

## Report from 13<sup>th</sup> ICPIC and 7<sup>th</sup> ASPIC: New Trends on Concrete-Polymer Composites

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**Abstract.** The field of polymers in concrete is consolidated in the construction industry. The future of polymers in concrete is governed by the synergic interaction between these materials, in order to contribute significantly towards a more sustainable construction. Concrete-polymer composites (C-PC) have excellent mechanical and durability properties. Appropriate combination of polymers and classical construction materials provides opportunities for innovative applications and systems. This paper highlights the innovations and new approaches presented at the 13<sup>th</sup> International Congress on Polymers in Concrete in Madeira, Portugal, 2010 and at the 7<sup>th</sup> Asian Symposium on Polymers in Concrete in Istanbul, Turkey, 2012. The new trends presented are related with the micro and nanostructure, properties, test methods and applications of concrete-polymer composites.

### Introduction

Cement concrete and polymers have long been considered to be complementary construction materials. Cement concrete is a material with high load bearing capacity. However, it is recognized that the synergetic action between polymers and concrete offers great opportunities for improvement and for new and innovative properties and applications in construction. Today the use of polymers is part of the search for more sustainable construction materials [1, 2].

The International Congresses on Polymers in Concrete (ICPIC) and the Asian Symposiums on Polymers in Concrete (ASPIC) aim to create a synergy between researchers and practitioners all over the world, dealing with the innovative possibilities of concrete-polymer composites (C-PC). The first international congress was organized in London in 1975. The 13<sup>th</sup> ICPIC in Madeira, Portugal, 2010, and followed by 7<sup>th</sup> ASPIC in Istanbul, Turkey, 2012 were forums for discussion and transfer of technology, exchange of experiences, opportunity for training of newcomers and to encourage research and development.

Although C-PC already has a long history of more than half a century, they still remain rather unknown and under-appreciated materials in the construction industry. However, the contribution of C-PC to sustainable construction materials and systems is important. Firstly, in renovation technology, structures can be restoring by using concrete-polymer composites as repair materials [3, 4]. The need for raw materials decreases and the environment is spared. Secondly, the synergetic interaction between the polymer film and the cement hydrates improves the properties of the modified system [5]. A material that is much more resistant against severe conditions is obtained [6 -8]. When field applications are considered, the main activities are on the Research and Development on overlay and repair, and the development of precast products [9].

In the future, the market share of Defined Performance Concrete that is concrete with performance values specific to a particular project, will continue to grow, as the industry becomes more sophisticated both in defining concrete properties of importance and in their measurement [10, 11]. In this context, the estimation of service life of C-PC is more and more important [12].

The innovation is realized by new products, improved processes or / and more efficient organization. Innovation becomes evident when a noticeable progress is achieved by implementing changes [13]. The family of C-PC came of age, but they are still very promising materials for new applications. They need theoretical basis to make the improvements more efficient and reliable. The study of nanosize effects and synergy is an important challenge in the future [14, 15].

### Micro and Nanostructure

The use of techniques like SEM, XRD and FTIR spectra, is very important in order to understand the micro and nanostructure of C-PC. Knapen and Van Gemert [16] used scanning electron microscope (SEM) to study the effects of the presence of water-soluble polymers in the microstructure of polymer-modified cement mortar. The polymers provide an improved dispersion of the cement particles in the mixing water. The tendency of certain water-soluble polymers to retard the flocculation of the cement particles minimizes the formation of a water-rich layer around the aggregate surfaces. They also provide a more uniform distribution of unhydrated cement particles in the matrix, without significant depletion near aggregate surfaces. Both effects enable to reduce the interfacial transition zone (ITZ).

In the presence of methylcellulose (MC),  $\text{Ca}(\text{OH})_2$  precipitates as stacks of layered crystals with an undistorted morphology. The crystal structure is strengthened by MC modification. At high magnifications, polymer bridges are detected between the layered  $\text{Ca}(\text{OH})_2$  crystals (Figure 1). The bridges are stretched between the layers, acting as an additional bond and gluing the layers together. Because  $\text{Ca}(\text{OH})_2$  crystals represent the weak phase in the binder matrix and the surfaces of those crystals form preferred cleavage sites, the strengthening by polymer bridges may improve the overall strength of the binder matrix.

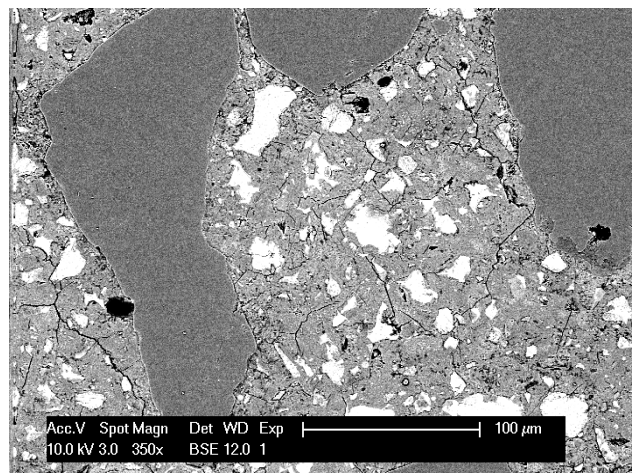
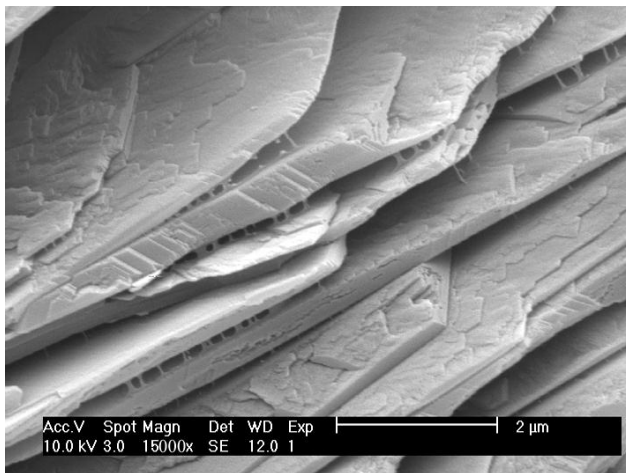


Fig. 1 Polymer films between layered  $\text{Ca}(\text{OH})_2$  crystals in 1 % MC modified mortar ( $w/c=0.45$ ) [16]. Fig.2 BSE image of crack pattern in unmodified mortar (left) and 1 % HEC modified mortar (right) [16].

The improvement of the cohesion of the bulk cement paste by polymer modification is observed in Figure 2 with the SEM images in the backscattered mode (BSE). The large dark areas represent the sand particles. Unhydrated cement particles are responsible for white areas. The epoxy-impregnated pores have low backscatter intensity and appear as black spots. A much higher amount of microcracks is detected in the bulk paste of unmodified mortar [16].

The decrease of cracks in polymer-modified cement mortars is also noticed by Bode and Dimmig-Osburg [17]. The shrinkage increases with the polymer content. This does not result from an increased drying shrinkage but depends considerably on the hydration shrinkage. As a consequence of an influence on the hydration caused by the polymers as well as by the increased water retentivity of

these mortars, it rather comes to a time-shift in the hydration. In the first days increased stresses in the mortar due to shrinkage can be taken by the increased tensile strength of the polymer-modified cement mortar.

Bezerra et al [18] used XRD and SEM techniques to show the presence of the most common phases of hydrated Portland cement in a polymer-modified cement concrete. Figures 3 and 4 show that the C-S-H gel is more abundant in the sample with 3 % of chitosan and 1 % of latex. This result is interesting because it explains why the concrete prepared with this concentration of polymers shows higher mechanical strength.

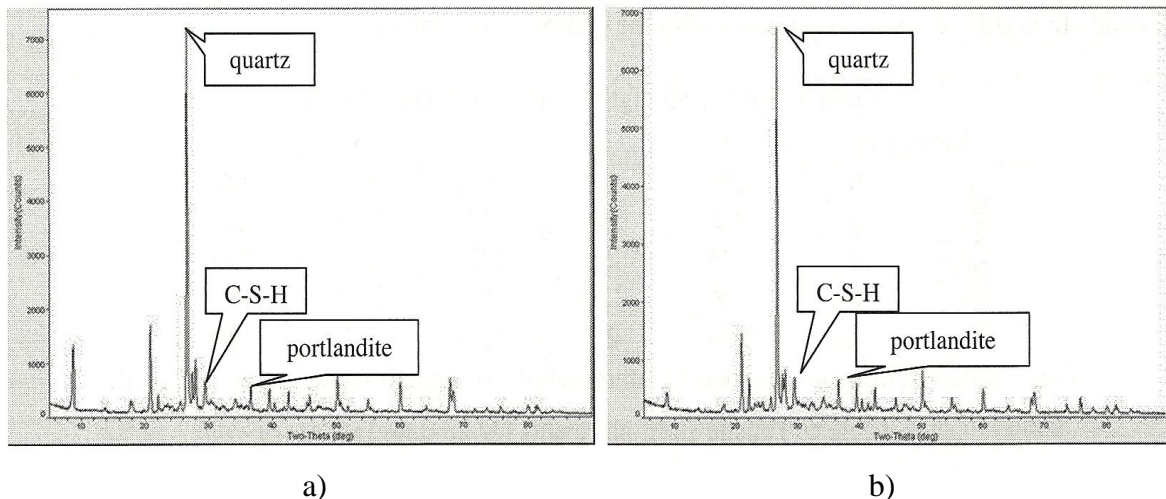


Fig. 3 XRD of (a) reference concrete; (b) concrete with 1 % latex and 3 % chitosan [18].

Pique et al [19] used FTIR spectroscopy and SEM techniques to monitor chemical transformation of polymer modified cement pastes. The determination of the activation energy of reactions showed that when polyvinyl alcohol (PVA) was added to the water-cement sample, the polymer shifted the hydration to higher times because the gel formed by the polymers hinders the hydration of cement with water. The results of the SEM investigation showed the cement hydration. Although water-soluble polymer was added at very low polymer-cement ratios, polymer film formation was detected. Figure 5 shows a polymer fraction localized in a cement hydrated grinded paste modified with PVA (2wt.%). The retardation effect on the formation of C-S-H and  $\text{Ca}(\text{OH})_2$  was also observed by Wang et al in the cases of styrene-butadiene rubber latex (SBR) [21] and styrene-acrylate copolymer (SAE) [22]. The polymer film was also detected in SEM images. By using energy dispersive x-ray spectroscopy (EDXS) analysis Wakasugi et al [23] demonstrated that even after 20 years unhydrated belite ( $\text{C}_2\text{S}$ ) and flux phase (aluminates and aluminoferrite) remained within the cement particles due to the inhibition of surrounding polymer film around cement particles.

Ekincioglu et al [24, 25] has been used atomic force microscopy (AFM) to reveal the morphological changes that occur on the surface of Macro Defect Free cement (MDF) samples (which have a composition of PVA/C: 0.07 and W/C: 0.11) during their immersion in water. The surface roughness has been calculated and 2-dimensional and phase images have been interpreted accordingly. When the MDF cements are immersed in water, some of the hydration compounds between the PVA film-coated cement particles are leached out, leading to the increase of the cements surface roughness, porosity and to the decrease of its biaxial flexural strength.

FTIR spectroscopy together with DTA have been used by Meng and Ping [26] to determine the type and content of polymer used in polymer concrete materials. Besides, FTIR spectroscopy has been used in order to monitor the structural changes of MDF cements that occur during exposure to water [25]. Chemical processes (carbonation, hydration) as well as the elimination of hydration products have been evidenced from the samples. It was noted both the bands shifting or disappearing and the appearance of new bands in the FTIR spectrum of the samples exposed to water by comparing with

that of the initial dry samples. Structural alterations of the MDF cement, determined by the hydration process, have been evidenced by FTIR spectra analysis. Similarly, the effect of Poly (ethylene-co-vinyl acetate) (EVA) on the polymer-cement mortar structure was identified by FTIR spectra interpretation [27].



Fig.4 SEM of chitosan (3%) and latex (1%) – presence of C-S-H gel [18].

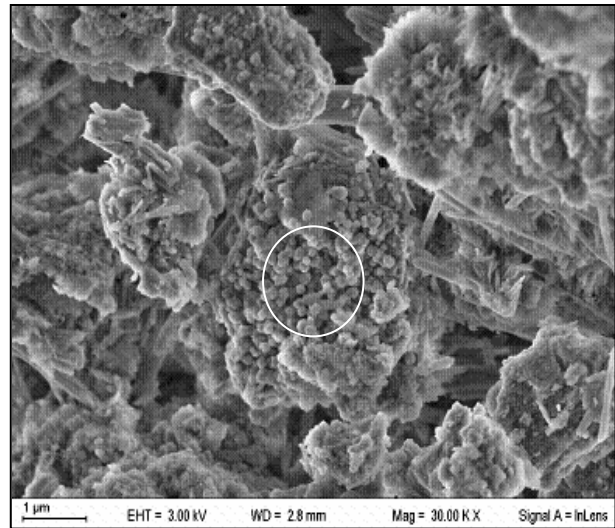


Fig.5 Nanoclay particles in between of polymer over a  $\text{Ca}(\text{OH})_2$  crystal surface polished sample [20].

## Properties

Concrete-polymer composites (C-PC) must present the appropriate properties related with the different possible applications. The study of the fresh and hardened properties of the concretes, mortars and pastes is extremely important for understand the behavior and performance of the developed mixtures.

Surfology is a scientific concept including all surface properties of materials and its influence on adhesion [28]. The analysis of the effect of surface parameters on polymer concrete composites is important for understand the wettability and thermodynamic properties. Taking account of one of the main C-PC applications, the repair or protection against corrosion, the analysis of the effect of surface parameters on adhesion is especially important. Courard et al [28] studied the surface properties of C-PC concluding that the interface quality depends on surface properties of materials, especially in the interface between repair systems and concrete substrate.

Workability and segregation resistance are important properties for fresh polymer-modified cement mortars. The effect of substitution degree [i.e., MeO% (Methoxyl%), HPO% (Hydroxyphoxyl%) and HEO% (Hydroxyethoxyl%)] of cellulose ethers on mortars were investigated by Choi et al [29]. It was reported that viscosity is increased with increasing MeO% and with decreasing HPO% or HEO%. The air content and slip decreased with an increase in MeO%, and with a decrease in HPO% or HEO%. In another study Han et al [30] investigated the influence of cellulose ether (CE) on the separation of latex from the latex-modified cement paste by using still standing test. The ratio of separated latex decreases with the CE increasing; when the dosage of CE is 0.5% no separation was observed for all polymer/cement ratios.

Schmidt et al [31] studied the effect of temperature on the performance of stabilising agents in cementitious systems and how performance changes affect fresh and hardening mortar or concrete properties. For the tests two differently stabilising agents were selected, one based on potato starch ether and other a microbial polysaccharide, in combination with superplasticizer. It was possible concluded that both stabilisers slightly retard setting without superplasticizers. On the other hand in presence of superplasticizer both stabilising agents cause retarded setting at 5°C and accelerated

setting at 20 and 30°C. This accelerating effect is stronger with higher temperature. Sandrolini and Manzi [32] studied the possibility of using in cement composite materials a strengthening epoxy resin with self-compacting properties. Concluded that the proper mix design outstanding increase the physical and mechanical properties of the cured composite. This encouraging the development of advanced concrete with high flowability in the fresh state together with high mechanical strength and better durability. Bezerra et al [33] studied the combination of two polymeric additives in concrete with the objective of improving the mechanical and durability performance. The polymers used were latex and biopolymer. Concluded that the combined used of both additives show an interesting interaction improving the compressive and tensile strength.

In addition to the mechanical properties, other properties such as thermal conductivity, shrinkage, creep and fire protection are beginning to make a great relevance.

The reduction of energy consumption, the production of thermally insulating materials and the solution of environmental problems by recycling of industrial and domestic waste are becoming a greater problem. Corinaldesi et al [34] investigated the mechanical behaviour and thermal conductivity of a lightweight building material containing either styrene butadiene rubber or polyurethane waste particles. This experimental investigation showed that the addition of rubber particles and polyurethane wastes particles reduces both the material unit weight and the thermal conductivity.

Shrinkage reducing admixtures (SRA) are being used more often in concrete structures in order to better control shrinkage cracks. Ribeiro et al [35] prepared different paste using two Portland cement from two sources and three SRA from different suppliers. One is a high molecular weight polyglycol, the other is hydroxyl combination (alkyl-ether) and glycol ether. Concluding that the influence of SRA products in chemical shrinkage of pastes depends on the cement used and the type of polymer. The values obtained indicate a small reduction of chemical shrinkage when using these polymers, mainly at early age. A consistent influence of SRA on pastes is a delay in the initial setting time and a compressive strength reduction, independent of the cement or SRA used. Bier and Bajrami [36] used ternary binders consisting calcium aluminate cement, ordinary Portland cement and calcium sulfate, and also a redispersible polymer powder as addition to produce high performance mortars. They observed that increasing polymer content increases the expansion for the ettringite binder system for free shrinkage measurements.

Karaguler and Yatagan [37] have studied the effect of polyisobutylene rubber addition in concrete on both free and restrained shrinkages. They reported that both shrinkages reduced in polymer added concretes with respect to plain concrete, furthermore the width of cracks for polymer concrete remained under that of plain concrete for the restrained shrinkage tests due to the bridging effect of polymer on cracks.

Fowler [38] investigated the possible reasons behind the falling of the ceiling panels connected to the top of the tunnel by epoxy anchors in a tunnel in Big Dig project in USA. It was concluded that the epoxy anchors were primarily responsible for the failure, because an epoxy that had high creep was used in anchors.

Fire protection is a characteristic of high importance, assuming an important role in maintaining the overall performance of the structure. However, it is considered that the fire-protection performance and thermal resistance of polymer-modified mortars are limited since they contain polymers which are thermally unstable and combustible. The noncombustibility behaviour of polymer-modified mortars is became an important aspect in the construction industry. Shirai et al [39], studied polymer-modified mortars using three different types of commercial polymer dispersions and flame retardants. The polymeric dispersions used were a styrene-butadiene rubber latex (SBR), poly(ethylene-vinyl acetate) (EVA) and polyacrylic ester (PAE), added to the ordinary cement mortar in different contents 5%, 10%, 15% and 20%. It also used three different flame retardant  $Mg(OH)_2$ ,  $Al(OH)_3$  and Calumite. It was possible to observe that all polymer-modified mortars without flame retardants and with polymer-binder of 5% and 10% are acceptable as noncombustible materials. For a content of 15 - 20% of polymer dispersions the polymer-modified

mortar presents a different behaviour. SBR-modified mortars can be improved only by use of Calumite, and they are acceptable as noncombustible materials. The EVA-modified mortars can be improved only by use of  $\text{Mg}(\text{OH})_2$ . The PAE-modified mortars can be improved by use of  $\text{Mg}(\text{OH})_2$ ,  $\text{Al}(\text{OH})_3$  or Calumite. Therefore it was possible to conclude that the fire-protection performance can be improved by the right choice of the appropriate flame retardants.

The high strength concrete not only allows an architectural structure to become very high, but also improves its durability and exhibits excellent performance in various aspects. However, the uncovered surfaces structures spall and fall off in fire, occurring with severe explosion. Han et al [40] studied the fundamental and spalling properties of high strength concrete, adding various types and contents of polymer resin. The polymers used were in powder and in fibers. The powder polymers were Ethylene Vinyl Acetate Copolymer (EVA-P) and Polyvinyl Acetate Copolymer (PVA-P). The polymeric fibers were Polyvinyl Acetate Copolymer (PVA-F) and Polypropylene Copolymer (PP-F). The fire tests allow to conclude that the concretes adding with EVA-P, PVA-P and PVA-F exhibited severe explosive spalling, regardless of their dosages. This is caused by the fact that polymer does not provide enough void net-works which is important in terms of evacuation of vapor to release steam pressure inside of concrete. However, when more than 0.10% of PP-F was added, spalling was effectively prevented. In order to prevent spalling of high strength concrete, it is concluded that the low melting point in polymer selection is critical.

Bacteria products have been developed as eco-friendly admixture for concrete. These biopolymers can be used to prevent the development of biofilms on surfaces and also as inhibitors of corrosion of rebars in concrete. Serres et al [41] reported that biopolymers increased the workability of cement mortar in the first 15 minutes; however they caused slight decrease in both compression and flexural strengths, but the strengths remained higher than the minimum requirements of the standard. Biopolymers act as corrosion inhibitors due to their slowdown of the cathodic reaction kinetic rather than modification of passivation layer [42].

### Test methods and applications

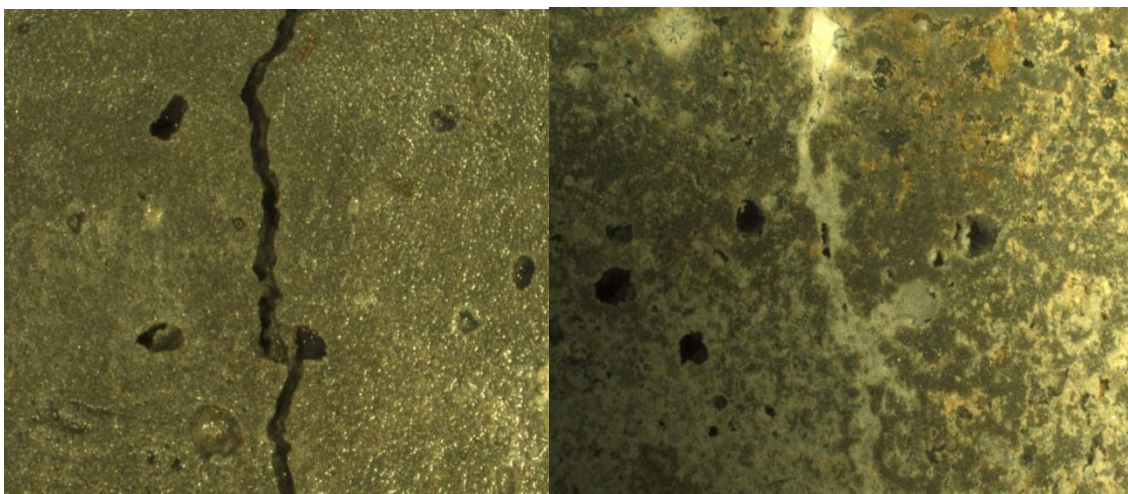
It is very important to protect the concrete structures from carbonation and the penetration of chlorides. Lohaus and Weicken [43] studied a new system for corrosion protection. This system was a layer made of high performance mortar of about ten millimeters thickness. The test program showed that the durability properties of corrosion protection mortars can be further enhanced by adding polymer dispersions. The depth of water penetration can be reduced to 90% by polymer modification by adding 20% of the styrene-acrylate. On other hand the resistance to chloride migration can be also increased, providing for a slower progress of the chloride migration front inside the mortar. The using of styrene-acrylate or styrene-butadiene not presented major differences. For that is possible concluded that a high concentration is not necessarily essential, which provides as option for saving costs.

The importance of testing fire-protection performance of polymer modified mortars was mentioned in the previous section. In the conventional test method with a heating speed of  $150^\circ\text{C}/\text{h}$  and 3% water absorption rate specimens generated spalling after 3 hours heating. For this reason Kim et al [44] used hot pressing test at a heating rate of  $100^\circ\text{C}/\text{h}$  and kept the water absorption of polymer-modified cement mortar (PCM) specimens at 1.5-2% in order to prevent spalling. PCM with EVA 5% (P/C) and polyacrylic ester (PAE) series in the range of 5-20% showed good mechanical properties in the range of  $200\text{-}400^\circ\text{C}$ .

Zhang et al [45] examined the effect of sizes of chemical admixtures, such as superplasticizers, polyacrylate latexes and asphalt emulsions, on the microstructure and permeability of cement pastes by applying the impedance spectroscopy measurement. The Nyquist plot of AC impedance spectra can be characterized by a semicircle and a straight line at the higher and lower frequencies, respectively. The semicircle diameter and the interception of semicircles with the real axis are related to porosity, average pore size and tortuosity while the diffusion coefficient can be obtained from the

straight line. The impedance response of mortars containing latex showed that the pore size become coarser at low latex contents but refined when the latex content is high. These findings were in good agreement with MIP results.

Self healing or self repairing means restoring of the properties totally or partially which were lost as a result of damage of a material. One type of self healing materials, which is also known as intelligent repair materials, is a hardener-free epoxy resin in a hydraulic cement system and developed by Ohama et al [4]. Lee et al [46] investigated the effect of dry curing followed the accelerated-cured-hardener-free-epoxy modified mortars. By using similar system, Lucowski and Adamezewski [47] studied the self repairing degree as a function of polymer/cement and binder/aggregate ratios. On the other hand, Ertug et al [48] tested a natural polymer-based hydrogel (chitosan) as a self healing material in cement mortars. Encapsulated hydrogel spheres in mortar were torn after cracking and showed considerable expansion, hence the cracks are sealed as seen in Figure 6.



(a)

(b)

Fig.6 The microphotograph of a crack on a sample from B series, x25 magnification, a) 7-day old sample, b) 28-day old sample (sealed) [48].

The enormous production of waste is a present problem. Expanded polystyrene is currently used as a popular packing or insulating material in various industrial fields in the world. A large quantity of expanded polystyrene is consumed and disposed as a waste. Žižoková et al [49] developed a study with the principal aim on the observation of the possibilities of use waste polystyrene for production cellular construction material. They concluded that is possible the application of polystyrene waste for screeding and adhesive materials designed for ETICS and the combination of polystyrene waste and fly ash combination as a substitution for a part of the filling. Gonzalez et al [50] studied the effects of adding polyamide powder waste (PAW) in mortar plasters. Polyamide powder was mixed with gypsum plaster in volume ratios of PAW/plaster up to 400%. They reported that higher the polyamide content in plaster higher the thermal resistance of plasters. Similarly Calderon et al [51] tested polyurethane (PU) foam waste with particle size less than 1 mm as a substitution of sand in Portland cement masonry mortars. It was concluded that these lightweight mortars obtained by PU addition exhibited similar behavior to conventional mortars in different aggressive environments.

Certain construction project applications may require a lightweight material that exhibits good all-round strength and durability characteristics. Kruger and Westhuizen [52] developed a ultra-lightweight thin filmed polymer modified concrete. They concluded that the addition of styrene butadiene rubber latex (SBR) or acrylic polymer, with small adjustments to the water cement ratio, it was possible to produce a high performance lightweight thin film concrete material for different

applications. Similarly, Kurbetci and Erdogdu [53] prepared pumice aggregate lightweight concretes by addition of SBR latex and reported that increasing SBR content improves the wear resistance, water absorption and sorptivity properties of concrete.

The incorporation of phase change materials (PCM) in lime-gypsum mortars has been investigated by Cunha et al [54] as an interior plaster of buildings for latent heat thermal energy storage. The effects of introducing PCM micro capsules in mortars on mechanical properties, shrinkage, aesthetic appearance and thermal behavior were explored. In order to make comparison a typical summer day was chosen and the indoor temperature change was monitored for two different rooms plastered with PCM and without PCM, respectively. Figure 7 shows that there is a temperature difference of 2.5°C between the peak points of these rooms indicating a more comfort condition for the former plastering.

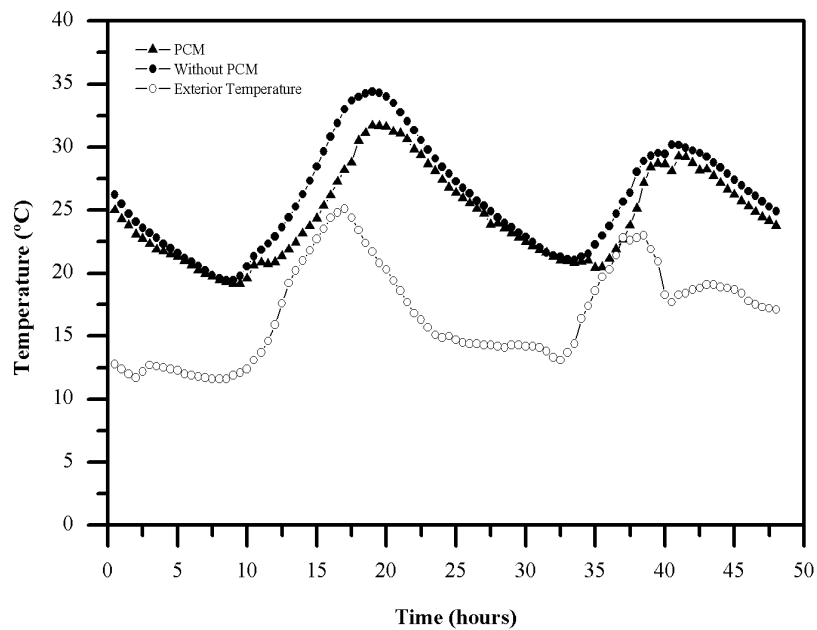


Fig.7 Monitored temperatures of the test cells with PCM and without PCM [54].

Heeger et al [55] used technical textiles for a new composite material, textile reinforced concrete, to produce concrete with better behaviour. In this study, alkali-resistant glass was applied in the concrete matrix for large-sized façade elements and sandwich panels. The textile reinforcement structures were coated with an epoxy resin. In comparison to an uncoated fabric, the tensile strength can be doubled with an epoxy resin coating. The concrete cannot penetrate into an uncoated roving and, thus, the inner filaments are not fully activated for load transfer. In contrast, the epoxy resin is able to penetrate into the roving and all filaments are activated resulting in a higher tensile strength. They concluded that the textile reinforced concrete broadens the application of concrete construction in the field of facade engineering.

A special fiber reinforced coating system was developed by Kawakami et al [56] to prevent concrete pieces falling from the surfaces of concrete structures. Thin vinylon fiber sheets and special epoxy resin with transparency after hardening have been used. Accelerated weathering test for the durability of transparency and punching test for sheet bonding and also accelerated carbonation and alkali-silica reaction tests were carried out.

The general requirement for repair of concrete structures is efficiency and durability. There are many factors affecting the bond quality. The basic operation during repair, that can increase or decrease bond strength between repair material and concrete substrate, is surface treatment. The impact-echo is treated as the most promising technique for bond strength evaluation. Piotrowski et al [57] studied the relation between bond quality and impact-echo frequency spectrum. The obtained results showed that bond strength estimation in repair systems can be made by impact-echo signal



analysis, especially using wavelet approach. Including into analysis the parameters characterizing concrete substrate quality, e. g. Surface Roughness Index and surface tensile strength can significantly improve this estimation.

The performance of a repairing system consisting of silane water repellent and flexible PCM coating after 20 years of application was investigated by Wakasugi et al [23]. They found that silane water repellent exists at the depth of 2-10 cm from the surface according to the results of DTA and still maintains water proofing and vapor permeability even 20 years after the repairing. Furthermore the flexible coating performed after 20 years even better than initial 5 years with respect to adhesive strength and crack elongation.

## Conclusion

The incorporation of polymers in pastes, mortar and concrete, has been increasingly common practice in the construction industry and consequently one of the areas with more research interest for the scientific community. There are numerous possibilities for incorporation of these materials, like fibers, dispersions or solid particles with different sizes.

The incorporation of polymers in construction products has as main objective the improvement of specific characteristics of the products developed in order to obtain a construction with a performance closer to the user needs, taking into account the functional and mechanical characteristics.

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