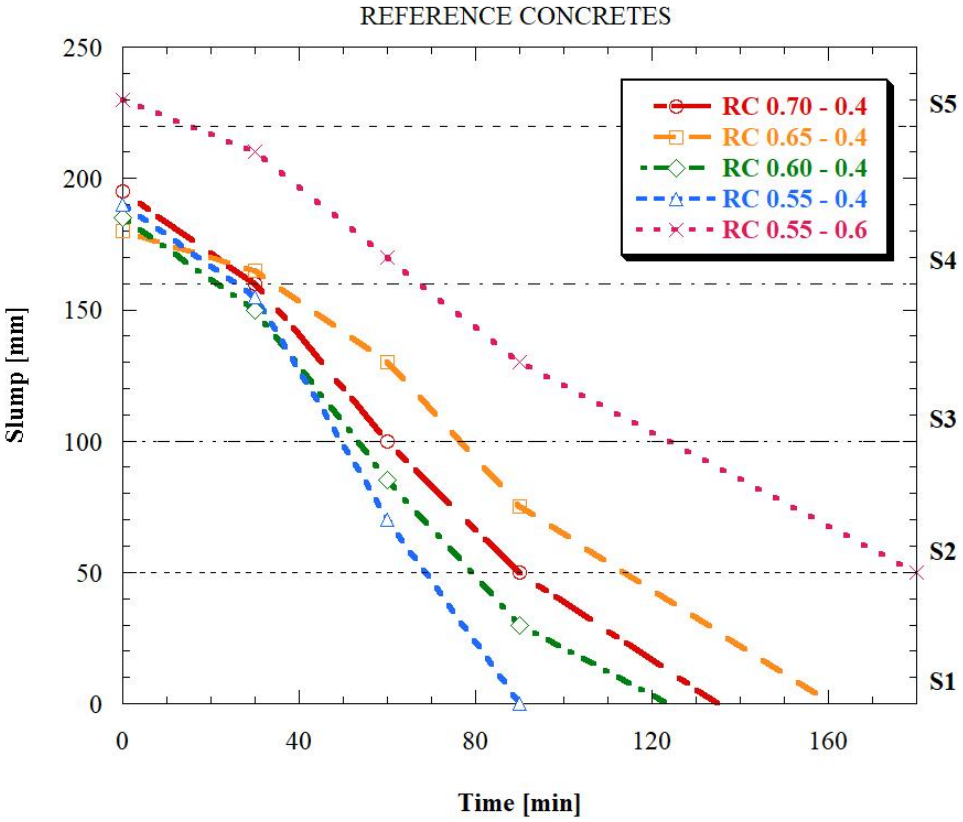

Figure 2 – Workability vs time of reference concretes (RC)

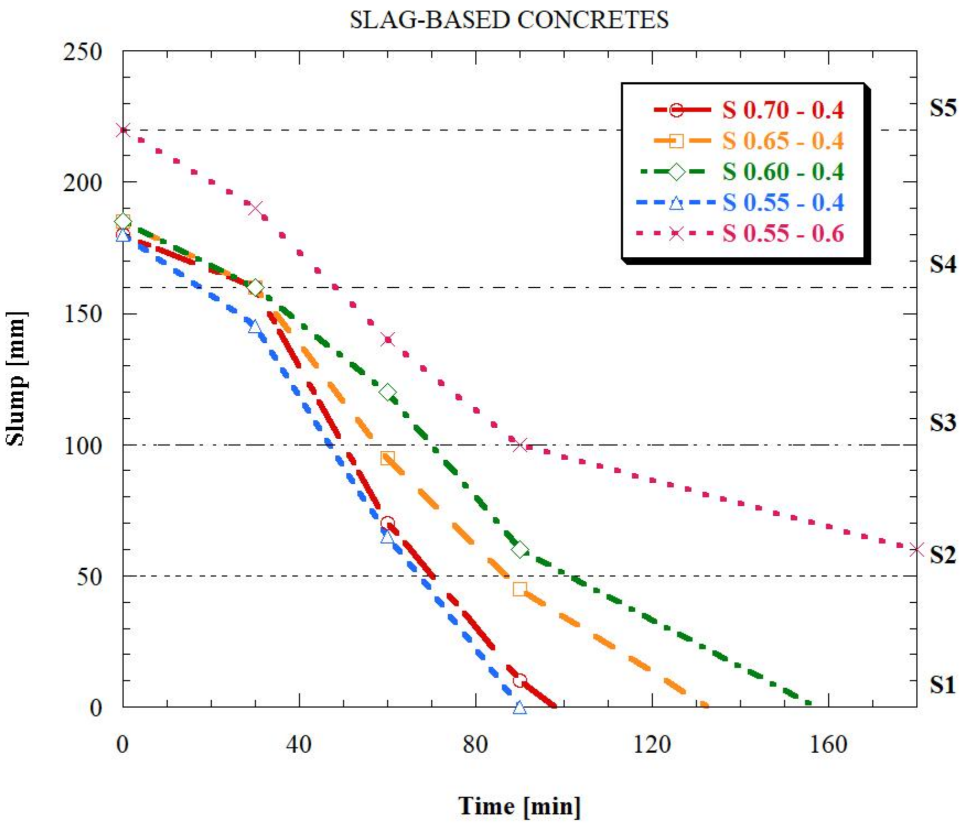

Figure 3 – Workability vs time of slag-based concretes (S)

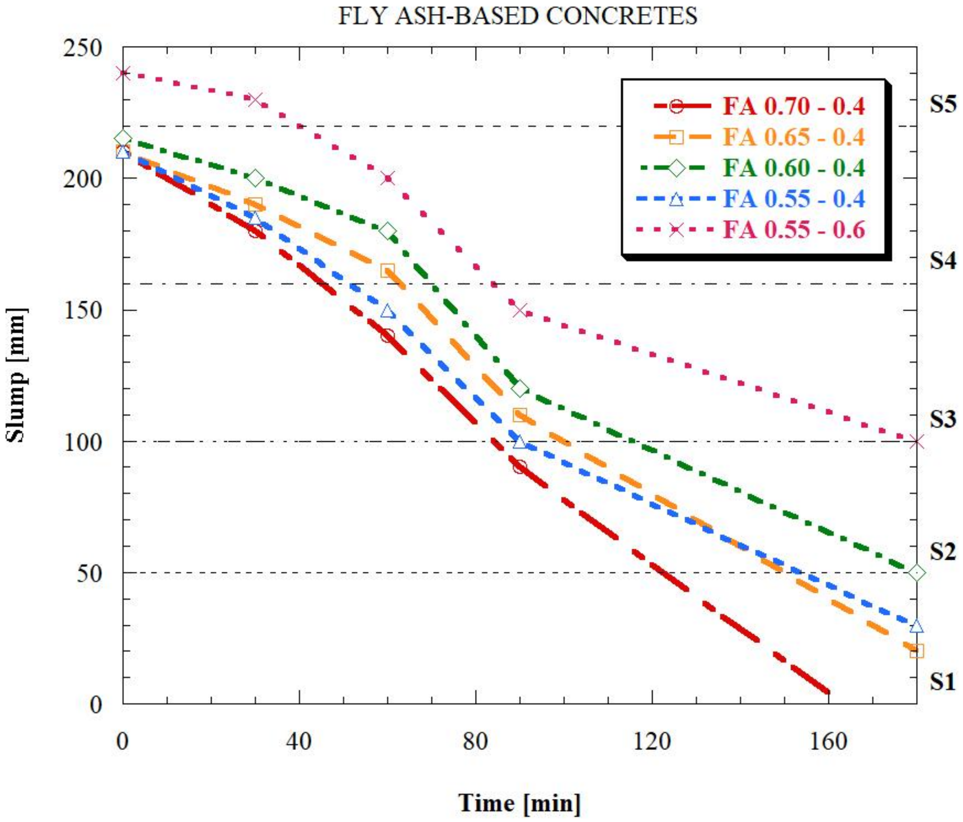

Figure 4 – Workability vs time of fly ash-based concretes (FA)

Moreover, variation in water/binder and tartaric acid dosage does not determine substantial changes of entrapped air (always between 0.8% and 1.5% by concrete volume) and specific mass in the fresh state. In particular, density is close to 2340 kg/m^3 for reference concretes (RC) while it attains values close to 2325 kg/m^3 for mixtures in which SCMs/lime have totally replaced ordinary Portland cement. On the contrary, the increase in water/binder ratio leads to a linear decrease in 28-day specific mass, independently of tartaric acid dosage and type of binder (Figure 5).

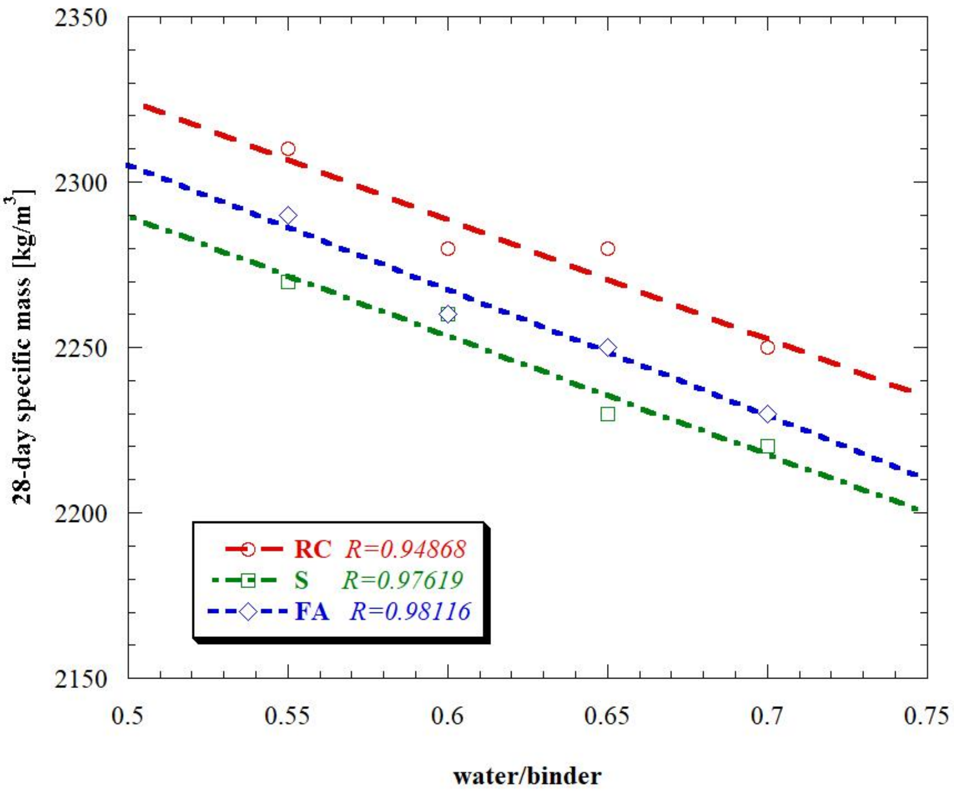

Figure 5 – 28-day specific mass vs water/binder ratio

Concerning compressive strength measured on cubic specimens cured under water, it is possible to note that the water/binder is a key factor (Figure 6-8). Indeed, similarly to Portland cement concretes [28], low w/b allows to obtain mixtures of excellent strength properties while increasing this parameter results in a general worsening of mechanical performances, regardless of binders employed and the age of concrete. Moreover, replacing OPC with hydrated lime and SCMs, negligible changes in 24-hour strength are noted. On the contrary, 30% reduction in compressive strength at 7 and 28 days were measured, independently of w/b. However, SCM-based concretes with w/b ratio from 0.55 to 0.70 exhibit 28-day compressive strength (25-40 MPa) suitable for reinforced slabs on grade.

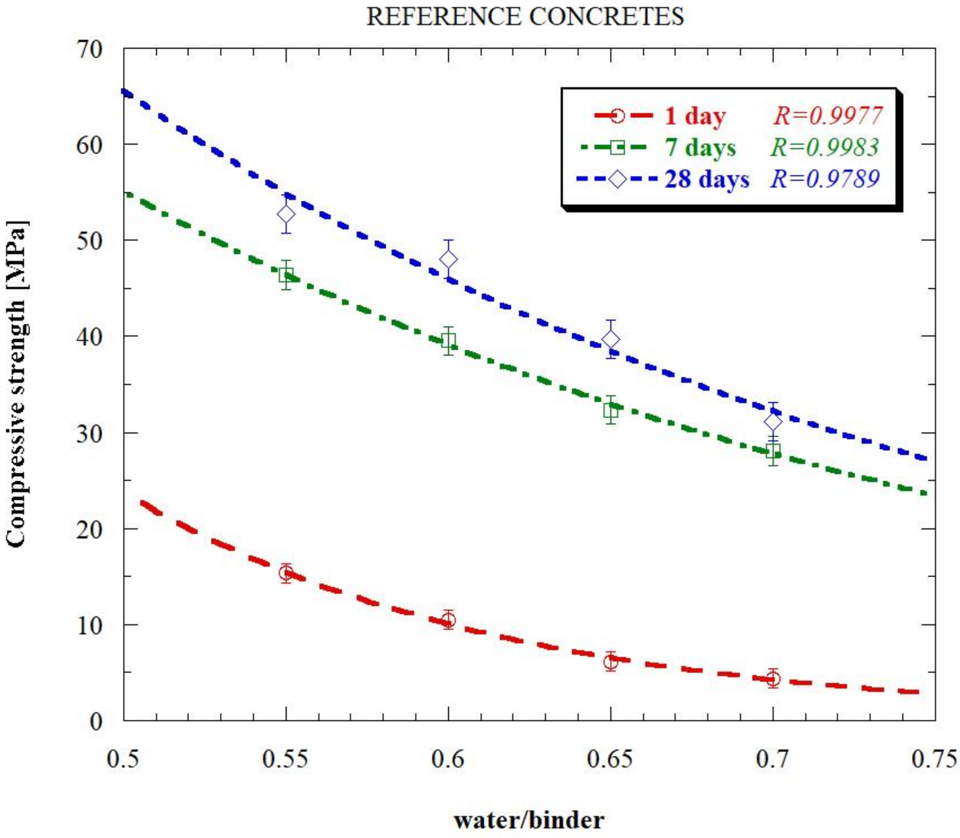

Figure 6 – Compressive strength of reference concretes (RC) vs water/binder ratio (wet curing)

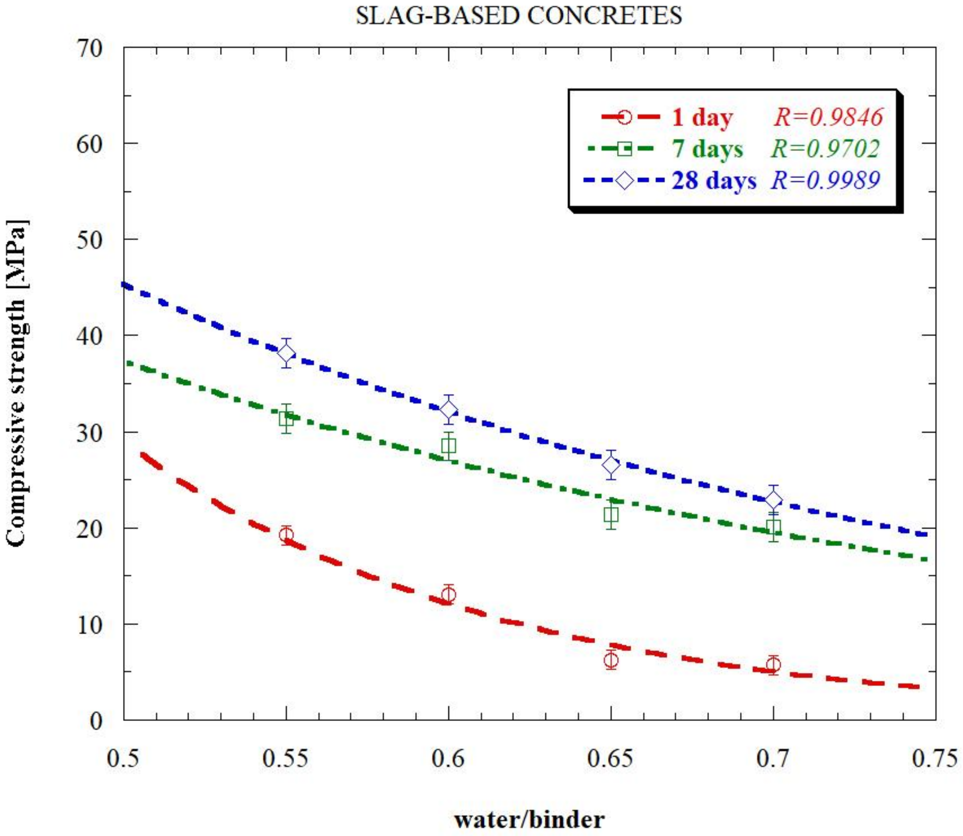

Figure 7 – Compressive strength of slag-based concretes (S) vs water/binder ratio (wet curing)

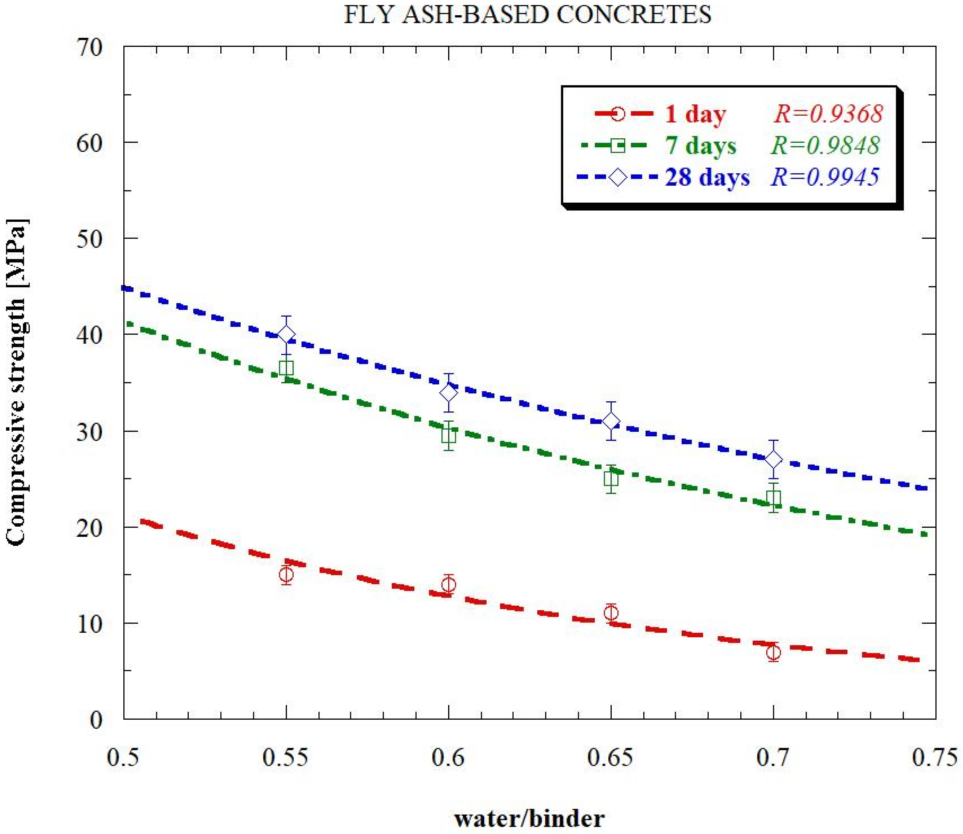

Figure 8 – Compressive strength of fly ash-based concretes (FA) vs water/binder ratio (wet curing)

Experimental data were used to determine the parameters A and B of Abram’s model (III) to estimate the compressive strength at 28 days of concrete manufactured with 0.4% vs binder mass of tartaric acid and cured underwater at 20°C:

$$f_{c,28} = {}^{A_{28}}\!\big/_{B_{28}^{x}} \qquad \text{(III)}$$

where ($f_{c,28}$) is the concrete compressive strength at 28 days, (A_{28}) and (B_{28}) are experimental parameters depending on the mixture composition and (x) is the water/binder ratio [29,30]. Results in Table 5 and Figure 9 show that concretes based on SCMs and lime have a mechanical behavior similar to that shown by traditional concretes manufactured with CEM I 52.5 R or CEM II/A-LL 42.5 R. On the contrary, compressive strength of reference mixtures CSA:OPC:$C\overline{S}$ is more affected by w/b ratio, even if at equal w/b ratio, compressive strength is significantly higher than that exhibited by CEM I 52.5 R mixtures. Finally, it should be noted that, by using sustainable CSA-based mixtures manufactured with FA or S and lime, it is possible to reach similar mechanical strength to those obtainable, at equal w/c ratio, with a traditional limestone Portland cement (CEM II/A-LL 42.5 R).

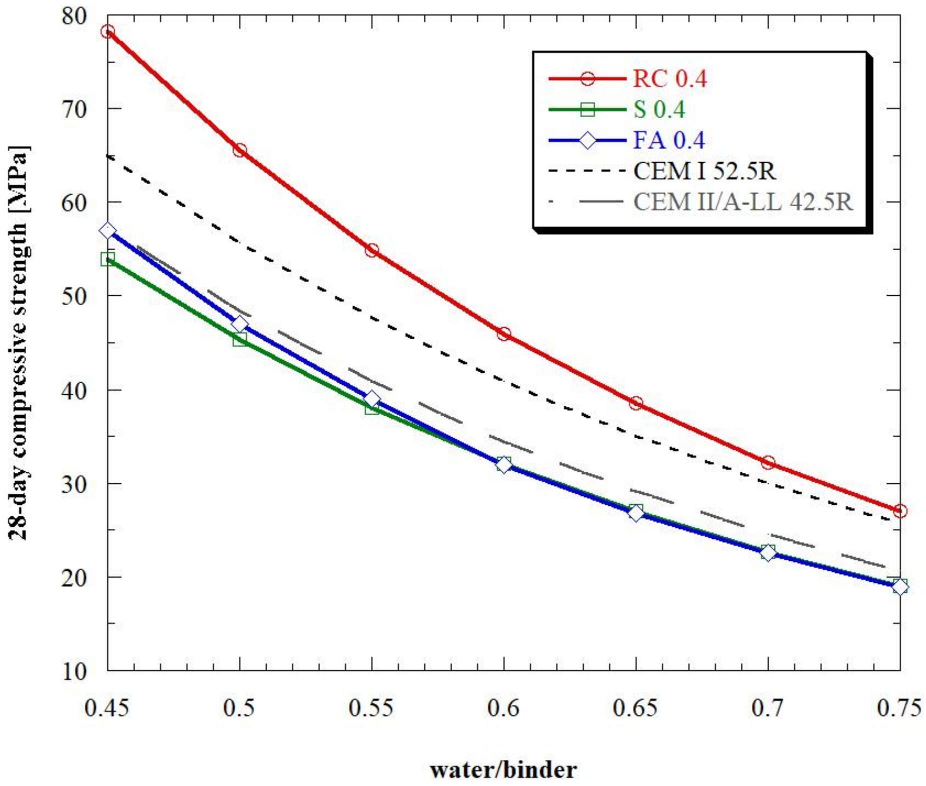

Figure 9 – 28-day compressive strength of concrete manufactured with different binders vs water/binder ratio (Abram's model, wet curing)

Table 5 – Coefficient of Abram's model for different mixtures

	RC 0.4	S 0.4	FA 0.4	CEM I 52.5 R	CEM II/A-LL 42.5 R
A_{28}	386.61	255.31	261.47	261.25	263.32
B_{28}	34.78	31.72	33.15	22.00	29.61

Also, the curing conditions strongly influence the mechanical properties of CSA-based concretes (Figure 10). In fact, the reference concrete (RC) cured in dry environment (T = 20°C, R.H. 60%) exhibited compressive strength approximately 15% higher compared to that of the same mixture cured under water. Concrete manufactured with SCMs/lime replacing OPC showed more marked differences, up to 30%, between wet and dry cured specimens. Furthermore, increasing the tartaric acid dosage up to 0.6% vs binder mass, all concretes (both references and those containing SCMs/lime replacing OPC) evidenced a general reduction in mechanical performances up to 25% both at early and long ages (Figure 11).

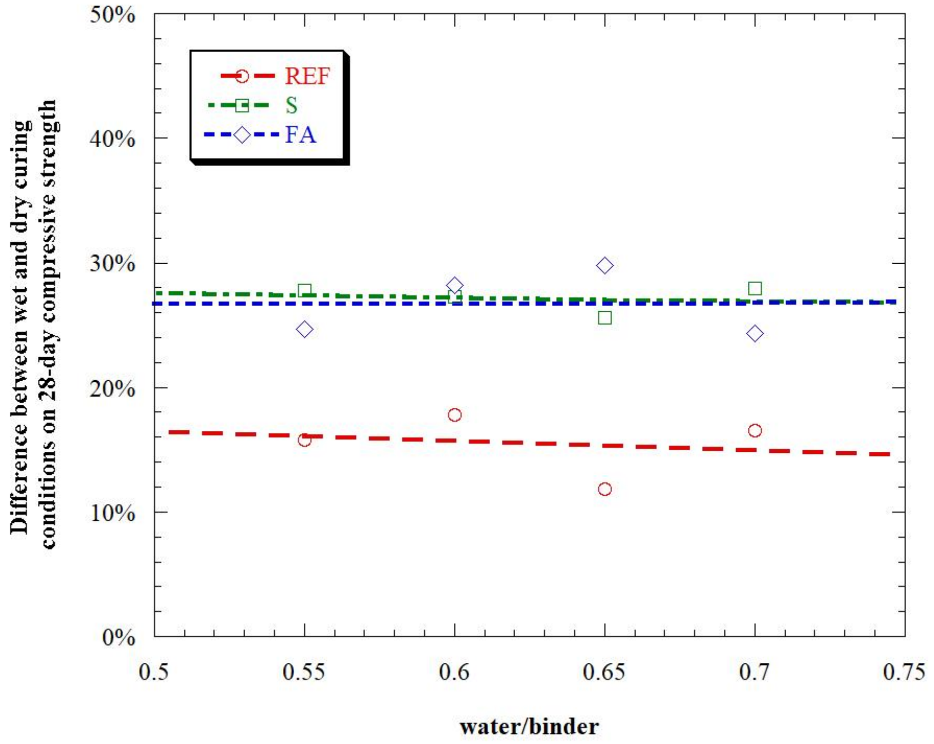

Figure 10 – Difference between wet and dry curing conditions on compressive strength at 28 days vs water/binder ratio (linear correlation)

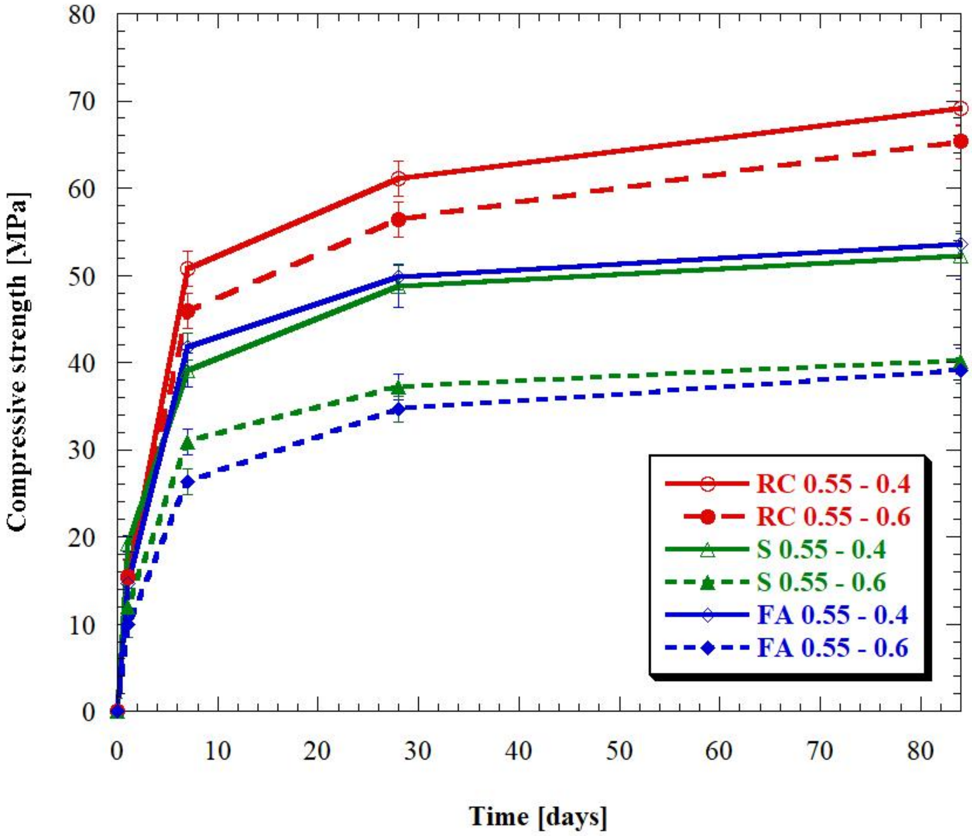

Figure 11 – Development of compressive strength over time on concretes (w/b=0.55) manufactured with different tartaric acid dosage (dry curing)

Total replacement of OPC with supplementary cementitious materials/lime and the underwater curing conditions determine a general worsening of both elasto-mechanical properties and watertightness of concretes. In FA- and S-based concretes, tensile strength decreases up to 40% compared to the reference mixtures (RC), independently of the curing conditions (wet or dry). However, tensile strength of CSA-based concretes (Figure 12) follows the equation proposed by Eurocode 2 (EN 1992-1-1) for ordinary Portland cement concretes (strength class lower than C50/60):

$$f_{ctm} = 0.30 \cdot f_{ck}^{2/3} \qquad \text{(IV)}$$

Young's modulus decreases, at the same w/b, replacing Portland cement with SCMs/lime due to the reduction of compressive strength caused by using FA or S (Figure 13). Nevertheless, elastic modulus of concrete based on calcium sulphoaluminate cement can be well approximated by the following equation proposed by Eurocode 2:

$$E_{cm} = k \cdot \left(\frac{f_{cm}}{10}\right)^{0.30} \qquad \text{(V)}$$

with k depending on the mineralogical nature of aggregates used.

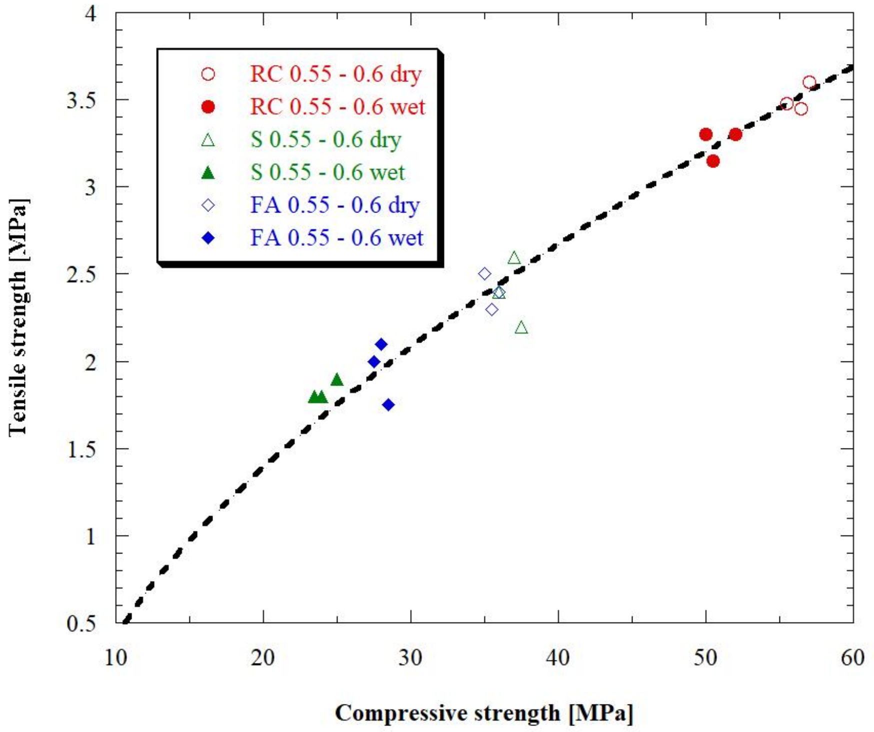

Figure 12 – Tensile strength of concrete vs 28-day compressive strength. In dash line, the correlation proposed by EC2 (IV)

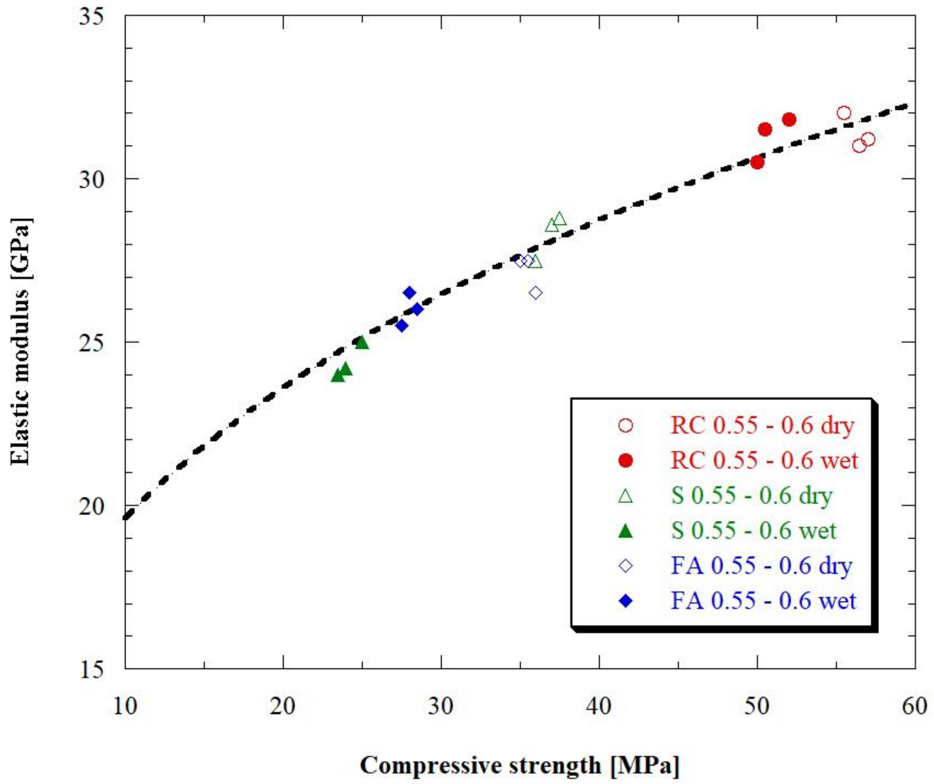

Figure 13 – Elastic modulus of concrete vs 28-day compressive strength. In dash line, the correlation proposed by EC2 (V)

Water penetration under pressure is influenced by the curing conditions of specimens. In general, concretes cured in dry environment show a lower water penetration respect to that of the same mixture stored underwater (Figure 14). This result is in good agreement with compressive strength data. Furthermore, water penetration in Portland-free concretes (S or FA) grows strongly compared to that detected for the reference mix (RC), independently of the curing conditions (wet or dry). In particular, water penetration in dry cured SCMs/lime based concretes was about 100 mm. This value is double compared to that of the reference mixture cured in the same conditions (D). In wet cured SCMs/lime mixtures water penetration was about 140 mm. This value is about two times and a half higher than the corresponding water penetration (60 mm) measured for the reference concrete (RC) containing OPC.

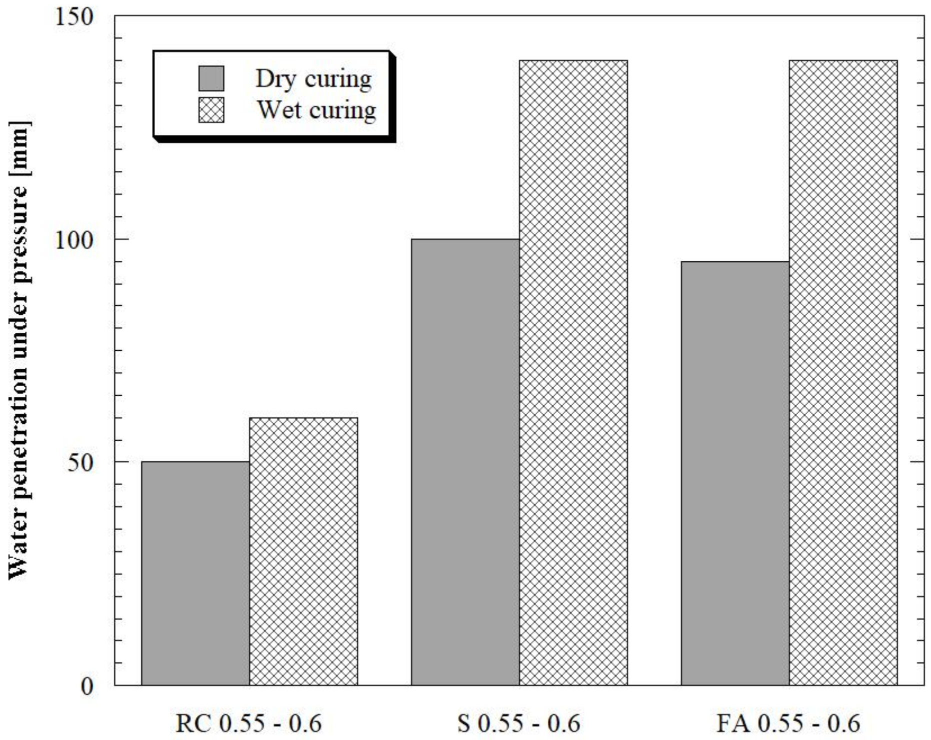

Figure 14 – Water penetration under pressure in different curing conditions (W or D)

Regardless of the binder used, shrinkage of CSA-based concretes is strongly influenced by curing conditions [31,32]. Indeed, free and restrained shrinkage tests show a stable behavior over time when specimens are stored in a climatic chamber at 20°C and 60% R.H (D). On the other hand, in concretes cured under water (W) an initial expansion was followed by a negligible shrinkage (Figure 15 -16). Total replacement of OPC with SCMs/lime modifies the shrinkage behavior of concretes. In fact, reference mixtures (RC) show more marked expansion underwater at early ages with respect to Portland-free concretes (S or FA), both in free and restrained conditions.

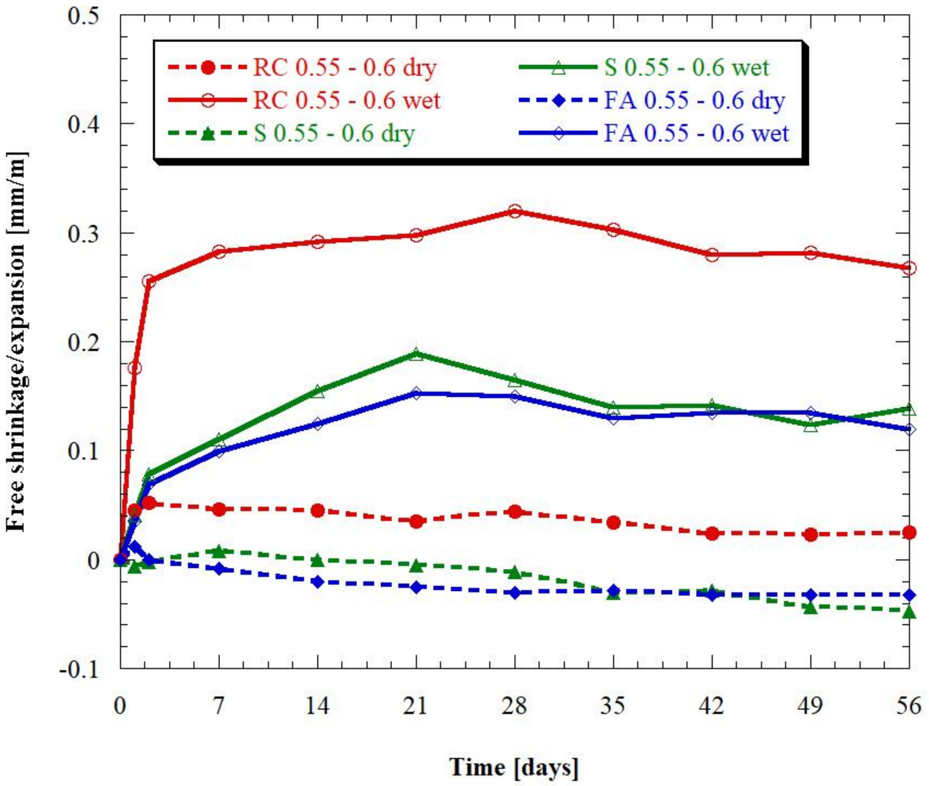

Figure 15 – Free shrinkage vs time in different curing conditions (positive values indicate expansion of concrete)

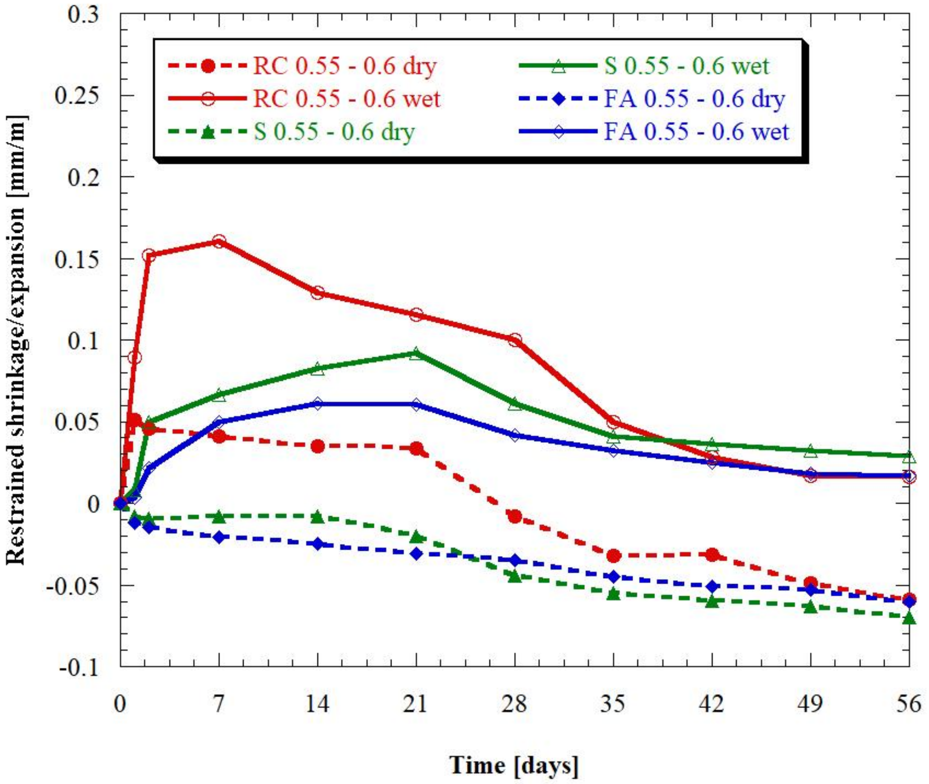

Figure 16 – Restrained shrinkage vs time in different curing conditions (positive values indicate expansion of concrete)

Figure 17 shows the environmental parameters GER (Gross Energy Requirement that correspond to the total energy necessary to produce 1 m^3 of concrete) and GWP (Global Warming Potential, related to the greenhouse gases emitted for 1 m^3 of cementitious mixture) for class C30/37 concretes manufactured with different type of binders calculated starting from the raw materials data reported in Table 1 (CEM II/A-LL 42.5R: 3.60 MJ/kg, $8.8 \cdot 10^{-1}$ kg CO_2/kg – aggregates: 0.13 MJ/kg, $2.4 \cdot 10^{-3}$ kg CO_2/kg). If the aim is to increase sustainability, reducing both the emissions of CO_2 (GWP) and the primary energy required (GER) for the production of one cubic meter of concrete, replacement of Portland cement type I with limestone Portland cement type II or with a ternary mixture, in which OPC and CSA are present in equal parts, is not a suitable solution to obtain a sharp reduction of the environmental impact in concrete production. In fact, improvements are rather limited, since reduction of GER and GWP is generally between 15% and 25%, due to both the high kiln temperatures required during Portland clinker production and the strong environmental impact of the extraction and grinding phase [33].

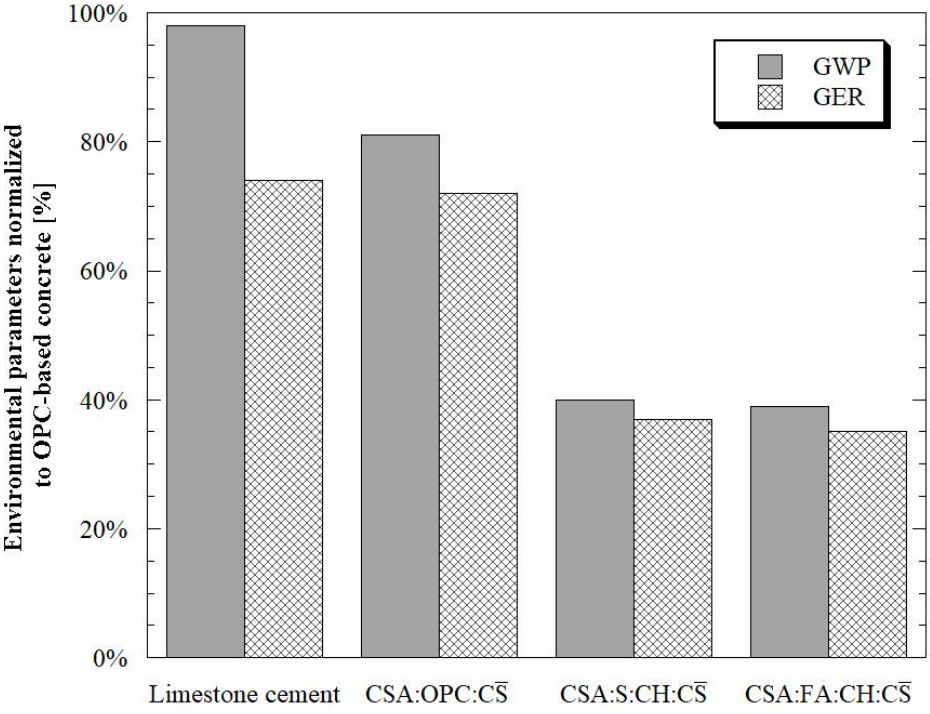

Figure 17 – GWP and GER parameters normalized to those of an OPC-based concrete at equal strength class C30/37

The best way to achieve a remarkable improvement in terms of sustainability is use of mixtures based on sulphoaluminate cement (CSA) in which OPC has been totally replaced by supplementary cementitious materials (SCMs) and hydrated lime (CH). In this case, it is possible to obtain, both for GHG emissions and consumption of energy, a reduction of about 60% at equal strength class due to the nature of the binders employed (generally wastes deriving from industrial process) that required limited processing before being used in mortars and concretes.

CONCLUSIONS

In this paper, the influence of water/binder ratio, dosage of a tartaric based set-retarding admixture and curing conditions on rheological, elastic and physical properties of environmentally friendly shrinkage-compensating concretes manufactured with calcium sulphoaluminate cement (CSA), anhydrite ($C\overline{S}$), lime (CH) and two different supplementary cementitious materials (fly ash: FA and slag: S) replacing totally ordinary Portland cement (OPC) was investigated. According to the experimental data, the following conclusions can be drawn:

- At equal mixing water, workability at the end of the mixing procedure (Figure 2-4) is not influenced by the type of the ternary binder and the water/binder ratio.
- The tartaric acid-based set-retarding admixture acts as a superplasticizer.
- OPC- or S- based mixtures require higher amount of tartaric acid-based set retarding admixture (0.6% vs binder mass) respect to that needed for concretes manufactured with FA (0.4% vs binder mass) in order to ensure a suitable workability retention.
- In general, by using Abram’s model, it is possible to note that Portland-free concretes have mechanical behavior close to that shown by traditional concretes manufactured with Portland cement or limestone Portland cement.
- Compressive strength values of reference mixtures (RC: CSA-OPC- $C\overline{S}$) are more affected by w/b ratio than those of Portland-free CSA-based concrete. However, reference concretes

(RC), independently of w/b, exhibited compressive strength values higher than those obtained for CEM I 52.5 R- based mixtures (Figure 9).

- Total replacement of OPC with supplementary cementitious materials and lime in underwater curing conditions determine a general worsening of elastic and mechanical properties (compressive and tensile strength, Young's modulus) and watertightness of concretes (Figure 10-14).
- Independently of binders employed, shrinkage of CSA-based concretes exhibit a stable behavior over time when specimens were cured at 20°C and 60% R.H. (D) while an underwater curing (W) determines an initial expansion of concretes followed by a negligible shrinkage (Figure 15-16).
- CSA-based concretes manufactured with SCMs and hydrated lime in place of OPC are very promising from an environmentally point of view since GER and GWP parameters decrease about 60% at equal strength class compared to traditional OPC or CSA-OPC-$C\overline{S}$ mixtures (Figure 17).

In future, durability issues of mortars and concretes manufactured with CSA-based Portland-free binders have to be thoroughly investigated, especially in chloride and sulphate-rich environments or in presence of freezing and thawing cycles. Finally, a great effort will be required to develop suitable admixtures – in particular superplasticizers – for these alternative blended binders.

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