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The Influence of Aggressive Environmental Conditions on the Adhesion of Applied Crystalline Materials

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Abstract: Crystalline coatings are waterproofing systems used for additional protection against increased moisture and subsurface water ingress. Even though these crystalline materials are commonly used in moisture-protective systems, they have not yet been sufficiently scientifically described. The weakest link in the chain of interaction between crystalline coatings and underlying concrete is the transition zone. To increase knowledge of the interaction between these materials, a series of experiments was prepared using a specially formulated protective mortar as the final surface layer, with the function of additionally waterproofing the structure. An experimental study of the adhesion of surface layers based on secondary crystallization to provide additional protection to concrete structures loaded with moisture or ground water exposure is presented in this paper. The series of experiments carried out consisted of an analysis of protective crystalline mortar adhesion to concrete samples of identical composition. A set of experimental measurements under the influence of various boundary conditions was carried out to determine the bond strength between two different materials. For the experimental measurements, the materials were exposed to aggressive environments for which durability verification had not yet been performed. A modified protective mortar with crystalline admixture was used as an overlayed material. This mortar worked similarly to a crystalline coating after application. Over time, there was penetration of the underlaying concrete and a secondary hydration of the cement matrix which resulted in the waterproofing of the structure. The test samples were exposed to aggressive environmental conditions in the form of freezing–thawing cycles and a carbonation process. Pull-off tests were carried out on every test sample to determine the strength of the surface layers. The penetration of the crystalline agent into the base concrete was confirmed with an SEM observation. The results of the experimental program showed that exposure to the aggressive environment further reduced the strength of the modified mortar containing the crystalline admixture. However, the bond strength between the concrete and the modified mortar exceeded the tensile strength of the concrete.

Keywords: crystalline coating; concrete; building; structures; bond strength; protective mortar; pull-off test; adhesion; surface layer

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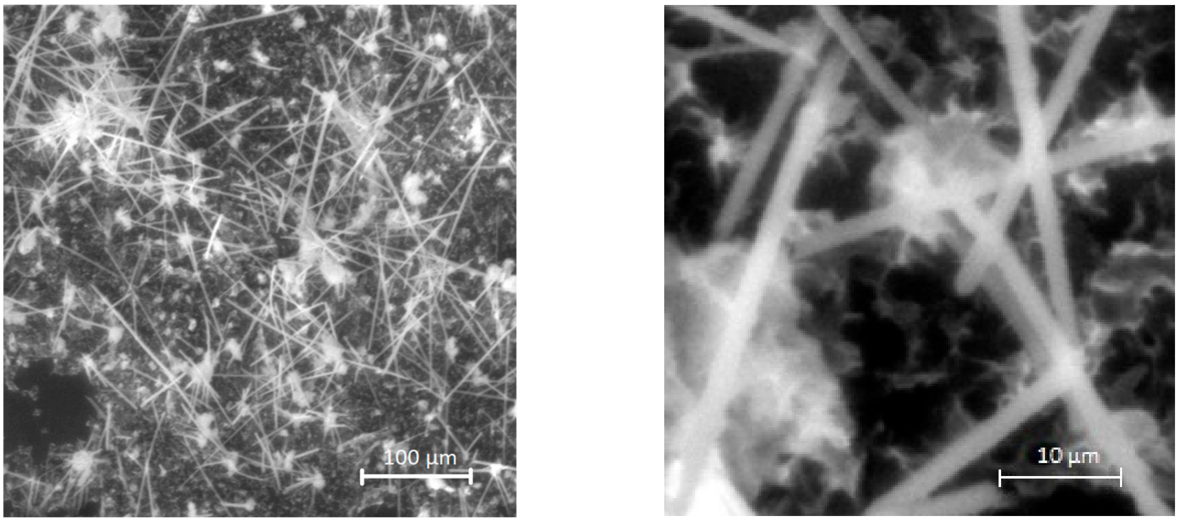
1. Introduction

Crystalline materials are commonly used in waterproofing systems in the construction industry. These materials are used in two basic ways: as a coating applied on the surface of a concrete structure or as an admixture applied directly into the concrete mixture. Protective systems based on crystalline materials are used for new structures as well as for the remediation of older structures.

Publications in the field of crystalline materials deal mainly with admixtures, meaning that much less attention is paid to coatings. At the same time, potential of the coatings for use in building structures is extensive, but there are some uncertainties that need to be studied.

Crystalline coatings are intended for application to new as well as existing or significantly old concrete structures. They are mainly used in the underground parts of buildings as an additional protection against increased moisture and subsurface water ingress. A crystalline coating is produced as a ready-mix powder, which is mixed with water in a precisely determined ratio before application. The coating is usually applied in several layers (two to three); alternatively, it is applied in the form of a screed. The entire thickness of the coating is approximately 5 mm. In addition to the dominant component, i.e., finely ground Portland cement, single-component crystalline agents also contain fine silica sand and special additives. The waterproofing effect of a crystalline coating on concrete is achieved through the reaction of the various chemical components contained in the solution when combined with the concrete matrix [1]. However, individual manufacturers keep the exact ratio of components and the specifications of the special components secret.

Crystalline materials, in essence, work in such a way that their chemical components react with the cement matrix during the hydration process, with the temporary formation of $Ca(OH)_2$ and the subsequent formation of disilicate and polysilicate anions. This cumulative process is usually accompanied by the formation of $3Ca{\cdot}2SiO_2{\cdot}3H_2O$ together with the formation of $3CaO{\cdot}Al_2O_3{\cdot}Ca(OH)_2{\cdot}12H_2O$ [2]. The product of this chemical reaction is the growth of new precipitants in the form of needle crystals inside the open-pore structure of the concrete. The needle-shaped crystals densify the pore structure of concrete, which results in a waterproofing effect. This process only occurs if there is a sufficient amount of water in the structure. If there is a lack of water in the system, the chemical reaction cannot take place in full and the effect of crystallization is limited [3,4]. The products of the chemical reaction are clearly visible from the microscopic images in Figure 1.



(**a**) (**b**)

Figure 1. Crystals in a cement matrix with crystalline admixture—SEM: (**a**) 2500× zoom and (**b**) 10,000× zoom.

The crystalline coating creates a waterproof zone in the structure of the treated concrete, which could reach up to a few centimeters in the long term [5]. This zone is formed when the crystalline agents contained in the coating penetrate the water-logged pore system of the concrete due to the concentration gradients. In the pore system, they cause the so-called secondary hydration of parts of the C-S-H gel non-fibrillar formations. As a result, the aforementioned zone with modified parameters is created. This technology helps to protect the concrete in the long term, prevents the penetration of water into and through the structure, and slows down the carbonation process. The resulting waterproofing effect of the crystallization coating does not lie only in its substance, but also in the modification of the surface layer of the concrete structure to which it is applied [6–11].

In the past, there have been many experimental laboratory measurements of the waterproofing ability of crystalline waterproofing systems aimed at verifying the waterproofing [12–15] and durability [16–19] of concrete. However, most laboratory measurements deal with crystallization additives, not coatings, and the area of crystalline coatings has not yet been sufficiently scientifically investigated.

It has been proven that a crystalline coating is able to change the concrete structure to a certain depth below the surface, thus creating a waterproof concrete structure. However, the process of secondary crystallization is relatively slow, even under ideal conditions, and the effective impact on the impermeability of the structure may only be noticeable after several months of application. The waterproofing effect of the crystalline coat is dependent on strict technological control, especially the thorough curing of the fresh coat. As mentioned above, if there is not a sufficient level of moisture inside the pore structure for at least two or three days after the coat is applied, the waterproofing effect is lost. The importance of curing fresh concrete is generally well known. Crystalline coatings have the ability to impregnate the substrate concrete over time. On the other hand, the effectiveness of the crystalline coating is mainly determined by the interaction of two different materials at the interface.

It is the interface between the concrete and the applied material that is considered the weakest link in this waterproofing measure and needs to be given sufficient attention. The performance of any method of concrete repair is highly dependent on the quality of the bond between the repair material and the underlying concrete. This is particularly true for repairs that are not anchored or tied back by encapsulating existing or new reinforcing steel or anchors, and thus rely totally on the durability of the bond to the substrate concrete for the long-term success of the repair. It is also evident that the durability of the repaired installation is only as good as the weakest link in the chain. When a repaired element or structure is subjected to excessive stresses, failure will occur in either the substrate concrete, repair material, or in the bond interface between them, depending on which plane is least able to resist the imposed stresses. Factors affecting the quality of this interface include friction; the wedging of aggregates; the direction of casting; the roughness, cleanliness, and strength of the surface; the humidity of the substrate at the time of placement of the repaired layer [20]; chemical adhesion or cohesion; use of binders; the methods of curing the applied material; and properties of the existing structure [21]. Applied surface layers often debond as a result of causes such as excessive shrinkage strains in Portland cement; excessive expansion in certain shrinkage-compensated repair materials; excessively high thermal expansions followed by cooling and shrinkage occurring during early setting and hardening reactions; and excessively high thermal expansion in repair materials during diurnal or seasonal temperature changes. Stresses in the overlay transition zone can be affected by factors such as the rate of shrinkage (plastic and drying, autogenous, carbonation), temperature stress from diurnal or seasonal temperature changes or external heat sources, changes in the humidity gradient, the amount of heat or hydration, dead loads, and changing live loads and dynamic loads [20].

The other important boundary condition is the optimal moisture level of the underlayer concrete for optimal bond compatibility. In the case of an excessively dry concrete surface, moisture can be absorbed from the repair material, causing drying at the interface and reduced bond strengths. On the other hand, repair materials should not be applied to surfaces with free surface moisture. The creation of a high water/cement ratio at the interface can have a damaging effect on bond strength [20,22].

Special attention should be paid to objects exposed to extreme environmental conditions. In cold regions or winter periods, alternating freeze–thaw cycles and subsequent expansions of microcracks in the concrete occur. This can lead to the breaking and flaking of the surface layers and progressive deterioration, or even complete structural failure in the case of the external parts of buildings. In rehabilitated concrete, the adhesive interface (as the weakest part) causes obvious initial defects in the repaired concrete before further exposure to freezing and thawing. Therefore, it is important to evaluate and increase resistance to freeze–thaw cycles [23].

2. Materials and Methods

A series of experiments were carried out consisting of the analysis of the adhesion of modified remedial crystalline mortars to concrete samples of the same composition under the influence of various boundary conditions. A mixture of remedial mortar with the addition of crystalline material was developed for this study. The mortar mix designed for additional water protection was also used as the final surface treatment of the concrete structure. Thus, both the waterproofing and final surface treatment of the structure were included in one single step in the construction process. The mortar was designed as a predominantly inorganic mixture based on Portland cement. The binding system was modified with silica fume and milled limestone. A polycarboxylate organic plasticizer was used to attain suitable workability. A sudden loss of moisture is an important aspect of such final surface treatments because it causes cracking due to the shrinkage resulting from drying; hence, an accelerator was used. The fresh mixture was workable for about 20 min. The applied water-to-mixture ratio was approximately 0.1. The basic composition of the applied mortar is shown in Table 1.

Table 1. The composition of the modified rehabilitation mortar.

Material	Quantity
CEM I 42.5 R	33–35%
Milled limestone	6–8%
Silica fume	2.5%
Sand 0.6–1.2 mm	28–31%
Silica sand 0.1–0.5 mm	14–15%
Silica sand 0.1–0.3 mm	10–12%
Accelerator	0.1–0.3%
Plasticizer	0.2–0.4%

The interaction between a crystalline admixture in a protective mortar has not yet been clearly confirmed. Previous research by the authors [5] confirmed a reduction in the permeability of the underlying concrete after the application of a crystalline coating, but the effect of the admixture in the mortar has not been systematically studied. To demonstrate the functionality of the presented application and to achieve secondary crystallization of the cementitious matrix, SEM analysis was performed on the reference specimens. This analysis was performed only to confirm the penetration between the modified mortar and the concrete substrate. Pull-off tests were used to quantify the experiment.

A series of measurements were performed to determine the strength at the interface of two different materials. Two types of aggressive environment were chosen for conducting the experimental measurements—freezing–thawing cycles and CO_2 exposure. Durability verifications for these exposures have not yet been performed. The selected aggressive environments represent some of the common environments to which structures are exposed. At the same time, these are marginal conditions that are critical to the design of the remediation measure as they could affect its durability. The samples were exposed to these environments for a predetermined period of time. A modified protective mortar with an added crystalline admixture was used as the applied material. As a result, this mortar works similarly to a crystalline coating after application. Over time, it penetrates the underlying concrete and provides secondary hydration to the cement matrix; thus, the creation of a waterproof structure is achieved.

Concrete blocks with dimensions of $300 \times 300 \times 37$ mm^3 (C20/25) were used as the base concrete, which was covered with a set of remedial mortar (see Figure 2). For the experimental program, a set of Portland cement-based mortars were designed. Pure silica sand was chosen as an aggregate, and the additive was dosed as a percentage weight of the cement. Sealing the additive positively affected the consistency of the fresh mortars, which is why the w/c ratio could be reduced. Three types of remedial mortars were applied to the concrete samples—a reference mixture, and mixtures with one and two percent of the crystalline admixture Xypex Admix C-1000 (dosed against the cement).

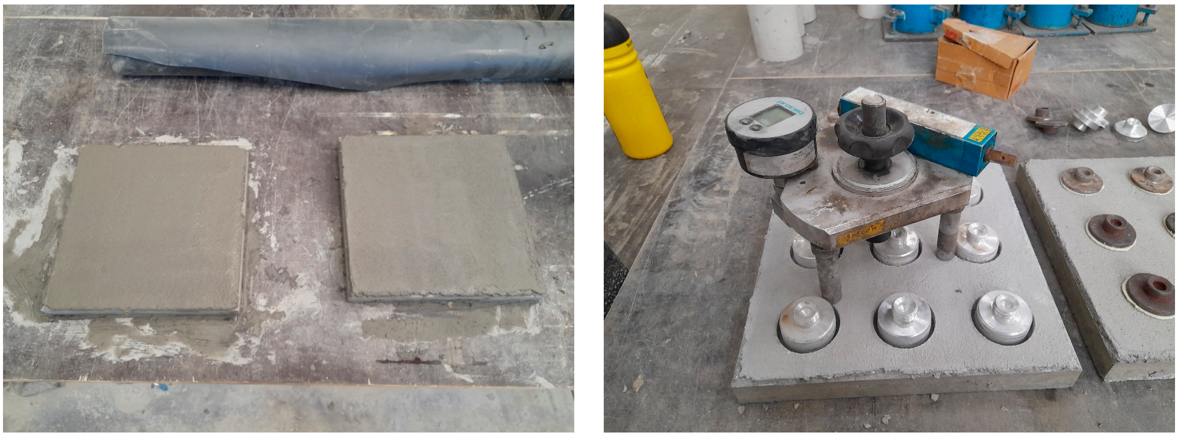

(**a**) (**b**)

Figure 2. Tested samples: (**a**) protective modified mortar applied; (**b**) test samples during the pull-off tests.

This modified protective mortar was applied to the underlying concrete surface at a thickness of 5 mm. These test specimens were stored in a humid environment for 28 days. Following this, pull-off tests were performed on these specimens. The common requirements for such repair mortars are focused on their resistance to severe external conditions.

2.1. SEM Analysis of the Interface between Concrete and Remedial Mortar

The interface between the concrete specimens and studied mortar with crystalline admixture was monitored using SEM. The purpose of this procedure was to confirm the penetration of the crystalline admixture into the base concrete matrix. The penetration effect of the crystalline coating was studied indirectly in previous research in terms of water permeability [5]. The physical interaction between new precipitants originating from the crystalline agent and the base concrete is a crucial aspect of the improvement in the functional properties of a developed mortar. The specimens used were cut from the produced slabs, saturated in epoxy resin, and then surface-treated with a platinum coating before SEM analysis for better contrast.

Only the effect of penetration of the crystalline material into the underlying concrete was monitored using SEM, not the effect of the aggressiveness of the environment. Pull-off tests were used to quantify the effect of the environmental conditions.

The microstructure of the contact zone between the base concrete and remedial mortar was documented using a Phenom XL SEM device with a CeB6 source operating at 15 kV with a BSE detector. The extracted samples were fixed in epoxy resin; after polishing, the studied areas were coated using a Mini Sputter Coater SC7620 (Au/Pd).

2.2. Cohesion of Repair Mortar with Concrete Blocks

The resistance of the studied modified protective mortars to selected exposures was studied through adhesion tests. Circular aluminum targets with a diameter of 50 mm were glued to the test surfaces using a two-component adhesive. To define the failure area, incisions were made with a core drill down to the underlying concrete around these targets. Subsequently, pull-off tests were carried out using a Proceq Dyna Z16 pull-off machine, where the pull-off stress value was automatically recorded. The targets and loading cell of the device used were equipped with hemi-spherical holds to ensure axial loading. The mode of failure was also monitored during the test, i.e., if the failure occurred in the adhesive, remediation mortar, or concrete. The procedure reflected the requirements of EN 1504-3. Nine pull-off tests were performed on every specimen with a given remediation mortar (see Figure 2).

2.3. Influence of Freezing–Thawing on the Adhesion of Crystalline Material

The second group of samples was subjected to the concrete freeze–thaw resistance test. The accelerated resistance test of the tested mortars was based on the procedure CSN 73 1322 [24], in which water-saturated samples are subjected to alternating freezing and thawing. The temperature of the freezing environment varied between −18 °C and 20 °C during each cycle. One freezing cycle consisted of 4 h of freezing and 2 h of thawing. A total of 100 freeze cycles were performed in this way. See Figure 3 for a schematic of one loading cycle. The actual temperature of the samples was not monitored. With respect to their thickness and relatively slow regime loading, it was assumed that the prescribed temperature was achieved. There was a set of six thermocouples in the test chamber. This set monitored the environmental conditions, based on which the internal temperature rise was controlled. The residual tensile properties of the concrete were monitored as standard during the test. In this case, due to the nature of the material, the tests focused on cohesion with the underground surface. After performing the freeze–thaw cycles, the pull-off tests were carried out on every samples. Nine pull-off tests were performed on each specimen with a given remediation mortar.

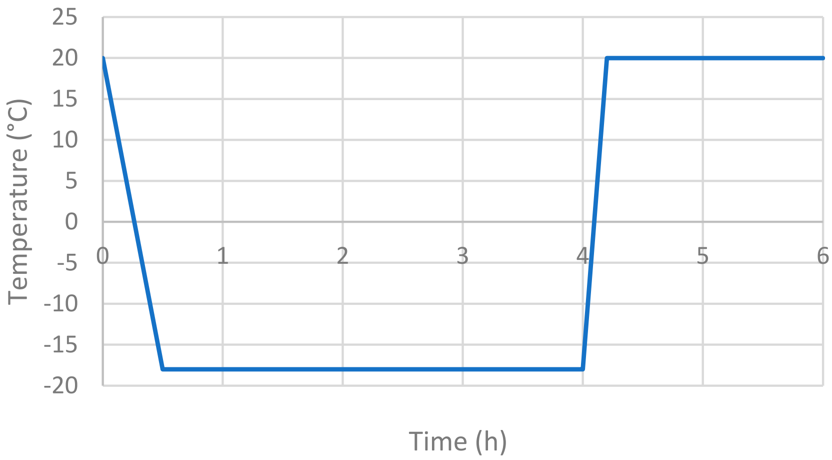


Figure 3. Scheme of one freeze–thaw cycle.

2.4. Influence of Accelerated Carbonatation on the Adhesion of Crystalline Material

The third group of test samples was subjected to an accelerated carbonation process. The test procedure was based on the test standard applied for remedial mortars according to CSN EN 13295 [25]. The exposure regime prescribes an environment in the testing chamber with a concentration of 1% CO_2 and 75–80% RH for 56 days. After the prescribed time period, the depth of the carbonation front was monitored. The concrete samples were coated on the bottom and sides with an impermeable one-component polymer dispersion-based waterproofing screed to ensure ingress only through the applied mortar. Thus, only the tested surfaces of the samples were exposed to the aggressive environment.

After being exposed to the environmental conditions, the pull-off tests were carried out on every sample. Nine pull-off tests were performed on each specimen with a given remediation mortar (see Figure 4).

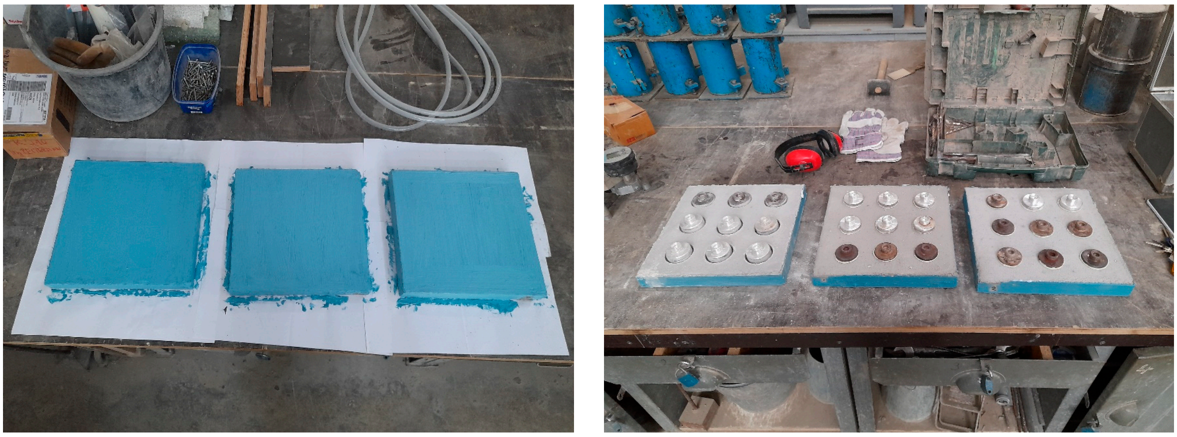

(**a**) (**b**)

Figure 4. Tested samples: (**a**) preparation of test samples before CO_2 exposure; (**b**) test samples ready for pull-off tests after CO_2 exposure.

3. Results

The influence of aggressive environmental conditions on the adhesion of applied crystalline materials was studied in the performed experimental program. The motivation was to expand knowledge about crystalline materials and to determine the influence of using crystalline admixture in protective mortars. The results of the tests have confirmed that the addition of crystalline admixture to protective mortar is able to change the adhesion of the applied surface layer. These results were achieved under various boundary conditions to provide the influence of carbonation and freeze–thaw cycles on the adhesion strength of surface layers.

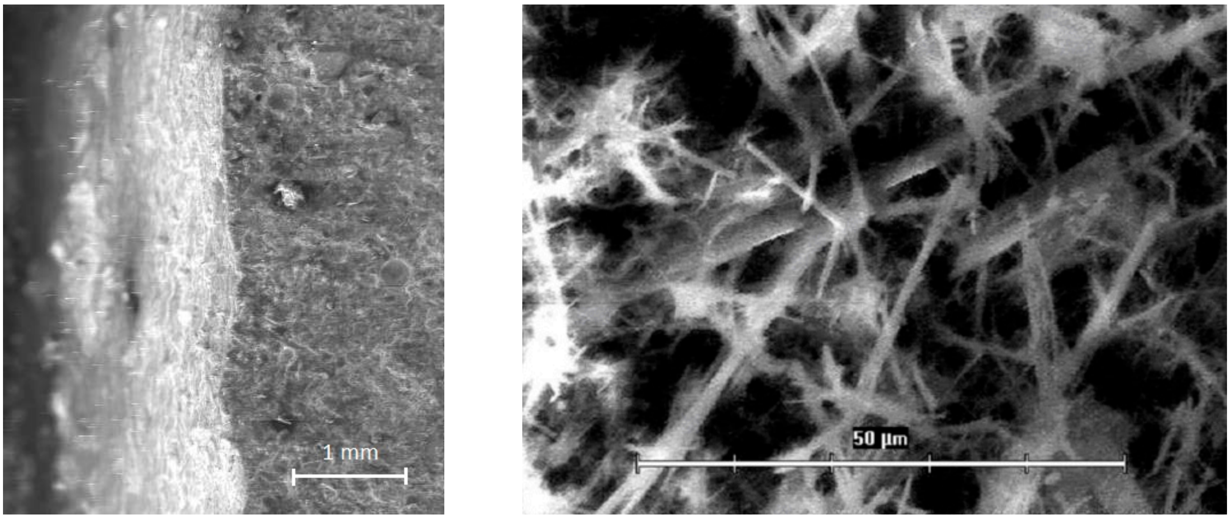
3.1. SEM Analysis

Microscopic images of the transition zone between the repair mortar and the underlying concrete were taken for the reference specimen (see Figure 5). It is clear that there is a penetration of chemicals from the mortar into the concrete structure, where material transformations occur. This results in the formation of a waterproof layer of concrete (as described above) and a possible improvement in the adhesion between the materials. Crystals resulting from the chemical reaction with the mortar also grow in the transition zone, and bonding improves between the layers of different materials. It can be seen from the image taken that the crystal growth thickens, rather than damages, the microstructure of each layer.

3.2. Adhesion of Crystalline Mortar to Reference Samples

It was found that the adhesion and strength of the modified protective mortar are so high that, in a number of cases, failure occurs in the concrete, not in the transition zone or in the mortar itself. A total of 81 pull-off tests were performed on 9 test samples, and only 28 samples failed in the mortar. The strength was so high that, in most specimens broken in mortar, there was no obvious failure in the transition zone. The strength of the modified protective mortar exceeds 3 MPa, which is the tensile strength of the commonly used concrete C30/37. An adhesion of refurbishment systems to the underlying concrete of at least 1.5 MPa is a general requirement. This requirement is fulfilled, and even surpassed, by using this modified mortar.

Figure 6 shows the values of the strengths of the surface layers according to the position of failure and the amount of crystalline admixture used. The resulting values of the surface layer strengths were obtained as the arithmetic average of the values obtained with the tear tests according to the position of failure.



(**a**) (**b**)

Figure 5. Crystalline coatings in detail: (**a**) the interface transition zone between the concrete and the coating containing crystalline admixture; 1000× zoom. (**b**) Detailed image of needle-shaped crystals inside the concrete structure (in the interface transition zone); 10,000× zoom.

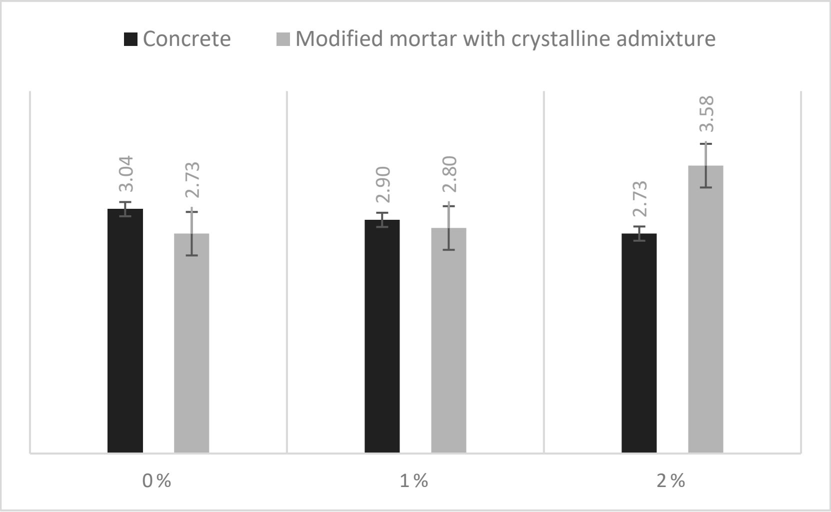


Figure 6. The strength of surface layer after 28 days without exposure to aggressive environment (MPa).

Due to the addition of crystalline admixture, an increase in mortar strength of up to 30% can be observed for the reference samples compared to the mixture without crystalline admixture.

Test samples were stored in a humid environment and the concrete surface was fully water-saturated before application of the crystalline mortar. Based on the high strength of the surface layers measured in these tests, and in relation to previous research results [21,23], it can be concluded that the moisture content of underlying concrete is a significant factor for the bond strength of surface layers.

3.3. Influence of Freezing–Thawing on the Adhesion of Crystalline Material

The results showed that the effect of freeze–thaw cycles is a reduction in the strength of the modified mortar with crystalline admixture. The strength of the modified protective mortar with crystalline admixture was reduced by more than 14% against the mortar without admixture. This is significantly more than that observed for the rest of the samples. Commonly applied requirements for mortars allow a reduction of up to 15% of the original value; the lowest limit for the residual properties of the mortar is 0.85.

This decrease in strength was evident even from the nature of the failure—2/3 of all samples failed in the mortar. The form of the specimens after pull-off tests can be seen in Figure 7. The strength of samples without crystalline admixture decreased minimally. The highest number of failures in the mortar itself occurred in samples with 1% of crystalline admixture.

(**a**) (**b**)

Figure 7. Rupture of samples after pull-off tests: (**a**) in mortar; (**b**) in mortar and concrete.

Figure 8 shows the values of the strengths of the surface layers according to the position of failure and the amount of crystalline admixture. The resulting values of the surface layer strengths were obtained as the arithmetic average of the values obtained with the tear tests according to the position of failure.

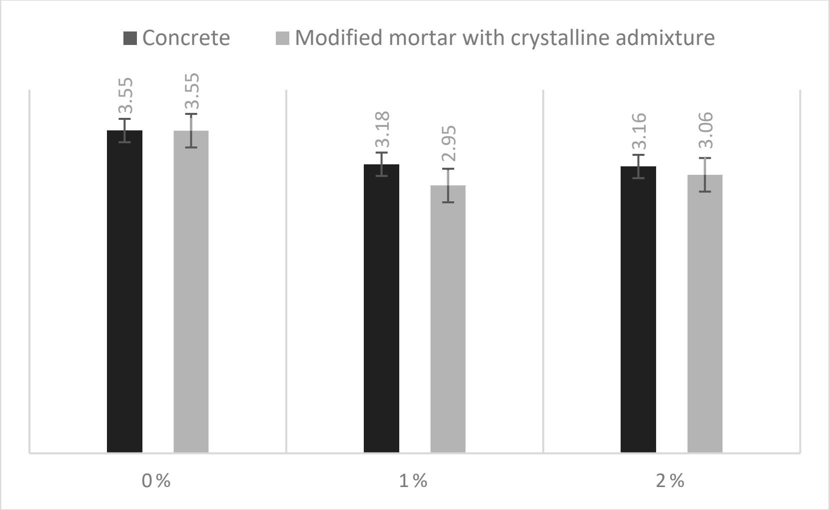


Figure 8. The strength of surface layer after the freeze–thaw cycles (MPa).

The test samples themselves could be a reason for the decrease in strength because dense concretes and mortars usually show signs of lower frost resistance. Blocks of dense concrete containing fine aggregate were used as the test samples and the modified refurbishment mortar contained only very fine aggregate.

3.4. *Influence of Carbonatation on the Adhesion of Crystalline Material*

The measured depth of the carbonation front was approximately 1 mm for all studied mortars. Hence, they demonstrated relatively high resistance to carbon dioxide ingress. However, the measurement of the carbonation front on the basis of the phenolphthalein test did not allow information about the influence of the real pH on the interface of the base concrete and the applied mortar to be obtained. With respect to the entire thickness of the mortar layer, the decline in the concentration of OH^- ions was assumed. Similarly to the samples exposed to freeze–thaw cycles, a decrease of more than 12% in the strength of the modified refurbishment mortar with crystalline admixture compared to the reference mortar without admixture could be observed after the carbonation test. The process of accelerated carbonation and secondary crystallization are, in these cases, competitive processes. In addition, the carbonation is in fact a natural process of ageing, which is accompanied by the decay of the original properties.

Figure 9 shows the values of the strengths of the surface layers according to the position of failure and the amount of crystalline admixture used. The resulting values of the surface layer strengths were obtained as the arithmetic average of the values obtained with the tear tests according to the position of failure.

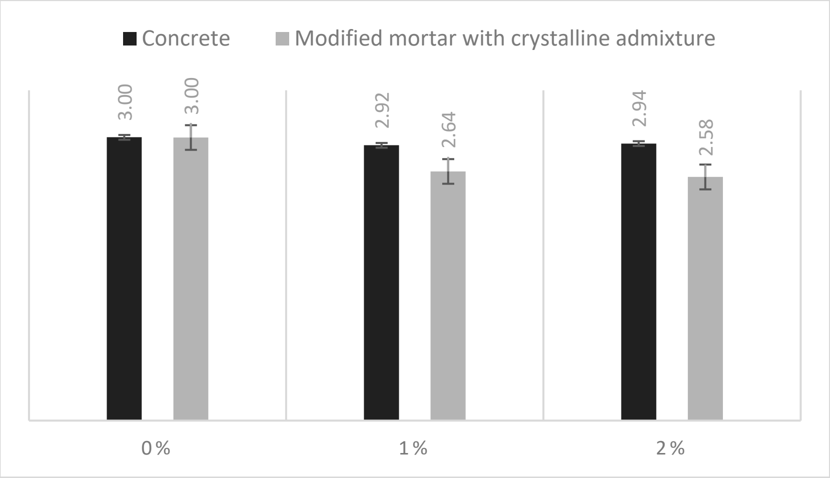


Figure 9. The strength of the surface layer after CO_2 exposure (MPa).

The strength of the samples without crystalline admixture decreased the least. The highest number of failures in mortar or glue occurred in samples with 2% of crystalline admixture.

3.5. Tear Strength Ratio of Surface Layers

The mentioned strength changes in the studied materials of different compositions (with or without admixture) after exposure are expressed as the ratio of the final tear strengths obtained after exposure to an aggressive environment to the original tear strength attained after 28 days of curing in a humid environment (see Table 2).

Table 2. Residual tear strength of surface layers (-).

Crystalline Admixture	0%	1%	2%
Freeze–thaw cycles	1.24	1.04	1.06
Carbonation	1.05	0.99	0.91

From the attained values, a decreasing trend for the strength values of the surface layers is clearly visible for the modified refurbishment mortar with crystalline admixture. The modified thin-film refurbishment mortar with crystalline admixture shows lower strength values after exposure to CO_2 than the reference mortar not exposed to an aggressive environment. The samples without crystalline admixture achieve higher strength surface layers after exposure to an aggressive environment compared to the reference samples after 28 days in a humid environment.

The strength of the surface layers decreases with the addition of crystalline admixture. However, the reduction in strength is still within the range of commonly applied requirements for mortar [26,27].

4. Conclusions

This study is focused on the change in the adhesion of an additionally applied remediation layer containing crystalline admixture under the influence of the surrounding environment.

The initial results attained using SEM observation confirmed that new precipitants from the crystalline coating penetrate the base concrete, which also has a physical influence on the properties of the interface between these two materials. The changes at the microscopic level had an impact on the technical properties, which were the core focus of the research.

The results of this experimental program showed that the tear strength of the surface layers produced from modified protective mortar is so high that, in a number of cases, failure occurs in the concrete, not in the transition zone or in the mortar itself. This knowledge is essential for determining the durability of such a remediation measure. The combination of the final concrete finish with the waterproofing function thus appears to be durable and functional. Exposure to freezing–thawing has a negative effect on the tear strength of surface layers with crystalline admixture. After exposure to freeze–thaw cycles, there is a reduction of more than 14% in the tear strength of the surface layers. After CO_2 exposure, a decrease in the tear strength of the surface layers when using protective mortar with crystalline admixture can also be observed. The results of the tests showed that the strength of the surface layers with crystalline admixture is more than 12% lower compared to the mortar without admixture.

It is important to note that even after the removal of mortar (or its damage), the concrete surface remains impregnated and the reduction in permeability is permanent (but of course, it is more effective to leave the mortar in place).

In view of these findings, it would be advisable in the future to also investigate the transport properties in the materials and to clarify the causes of these changes in strength, which are probably dependent on the content of crystallization impurities. The obtained results could help in finding an optimal method to ensure the long-term durability of structures. A reduction in water transport in building materials could improve a number of utility properties and increase the comfort of a building's inner conditions for human life. Next, extending the lifetime of building materials could improve the visual impression of used materials through a reduction in efflorescence, which is closely related to water transport. Adhesion at the interface of the materials is essential to achieve the required service life and durability.

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