

REDUCTION OF CONCRETE SURFACE PERMEABILITY BY USING CRYSTALLINE TREATMENT

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Maintenance of the present concrete infrastructure is of great concern, because a great many concrete structures are approaching the end of their service lives. Considerable attention in contemporary research is focused on advanced technologies, such as self-healing processes. One the oldest is the use of crystallizing treatments, which improve the impermeability of the concrete and contribute to prolonged durability. This work deals with the reduction of concrete surface permeability properties by using a repair mortar with a crystallizing admixture for penetrating the concrete. The evaluation was based on the chloride migration test, water penetration test and initial surface absorption test (ISAT). The mortar was polished prior to testing to eliminate the barrier effect of the applied mortar and assess the quality of the penetration. The performed experiments confirmed significant improvement of the surface concrete layer based on all the permeability procedures over time. After a year of the program, the permeability of the treated concrete surface was decreased by approximately by 40 %, but the greatest sealing effect was recorded during the first two weeks.

Keywords: Crystallizing admixture; surface permeability; chloride migration test; water permeability. .

1. Introduction

Reinforced concrete structures form number of key elements in the civilian infrastructure; however, most of them are currently approaching the end of their service lives [1]. Removal and subsequent construction of new concrete structures is extremely expensive and time-demanding; in addition, prolonging the service lives of current concrete structures is also preferable in terms of sustainability. On the other hand, annual maintenance costs constitute an essential part of the construction budget [2]. Hence, the development of new advanced technologies for improving concrete durability has become a focus of contemporary research [3,4].

Self-healing technologies seem to be very promising [3,5]. A number of procedures have been developed for concrete recovery. A modern solution entails the use of bacteria [6,7] which are able to seal the cracks. This technology has been partially transformed to preventive protection consisting of encapsulation, where the biologically based technology is activated after the crack occurs [8]. Alternatively, various types of active treatment could be used in case of encapsulation, such as 2-part epoxy, cyanoacrylates, colloidal silica or sodium silicate [9-11].

A specific part of self-healing technologies consists in the use of crystalline admixtures, which can be incorporated directly into the concrete mix or as later surface treatment. Crystalline admixtures have been thoroughly studied in recent years by a number of researchers [12- 15]. Their work was

concerned mainly with quantitative assessment of the effectiveness of the crystalline admixture.

Ferrara et al. [16] derived a model for the quantitative analysis of concrete recovery by using fracture mechanics. They studied the effect of a crystalline admixture in terms of the three-point bending test using pre-cracked specimens. The description of self-healing in terms of a numerical model was greatly extended in subsequent works [11,17].

A thorough study dealing with the process of healing surface cracks was conducted by Sisomphon et al. [13], who studied the sulfoaluminate additive and crystalline additive self-healing ability in terms of a complex set of permeability tests. They reported that calcium carbonate is the main healing product which, during 28 days, is able to close cracks up to 400 µm in width. On the other hand, a control mixture healed cracks with a width of 150 µm. The autogenous healing ability of concrete was predominantly reported for the use of active mineral additives [18-21]. The healing of cracks induced by freezing-thawing was observed by Chung et al. [22] and also in previous research, which was focused on the freezing-thawing resistance of mortars containing supplementary cement materials [23,24].

The application of the cement-based coating containing crystalline additives is used worldwide to repair new and existing structures. This approach prevents the penetration of water into the substrate concrete [25,26]. Previous research has confirmed the ability of a crystalline coating to seal a structural

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joint [27] or to repair an old substrate concrete [28]. Lim and Kawashima [26] studied the healing products of surface treatment and admixtures, both containing crystalline admixtures. They identified calcium carbonates as the main healing products with slight penetration into the substrate concrete, which subsequently exhibited increased impermeability. Teng et al. [29] reported the penetration of crystalline products to a depth of 10 mm.

This work was carried out to study the impermeable zone formed by the penetration of a crystalline admixture in the applied repair mortar. This type of repair mortar is widely used. However, the additional application of a surface coating is frequently undesirable, because it changes the original appearance of the concrete structure and this is especially true for historical monuments. Hence, this work is focused on the quality of this zone after subsequent abrasion of the repair mortar in terms of permeability as a factor in the durability.

2. Experimental program

The present experimental program was focused on assessment of the sealing effect due to the additionally applied surface treatment containing a crystallizing admixture. The sealing ability was investigated in terms of selected permeability methods in time. Concrete C20/25, whose composition is given in Table 1, was used. The employed crystalline admixture of this product is based on calcium silicate, whose precipitation of healing products according to ACI [30] is described in Eq.1. M_xR_x is a crystalline promoter reacting with tricalcium silicate and water to form modified calcium silicate hydrate and pore-blocking $M_xCaR_x - (H_2O)_x$.

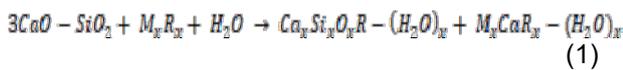


Table 1

Composition of the employed concrete.

CEM I 42,5 R	w/c ratio	Sand 0-4 mm	Crushed agg. 4-8 mm	Crushed agg. 8-16 mm
1.00	0.50	3.09	0.90	1.55

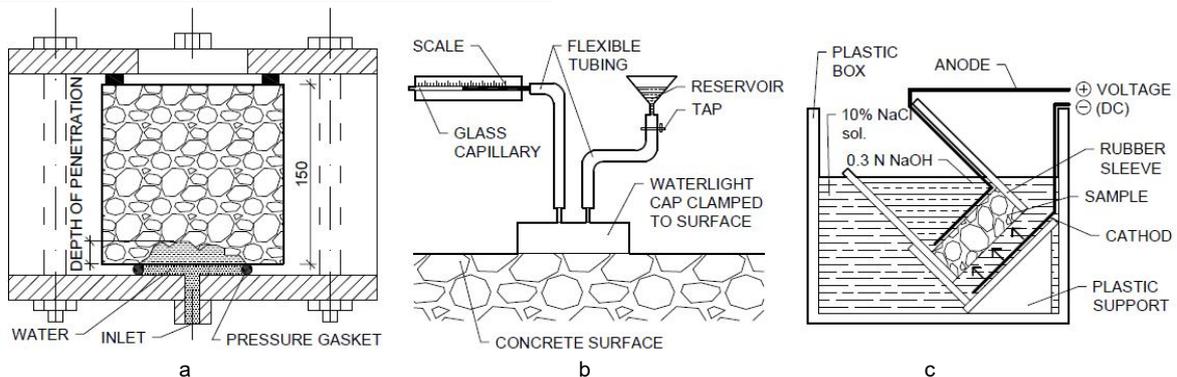


Fig. 1 - Illustration of the conducted permeability tests; a) water penetration test, b) ISAT, c) chloride migration test.

A number of commercial crystalline admixtures, which react in different ways, are available. Sisomphon et al. [13] described the hypothesis of ettringite formation during the initial phases of healing and ettringite was identified by Escoffres et al. [31]. The published works dealing with crystalline admixtures are focused on identification of the precipitated healing products and not on the chemistry of the crystalline promoter.

The produced specimens were kept for 28 days under humid conditions; subsequently, a layer of the repair mortar Xypex Concentrate with a thickness of 3 mm was applied. Then, the treated and control samples were partly immersed in a water basin, so that the applied mortar was approximately 40 mm above the water level. Treated and control samples were kept separately. Sets of treated samples were extracted at selected time intervals and the mortar was removed with a spatula to obtain the original concrete surface, which was penetrated by the crystallizing admixture contained in the mortar. These samples were subjected to subsequent permeability tests together with control sets of untreated samples. The procedures employed are depicted in Figure 1.

The compressive strength was monitored according to CSN EN 12390-3 [32] using standard cubic specimens with an edge of 150 mm at selected time intervals. Three samples were used for each single measurement.

A water penetration test was carried out at selected time intervals using cubic specimens. The treated area of the sample was exposed to the penetration of water with a pressure of 0.5 MPa for 72 hours and then the sample was broken by a split test and the maximal depth of water penetration was recorded. The average value of three measurements is given. This is the standard procedure described in CSN EN 12390-8 [33].

The initial surface absorption test (ISAT) was conducted on a cube with an edge of 150 mm. The testing apparatus was equipped with a glass capillary with a scale, allowing the recording of water flow in time in $[ml \cdot m^{-2} \cdot s^{-1}]$. The value after 10 minutes is usually used as the basic surface quality indicator [34,35]. Three tested samples of a

Table 2
Indicative assessment of the concrete permeability using ISAT [34,35].

ISAT _{10min} [g·m ⁻² ·s ⁻¹]	Permeability classification of the concrete		
	low	average	high
	< 0.25	0.25 – 0.50	> 0.50

single set were dried at 80°C for 24 hours prior to testing.

The chloride migration test was carried out in accordance with procedure NT Built 492 [36] by utilising three cylindrical specimens with diameters of 100 mm and heights of 50 mm, which were cut from the original cylinders with similar diameter and a height of 200 mm. The non-steady state chloride migration coefficient is calculated according the NT Built 492 manual [36] on the basis of the obtained depth of chloride penetration determined using a 0.1 M solution of silver nitrate and a selected test duration, applied voltage and temperature. The applied voltage and test duration are depended on the initial response of each sample.

3. Results and discussion

The performed program dealt with the sealing effect of the crystalline admixture penetrating the concrete from the previously applied repair mortar, which was removed prior to testing. The investigation was conducted on concrete with strength class C20/25. After 28 days of curing, samples covered with the repair mortar containing a crystalline admixture to penetrate the original concrete were selected. The sealing effect was evaluated on the basis of three permeability tests.

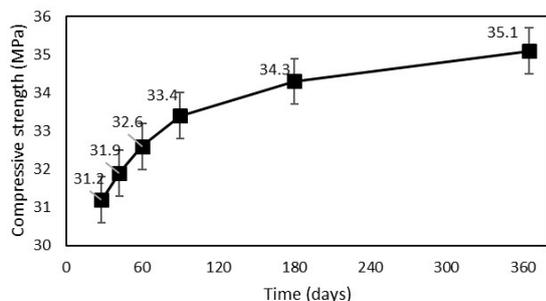


Fig. 2 - Evolution of the compressive strength in time.

The employed concrete exhibited a partial increase in the compressive strength over time, which is well illustrated in Figure 2; the gradual increase in compressive strength was 2.2, 4.5, 7.1, 9.9 and 12.5% during a year. This progress is related to gradual hydration of the employed Portland cement, which is accompanied by a reduction in the porosity as a result of growth of the hydrated phases [37,38]. Blended cements containing active mineral additives could attain greater improvement in time; however, it would complicate the assessment of the studied

admixture. In addition, the presence of active mineral additives causes considerable chloride binding, which would lead to misrepresentation of the employed chloride migration test.

The water penetration test is a standard procedure for concrete permeability measurements contained in EN 206 [39]. The requirements on the maximal level of water penetration vary according to the expected environment of the single exposure classes; nevertheless 50 mm is commonly accepted as an acceptable level of water-tightness. The obtained results are depicted in Figure 3. It is clearly visible that the effect of penetration by the crystalline admixture is apparent just 14 days after the application. Thus, the improvement was approximately 25% shortly after application, which increased slightly to 30% after one year. On the other hand, the improvement caused by the subsequent cement hydration reached approximately 16% after one year.

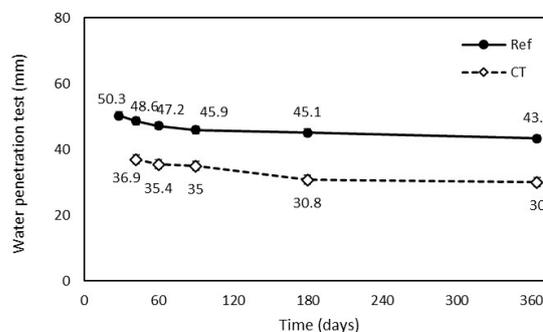


Fig. 3 - The water depth during the penetration test.

The initial surface absorption test (ISAT) is a relatively simple and rapid method for assessment of the quality of the concrete surface layer. The test involves recording of the actual flow (ml/s) of penetrating water using a glass capillary; whereas the area is constant during the test, the total mass of penetrated water could be readily determined. If the time is expressed as the square root, the shape of the resulting curves is almost linear due to the limited depth of penetration [40]. The obtained curves of a control set of samples over time are shown in Figure 4, where the gradual improvement is clearly visible. These results correspond to evolution of the compressive strength of the employed concrete.

The improvement in the surface impermeability due to penetration of the crystalline admixture is clearly visible in Figure 4, which depicts the attained evolution of the mass of penetrated water in time. It is evident that the crystalline treatment contributed to reduction of the permeability over time. The initial reduction of the mass of penetrating water is substantially lower due to application of the crystalline admixture compared to the natural densifying caused by the cement hydration. The obtained values of

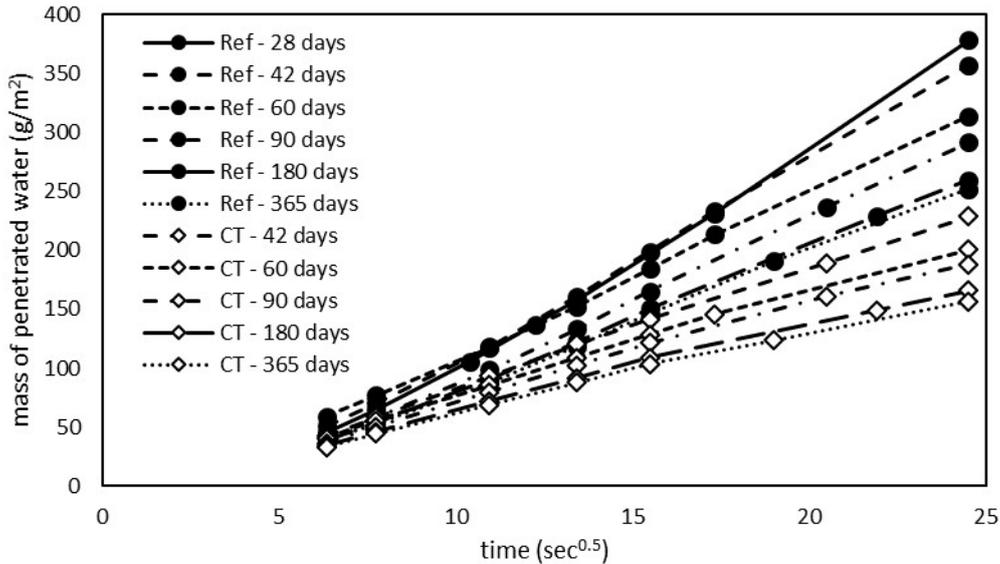


Fig. 4 - Surface water absorption of control specimens determined by ISAT.

ISAT_{10minutes}, which are commonly used for the concrete surface quality classification, are introduced in Figure 5.

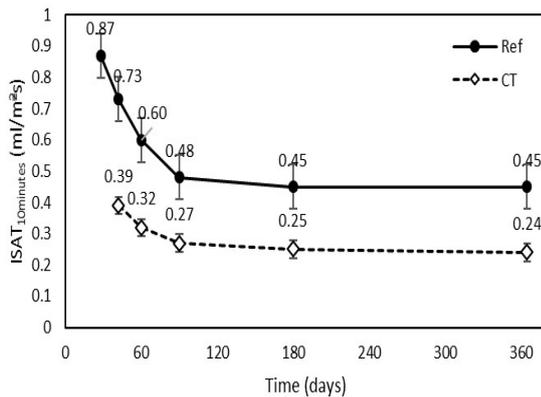


Fig. 5 - The surface absorption in time obtained by ISAT.

These results correspond to the water penetration test, which indicates a significant shift in a single set of samples due to the sealing effect of the crystalline admixture. In terms of the recommended classification introduced in Table 2, the permeability was improved by one degree, from average to low surface permeability. These results also clearly emphasise the need for curing because cement-based concrete has a substantial natural potential in this respect.

The improvement in the concrete surface impermeability was also confirmed by the chloride migration test, introduced in Figure 6. After 14 days of penetration, corresponding to an age of 42 days, a decrease in the surface permeability of approximately 20% was recorded. On the other hand, it should be noted that, from the long-term point of view, the natural decrease in the permeability due to cement hydration, in terms of the chloride migration test, is significantly lower

compared to the initial phases of the treated concrete surface. This can be partly caused by the curing regime and the process of carbonation. This is because the formed carbonates could fix part of the migrated chloride ions during the accelerated test, which has a direct influence on the resulting depth of chloride ion penetration. The problem of chloride binding in carbonated concrete was thoroughly studied in [41, 42]. On the other hand, Sisomphon et al. [13] reported that calcium carbonate is the main product of crystalline admixtures.

The final evaluation of the sealing effect of crystalline admixture penetration in terms of the conducted permeability tests is illustrated in Figure 7. The levels of the attained impermeability improvement are influenced by the testing regime. Relatively high values exhibited by ISAT are caused by the short duration of the test and proportionally small mass of concrete, which was penetrated by the acting water. ISAT clearly documents that a crystalline admixture penetrated the surface of the substrate concrete to form a thin impermeable layer. Hence, with respect to the test duration, no significant progress of the sealing effect was observed over time in terms of ISAT.

On the other hand, a substantially greater mass of concrete is penetrated during the water penetration test, leading to the observed improvement in time. This gradual improvement of the impermeability indicates crystallization of the studied admixture at greater depth. This hypothesis is supported by the results of the chloride migration test, which also exhibited improvement in time. These results confirmed previous findings that the crystalline coating is able to penetrate the substrate concrete and form a thin impermeable layer [27]. However, the major part of the penetration and subsequent sealing occurs within 14 days after application of the coating. This

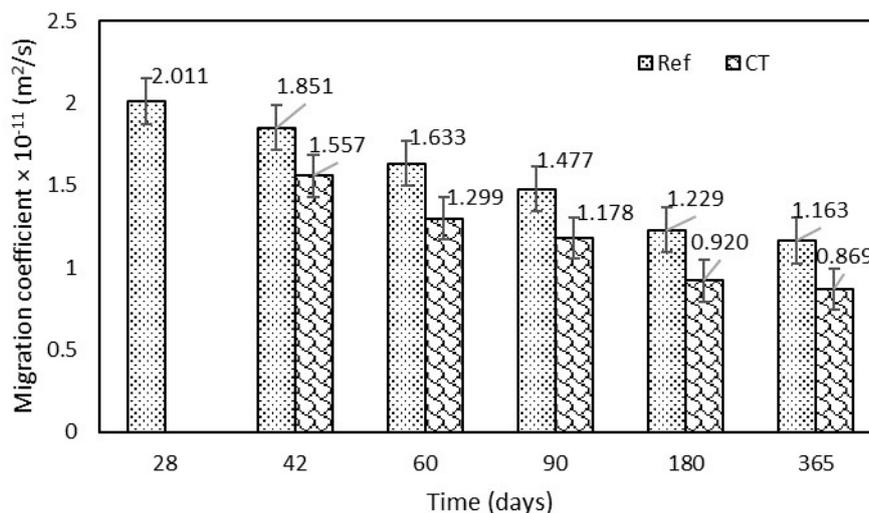


Fig. 6 - The obtained values of the coefficient of chloride migration at non-steady state in time.

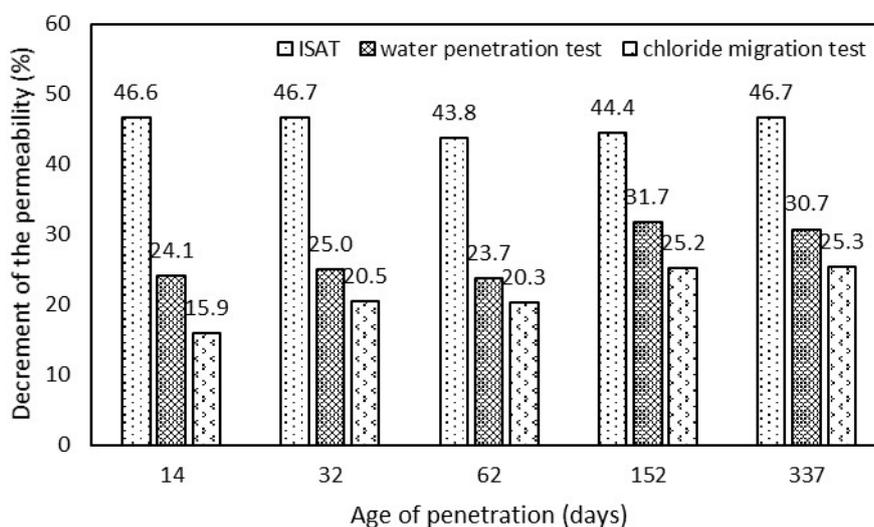


Fig. 7 - The attained improvement in the concrete impermeability.

corresponds to the finding of Al-Kheetan et al. [15]. Relatively high kinetics of concrete recovery when using the crystalline treatment were also reported by Ferrara et al. [16] and other authors [13, 29].

4. Conclusion

The performed experimental program was focused on the experimental assessment of the ability of repair mortar containing a crystalline admixture to penetrate the substrate concrete and to improve the impermeability of the concrete surface. The evaluation was conducted using three different permeability tests over time to omit natural improvement in the concrete structure due to the hydration process. The tests confirmed the ability of the repair mortar containing a crystalline admixture to penetrate the concrete surface and form a thin layer with reduced permeability. On the basis of the obtained results, it is evident that the penetration of the underlying concrete mostly takes place during the initial phases, while later

improvement corresponds to natural improvement due to subsequent cement hydration.

The implemented program confirmed the effectiveness of the crystalline admixture and its favourable impact on reduction of the surface permeability, which is closely related to the concrete durability. The obtained results are encouraging because they indicate the possibility of the removing the crystalline coating with simultaneous conservation of the achieved improvement. This aspect is important for historical monuments, where additional layers cannot be applied.

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REFERENCES

- [1] M. Alexander, H. Beushausen, Durability, service life prediction, and modelling for reinforced concrete structures – review and critique, *Cement and Concrete Research*, 2019, **122**, 17.
- [2] X. Wang, J. Zhang, R. Han, N. Han, F. Xing, Evaluation of damage and repair rate of self-healing microcapsule-based cementitious materials using electrochemical impedance spectroscopy, *Journal of Cleaner Production*, 2019, **235**, 966.
- [3] A. Sidiq, R. Gravina, F. Giustozzi, Is concrete healing really efficient? A review, *Construction and Building Materials*, 2019, **205**, 257.
- [4] K. Van Tittelboom, E. Tsangouri, D. Van Hemelrijck, N. De Belie, The efficiency of self-healing concrete using alternative manufacturing procedures and more realistic crack patterns, *Cement and Concrete Composites*, 2015, **57**, 142.
- [5] A. Sidiq, R. J. Gravina, S. Setunge, F. Giustozzi: Microstructural analysis of healing efficiency in highly durable concrete. *Construction and Building Materials*, 2019, **215**, 969.
- [6] E. Tziviloglou, V. Wiktor, H. M. Jonkers, E. Schlangen, Bacteria-based self-healing concrete to increase liquid tightness of cracks, *Construction and Building Materials*, 2016, **122**, 118.
- [7] K. Vijay, M. Murmu, S. V. Deo, Bacteria based self healing concrete – A review, *Construction and Building Materials*, 2017, **152**, 1008.
- [8] M. Alazhari, T. Sharma, A. Heath, R. Cooper, K. Paine, Application of expanded perlite encapsulated bacteria and growth media for self-healing concrete. *Construction and Building Materials*, 2018, **160**, 610.
- [9] B. Van Belleghem, S. Kessler, P. Van den Heede, K. Van Tittelboom, N. De Belie, Chloride induced reinforcement corrosion behavior in self-healing concrete with encapsulated polyurethane, *Cement and Concrete Research*, 2018, **113**, 130.
- [10] S. K. Ghosh, in *Self-Healing Materials* (Wiley-VCH Verlag GmbH & Co. KGaA, n.d., 2008), pp. 1–28.
- [11] N. De Belie, E. Gruyaert, A. Al-Tabbaa, P. Antonaci, C. Baera, D. Bajare, A. Darquennes, R. Davies, L. Ferrara, T. Jefferson, C. Litina, B. Miljevic, A. Otlewska, J. Ranogajec, M. Roig-Flores, K. Paine, P. Lukowski, P. Serna, J.-M. Tulliani, S. Vucetic, J. Wang, H. M. Jonkers, A Review of Self-Healing Concrete for Damage Management of Structures, *Advanced Materials Interfaces*, 2018, **5**(17), ID1800074.
- [12] F. ul R. Abro, A. S. Buller, K.-M. Lee, S. Y. Jang, Using the Steady-State Chloride Migration Test to Evaluate the Self-Healing Capacity of Cracked Mortars Containing Crystalline, Expansive, and Swelling Admixtures, *Materials*, 2019, **12**(11), 1865.
- [13] K. Sisomphon, O. Copuroglu, E. A. B. Koenders, Self-healing of surface cracks in mortars with expansive additive and crystalline additive, *Cement and Concrete Composites*, 2012, **34**(4), 566.
- [14] M. Roig-Flores, S. Moscato, P. Serna, L. Ferrara, Self-healing capability of concrete with crystalline admixtures in different environments, *Construction and Building Materials*, 2015, **86**, 1.
- [15] M. J. Al-Kheetan, M. M. Rahman, D. A. Chamberlain, A novel approach of introducing crystalline protection material and curing agent in fresh concrete for enhancing hydrophobicity, *Construction and Building Materials*, 2018, **160**, 644.
- [16] L. Ferrara, V. Krelani, M. Carsana, A 'fracture testing' based approach to assess crack healing of concrete with and without crystalline admixtures, *Construction and Building Materials*, 2014, **68**, 535.
- [17] G. Di Luzio, L. Ferrara, V. Krelani, Numerical modeling of mechanical regain due to self-healing in cement based composites, *Cement and Concrete Composites*, 2018, **86**, 190.
- [18] M. Sahmaran, G. Yildirim, T. K. Erdem, Self-healing capability of cementitious composites incorporating different supplementary cementitious materials, *Cement and Concrete Composites*, 2013, **35**(1), 89.
- [19] D. Józwiak-Niedźwiedzka, Microscopic observations of self-healing products in calcareous fly ash mortars, *Microscopy Research and Technique*, 2014, **78**(1), 22.
- [20] H. Huang, G. Ye, D. Damidot, Effect of blast furnace slag on self-healing of microcracks in cementitious materials, *Cement and Concrete Research*, 2014, **60**, 68.
- [21] J. Qiu, H. S. Tan, E.-H. Yang, Coupled effects of crack width, slag content, and conditioning alkalinity on autogenous healing of engineered cementitious composites, *Cement and Concrete Composites*, 2016, **73**, 203.
- [22] C.-W. Chung, C.-S. Shon, Y.-S. Kim, Chloride ion diffusivity of fly ash and silica fume concretes exposed to freeze–thaw cycles, *Construction and Building Materials*, 2010, **24**(9), 1739.
- [23] P. Reiterman, O. Holčapek, O. Zobal, M. Keppert, Freeze-Thaw Resistance of Cement Screed with Various Supplementary Cementitious Materials, *Reviews on Advanced Materials Science*, 2019, **58**(1), 66.
- [24] P. Reiterman, Influence of metakaolin additive and nanoparticle surface treatment on the durability of white cement based concrete, *European Journal of Environmental and Civil Engineering*, „article in press“.
- [25] M. J. Al-Kheetan, M. M. Rahman, D. A. Chamberlain, Influence of early water exposure on modified cementitious coating, *Construction and Building Materials*, 2017, **141**, 64.
- [26] S. Lim, S. Kawashima, Mechanisms Underlying Crystalline Waterproofing through Microstructural and Phase Characterization, *Journal of Materials in Civil Engineering*, 2019, **31**(9), ID4019175.
- [27] P. Reiterman, J. Pazderka, Crystalline Coating and Its Influence on the Water Transport in Concrete, *Advances in Civil Engineering*, 2016, **2016**, 1.
- [28] P. Reiterman, J. Pazderka, Czech WW2 concrete fortifications: Corrosion processes and remediation method based on crystallizing coating, *Acta Polytechnica*, 2019, **59**(4), 359.
- [29] L.-W. Teng, R. Huang, J. Chen, A. Cheng, H.-M. Hsu, A Study of Crystalline Mechanism of Penetration Sealer Materials, *Materials*, 2014, **7**(1), 399.
- [30] ACI (American Concrete Institute), 212-3R-2010. Report on chemical admixtures for concrete.
- [31] P. Escoffres, C. Desmettre, J.-P. Charron Effect of a crystalline admixture on the self-healing capability of high-performance fiber reinforced concretes in service conditions, *Construction and Building Materials*, 2018, **173**, 763.
- [32] CSN EN 12390-3: Testing hardened concrete – Part 3: Compressive strength of test specimens, Czech Republic (2009).
- [33] CSN EN 12390-8: Testing hardened concrete – Part 8: Depth of penetration of water under pressure, Czech Republic (2009).
- [34] S. Y. Chan, X. Ji, Comparative study of the initial surface absorption and chloride diffusion of high performance zeolite, silica fume and PFA concretes, *Cement and Concrete Composites*, 1999, **21**(4), 293.
- [35] P. A. Claisse, in *Transport Properties of Concrete* (Elsevier, 2014), pp. 26–42.
- [36] Nordtest Method NT Build 492. Concrete, mortar and cement-based repair materials: Chloride migration coefficient from non-steady-state migration experiments, Finland (1999).
- [37] L. Bágel, V. Živica, Relationship between pore structure and permeability of hardened cement mortars: On the choice of effective pore structure parameter, *Cement and Concrete Research*, 1997, **27**(8), 1225.
- [38] T. Luping, L.-O. Nilsson, A study of the quantitative relationship between permeability and pore size distribution of hardened cement pastes, *Cement and Concrete Research*, 1992, **22**(4), 541.
- [39] EN 206-1. (2006). Concrete. Specification, performance, production and conformity.
- [40] Y. Mualem, A new model for predicting the hydraulic conductivity of unsaturated porous media, *Water Resources Research*, 1976, **12**(3), 513.
- [41] M. D. A. Thomas, R. D. Hooton, A. Scott, H. Zibara, The effect of supplementary cementitious materials on chloride binding in hardened cement paste, *Cement and Concrete Research*, 2012, **42**(1), 1.
- [42] W. Wongkeo, P. Thongsanitgarn, A. Ngamjarrojana, A. Chaipanich, Compressive strength and chloride resistance of self-compacting concrete containing high level fly ash and silica fume, *Materials & Design*, 2014, **64**, 261.
