SUSTAINABLE DEVELOPMENT AND DURABILITY OF SELF-COMPACTING CONCRETES

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ABSTRACT

Self-compacting concretes (SCC) represent a move toward a sustainable material since they encourage the use of waste and recycled materials. The high volume of very fine powder necessary to achieve deformability and passing ability properties, in fact, permits SCC to consume large amount of fly-ash, very fine particles generated by the recycling of demolished concrete structures, and huge amount of calcareous filler available from the marble quarries. Moreover SCC turn out to be materials with an extended durability with respect to conventional concretes. Since fresh properties of self-compacting concretes (SCC) are significantly different from those of conventional concretes (CC) durability can be significantly improved when a SCC is used due to a modification of the microstructure of the interfacial transition zone between aggregates and cement matrix.

This paper presents results of an experimental study carried out to evaluate changes in microstructure of interfacial transition zone (itz) and of bulk paste for both SCC and CC. Data on the influence of the calcareous filler, a fundamental ingredients to achieve self-compactability, on the hydration process of cement are also presented.

Data indicate that the decrease in internal bleeding, when self-compacting concrete is used, seems to favour the formation of a stronger transition zone characterized by a less porous structure and with a limited amount of microcracking responsible for higher compressive strength values for SCC with respect CC. No differences were detected by EDAX analysis in the chemical nature of itz with respect the bulk matrix both for SCC and CC.

Finally, observations of the cement hydration by analysis of the temperature profile vs time seem to indicate the calcareous grains promote formation of heterogeneous nucleation responsible for the increased crystallinity of ettringite, for a shorter normally dormant period and, hence, for higher strength values at early ages, when the calcareous filler is used.

1 INTRODUCTION

According to a study that has been carried out recently [1], in Europe innovation in construction is concentrated in 4 main directions, which can be defined simplistically as follows:

- to build in a sustainable manner;
- to schedule appropriate building maintenance;
- to build safe and comfortable buildings;
- to build well and efficiently.

Of these directions, the first two ones are prevalent and are destined to represent the “new construction ideology” of this millennium, at least for the next 30-50 years. In particular, to build in a sustainable manner means to focus great attention on the following features:

- physical, environmental and technological resources;
- problems related to human health;
- energy conservation of new and existing buildings;
- control of construction technologies and methods.

Moreover, sustainability requires is to consider a building “life-cycle” extended over the useful life-time. This includes consideration of the building maintenance, demolition and recycling. This means that in our country, and generally speaking in “our ancient continent”, to build sustainably imposes dramatic changes to our construction methods and a need to avoid provoking any “destabilizing” impact on our environment. Therefore, this request for sustainability requires a change of our thousand year-old European and Italian philosophy of construction buildings as:

- monolithic;
- realized by means of irreversible assemblies;
- based on rigid connections;

- based on “non-renewable resources”.

As a matter of fact, reinforced concrete structures represent the real antithesis of sustainable development since they are:

- monolithic;
- based on irriversible reinforced concrete conjunctions;
- based on portland cement concrete, the production of which demands consumption of large amount of natural resources (such as clay, limestone and, generally, stony materials from quarries or riverbeds).

Taking into account the above mentioned features the question is: “What direction must the concrete industry follow to satisfy the need for sustainability in constructions?” *By optimizing the composition of new concretes using different ingredients from those utilized in traditional concretes, capable of assuring sustainability on the basis of “innovation”.*

Innovation in concrete construction means:

- production of precast reinforced concrete elements based on hydraulic materials using renewable or recycled ingredients;
- to increase perfomance in service of reinforced concrete elements in order to reduce cross-sections and, hence, volume of concrete produced;
- to increase durability of concrete structures to prolong the maintenance time reducing the amount of deteriorated concrete to be recycled and consumption of special repair materials based on non-renewable resources.

1.1 Precast Concrete

In the near future the concrete industry is destined to serve the precast element plants, more than placement on job-sites. In this way the realization of elements that can be assembled in the job-site will allow the realization of components that can be mounted easily and, at the same time, can be replaced and recycled in a very simple manner.

The production of these reinforced or prestressed concrete elements will be located close to urban areas, in order to meet prevalent methodology of the construction works in the next 20-30 years (Table 1), which will be made up of dispersed small projects, in form of buildings for commercial use and which will be in the centres of the towns subjected to a strong urban renewal. This methodology of building works will encourage the production of precast elements located close to urban areas requiring a small impact on the environment. From this point of view, self-compacting concretes can satisfy sustainability since they can drastically reduce acoustic environmental pollution, due to the possibility of casting precast elements without vibration.

Self-compacting concretes (SCC) also provide a significant opportunity for the use of waste and recycled materials. The higher volume with respect to conventional concrete (CC) of very fine powder necessary to achieve deformability and passing ability capacities permits them to incorporate high volumes, for instance, of fly-ash [2]. This will represent a key-point since the amount of fly-ash available in the near future will increase. In fact, some governments (i.e. Italy, USA, etc.) will build new carbon-based thermoelectric power generation plants to resolve problems related to the increasing demand of energy. These demands partially caused the black-outs in Italy and USA during the summer of 2003.

Finally, SCC are also “large volume consumers” of very fine particles generated by the recycling of demolished concrete structures not easily usable in traditional mixtures [3, 4, 5] and the huge amount of calcareous fillers available from the marble quarries. From this point of view SCC represent the “most voracious Dea Khali” as Metha defined in sustainable concretes [6].

1.2 Increased Performance

Use of large amounts of pozzolanic materials and limestone, together with the acrylic-based admixtures, is more effective in reducing the mixing water with respect traditional napthalene-based superplasticizers and allows the manufacture of high performance self-compacting concretes. The greater concrete homogeneity in the job-site and the smaller dependence of the structural performances on the workmanship quality will allow in the future to design self-compacting concrete structures, with a lower partial safety coefficient of the material. This will mean inevitably a reduction of the structure cross sections and consequently of the amount of the concrete used. Moreover, self-compacting concretes can be made easily and, thanks to their rheological properties, they may be used not only precast elements, but also in concrete cast on the job-site, with the respect for both the workers' health (because of the absence of vibration) and for the environment (because of the drastic reduction of the acoustic noise).

1.3 Improved Durability

Finally, self-compacting concretes turn out to be materials for the realization of building structures and components, with a much durability than the one of the standard concretes. The greater durability of self-compacting concretes satisfies the request for sustainability, because it will be possible to delay the maintenance and, therefore, limit the volumes of deteriorated concrete to be disposed of and lower use of repair mortars based on special mixtures big consumers of non-renewable resources.

Extension of durability of self-compacting concrete structures and elements is due to:

the improved durability of self-compacting concretes themselves, as direct consequence of the better quality of the interfacial transition zone and of the lower tendency to crack in comparison with the standard concrete mixtures;

a better quality of the concrete in the job-site, due to the reduced dependency on the procedures of casting and compaction.

2 PARAMETERS AFFECTING DURABILITY

Many factors influence physical and mechanical properties of concrete, such as water/cement, type of cement, use of materials having pozzolanic activity, type and particle size distribution of aggregates, type and dosage of admixtures. But even other important parameters may have a strong influence on the microstructure and, hence, on elastic and mechanical properties of concrete. These factors, less generally focused on, are related to the fresh state of concrete and concern rheological properties (cohesion and plastic viscosity), transportation, handling and placing of fresh concrete, segregation and bleeding, plastic settlement and curing. Rheological properties, in fact, can significantly modify nature and structure of the concrete interfacial transition zone (itz), between aggregate and concrete paste, which is responsible for the mechanical properties, watertightness, fire resistance and durability of prestressed and reinforced concrete structures [7].

Since fresh properties of self-compacting concrete (SCC) are significantly different from those of conventional concrete (CC), formation, nature and microstructure of itz can change significantly when a SCC is used instead of CC. For these reasons properties of hardened SCC can dramatically differ from those of CC even for the same water/cement ratio, type of cement, type and maximum size of the aggregates.

This paper presents results of an experimental program carried out to evaluate the main changes of the itz in SCC with respect that of CC.

3 INTERFACIAL TRANSITION ZONE

In freshly conventional compacted concrete, due to the presence of coarse aggregate, water films, initiated by vibration, tend to accumulate close to the aggregate surface. These films determine an increase in the w/c next to the aggregate compared to the bulk mortar [8]. As a consequence of the higher water/cement, the crystalline hydration products ettringite and calcium hydroxide (CH) in the vicinity of the coarse aggregate consist of larger crystals and therefore they form a more porous structure than in the bulk mortar matrix [9]. For these reasons, even for low w/cm, amount and size of voids in the transition zone will be larger than in bulk mortar. Moreover, CH crystals tend to locate themselves with the c-axis perpendicular to the aggregate surface. This results in a less adhesion capacity both for the lower surface area, and consequently weaker Van der Walls forces, and also because of preferred failure sites for the oriented structure [10].

In addition to larger amount and size of voids, the presence of microcracking is the major reason for the weakness of itz. Extension of microcracking depends on many factors, but the most important is related to concrete tendency to segregate which, in turn, is related to the aggregate size and grading, cement factor, type and dosage of admixtures, amount of water, workability, method of placing and compaction. For instance, a poorly graded aggregate can favour segregation; the larger the maximum size of the aggregate, the higher the internal bleeding. Moreover, for the same workability, the higher the amount of water the weaker the itz.

Due to the presence of microcracks, the transition zone significantly influences the durability of concrete, since it is more permeable than the bulk mortar. This means that concrete durability cannot be exclusively related to the w/c, type of cement and pozzolanic material, but should be attributed even to parameters, such as size and grading of aggregate, cohesion, etc, that can influence properties of itz.

Self-compacting concretes with high content of very fine particles (size lower than 0.150 mm), a lower volume and maximum size of coarse aggregates with respect those of conventional concrete, a more stable conveyor phase, improved rheological properties due to effective high-range water reducers and viscosity modifying agents, are expected to affect in a positive manner the itz, promoting a less porous microstructure.

In order to investigate these effects on itz, microstructure of self-compacting concretes and conventional concretes were analyzed by using the environmental Scanning Electron Microscope.

4 EXPERIMENTAL INVESTIGATION

Two conventional concretes, two self-compacting concretes with limestone filler (av.size = 30 μm) and two SCC containing pulverized fly-ash were manufactured. Mixture compositions are given in Table 2.

Water/cement was kept constant for CC and SCC: 0.40 for the first set of mixtures and 0.60 for the second one, respectively. Mixtures proportions were approximately the same: the cement content and the amount of water were constant. Dosage of superplasticizer, content of viscosity modifying agent, and the amount of coarse aggregate were adjusted to achieve self-compactability properties.

Concrete specimens were manufactured both to study microstructure and to determine compressive strength at early and long ages (cured at T = 20°C; R.H. = 95%).

Figures 1-4 show micrographs of self-compacting concretes and conventional concrete with a w/c of 0.60. Figure 1 highlights presence of a very porous microstructure in itz of conventional concrete as confirmed by the large dark area. On the contrary a more homogeneous and less porous structure of itz was detected for SCC containing both FA and $CaCO_3$.

Figure 2 indicates the presence of microcracks in itz and the matrix not well-bonded to the aggregate surface, for the CC. On the other hand absence of evident microcracks and a matrix well-bonded to the aggregate were found in SCC containing both FA and $CaCO_3$. The denser microstructure of SCC was confirmed by analysis of Figure 3, where the porosity of itz in CC concrete is significantly higher with respect that of SCC.

The higher porosity and the presence of microcracks in itz were responsible (Fig. 4) for the larger crystals of hydration products in CC with respect those of SCC [11].

These results seem to confirm the beneficial effect of a more stable paste in self-compacting concretes that is capable of decreasing internal bleeding and, hence, porosity of itz. This effect was independent of the type of filler: in SCC both with limestone and fly-ash, porosity was consistently lower than that in conventional concrete. Reduction in microcracking and in porosity of itz with respect CC were more pronounced in self-compacting concrete with w/c equal to 0.60: in conventional concrete, in fact, the higher w/c, the higher tendency of the mixture to segregate. Same results, not shown, were obtained for concrete with w/c equal to 0.40, even though differences were not as evident as detected in mixtures with w/c equal to 0.60.

Edax analysis indicated that no differences exist in terms of chemical structures between crystal phases in itz with respect hydration products in the bulk matrix for CC or SCC. In particular, data did not reveal any differences of the amount of sulfur in itz compared with the bulk matrix for CC or SCC. These results seem to be not in agreement with some experimental data available in literature [10, 12] which indicated in CC, near to itz, elongated needle-like crystal phases with high amounts of sulfur.

Data on microstructure seem to be in agreement with compressive strength values (Table 3). Independent of the nature of the very fine material, the compressive strength of SCC is higher than that of CC. In particular, compressive strength of SCC containing large amount of calcareous filler was higher than the corresponding value measured for both CC and SCC with fly ash, independent of the w/c and the age. At age of 90 days the gap in compressive strength value is partially bridged in SCC with fly-ash due to the pozzolanic reaction. Data confirm the positive role exerted by a stable and cohesive paste in SCC in reducing internal bleeding. This limits microcracks formation and reduces porosity of itz increasing compressive strength values of SCC with respect CC. Same results on the influence of SCC and limestone filler on properties of itz and, consequently, on compressive strength of concrete are reported in [11, 12, 13]: data available in literature seem to confirm that, at the same w/cm, SCC will produce higher compressive strength than CC [14]. In particular, when limestone filler is used higher compressive strength values were detected even at early age [15, 16]. Finally, data seems to indicate that limestone filler affects cement hydration process. In order to investigate this effect, if any, the temperature profile of the hydration process of cement pastes with and without calcareous filler was studied. Three different pastes were manufactured by using a CE I 52 R portland cement according to EN 197/1, two types of calcareous fillers with different granulometry (30 μm and 150 μm the average size for CF1 and CF2, respectively) and a siliceous filler with the same average size of CF1. The water/cement and the cement/filler were kept constant and equal to 0.35 and 0.30 respectively.

Figure 5 shows the instantaneous peaks in the temperature profiles for the three different pastes as a consequence of the hydration of the alumina phases. Figure 5 seems to indicate that the paste containing the finer (CF1) calcareous filler is characterized by a lower maximum temperature compared with that of cement pastes with the coarser limestone filler or the siliceous filler. Moreover, as clearly indicated in Fig. 6, the cement paste with the finer limestone filler is responsible for a shorter dormant period with respect to those of the other cement pastes studied. This means that the finer limestone filler is capable to accelerate the hydration process of the silicate phases.

The changes in temperature profile of hydration process of paste containing the finer $CaCO_3$ filler seems to indicate an increased crystallinity of ettringite due to the presence of the calcareous grains acting as nucleation site responsible for the lower temperature of the instantaneous peak. Formation of sites of heterogeneous nucleation is responsible for the shorter dormant period, the acceleration of the hydration process and, hence, for the higher strength values at early ages (Table 3).

This hypothesis on calcareous grains acting as nucleation sites is also reported in a paper by Kadri et.al. [17].

Several authors mention that the presence of calcareous filler reduces the normal dormant period and accelerates the hydration process [18-21].

These data on the effect of limestone filler seem to be in agreement with the results of adiabatic hydration tests [22]: the maximum temperature rise was systematically higher for self-compacting concrete containing calcareous filler with respect CC.

5 PLASTIC SETTLEMENT

The beneficial effect in reducing internal bleeding is confirmed by the reduction of plastic settlement in SCC with respect to conventional concrete. Sonebi et al [23] found that SCC exhibit greater stability than the traditional concretes: at about the same w/c (0.68 and 0.71 for CC and SCC, respectively) plastic settlement was 0.24% and 0.21% for CC and SCC respectively. Same results were obtained by Khayat [24, 25].

These data are in agreement with the bond strength between deformed steel bar and concrete matrix: at 28 days τ_{max} of SCC [23] was about 16-40% higher than the conventional mixture. Higher bond strength in SCC, in fact, should be related to a more stable paste capable of reducing the water film beneath the reinforcement and, consequently, to improve the itz mechanical properties.

6 PLASTIC SHRINKAGE

Absence of internal and external bleeding in SCC, however, is one of the causes for their higher plastic shrinkage with respect conventional mixtures. Turcry et al [26] found that, for the same loss of water, plastic shrinkage of SCC is at least 2 times higher than that of CC. The lower water/fine material and the finer capillary pore structure are responsible for a faster development of the negative pressure causing plastic shrinkage. Even with a delay in the setting time of SCC, due to the higher dosage of superplasticizer, the higher plastic shrinkage of SCC occurs with respect CC. This means that, in order to gain beneficial effect on strength and durability due to the improvement in properties of itz, in real structures curing of fresh SCC surface requires great care.

7 GAS PERMEABILITY, RESISTANCE TO FREEZING AND THAWING AND CHLORIDE DIFFUSION

The positive role of SCC in decreasing microcracking and porosity of itz is also responsible for a more durable concrete. Trägardh [13] found a decrease in the gas diffusion coefficient for a self-compacting concrete ($9.8 \bullet 10^{-18}$ m^2/s) with respect a conventional concrete ($13.6 \bullet 10^{-18}$ m^2/s) with the same w/cm (0.40).

Results obtained by Assiè et al. [27] reveal that SCC (w/cm = 0.53) shows lower permeability to oxygen ($0.6 \bullet 10^{-16}$ m^2) than CC ($1.5 \bullet 10^{-16}$ m^2) with about the same w/c (0.50).

Similar results were obtained by De Schutter et al. [28]: for the same w/cm, gas permeability of SCC was from ½ to $^1/_6$ of the corresponding value for CC whereas water permeability was somewhat lower for SCC ($1.44 \bullet 10^{-11}$ m/s and $1.66 \bullet 10^{-11}$ m/s for SCC and CC, respectively).

Resistance to freezing and thawing in the presence of deicing salts is essentially the same for SCC and CC [15, 28]. This means that in order to prevent concrete deterioration a water/cement lower than 0.50, blastfurnace slag cement (CE III/B according to EN 197/1) and air entraining admixtures must be used [29, 30].

Chloride diffusion coefficient of SCC ($16.1 \bullet 10^{-12}$ m^2/s) was found lower (13) than that ($19.2 \bullet 10^{-12}$ m^2/s) of CC with the same w/c (0.40). Audenaert et al. (31) found that for the same w/c chloride diffusion coefficient was slightly lower for SCC with respect CC. However, type of cement (blastfurnace slag cement replacing ordinary Portland cement) and use of fly-ash seems to have more influence than self-compactability on the reduction of chloride diffusion. This means that the capacity of hydration products in the presence of pozzolanic addition to absorb free chloride ions is more important in slowing down chloride penetration than the beneficial effect exerted by SCC on properties of itz.

Capillary suction is significantly lower in SCC with respect CC: Audenaert et al. (31) found a penetration height in CC almost twice as high as that for SCC, independently of cement and filler type. This result is to be ascribed to the denser itz of SCC.

8 MECHANICAL PROPERTIES

Due to the improvement in homogeneity and denser microstructure in the vicinity of itz, mechanical properties and, in particular, tensile strength of SCC may be higher than for conventional concrete.

Also, due to a less porous microstructure of the itz tensile strength of SCC should be higher than that of CC. The transfer of tensile stress, in fact, is based on the adhesion of the cement matrix with the aggregate. Values available in literature [14] seems to indicate higher splitting tensile strength for SCC with respect CC.

The lower maximum size of coarse aggregate and the reduction in coarse aggregate volume of SCC is responsible for a lower modulus of elasticity compared with that of CC. Data available in technical literature indicate that modulus of elasticity of SCC is within the lower half of the scattering range according to the Model Code 90. In particular, the average value of the elastic modulus for CC represents the upper limit for SCC [13].

Therefore, data concerning tensile strength and elastic modulus for SCC suggest a lower tendency to form cracks with respect a CC with the same time-dependent deformations.

9 CONCLUSIONS

Results of the experimental study carried out to evaluate major changes in microstructure of interfacial transition zone and bulk paste for both SCC and CC indicate that:

- reduction of internal bleeding when SCC is used is responsible for a denser and stronger itz with respect that of CC;

- large amount of fly ash or limestone filler in SCC favours formation of a less porous and, hence, a stronger transition zone due to a limited amount of microcracking in the vicinity of the itz;
- calcareous filler in SCC seems to act as nucleation sites promoting the formation of ettringite with enhanced crystallinity;
- calcareous filler in SCC seems to be responsible for a short dormant period and, hence, for higher compressive strength values both at early and long ages;
- the denser microstructure of the itz in SCC may contribute for a lower plastic settlement, higher bond between steel and concrete matrix, lower permeability to oxygen and lower chloride diffusion coefficient with respect to corresponding values for conventional concretes;
- higher tensile strength for SCC is due to improvement in the homogeneity and denser microstructure of the itz;
- lower elastic modulus for SCC is due to the lower size and amount of coarse aggregates with respect corresponding values for CC.

As a consequence of the experimental results and on the basis of data available in technical literature, self-compacting concretes should provide a higher intrinsic durability and a lower tendency to crack. In other words, structures with SCC could be significantly more durable than those manufactured with conventional concretes and, hence, they can answer the request for sustainability in future construction.

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Table 1 – Comparison of construction works and job-sites [1]

1960	2020
Huge projects	Very small projects
Concentrated	Dispersed
For residential use	For commercial use
New buildings	Urban renewal
In the suburban areas	In urban areas

Table 2 – Mixture proportions of conventional and self-compacting concretes (w/c=0.40 for set 1; w/c = 0.60 for set 2)

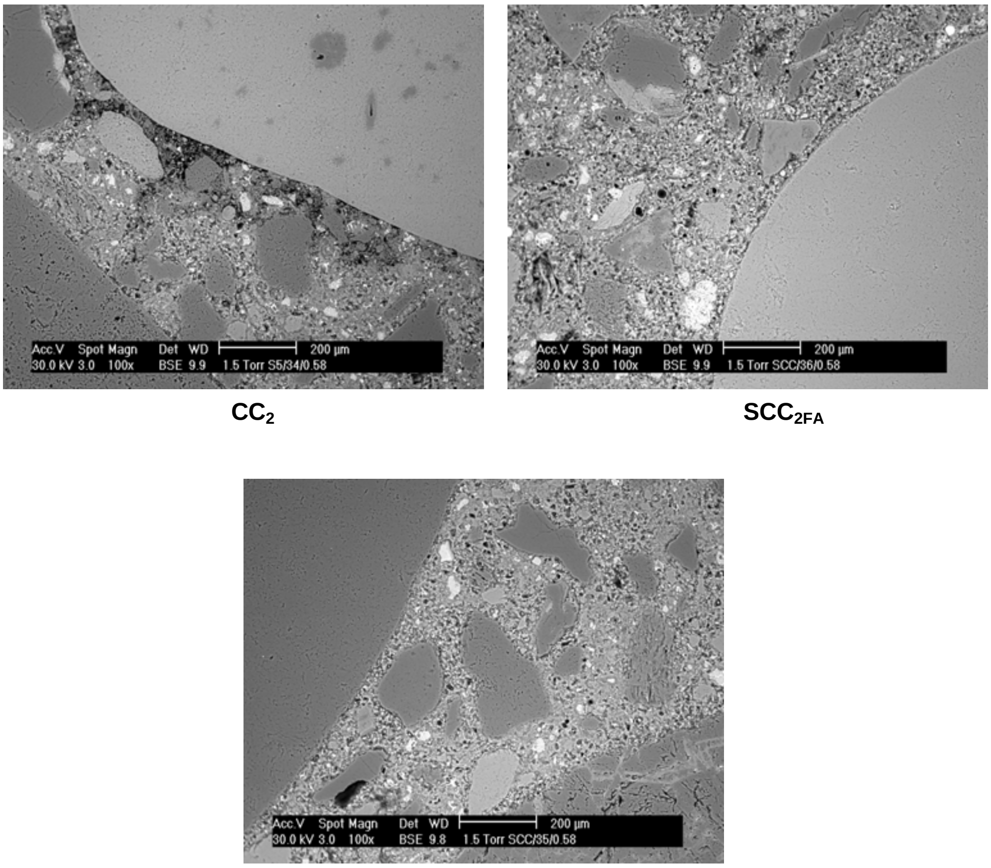
	CC1	**SCC1$_{CF}$**	**SCC1$_{FA}$**	**CC2**	**SCC$_{2\ CF}$**	**SCC$_{2\ FA}$**
Cement CE I 42.5 R (kg/m^3)	430	430	430	300	300	300
Water	172	175	170	175	174	172
CAE – HRWR (% by weight of cement + filler)	0.8	1.0	1.0	0.7	0.9	0.9
VMA (kg/m^3)	-	0.5	0.5	-	0.5	0.5
Limestone Filler (kg/m^3)	-	170	-	-	240	-
Fly ash (kg/m^3)	-	-	170	-	-	240
Sand 0-4 mm (kg/m^3)	875	795	775	930	810	785
Aggregate 4-8 mm (kg/m^3)	440	400	385	465	400	390
Aggregate 8-16 mm (kg/m^3)	435	395	390	465	410	390

Table 3 – Compressive strength (R_c) of CC and SCC with limestone and fly-ash powder

Rc (N/mm^2) at:	CC1 (w/c = 0.40)	SCC1$_{CF}$ (w/c = 0.40)	SCC1$_{FA}$ (w/ = 0.40)	CC2 (w/c = 0.60)	SCC$_{2CF}$ (w/c = 0.60)	SCC$_{2\ FA}$ (w/c = 0.60)
1 d	21	28	22	8	11	8
7 d	50	65	52	26	34	27
28 d	68	82	73	36	45	39
90 d	77	87	81	43	53	48

Table 4 – Cement pastes composition

Cement Paste No.	Cement CE I 52.5 R (EN 197/1)	Water	Calcareous filler 1 (CF1) (av.size = 30 μm)	Calcareous filler 2 (CF2) (av.size = 150 μm)	Siliceous filler (av.size = 30 μm
1	100	35	30	-	-
2	100	35	-	30	-
3	100	35	-	-	30

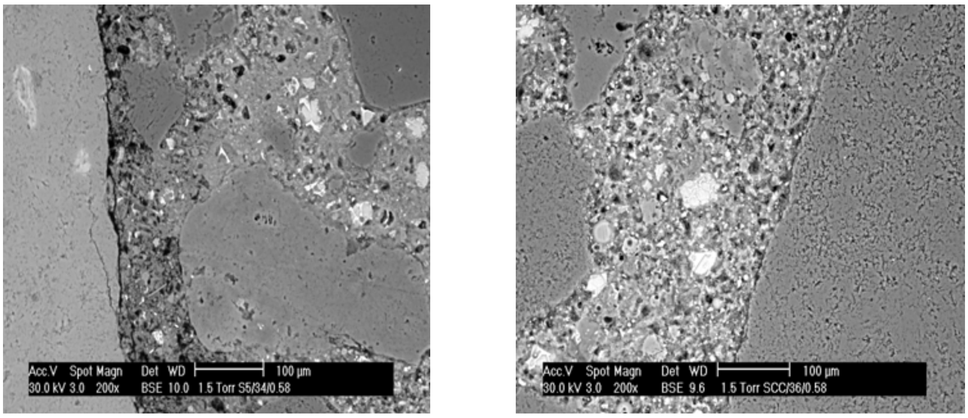


CC_2

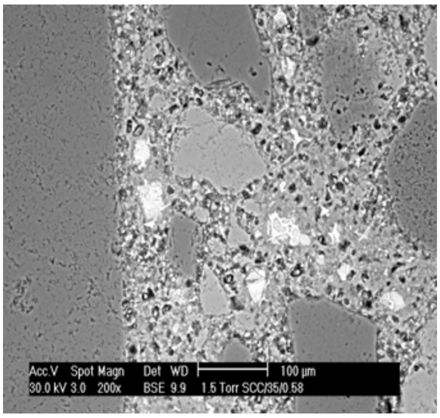
SCC_{2FA}

SCC_{2CF}

Fig. 1 – Scanning electron microscope of the interfacial transition zone in conventional concrete (CC_2) and in self-compacting concretes containing fly-ash (SCC_{2FA}) and calcareous filler (SCC_{2CF}) with the same w/c of 0.60 (200 μm).

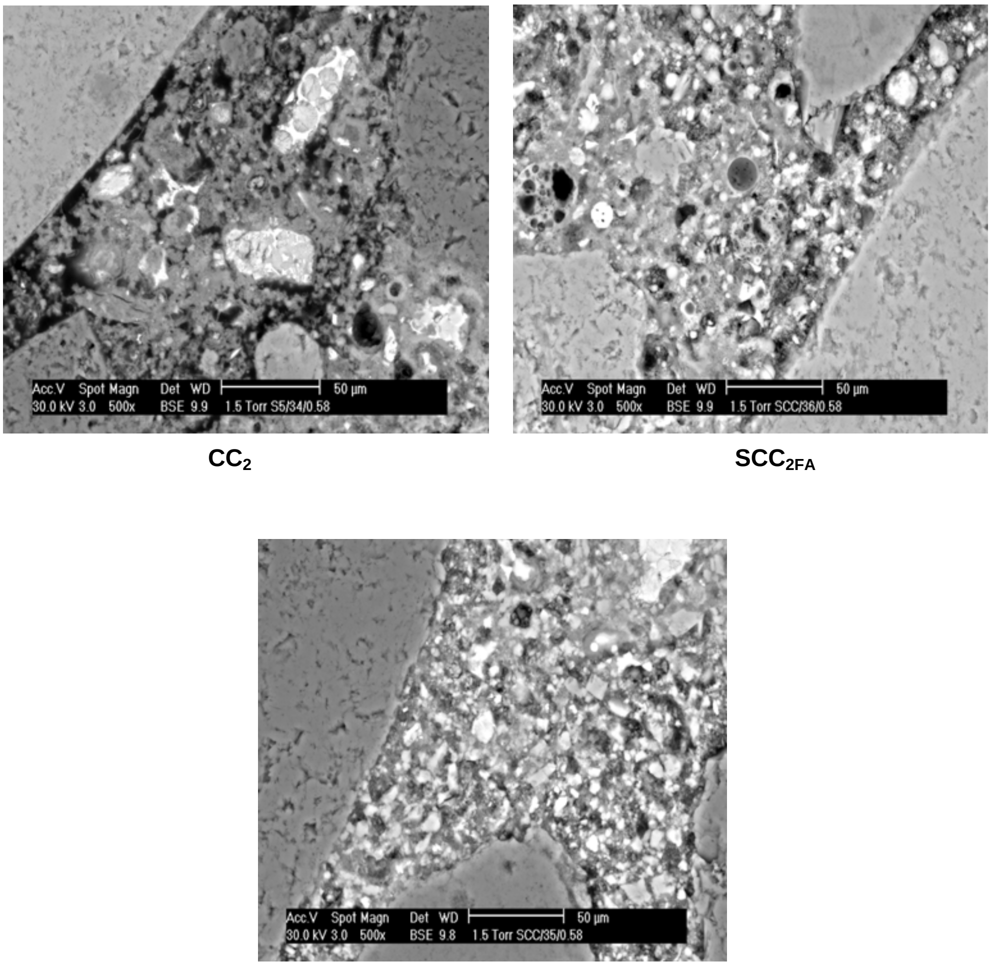


CC_2 **SCC_{2FA}**



SCC_{2CF}

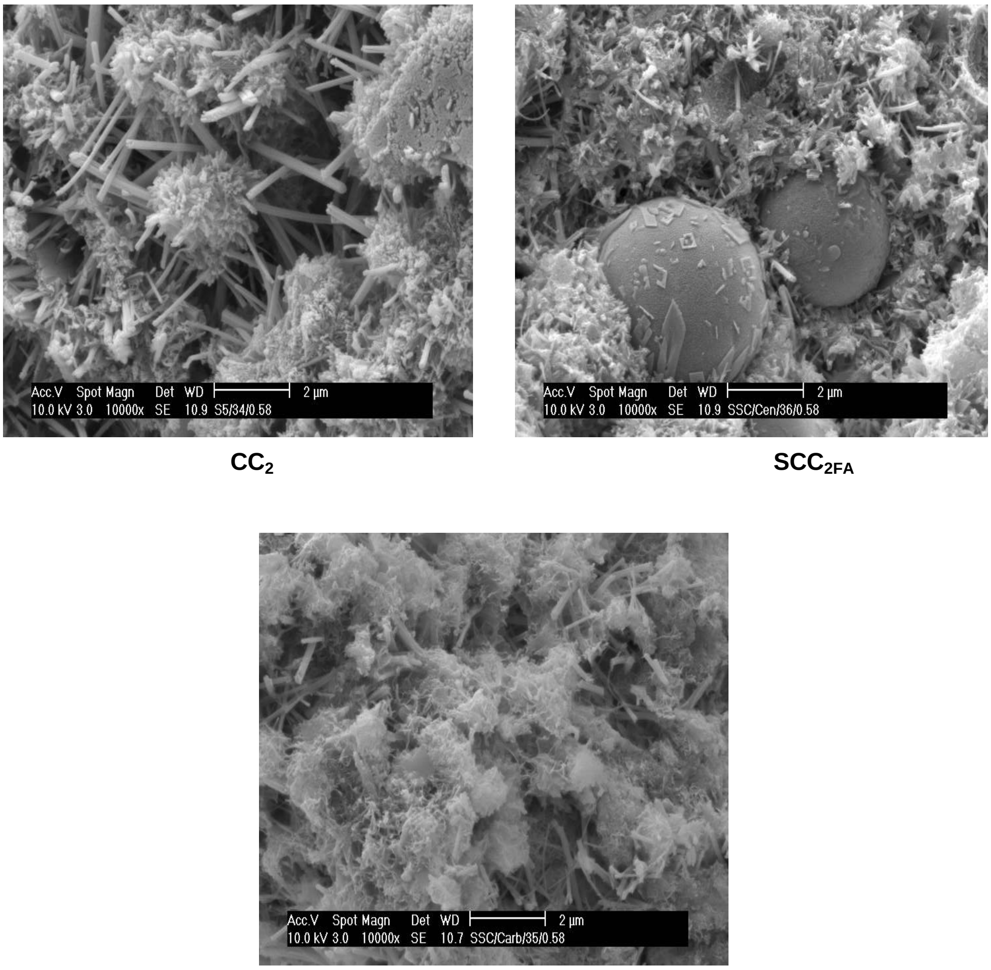
Fig. 2 - Scanning electron microscope of the interfacial transition zone in conventional concrete (CC_2) and in self-compacting concretes containing fly-ash (SCC_{2FA}) and calcareous filler (SCC_{2CF}) with the same w/c of 0.60 (100 μm).



CC_2 **SCC_{2FA}**

SCC_{2CF}

Fig. 3 - Scanning electron microscope of the interfacial transition zone in conventional concrete (CC_2) and in self-compacting concretes containing fly-ash (SCC_{2FA}) and calcareous filler (SCC_{2CF}) with the same w/c of 0.60 (50µm).



CC_2 **SCC_{2FA}**

SCC_{2CF}

Fig. 4 - Scanning electron microscope of the interfacial transition zone in conventional concrete (CC_2) and in self-compacting concretes containing fly-ash (SCC_{2FA}) and calcareous filler (SCC_{2CF}) with the same w/c of 0.60 (2 μm).

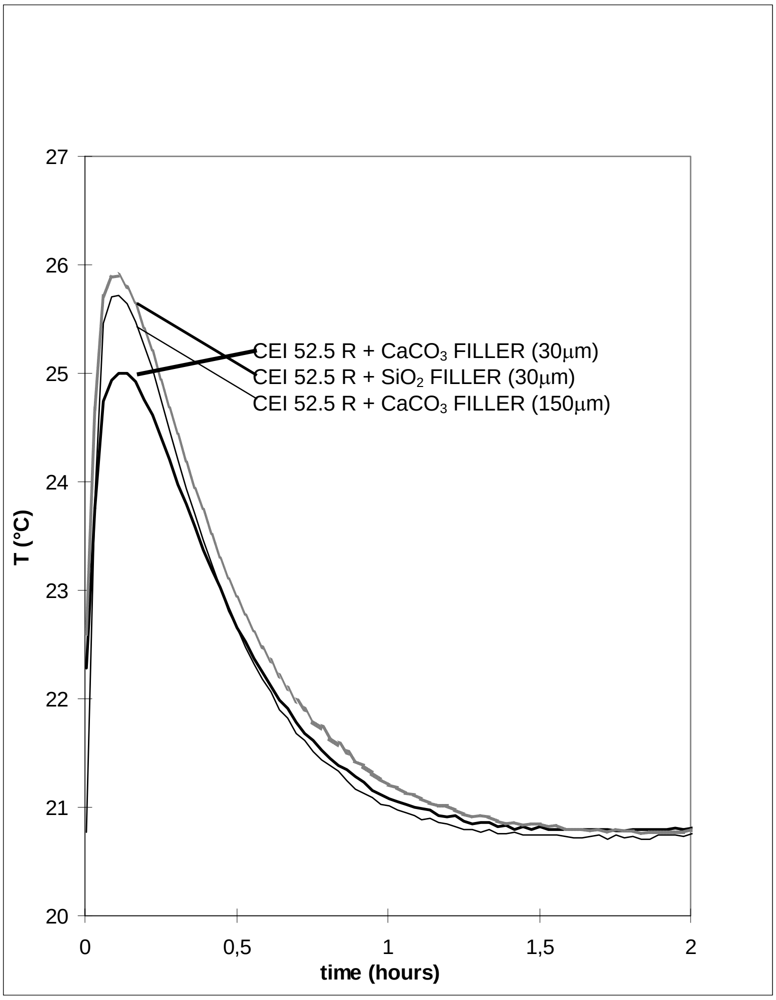


Fig. 5 – Istantaneous thermal peaks of cement pastes (w/c = 0.35) with different fillers.

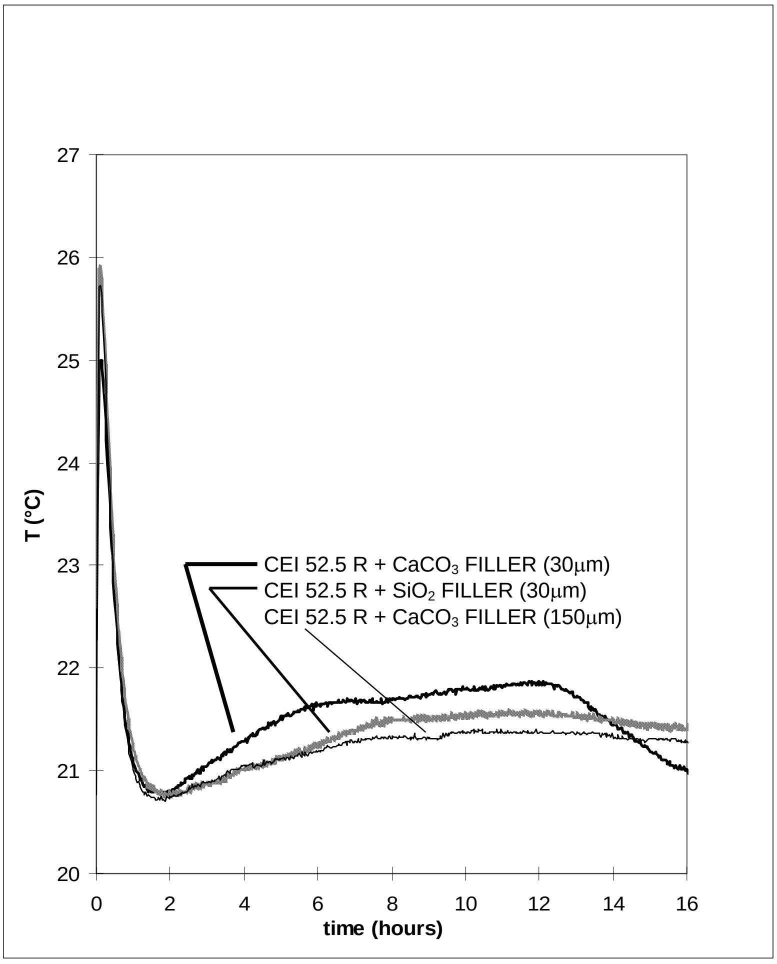


Fig. 6 – Temperature profiles versus time for different cement pastes containing calcareous or siliceous fillers.