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Cement concrete and concrete–polymer composites: Two merging worlds. A report from 11th ICPIC Congress in Berlin, 2004

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Abstract

The search for durable and sustainable construction materials inspires the developments in the world of cement concrete, as well as in the world of concrete—polymer composites. Both worlds recognize, strive for and accept each other's contribution to the synergetic effects that are realized by the combination of classical building materials and polymers. A better knowledge of materials behaviour, especially in the field of admixtures, and a better understanding of curing processes allowed the development of highly performing mineral or modified mineral concretes, mortars and grouts. CPC-science becomes an invaluable element in the development of sustainable construction materials. ICPIC brings together practitioners and scientists, dealing with concrete—polymer composites in all industrial fields, but with emphasis on construction industry. The 11th International ICPIC Congress took place in Berlin, 2–4th June 2004. New trends and evolutions have been presented and discussed. The highlights of the Congress, and the synergies for the construction world that emerge from this congress on polymers in concrete in combination with cement concrete, are presented.

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1. Introduction

The International Congresses on Polymers in Concrete (ICPIC) aim to create a synergy between researchers and practitioners all over the world, dealing with the spectacular possibilities of concrete–polymer composites. During the three decades from the first congress

in London in 1975 to the 11th congress in Berlin in 2004 [1], there have been dramatic changes in the way of thinking about industrial processes and the approach and evaluation of new and innovative materials.

In the industrialised world, the sixties and seventies of last century were characterised by an unlimited belief in new and modern materials and techniques. The use of polymers was considered to be a sign of progress and modern attitude in construction and in industry. Glass fibre reinforced polyester panels, polyester resin and polyester mortar were known as inexpensive plastics. Epoxy glues were used as highly performing adhesives in concrete prefabricated applications, epoxy resins were

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used as binders in chemically resistant coatings and flooring systems as well as in electrical insulation applications, acrylic resins were used in machine frame construction. Research aimed at developing new, improved polymers was continuously done, and new applications for polymers and concrete-polymer composites (CPC) were constantly developed. In building industry the use of polymers gradually extended from concrete crack injection to repair mortars for concrete and stone, consolidation of masonry, repair of timber structures, not to forget all finishing, piping and waterproofing materials. At that time, building materials science was an underdeveloped field of construction industry and science. Therefore, the extended use of pure polymers led to inherent chemical and physical incompatibility problems, mechanical malfunctioning and durability problems.

In the early seventies the oil crises learned that mineral oil as a cheap basis for polymer production was not longer available. It was also found that oil reserves were limited. As a consequence all uses of fossil resources were questioned. First of all the waste of fossil oil for energy production, and secondly the massive use of polymers as alternative for classical building materials such as concrete, masonry, timber and metals. The use of polymers became part of a search for durable and sustainable construction materials. Polymers should only be used in these areas were their specific properties are needed. The synergetic action between polymers and classical construction materials offers great opportunities for improvement and a wide range of new and innovative properties and applications of e.g., polymer modified repair mortars for concrete, stone and masonry. Pure polymer concrete building components can be partly replaced by CPC, as in flag-stones and building panels. A better knowledge of materials behaviour, especially in the field of admixtures, and a better understanding of curing processes allowed the development of highly performing mineral or modified mineral concretes, mortars and grouts. CPC-science is now an invaluable element in the development of sustainable construction materials.

The topics of the 11th ICPIC Congress reflect new approaches and findings in CPC-research. The microstructural models are subject to changes, polymers in solution are subject of research and might become valuable alternatives for polymer emulsions and polymer dispersions, biological effects are taken into account in degradation processes but might also be turned into positive performance contributions. Sustainability, and reversibility and recycling as subtopics of it, dominate the developers' minds. The scientific program of the congress was managed by BAM, the Federal Institute for Materials Research and Testing (Germany), in collaboration with Warsaw University of Technology (Poland) and Katholieke Universiteit Leuven (Belgium).

2. Importance of polymers in European construction industry

During the European Colloquium Orgagec'02 organised by Laboratoire Central des Ponts et Chaussées in Poitiers, France, in March 2003, M. de Longcamp presented figures of consumption of polymers in the European Union (15 members; 330 million inhabitants) in 2000 [2]. Some data for the largest components in polymer consumption are listed in Table 1.

On average about 8% of polymer consumption concerns thermosets, the rest being thermoplasts [3]. In 2003, 7350000 tons or 18.5% of the total market of polymers in Western Europe (European Union, Norway and Switzerland) were used in building and construction industry [4]. The relation to other industries is shown in Table 2. However, if the consumption of polymers is compared to the consumption of other construction materials, its relative share is only about 1%, Table 3. The share in weight is only about 1%, but in financial turnover polymers represent more than 10% of construction industry. It is expected that in about 10–15 years polymers in construction will be the prime part of the polymer market. The amount of polymers used

European consumption of polymers in construction

Application field	Type	Consumption
Textiles in architecture	Polyester/glass PVC/polyester Carbon/Kevlar	28 000 tons 100 000 tons Not significant
Highway noise barriers	System concrete-timber Recycled plastics Transparent	±30% ±2% ±5%
	plastics Concrete Timber Metal Vegetation screens	$\pm 10\%$ $\pm 15\%$ $\pm 10\%$ $\pm 10\%$ Total 1970000 m ² , 15% increase/year
Impermeable membranes	PVC PE (HD + LD) EP, PU, UP resins SBS, APP bitumen modif.	200 000 tons 250 000 tons 50 000 tons 88 000 tons
Road paintings	Liquid, hot melt, strips	280 000 tons
Tubing for optical fibres	PEHD	200 000 tons (2001) 600 000 tons (exp. 2010)
Tubes for sewers, gas, water	PVC, PE, PP, UP	2761 000 tons
Concrete modification		486 000 tons

Table 2 Polymer consumption by industry sector in Western Europe (2003) [4]

Industry	Polymer consumption (%)			
Packaging	37.2			
Construction	18.5			
Large industry	5.8			
Electrical/electronic	8.5			
Other household/domestic	20.1			
Automotive	8			
Agriculture	1.9			

Table 3
Relative share of building materials in EU-construction industry (year 2000)

Construction material	Consumption (tons)	Ratio (%)	
Concrete and cement based	503 000 000	71	
Tiles and bricks	73 000 000	10	
Timber	54000000	7	
Iron and steel	24000000	3	
Stone, quarry	16000000	2	
Asphalt and bitumen	16000000	2	
Polymers	6850000	0.97	
Flat glass	5 200 000	0.73	
Mineral wool	2000000	0.3	
Copper	1 300 000	0.2	
Aluminium	900 000	0.1	

in concrete-polymer composites, is only a minor part in polymers for modification of concrete, which also include water reducing agents (121500 tons of superplasticizer in EU in 1998). However, due to the synergetic action between polymers and the cementitious matrices, the impact on performance of building materials largely overpasses the weight ratio.

The Construction Products Directive 89/106/EEC (CPD) defines the essential requirements for construction products as follows:

- 1. Mechanical resistance and stability.
- 2. Safety in case of fire.
- 3. Hygiene, health and the environment:
 - dangerous substances,
 - global environment impact.
- 4. Safety in use.
- 5. Protection against noise.
- 6. Energy economy and heat retention.

The European Legislation assumes that a producer is responsible for knowing and complying with all applicable legislation. This is a difficult area, because up to now only national regulations exist, and chemicals and dangerous substances are also dealt with by several DGs (Directorate General), e.g., DG Enterprise, DG Environment, DG Agriculture, DG Health, DG Consumer Protection, etc. The European Commission for Standardisation CEN is now harmonising the regulations

and a database on dangerous substances regulations is available [5].

With respect to global environmental impact, the European Union is striving at an integrated product policy, to reduce the environmental impact of the whole life cycle of products. The Union will use market forces efficiently to reach environmental policy objectives, in a way that prices should reflect also the environmental costs, demand should be more oriented towards ecological products, and supply should adapt and even promote ecological products.

All developments in using concrete—polymer composites should take into account this policy of the European Union. As engineers, we tend to stress on most of the 6 essential requirements of the construction products directive, but we have to admit that our interest in the 3rd one for hygiene and health is rather limited, presumably because of lack of knowledge in the field. Producers and construction engineers will have to collaborate with the medical world, to meet also those essential requirements.

3. Concrete-polymer composites

Polymer modified concrete or mortar is a composite material consisting of two solid phases: the aggregates which are discontinuously dispersed through the material and the binder which itself consists of a cementitious phase and a polymer phase. According to the volume fraction of the polymer in the binder phase the material shifts from PCC, i.e., polymer cement concrete, to PC, i.e., polymer concrete.

In the case of PCC, the binder consists of a polymercement co-matrix. The polymer is added to the fresh mixture as dispersion or as redispersible polymer powders. During hardening and curing cement hydration and polymer film formation take place resulting in a co-matrix in which polymer film is intermingled with cement hydrates.

A special group of materials in which polymers are used in combination with concrete is PIC, polymer impregnated concrete. Here, low-viscosity monomers are injected in the pores of the hardened concrete and subsequently polymerized. The resultant polymers form a second matrix if the pores are interconnected throughout the material. The hardened concrete may be a cement concrete, a PCC or a PC. The properties of the composite material derive not only from its constituents, but there is also a synergetic effect.

Applications of concrete-polymer composites cover a wide range of applications in different industries: concrete and stone repair materials; polymer modified cement adhesives; prefabricated building components like flag stones, tubes, panels; porous and eco-concrete; machine base elements; insulators for electrical and chemical industry; fibre reinforced materials; chemically resistant materials; industrial floors; recycling of polymer wastes; liquid applied waterproofing materials....

4. Integrated model for microstructure building in polymer cement concrete [6]

Cement hydration in polymer modified material is influenced by the presence of polymer particles and polymer film in the fresh state, during hydration as well as in the hardened state. The properties of the fresh mixture are influenced to a large extent by the surfactants, present at the surface of the polymer particles. The cement particles are better dispersed in the mixture and a more homogeneous material is formed. The hydration of the cement is reflected in the strength evolution of the material.

The influence of the polymer modification is twofold. Due to the presence of the polymers and the surfactants, a retardation of the cement hydration can be noticed. This is especially visible in the compressive strength of the mortar beams. On the other hand, due to the film formation or due to the interaction between the cement hydrates and the polymer particles, the tensile strength of the binder matrix as well as the adhesion strength between the aggregate and the binder increase. This is especially seen in the flexural strength of the mortar beams. The results are presented in [7–9].

From the results of the compressive strength and flexural strength tests, conclusions towards film formation mechanism and especially towards the time at which film formation takes place may be drawn. After 7-day dry curing, the flexural strength is increased in relation to the strength of the unmodified mortar. Nevertheless, the cement hydration is retarded when the polymercement ratio is increased, illustrated by a decrease of compressive strength. This points at the existence, already at an early stage of curing, of a polymer film or at least at the interaction between polymer particles and cement particles. However, the incapacity of the modified "porous" mortar specimens to overcome the large shrinkage stresses after 7-day or 28-day moist cur-

ing indicates that the continuous polymer film is not yet formed in the case of water saturated conditions. No influence of the polymer modification on the flexural strength is noticed in the case of standard cured and water cured samples as long as no dry curing period is applied.

Accordingly, one may conclude that at high relative humidity, the influence of polymer modification on the flexural strength at short term is limited. From the moment a dry curing period is introduced, a polymer film starts to build up through the binder phase and an increase in flexural strength is observed with increasing polymer–cement ratio. The influence on the flexural strength of the retardation of the cement hydration is compensated by the presence of the polymer film. When long-term behaviour is considered, a maximum of flexural strength is established around a polymer–cement ratio of 15%.

The mutual influences between the cement hydrates and the polymer particles and film are incorporated in an integrated model of structure formation. The model is based on the three-step model as proposed by Ohama [10], but stresses the positioning of the mechanisms on the time scale and the interaction between the different components. The findings are supported by images taken with an environmental scanning electron microscope at the University of Dresden [11].

The formation of the polymer film can take place from the moment two polymer droplets have sufficient energy to overcome the repulsion forces originating from the surfactants. In other words, if the temperature is high enough to cause sufficient Brownian motion, or if additional forces are working on the liquid layer around the polymer droplets, such as capillary forces or water withdrawal by further cement hydration, two droplets can come close to each other and can coalesce into each other and a polymer film is formed. This process simultaneously can take place with the cement hydration mechanism, especially in the case of dry curing conditions. Therefore, partial or full encapsulation of the cement hydrates is possible, which retards the hydration process. The different steps of the conclusive model are presented in Figs. 1-4.

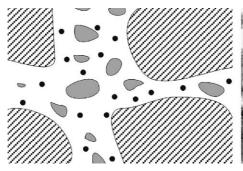




Fig. 1. Step 1, immediately after mixing, aggregates, cement particles, polymer particles and mixing water—small ettringite needles are formed.

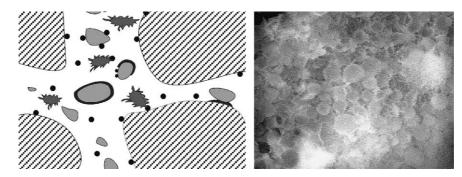


Fig. 2. Step 2, after mixing, the polymer particles interact with the cement particles and the aggregates. In the case a dry curing period is introduced, a continuous film may be formed—polymer particles flocculate together, on restricted places, no coalescence has taken place at this stage.

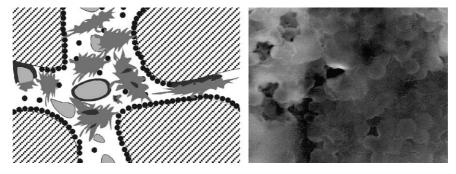


Fig. 3. Step 3, cement hydration proceeds, polymer film formation starts on specific spots—polymer particles coalesce together into a continuous film.

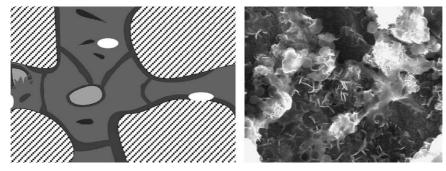


Fig. 4. Final step, cement hydration continuous, the polymer particles coalesce into a continuous film—cement particles are hydrated.

Immediately after mixing, the cement particles and polymer particles are dispersed in the water. The first hydration of the cement takes place, which results in an alkaline pore solution. This is indicated as Step 1, Fig. 1.

The second step is presented in Fig. 2. A portion of the polymer particles is deposed on the surface of the cement grain and the aggregate. The polymer–cement ratio determines the amount of polymers present in the pore solution and present at the aggregate surface. Part of the polymer particles may coalesce into a continuous film. This preferably takes place at the surface of the

cement hydrates where extra forces are exerted on the polymer particles due to the extraction of water for cement hydration. The polymer film can partly or completely envelop a cement grain, which results in a retardation or even a complete stop of the hydration of the cement grain.

The following step, Fig. 3 consists of cement hydration, polymer flocculation and possibly polymer coalescence into a film. The processes that take place depend on the curing conditions. If no dry curing period, i.e., curing at a lower relative humidity, is included, the overall film formation is retarded and the influence on the

properties of the fresh mixture is limited at this stage. If a dry curing period is included, polymer film formation takes place during this step, which influences the cement hydration process as well as the strength development at early ages. In the bulk liquid phase, hydrates are present, which form a combined inorganic and organic product. The fractions of the different types of product formed depend on the polymer-cement ratio used. The polymer fractions included in these hydration products do not contribute to the strength development of the specimen [12].

The final step, Fig. 4 includes further hydration and final film formation. Through the cement hydrates, a continuous polymer film forms as water is further removed from the pore solution. The part of the polymer particles, that is still present in the dispersion, is restricted to the capillary pores and at the interface of the aggregates and the bulk polymer-cement phase. It is this part which contributes the most to the elastic and final strength properties. The continuity of the polymer phase through the binder matrix is more pronounced in the case of a higher polymer-cement ratio. In the case the minimum film-forming temperature (MFT) of the polymer dispersion is much more elevated than the curing temperature, the polymer particles may not coalesce into a continuous film, but remain as closely packed polymer particles.

The use of the integrated Beeldens-Ohama-Van Gemert model can be illustrated with the different curing conditions. From the results, it is concluded that optimal conditions towards the strength development are a wet curing period followed by a dry curing period. The longer the moist and water curing period is, the higher the final flexural strength will be if shrinkage is prevented and if a curing period at lower relative humidity is introduced. This means that first cement hydration takes place and only limited film formation. Therefore, the polymer particles remain in the pore solution and a larger amount of polymer particles will be incorporated into the continuous film, which is formed in the final stage. If the drying period is introduced earlier in the process, the film formation will start sooner, i.e., before and simultaneously with the cement hydration, resulting in enlarged encapsulation of the cement hydrates as well as incorporation of the polymer phase in the hydration product precipitated from the pore solution.

The relative humidity of the surrounding atmosphere has a large influence on the film formation and especially on the drying rate. The higher the relative humidity of the surrounding atmosphere, the lower the drying rate becomes. This influences to a large extent the film-forming temperature of the dispersion. The lower the drying rate, the lower the amount of energy needed for the polymer particles to coalesce into a continuous film. Therefore, the MFT is reduced with a reduced dry-

ing rate. Tests indicated that even at laboratory circumstances, i.e., 20 °C, a poly(styrene-acrylic ester) (SAE) dispersion with a MFT of 32 °C, could form a continuous film, as long as the drying rate was low enough.

The cement hydration is also influenced by the fact that water is longer retained due to the presence of the surfactants at the surface of the polymer particles. This results in a better dispersion of the polymer particles and the cement hydrates, but also retards the cement hydration. The influence increases with increasing polymer—cement ratio.

This model accentuates two important changes towards the original model of Ohama. First of all, a relation to the time scale of the different processes is made. When a dry curing period is included, cement hydration and polymer film formation coincide and encapsulation of cement particles is possible. Further, the formation of an interstitial phase, consisting of inorganic and organic precipitates in the bulk phase is pointed out. This is important towards an optimal benefit of polymer modification since the polymers present in this phase are contributing less to the final properties of the material. The optimum conditions come forward from these findings, i.e., a long period of water or moist curing (up to 28 days) during which the cement hydrates develop followed by a period of curing at lower relative humidity during which the polymer film formation is promoted.

5. Polymers as microcrack stopper in cement concrete [13]

At the Building Materials Institute of Technical University of Dresden, the new technology of environmental scanning electron microscopy is being used in combination with a specially developed loading device, that allows to apply tensile load on small specimen inside the microscope chamber. They are now able to show the effects of microcrack propagation in the loaded specimen. Opening increase of microcracks can be followed and

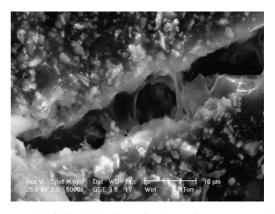


Fig. 5. Crack bridging behaviour of polymers in concrete, maximum crack width ${\sim}9~\mu\text{m}.$

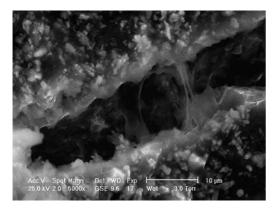


Fig. 6. Crack bridging behaviour of polymers in concrete, maximum crack width ${\sim}11~\mu m.$

investigated from a width of about 300 nm up to about $20 \mu m$, allowing to study the behaviour of all the particles, which are broken by the crack. Especially the polymer action as microcrack stopper can be observed by visualization of the stretching of the polymer particles over the microcrack.

The power of this innovative investigation technique can be illustrated with some pictures of a microcrack in a polymer cement concrete, with a polymer–cement ratio of 30%, at consecutive crack widths during tensile loading. Not only the crack bridging effect can be seen, but also the moment of rupture of the polymer bridges can be observed, Figs. 5–7. This investigation technique

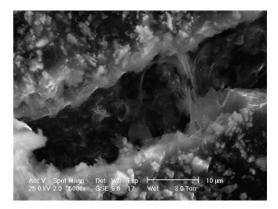


Fig. 7. Crack bridging behaviour of polymers in concrete, maximum crack width ${\sim}13~\mu m.$

provides spectacular features for the study of hydration and hardening reactions, and for the study of polymer–cement hydrate interactions.

6. Results and expectations of 11th ICPIC

The novelties and significant progress, reported at the 11th ICPIC Congress in Berlin, are summarized in Table 4. The paper number refers to the reference number of the paper in the proceedings [1]. For each topic, we gave our appreciation of the relevance field, or the state of progress of the matter.

Table 4 Novelties and significant progress, reported at 11th ICPIC, Berlin 2004

Description	Reference	Paper no.	Test method	Research	Application
Integrated model	Beeldens et al.	1		+	
2. Nanomonitoring interfaces	Schorn et al.	2		+	
•	Benzarti et al.	67		+	
3. Soluble polymer modifiers	Knapen et al.	11		+	
4. Slag PCC	Joo, Ohama et al.	13			+
Fly ash PCC	Jolley, Kruger	60			+
5. Ultrasonic evaluation methods	Garbacz	15	+		
6. Profilometry and surfometry analysis	Courard et al.	16		+	
7. CPC standardisation	Ohama	18			+
8. Sound absorbing materials	Knapen et al.	24			+
9. Quantification of synergy	Czarnecki et al.	25		+	
10. High strength ECC without hardener	Ohama et al.	30		+	
11. Ultra-lightweight polymer concrete	Sung et al.	35,36			+
12. GFRP-PC	San-José et al.	37			+
13. Electrical insulators	Gunasekaran	40,41			+
14. Preplaced aggregate epoxy concrete	Murray	43			+
15. Ecological coatings	Erkens et al.	44			+
16. Optimization thin overlays	Šušteršic	54			+
17. Textile reinforced PCC	Dilthey et al.	58			+
18. Waste reuse	Bignozzi et al.	72			+
19. Intelligent PC	Blair et al.	31		+	
20. FRP strengthening	Frigione et al.	61			+
-	Ignoul et al.	63		+	+
	Byun et al.	64		+	+
21. Admixtures	Pourchez et al.	69		+	
	Grübl et al.	75		+	

7. Conclusions

The use of polymers in construction industry is steadily growing. The synergetic action of polymers and cement mortar and concrete offers great opportunities for improvement and a wide range of new and innovative applications. Society and environment require corrective actions to be taken continuously. The use of polymers should be well-considered to guarantee better performance and improved sustainability.

Polymers are no longer special construction materials that replace classical mineral or organic building materials. They are now one vital component in the production of composite and sustainable building materials. They will further allow the development of new and durable constructions, as well as new and durable restoration and retrofitting techniques.

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