

Improvement of bond behaviour and durability of AR-glassfibre-reinforced concrete by polymer-fibre coatings

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1. Introduction

The durability and mechanical performance of textile-reinforced concrete (TRC) made with alkali-resistant (AR)-glass yarns are mainly determined by both the adhesion behaviour between the fibres and the cement matrix as well as the interaction of filaments in the roving. Furthermore, the resistance of AR-glass yarn against alkaline attack influences the durability of TRC^[1].

The bond between AR-glass yarns and the cement matrix arises due to the penetration of the water-dissolved cement particles into the outer areas of the yarns. The hydration products (mainly CSH phases) interconnect the filaments, and the so called 'fill-in' zone is formed in the hardened state of concrete. This fill-in zone of the yarn realises mainly the load transfer from the concrete to the textile reinforcement. The inner filaments of the yarn are not fully reached by cement particles and hydration products and are also not connected with the filaments in the fill-in zone. They take part in the load transfer due to either friction forces or at higher loads due to lateral compressive forces, additionally^[2, 3].

An additional coating of the original yarns using polymer dispersions is an appropriate method for improving the bond behaviour in the fill-in zone and to interconnect the inner filaments of the yarn in order to use the entire roving for the load transfer. Due to the small size of the polymer particles (150–200 nm), they can penetrate the internal region of the yarns and connect the internal filaments. However, the coating leads to a reduction of the whole surface area, which can be utilised for the bond with the cement matrix. Thus, the complete activation of the reinforcement is shifted to larger crack widths. This disadvantage (in contrast with non-coated yarns) can be compensated as far as possible by an appropriate composition of the polymer dispersion, e.g. the addition of nanoclay particles. Furthermore, the coatings provide an additional protection of the fibres against the alkaline environment as well as against the mechanical impact due to sharp crystalline formations of the cement matrix^[4, 5].

The enhancement of the working capacity of TRC made with polymer-coated AR-glass yarns depends not only on the composition of the polymer coatings but also on the interaction with the matrix constituents and on the ageing behaviour of the fibre–matrix interface. These parameters influence mainly the durability of the TRC. This paper describes investigations concerning the time-dependent development of the bond between AR-glass yarns, which have been coated with polymeric dispersions, and the matrix. The results compare reinforced coated samples with original (non-coated) AR-glass yarn.

The investigations are part of the complex research programme within the collaborative research centre 528 at Technische Universität Dresden.

Keywords: TRC, AR-glass fibre, durability, fibre coating, glass fibre corrosion, nanoparticles, accelerated ageing tests



2. Methods

For verifying the improvement and the time-dependent development of the working capacity of concrete reinforced by AR-glass yarns coated with polymeric dispersions (in contrast to original yarns), the specimens were stored in a fog chamber at 40°C and 99% relative humidity (RH). This storage is used as a common accelerated ageing test method.

After 28, 90 and 180 days, respectively, the specimens were tested using a centric tensile test to measure the bond behaviour. To compare the results of the fog chamber storage, specimens stored at standard climate (20°C and 65% RH) were also tested at concrete age of 28 days.

Afterwards, the fibre–matrix interface of the tested specimens was inspected using a scanning electron microscope in ESEM mode (REM/ESEM).

3. Materials

3.1. Specimens – geometry, fabrication and storage

For the centric tensile test, a special-shaped, double symmetrical tensile test specimen has been developed and provided with a small notch as predetermined breaking point to improve reproducibility and accuracy of the test results. The thickness in the notch region was 5 mm. The specimens were manufactured using all-side closed moulds to prevent inequalities of matrices and unwanted deformations. Three parallel stretched multi-filament yarns were embedded in the longitudinal axis of the specimen at equal spacing of 10 mm. The low number of the reinforcing yarns was chosen to exclude multiple crack formation during the tensile tests. Therefore, the deformation behaviour is represented only by a single crack generated in the notch of the specimen. This arrangement results in a volume content of the unidirectional reinforcement of 0.105 vol% in the notch region. A schematic of the specimen including the geometrical dimensions is shown in Figure 1.



Figure 1: Schematic drawn of the specimen (dimensions in mm)

One day after casting, the specimens were de-moulded and the stretched yarns were separated from the clamping fixture. Subsequently, the samples were stored under water at 20°C for 7 days. Afterwards, to investigate the ageing behaviour of the bond, the specimens were stored in the fog chamber. The samples used for comparison were stored at standard climate.

3.2. Concrete mixture

The mixture used to fabricate the test specimens is a typical fine-grained glassfibre concrete with maximum grain size of 1 mm. This mixture is especially developed for investigations within the collaborative research centre 528. The alkali level is reduced due to the addition of pozzolans (fly ash, micro-silica suspension). The mixture contained the components as given in Table 1.



Component	Volume, kg/m ³
CEM I 32.5	622
Fly ash	280
Micro-silica suspension	62
Sand 0–1	933
Water	280

Table 1

3.3. AR-glass yarn and polymer coatings

The AR-glassfibre yarn used for the unidirectional reinforcement of the specimens was a VETROTEX roving, 640 tex, sized with an unknown sizing composition. The yarns have been additionally coated with polymer dispersions designed by the Leibniz Institute of Polymer Research in cooperation with the Institut for Building Materials within the collaborative research centre 528.

One formulation of polymer coating has been applied to the sized glass fibres as polymer dispersion consisting of self cross-linking carboxylated styrene butadiene copolymers. The total weight gain due to the coating is 7.7 wt%, determined by pyrolysis (600°C, 60 min) of the coated fibres.

In a further formulation, nanoclay particles (Nanofil 15, Süd-Chemie AG, Moosburg) having a particle size of 60–130 nm were added to the dispersion. An effective utilisation of montmorillonites requires a sufficient intercalation/exfoliation (dispersion of the layered silicates). Therefore, a quaternary ammonium surfactant and a non-ionic surface-active agent were added to the dispersion for homogeneous distribution of the constituents. The amount of the nanoclay particles for the formulation investigated was 5 wt%. The nanoclay particles were investigated to realise multifunctional interphases (improved adhesion strength, reduction of the diffusion coefficient^[5] in TRC).

3.4. Test set-up

Both ends of the specimen were encapsulated by steel plates fixed by epoxy resin on the sample. The specimens were clamped by the steel plates in the test machine. This stiff clamping prevents a mutual rotation of specimen parts after the matrix rupture. Figure 2 shows the test specimen arrangement in the test machine.



The test procedure was performed in a displacement-controlled manner. During the uncracked state the displacement of the hydraulic jack was limited to a rate of 0.05 mm/min. After concrete rupture the speed was increased to a rate of 1 mm/min. The deformation process was stopped when a crack width of 2.5 mm was exceeded. Load, deformation path and crack width were recorded.

4. Results

4.1. Investigated rovings

The following differently surface treated rovings were investigated within this work:

- non-coated (original) ARG-VETROTEX roving (F 500)
- ARG-VETROTEX roving coated with polymer dispersion (F 501)
- ARG-VETROTEX roving coated with polymer dispersion F 501 and improved by 5 wt% nanoclay particles (F 502).

The results of the centric tensile tests are presented in form of graphs showing the load (= fibre pull-out force) in kN versus the deformation (= crack width) in mm. The first part of the curve, displaying the increasing load of the matrix and the matrix rupture, is not presented. The load–deformation graphs show the fibre pull-out occurring after the matrix failure. For each parameter combination, a characteristic curve is selected.

4.2. Original ARG-VET, F 500

Figure 3 shows the load–deformation curves of the specimens with the original ARG-VETROTEX yarn, 640 tex, after 28 days of storage at standard climate and after different durations of fog chamber storage.



Figure 3: Load-deformation curves of specimens reinforced by the original ARG-VETROTEX, 640 tex (F 500) after storage at standard climate and fog chamber storage

The curve for 28 days' storage at standard climate represents the typical load–deformation behaviour of concrete reinforced by unidirectional embedded AR-glass rovings. The increasing part of the curve displays the load transfer from the matrix into the roving (after the matrix failure) up to a maximum value of the load and the corresponding crack width. The decreasing part of the curve displays the successive failure of the filaments.

The accompanying microscopic investigations of the fibre–matrix interface revealed that the filaments located in the fillin zone are enveloped and connected by slender hydration products (CSH phases), which ensure the load transfer from the matrix into the yarn (Figure 4).



During storage in the fog chamber, the level of working capacity decreased continuously. The reduction of the maximum load and the working capacity, due to the storage in the fog chamber, can be explained as follows. Due to the impact of the humidity and alkalinity of the pore solution, a partial dissolution of the fibre sizing is possible. Thus, the primary surface flaws are unprotected and could act as sources for high localised stresses. Subsequently, lower stresses can lead to a failure of the filaments. A further reason for the reduction of the maximum force is the obviously increased formation of portlandite $(Ca(OH)_2)$ during the fog chamber storage (Figure 5). Compared to the CSH phases, the portlandite formations are about 100 times bigger. They displace the CSH phases, disturb the bonds between the filaments and decrease the working capacity. Corrosive changes on the fibre surfaces as cause of the loss of working capacity due to fog chamber storage could not be found^[6].



Figure 6: Load-deformation curves of specimens reinforced by the ARG-VETROTEX, 640 tex, additionally coated with the polymer dispersion (F 501), after storage at standard climate and fog chamber storage

4.3. ARG-VETROTEX coated with polymer dispersion, F 501

The additional coating of the ARG-VETROTEX roving using the polymer dispersion see section 3.2) led to distinctly changed working capacities, as presented in Figure 6.

Following 28 days' storage in a standard climate and in comparison to the results of the specimens reinforced by the original (non-coated) yarn, the slope of the load–deformation curve after the matrix failure was slightly less, and the maximum fibre pull-out load was reached at increased deformation/crack width. Therefore, the working capacity was higher but connected with larger deformations as well.

The increase of the working capacity was caused by the homogenisation of the yarn cross-section and by the improved fibre–matrix interface. In addition, the inner filaments of the yarn are involved in the load transfer to a higher extent due to the polymeric coating. The successive failure of the filaments was reduced and the deformation increased almost at an equal load level compared to the non-coated yarn (F 500). On the one hand, the observed lower slope of the load–deformation curve was caused by the decrease of the lateral surface of the yarn. The spaces between the external filaments are filled with the polymer. This leads to a reduction of the contact areas of the yarn (in contrast to many single filaments). On the other hand, less stiffness of the fibre–matrix interface due to the polymeric coating leads to the decrease of the curve slope and to a shift to larger crack widths. ESEM images in Figures 7 and 8 show the internal area of the coated yarn and the polymer-filled interspaces of the filaments in the fibre–matrix interface, respectively.





Figure 7: Polymer coated yarn (F 501) after 28 days of storage at standard climate, internal filaments are interconnected by the polymer coating



Figure 8: Interspaces between filaments filled by the polymer dispersion

The continuous degradation of the working capacity, during the 180 days of storage in the fog chamber, is due to the decomposition and dissolution of the polymer coating. The permanent humidity in the fog chamber caused the polymer to swell and enabled the diffusion of the highly alkaline pore solution. Simultaneously, a deterioration of the polymer linkages and an increased brittleness of the polymer coating occurred and were followed by a decrease of maximum load and deformation of the coated rovings. The hydration products (CH and CSH phases) could grow into the penetrable polymer coating. Due to the intensification of the bond between the coated yarn and the cement matrix, the slope of the load–deformation curve increased. Figure 9 shows the sample after 28 days' fog chamber storage. The swelled polymer coating is distinctly visible. The described intensified envelopment of the filaments by the hydration products after 180 days' storage in the fog chamber is presented in Figure 10.



Figure 9: Swelled polymer dispersion on the filament surfaces and interspaces after 28 days of fog chamber storage



Figure 10: Intensive envelopment of the filaments by the CSH phases after 180 days of fog chamber storage

4.4. ARG-VETROTEX coated with nanoclay improved polymeric dispersion, F 502

The AR-glass rovings coated with the polymeric film former (F 501) with 5 wt% nanoclay particles led to the bond behaviour displayed by the load–deformation curves in Figure 11.







Figure 12: Interface fibre/matrix, polymer dispersion with 5% nanoclay particles (F 502), 28 days storage of standard climate, homogeneous embedding of the filaments

Figure 11: Load-deformation curves of specimens reinforced by the ARG-VETROTEX, 640 tex, additionally coated with the nanoclay doped polymer dispersion F 502, after storage at standard climate and fog chamber storage

After 28 days' storage at standard climate, the slope of the load–deformation curve is steeper than in case of the polymer dispersion without nanoclay, and corresponds to the load–deformation behaviour of the non-coated (original) yarns. Already at small crack widths, the maximum fibre pull-out forces were reached at higher load levels. Figure 12 shows the formation of the microstructure at the fibre–matrix interface characterised by homogeneous CSH phases grown on the fibre surface. They enable an intensive connection of the coated fibres with the cement matrix. Thus, a high level of load transfer can be realised at small crack widths. The intensive contact between yarn and matrix avoids the slip in the interface as far as possible and explains the nearly abrupt failure of the yarn. Besides the swelling effect of the nanoclay particles, which enhanced the contact between yarn and matrix, the nanoclay particles work obviously as nucleating species for an intense crystallisation and growth of the hydration products on the fibre surface.

With increasing duration of the fog chamber storage, also a decrease of the working capacity has been observed. Nevertheless, up to the 90th day of fog chamber storage, the level of the reduced working capacity was higher than for the other formulations investigated. After 180 days' storage at 40°C and 99% RH, an almost total loss of the mechanical performance of the yarn and the working capacity was observed.

The electron microscopic investigation of the fibre–matrix interface enabled an explanation of the phenomena as shown in Figures 13 and 14. The filaments are encased by extreme dense and thick layers of CSH phases, avoiding a relative movement of the single fibres and the load transformation. Due to the extreme intensive penetration of the CSH phases into the polymer layer, the coating became stiff and brittle and therefore elasticity was lost. The yarn acts like a compact bar and fails at minimal crack widths. The reduced maximum fibre pull-out load indicates a minimisation of the efficient yarn cross-section, due to the increased zone of stiffly embedded filaments. Furthermore, the polymer coating on the internal filaments is attacked and partly dissolved (Figure 15).





Figure 13: Interface fibre/matrix after, 180 days of fog chamber storage very dense and stiff envelopments of the filaments



Figure 14: Thick layer of CSH phases covering the filaments, brittle polymer without effect



Figure 15: Partly dissolved polymer dispersion on the surface of the internal filaments

The primary positive effect of the nanoclay particles to enhance an intensive and homogeneous growth of the hydration products in the fibre–matrix interface seems to be disadvantageous after the longer impact of the fog chamber climate. After 180 days' storage in the fog chamber, the formulation with the polymer dispersion improved by nanoclay particles showed a worse performance compared with the polymer dispersion without nanoclay.

In consequence of this unexpected development of the working capacity under accelerated ageing conditions, yet determined to use the full performance of the nanoclay improved polymeric coatings, our future research work will focus on the following questions:

- Does a reduced content of nanoclay particles lead to better long-term results of working capacity?
- Does a variation of the cement matrix (binder, pozzolans) lead to an advantageous formation of the fibre-matrix interface?



5. Conclusions

The bond strength in cement matrix composites is mainly influenced by the load transfer in the fill-in zone, if non-coated (original) AR glass rovings are utilised as reinforcement for TRC. However, the working capacity is limited because the internal filaments of the rovings hardly participate in the load transfer.

With increasing duration of storage under accelerated ageing conditions, the working capacity decreases. This decrease is caused by a partly successive degradation of the sizing and an increased portlandite formation, which disturb both the fibre–matrix and fibre–fibre bond.

Polymeric coatings of rovings applied by polymer dispersions are an appropriate method to activate the internal filaments and to enhance the bond behaviour due to the changed formation of the fibre–matrix interface. The utilisation of polymeric dispersions without nanoclay particles leads to higher working capacities at increased deformations. During the long-term ageing in the fog chamber, the effect of the coating decreased due to dissolving of the polymer and the subsequent change of the fibre–matrix interface. However, after 180 days of fog chamber storage, the working capacity of specimens was still improved, compared with non-coated rovings.

The addition of nanoclay particles to the polymeric dispersion led to a distinct enhancement of the working capacity at lower deformations due to a homogeneous formation of the fibre–matrix interface. Under fog chamber conditions, the working capacities decreased as well. Until 90 days of storage, the performance of these specimens was still improved, compared with coated specimens without nanoclay particles, whereas a long-term impact of the fog chamber climate caused the formation of very thick and stiff encasements of hydration products and led to an embrittlement and stiffening of the polymer. Therefore, the flexibility of the filaments is extremely restricted and the yarns fail like a bar.

In further test series, the changed compositions of the matrix and modified formulations of the nano-reinforced polymeric coatings are planned to be further developed in order to minimise this effect and to ensure durability.

6. References

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