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Case Studies in Construction Materials

journal homepage: www.elsevier.com/locate/cscm

Dual-crystallization waterproofing technology for topical treatment of concrete

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ARTICLE INFO

Article history:

Received 20 May 2020

Received in revised form 14 July 2020

Accepted 17 July 2020

Keywords:

Concrete

Durability

Protection

Waterproofing

Crystallization

Hygroscopic

Hydrophilic

Hydrophobic

ABSTRACT

Concrete structures and pavements are subjected to various durability problems associated with moisture penetration through the pores and the capillary networks. Published data on waterproofing has provided conflicting and inaccurate information on crystalline-based technologies. Chem-Crete Pavix DCE is a patented, dual-crystalline waterproofing technology with performance characteristics which are materially distinct from other crystalline-based technologies. It combines hygroscopic crystallization, hydrophilic crystallization and hydrophobic characteristics. This publication presents the results of experimental research work on the significant characteristics of the dual-crystallization waterproofing technology as a treatment for existing or fully cured concrete. A program of laboratory testing has been completed in compliance with applicable ASTM and AASHTO standards using common concrete design mixes, in addition to cored specimens obtained from existing concrete pavements. The investigated experimental parameters included pore size, density, permeability, water absorption, resistance to cycles of freezing and thawing, chloride ion penetration, and mechanical properties of the surface, including pull-off strength, abrasion resistance and the coefficient of friction.

Treated concrete has been found to have higher density, less voids, less surface area, and lower permeability than untreated specimens. These characteristics yielded significant reduction in water absorption, significant increase in resistance to damage from freezing and thawing and to biological attack, and significant reduction in chloride ion penetration, without negative impacts on mechanical properties.

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1. Introduction

Concrete is one of the main construction materials for road pavements, that is continuously growing, worldwide [1]. Due to its porosity, concrete pavement is subjected to several durability problems, associated with various chemical and physical interactions with water. Consequently, the costs of highway pavement construction, maintenance and rehabilitation are rising dramatically [2]. Thus, the service life of reinforced concrete subjected to water and aggressive environments [3–5] is governed by the composition of concrete, cement composition and additives, as well as its waterproofing technology. In some pavement applications, pervious concrete is used, which contain continuous voids which are intentionally incorporated into concrete [6]. The purpose of such a special concrete type is to allow the penetration of water runoff into the lower pavement layers in order to enhance recharge of underground water. According to ASTM C125-15b [7], pervious

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concrete is proportioned with sufficient, distributed, interconnected macroscopic voids that allow water to flow through the material under the action of gravity alone. However, most concrete pavements are of the typical concrete type. Pervious concrete this is another technological approach applicable in certain cases and is beyond the scope of this introduction.

Concrete durability problems include thermal, chemical and biological attacks. The deterioration process of concrete is strongly dependent on the penetration of water and moisture through the pores and the capillary networks. Factors that affect concrete degradation includes permeability and other transport properties, which are covered extensively in various publications [8–10]. The most important factors include pore size and distribution, pH of the pore solution, concrete content of other supplementary cementing materials such as fly ash, weather conditions, air content, spacing factor and permeability of concrete. Concrete permeability is governed by the structure of pores, i.e. size and size distribution [11]. A limited degree of porosity is essential for the escape of water vapor from concrete, and for promoting concrete breathability, by allowing the transmission of water vapor through capillaries [12]. The concrete pore structure has a wide range of pore sizes ranging from less than 1 nm to 1 μm , or more [11]. The pores are categorized into gel pores (with a size range of 0.5 and 2.5 nm) [13], and capillary pores with a certain level of connectivity, including micropores (with a size range of 10–50 nm) in pastes with a small water/cement ratio and macropores (up to 5 μm) in pastes with a large water/cement ratio, and at early ages of cements hydration [14]. Increasing the porosity of concrete will increase the flow and diffusional fluxes of harmful and reactive water-soluble components into concrete structure, which causes various water-associated problems. These include concrete deterioration as a result of repeated cycles of freezing and thawing [15], corrosion of steel as a result of chloride ion diffusion within water filled capillaries [16], deterioration resulting from alkali-silica reactions [17], and from mold growth on concrete [18]. These water related problems cause further concrete distresses such as spalling, shrinking, dusting . . . etc [3–5].

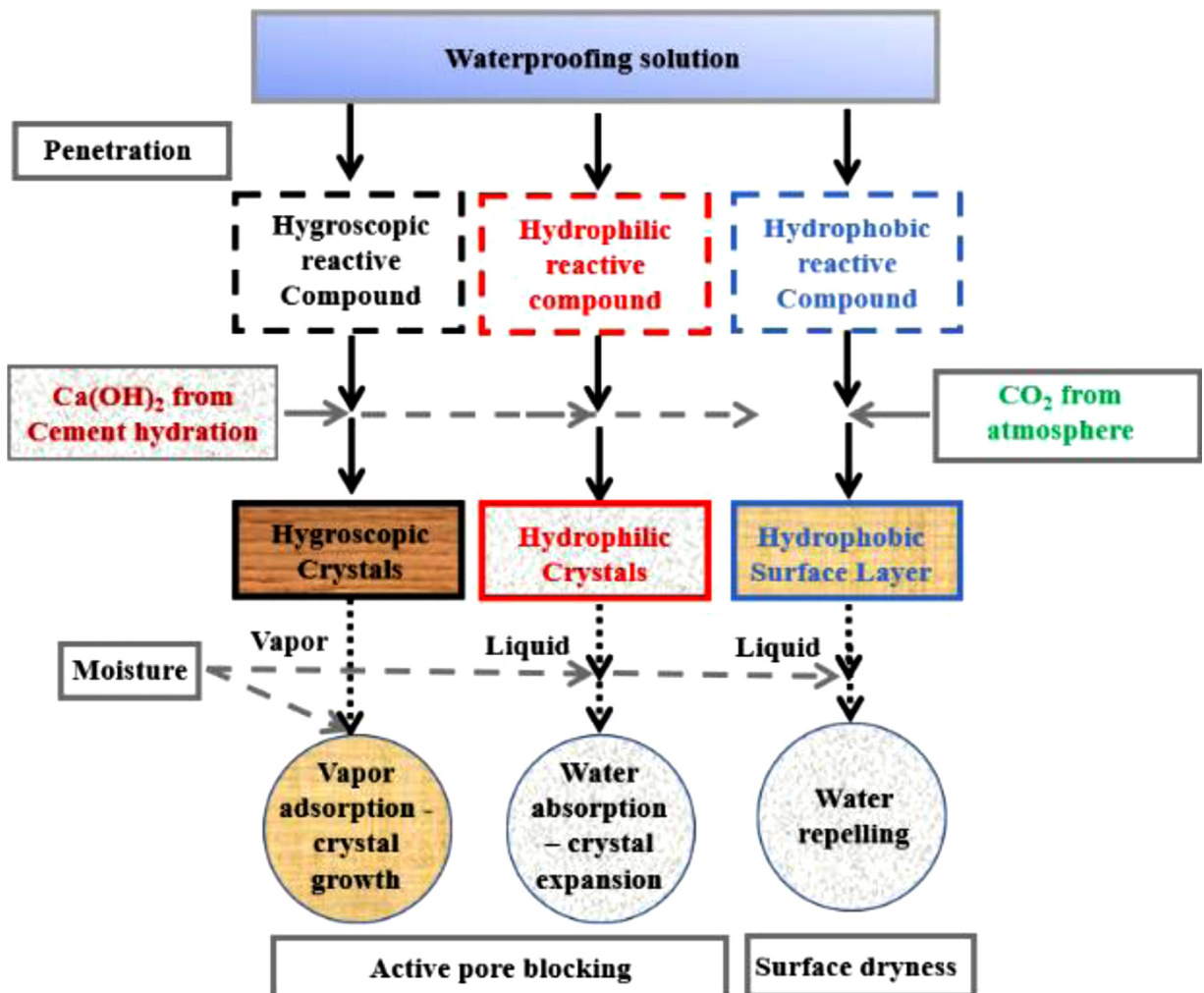


Fig. 1. A schematic representation of the formation of the DCE system and its performance mechanism.

There are several solutions for concrete durability problems, that extend the service life of concrete structures. These include traditional and innovative approaches that are based on material selection and proportioning for concrete mixtures [19,20], e.g. by modifying concrete mix design using supplementary cementitious materials such as fly ash and slags [21–24] and using chemical admixtures such as shrinkage reducing admixtures [25,26]. For existing concrete, various waterproofing technologies are used for minimizing water-associated problems. These include barrier waterproofing systems [27,28] and integral waterproofing systems [29]. Barrier polymeric systems include liquid-applied films or sheets made of acrylic resins, bitumen, styrene butadiene–styrene copolymer, ethylene propylene diene monomer (EPDM), acrylics, polyethylene . . . etc [27,28]. However, they have major limitations resulting from insufficient adhesion, acidic and sulfate attacks, which cause a limited service life, and economic losses. Cementitious coatings [30–38] are not suitable for pavements due to negative impacts of traffic conditions on their performance and service life.

On the other hand, hydrophobic waterproofing solutions such as silane and siloxane products [28] have environmental limitations since they are solvent-based, and are hazardous to the environment. In addition, such a single hydrophobic treatment cannot resist hydrostatic pressures and is not efficient for the long-term effect. Recent publications on single waterproofing systems focused on the utilization of silicate based and mineral compounds [39–47]. These systems are based mainly on the hydrophilic effect of these compounds. The single hydrophilic function is not sufficiently efficient for eliminating most of concrete durability problems. In recent publications, hygroscopic crystallization waterproofing receives great attention, and Chem-Crete products have been featured in previous papers [48]. In addition, a single-hygroscopic crystallization system based on sodium acetate has been tried on the lab scale [49]. However, these papers presented the crystallization system mainly as a single hydrophobic waterproofing, which is not a well description of the technology. Thus, the mechanism of hygroscopic crystallization system requires experimental demonstration.

The dual crystallization waterproofing engineered treatment (DCE) of Chem-Crete is based on combining hydrophilic and hygroscopic crystals, with hydrophobic characteristics, in a single treatment using an aqueous mixture [50]. This is the first paper that investigate experimentally this aqueous chemical mixture for hindering water associated problems in concrete. Its objective is to demonstrate the DCE technology and to test the durability of concrete treated with Chem-Crete Pavix CCC100 DCE, comparing treated and untreated concrete specimens. Experiments included measurements of permeability, water absorption, stability under freezing and thawing, chloride ion penetration and petrographic analysis as well as biological attack of concrete.

2. The dual-crystallization technology

The DCE technology is based on a dual-component reactive solution that is sprayed on fully cured or old concrete and undergoes various chemical reactions and physical interactions with water. The solution is water-based with low viscosity that allows its penetration into the concrete structure [50]. Measurements of penetration depths for 10 concrete specimens, indicated that the product reaches between 0.13–0.254 inch (33–65 mm), depending on the variability of the coarse aggregates located near the surface of the specimen. Within such a depth, the multiple, simultaneous chemical reactions that take place within the concrete capillaries are triggered and/or accelerated by the increase in concentration of the chemicals as a result of water-carrier evaporation. A schematic representation of the formation and the performance mechanism is shown in Fig. 1. The resulting crystals have hygroscopic [51], hydrophilic and water repelling characteristics, and thus minimize water transport through the concrete matrix, by generating a crystallization system filling and blocking the concrete voids and capillary network.

The hygroscopic crystals are produced within the concrete pores from the active ingredient of the system, through active and dynamic crystallization mechanism. These crystals block the pores of concrete and reduce the available free space for water penetration. In case of partial water penetration, they consume water by adsorption and growth. It is well documented in the literature that hygroscopic crystals are interactive with water vapor, and grow with moisture in a dynamic way [52]. Such crystal growth enhances pore blocking and consumes water vapor, resulting in a lower-moisture condition.

In addition to these hygroscopic crystals, the reactivity of the ingredients of the dual-crystallization system, with cement hydration by-products, yields hydrophilic crystals that swell, preventing water from penetrating through the large capillaries. The reaction is believed to take place on the surfaces of $\text{Ca}(\text{OH})_2$ particles, converting them to hydrophilic crystals. This reactivity mechanism consumes the undesired $\text{Ca}(\text{OH})_2$ [53] and produces a protective material. The increase in crystal size through swelling provides further moisture protection. In addition, these crystals extend toward the water source, effectively sealing it off. When the concrete dries, the crystals release the moisture and shrink, allowing the essential process of concrete breathing, which ensures concrete dryness and reduces durability problems such as biological attacks [18].

In addition to these hygroscopic and hydrophilic crystals, the ingredients of the system react with the external surface of concrete and with the internal surface of pores in the presence of carbon dioxide from atmosphere, forming an insoluble water-resistant treatment. The treatment becomes an integral part of the concrete structure through a chemical bond that is highly resistant to strong acids and alkali solution, as well as to most aggressive chemicals. Such a reaction involves carbon dioxide from air and results in the adsorption of hydrophobic [54], invisible, resin-like materials on these surfaces, ensuring better concrete dryness. They increase the surface tension of water to a degree that a further reduction in water penetration in concrete pavements through capillaries is achieved. This contribution of the system ensures a dry sub-surface within the concrete pavement and prevents moisture build up within the concrete.

Through these multi-function characteristics, the system provides a long-term sustainable solution for the major water related problems in concrete, and increases concrete resistivity to water penetration [50]. The system protects concrete pavement, concrete bridges, concrete highways, airport runways and concrete taxiways. It provides permanent, internal waterproofing and moisture blocking from positive and negative sides, preventing water, jet fuel, and oil penetration from the surface and resists aggressive chemical such as acids, caustics jet fuels and oil [50].

Overall, decreasing the pore sizes within concrete structure by crystallization, seals and protects hairline and thermal cracks and minimizes water penetration and moisture associated problems in concrete [11]. It increases the concrete resistivity to cycles of freezing and thawing. It hinders chloride ion penetration and thus solves the corrosion of steel reinforcing bars, and minimizes the attack of the concrete matrix which breaks the cement paste bond. The consumption of calcium hydroxide reduces the alkali-silica reactions [55]. The dry condition obtained by the system eliminates the concrete durability problems associated with mold growth [18].

The WP approach presented in this paper is limited to surface treatment of fully cured or existing old concrete. This usually requires additional construction management activities after curing. For existing concrete, the treatment is made as a part of the maintenance operations, which requires the shutdown of traffic. However, the application is usually made using spraying machine and finished in a short period of time. Then, the material penetrates into concrete surface layer, and the pavement becomes available for operation, without typical curing times, as required by other cementitious and barrier systems. Other alternative approaches of multiple crystallization technology, which reduces operational cost, are based on applying the surface treatment on freshly casted concrete, or adding the crystallization material as an enhancer to concrete mixture at the time of batching. This will be the subject of subsequent papers. Using supplement cementitious materials as mineral additives to concrete mixture are widely practiced in the construction of pavements. They are used for reducing concrete permeability and enhancing its durability, and from economic and environmental perspectives. This DCE surface treatment can add additional protection in such cases, or becomes more useful in maintenance works

3. Experimental

3.1. Materials

The materials used in this work included Portland cement (mainly type I) meeting ASTM C150 specifications. The coarse aggregates were crushed limestone with a maximum size of 25.4 mm, meeting the ASTM C33 specifications, supplied by Hanson Aggregates, from the Bridgeport Pit located at Bridgeport, Texas. They had a bulk specific gravity of 2.69, a fineness modulus of 3.20 and a moisture content of 0.176. The fine aggregate was natural sand, smaller than mesh #4, meeting the ASTM C 33 specifications, supplied by Hanson Aggregates, from the Ferris Pit located at Ferris, Texas. They had a specific gravity of 2.65, a fineness modulus of 2.6 and a moisture content of 0.067. Admixtures included air entraining admixture meeting the ASTM C 260, and water-reducing admixture meeting the ASTM C 494 Types A and D, manufactured by Pro Mix Technologies, Rockwell, Texas.

The used DCE aqueous material is Chem-Crete Pavix CCC100 DCE (International Chem-Crete Corporation, Richardson, Texas, U.S.A.). It is a patented solution [50] with a total solid content close to 15 %, composed of a system of active ingredients of hygroscopic, hydrophilic and hydrophilic materials. It has a specific gravity of 1.1, a viscosity of 2.4 cP, and a freezing temperature of 28 °F (2.2 °C). The test organisms used for investigating the fungal resistance of concrete included *Aspergillus niger* ATCC 9642, *Penicillium funiculosum* ATCC 11797, *Aspergillus flavus* ATCC 9643, *Aspergillus versicolor* ATCC 11730 and *Chaetomium globosum* ATCC 6205.

3.2. Concrete mix design

In order to study the effect of DCE treatment on old and fully cured concrete, two types of concrete specimens were used. These included cored specimens from old concrete and laboratory prepared specimens. Table 1 lists the two types of the used mix designs. The mix design used in preparing fully cured concrete was chosen from the range of standard concrete, also categorized as a moderate-strength concrete, which is typically used for multi-purpose concrete mix for construction, and suitable for pavements according to Texas department of Transportations (TXDOT), with a compressive strength of class C. The water to cement ratio (w/c) was 0.5. Such a high w/c ratio was selected in order to test the performance of the product on

Table 1
The used mix designs.

Component	Type 1		Type 2	
	British Units	SI units	British Units	SI units
Cement	517 lb	234.7 kg	529 lb	240.2 kg
Coarse aggregates	1850 lb	839.9 kg	1773 lb	804.9 kg
Fine aggregates	1286 lb	583.8 kg	1130 lb	513.0 kg
w/c	0.5 and 0.35	0.5 and 0.35	0.43 lb	0.43
slump	5 in	12.7 cm	2.75 in	7 cm

highly permeable concrete. This mix design was conducted for a target slump value of 5 inch (12.7 cm) and air content of 5%. The water reducing admixture was used at 3.0FL.Ozs/100 cement weights and the air entrainment agent was used at 0.4FL.Ozs/100 cement weights. In some experiments, particularly for water absorption tests, additional concrete specimens with a w/c ratio of 0.35 were prepared, for the purpose of comparison. The specimens were demolded approximately 24 h after casting, and immersed in water for 28 days until tested.

For high integrity of concrete in the abrasion resistance experiments (Section 3.9), the second design mix was used, with a higher cement content and a low w/c as listed in Table 1. The water reducing admixture was used at 3.9 Ozs/100 cement weights and the air entrainment agent was used at 0.3 F.L.Ozs /100 cement weights. The slump was 2.75 in. (7 cm) and the air content was 5.1 %. The specimens were moist cured for 28 days according to ASTM944-99. Then, they were dried in lab conditions for 14 days. The DCE was then applied at a coverage rate of 150 ft²/gallon (3.67 m²/L), then cured for 15 days.

3.3. Concrete characterizing experiments

In order to confirm the validity of the chosen mix design, experiments were performed for measuring the compressive strength and the flexural strength. The compressive strength of the design mix for the laboratory prepared specimens was measured according to ASTM C 39-01; Three concrete cylinders with 6 in. (15.2 cm) diameter and 12 in. (30.5 cm) height, were tested at standard 28-day, and their average value was calculated and recorded. The target 28-day compressive strength of the mix design was 3500 psi (24.1 MPa). The measured compressive strength of the mix was within the range of $\pm 10\%$ of the targeted compressive strength. The flexural strength of the design mix was measured according to ASTM C 78-00: Three concrete beam specimens with 6 × 6 × 20 (by in.) (15.2 × 15.2 × 0.5 by cm) were used. The third point loading standard test method was conducted to determine the modulus of rupture, as a measure of flexural strength. This test was conducted at the same time as compressive strength tests (28 days). The average flexural strength of the three samples was 573 psi (3.95 MPa), which was well above the minimum requirement of 555 psi (3.83 MPa) required by the Texas Department of Transportation. Then, the concrete mix design was identified acceptable to be used for further laboratory testing.

3.4. Application of the aqueous product on concrete samples

After 28 days of curing period, half of the specimens were treated with the DCE waterproofing substance, according to technical data sheet of the product, 200 ft²/gallon (4.91 m²/L), as documented in the Pavix patent. For the abrasion resistance experiments (Section 3.9), the DCE material was applied at a coverage rate of 150 ft²/gallon (3.67 m²/L), then cured for 15 days.

3.5. Permeability and water absorption experiments

In order to demonstrate the effectiveness of DCE technology in reducing moisture permeability and effectively managing free moisture, experiments for measuring specific gravity, percent water absorption, and percent voids in hardened concrete were conducted according to ASTM 642-97. Six laboratory prepared concrete specimens of 4 × 8 (by in.) (10.2 × 20.3 by cm) cylinders and 6 × 6 × 20 (by in.) (15.2 × 15.2 × 0.5 by cm) beam specimens were used for testing. The standard test method for determination of water absorption of hardened concrete treated with a water repelling coating (ASTM C 6489–99) was used. The percentage water absorption was estimated by using the values for concrete mass determined in accordance with the procedures described in ASTM C642, using following equation:

$$\text{percentage water absorption} = (W2 - W1)/W1$$

where W1 is the oven dry weight of the specimen, and W2 is the saturated surface dry weight.

3.6. Freeze-thaw cycle experiments

In order to investigate the effectiveness of DCE in resisting the thermal attack, the standard test method for “Resistance of Concrete to Rapid Freezing and Thawing” (ASTM C 666-97) was performed. The percentage mass loss and percentage change in length were reported with the number of cycles.

3.7. Chloride ion penetration experiments

In order to investigate the effectiveness of DCE in resisting the chemical attack, the rapid chloride ion permeability standard test method was performed according to ASTM C 1202-91 and AASHTO T 277-93. The chloride ion permeability was measured in terms of electrical charge passing, measured in coulombs, through a two-inch section of concrete specimen. The test was conducted on the top two inches of concrete specimens since they were subjected to more environmental action. All tests were conducted by maintaining the potential difference of 60 V DC for 6 h across the ends of the specimens as per ASTM C 1202-91. Test data was collected at five minutes intervals throughout the 6-h duration of the test. Since the age and the type

of the specimens have significant effects on the results, these tests were performed using cored specimens (taken from a parking lot which were two years old) and laboratory prepared specimens.

In addition, the chloride ion content was measured and compared for treated and untreated fully cured concrete using slab specimens, using a commercial ready-mix concrete (obtained from a local supplier in Maricopa Country, Arizona). It was prepared using Type II cement, with a slump of 5.5 in., w/c of 0.58, 4.2 % entrained air content and a compressive strength of 3500 psi (24.1 MPa). Chem-Crete Pavix CCC100 DCE was then applied at rate of 150 ft²/gallon (3.67 m²/L), then cured in laboratory air for 14 days. Each slab was then tested according to AASHTO T 259-00 "Resistance of Concrete to Chloride Ion Penetration". Then, the chloride ion content was measured in accordance with AASHTO T 260-97.

3.8. Petrographic analysis for air content

The standard test method for "Microscopic Determination of Parameters in Hardened concrete" (ASTM C457-98) was performed. The petrographic analysis was conducted for the same specimens used for the chloride penetration test. The magnification was 100×. For the petrographic analysis, procedure A, the linear-traverse method was performed. The test was performed on both treated and untreated cored and laboratory specimens. The data collected from this test was used to determine the air content and other parameters of the air-void system of hardened concrete such as the spacing factor of air voids through the specimens.

3.9. Mechanical properties

The impact of applying the dual crystallization on the mechanical properties was investigated. These include experiments for bond strength and surface hardness.

The standard test method for "Pull-Off Strength of Coating Using Portable Adhesion Testers" (ASTM D 4541-95) was performed using core and laboratory prepared specimens. The used core specimens, in these tests, were about 10 years old. The specimens were coated with Chem-Crete Pavix at a rate of 150 ft²/gallon (3.67 m²/L). The coated concrete specimens were cured for 10 days.

The standard test method for "Abrasion Resistance of Concrete or Mortar Surfaces by the Rotating-Cutter Method" (ASTM C 944-99) was performed. The abrasion resistance of the concrete for treated and untreated concrete specimens was measured in terms of weight loss.

The effects of surface treatment by the DCE on foot and tire heavy traffic was investigated: The standard test method for "Static Slip Resistance of Footwear Sole, Heel, or Related Materials by Horizontal Pull Slipmeter (HPS)" (ASTM F609) was performed. HPS-III Slip Master Horizontal Pull Slipmeter made by Whiteley Industries, Inc., USA, was used. The test procedures measure the static coefficient of friction between a walkway surface and standard footwear sole materials. The average value and the standard deviation from four test replicates were reported. In addition, the standard test method Using the British Pendulum Tester (ASTM E303) was performed. Six slabs of concrete, 12 × 12 in. (30.5 × 30.5 cm) were casted. Half of them were treated with DCE and the other half were used as control samples.

3.10. System toxicity testing

In order to confirm the environmental safety of DCE solution, static acute 48-h definitive toxicity tests were performed for the DCE material using two types of freshwater aquatic indicator organisms. They included Ceriodaphnia Dubia (water flea) and Pimephales Promelas (fathead minnow), frequently used to evaluate the potential toxicity of a compound or an effluent. The test procedures followed recommended methods contained in "Methods for measuring the Acute Toxicity of Effluents and Receiving Waters to Freshwater and Marine Organisms, Fifth Edition", EPA-821-R-020012, October 2004. The testing system was composed of a multi-concentration system consisting of a control laboratory water and five increasing concentrations of product added to control water. Laboratory water/control water was distilled, deionized water reconstituted with reagent grade chemicals to a hardness of 160 mg/L as CaCO₃ and a pH of 8.26.

The tests were designed to provide dose-response information, expressed as the concentration that is lethal to 50 % of the test organisms. The product was initially prepared for definitive range-finding testing by adding wide concentrations of product to water including 5, 10, 25, 50, 100 and 200 mL/L. Based on the results of the range-finding tests, definitive test solutions were prepared using a smaller range of concentrations including 5, 10, 20, 30, 40 and 50 mL/L. The pH exceeded 9.0 in the 10 ml/L to 50 ml/L solutions. An aliquot of each solution was pH adjusted to 8.5 and also tested. The 48-h static, non-renewal, definitive Ceriodaphnia Dubia test was conducted in 20 mL beakers containing 10 mL of test solution. Five Ceriodaphnia Dubia neonates, less than 24-h old, were added to each of the four replicate beakers per concentration. Neonates were fed a concentration of Selenastrum Capricornutum plus cerophyll extract while in holding prior to test initiation. The tests for Pimephales promelas were conducted in 250 mL beakers containing 200 mL of test solution. Ten Pimephales promelas larvae were added to each of four replicate beakers per concentration. Larvae originated from laboratory cultures and were six days old at test initiation. Larvae were fed laboratory cultured Artemia nauplii during test exposure. The test was conducted for 48-h during which survival was recorded daily. A control of four replicates beakers for each organism (containing five Ceriodaphnia dubia or ten Pimephales promelas) neonates each in laboratory water was conducted concurrently with the test. Survival data was statistically analyzed using the Trimmed Spearman-Kärber point

estimate test to determine the lethal concentration to 50 % of the test population (LC50). The percentage survival was determined.

3.11. Experiments for investigating the fungal resistance

In order to investigate the effectiveness of DCE in resisting the biological attack, experiments for investigating the fungal resistance of concrete specimens were performed according to MIL-STD 810G, Method 508.6. One sample was inoculated after MCE treatment was applied to the surface of concrete specimen and dried overnight. The second sample was inoculated first, then the MCE treatment was applied after inoculant dried on the surface of the concrete specimen. Specimens and cloth control were inoculated by spraying with mixed spore suspension. Prior to inoculation, each test organism was plated on potato dextrose agar. Sterilized unbleached plain weave 100 % cotton dipped in a glycerol solution was used as the viability control. The samples were placed between the control items in the incubator. The conditions for incubation were 30 \pm 1 \circ C and a relative humidity between 90 % and 99 % for 28 days. Generally, mold only need a relative humidity over 60–70 % and temperature between 25–32 \circ C for ideal growth conditions. The cloth control strips were observed after 7 days and exhibited visible growth confirming the viability of the spore suspension and verifying environment conditions in the chamber are suitable for growth. All the fungi species in the spore suspension exhibited typical growth on the potato dextrose agar after 7 days of incubation. The samples were observed at 28 days of incubation and images were taken using a microscope, at various magnifications. The level of growth of fungus was observed.

4. Results and discussion

4.1. Effect of DCE on concrete density and permeability

The experimental results for concrete density and permeable pore space of the laboratory prepared concrete specimens used in this research are presented in Table-1. Obviously, the density of the treated samples increased by about 6–7 % over that of the control specimens. This is attributed to the formation of crystals as a result of DCE reactions that add material mass within the pores and capillary network. This confirms the effectiveness of the crystallization system in filling concrete pores. As a result of such a pore filling, a reduction in the volume of permeable pore space by approximately 45–60 % was obtained as indicated in Table 2. These findings confirm the pore blocking mechanism and the effectiveness crystallization mechanism in decreasing the pore sizes within concrete structure. The pore blocking effect of the dual crystallization technology and the decrease in the size of the capillary network assist in maintaining a fraction of water molecules within the concrete structure. The entrapped vapor and liquid water molecules interact with the crystalline system by two mechanisms: bonding with the hygroscopic crystals causing their growth, and swelling within the hydrophilic crystals, yielding their expansion. These two mechanisms minimize free water in concrete subsurface.

Fig. 2 presents results of water absorption indicated as measurements of percent water absorption and percent water absorption after immersion and boiling. Obviously, there is a major reduction in concrete permeability: the percentage water absorption was reduced by about 60–75 %. This is due to the mechanisms of crystal formation and growth with moisture, which results in a major reduction in water penetration through concrete, in all water phases. Besides the above-mentioned pore blocking by the hygroscopic and hydrophilic crystals, the hydrophobicity of the formed invisible resin-like, increases the surface tension at the concrete surfaces and creates more resistant to water penetration through concrete capillary network, and thus decreases water penetration in concrete. The observed variations in the effectiveness (%water absorption) between the two types of specimens is explained by the variations in surface area. The system remained effective in the case of immersion and boiling, with a reduction of 47–64 % in percent water absorption. This is because the products of the crystallization reaction are stable at boiling temperature and thus, they retain their integrity within the pores, under this condition.

The observed minor variations for results from beam and cylindrical specimens are explained by the difference in the specific external surface (external surface area per unit volume). This is because when the specific surface area increases, the water absorption per unit volume increases.

Table 2
Experimental results of specific gravity and percent voids in hardened concrete.

Parameter	From tests on beam specimens			From tests on cylindrical specimens		
	Untreated Specimen	Treated Specimen	% Difference	Untreated Specimen	Treated Specimen	% Difference
Specific gravity	2.1	2.25	7.1	2.11	2.24	6.2
Volume of permeable pore space (%)	14.46	5.66	–60.9	14.69	8.33	–43.3

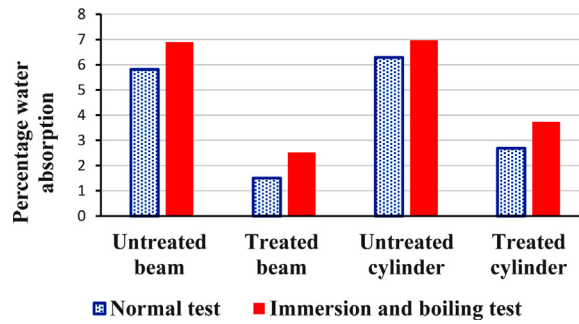


Fig. 2. Experimental results of percentage water absorption of hardened concrete according to ASTM 642-97 ($w/c = 0.5$), comparing treated and untreated concrete with beam and cylindrical specimens.

In addition, additional water absorption experiments for a similar mix design but with a water to cement ratio of 0.35 (with beam specimens) has resulted in 77.6 % reduction in water absorption (3.96 % and 0.89 % for untreated and treated specimens, respectively). The water absorption for this case is less than that for $w/c = 0.5$ for both cases of untreated and treated samples. This observation occurs because increasing w/c ratio increases the permeability of concrete, which consequently increases water penetration and absorption. Due to this impact of w/c , a high w/c ratio (0.5) was used in most of the experiments in order to test product efficacy of DCE with highly permeable concrete, i.e. under more severe situation for chemical and thermal attacks.

Preventing or minimizing water penetration through concrete is the main reason for increasing its resistance against the chemical and thermal attack. In fact, the dynamic crystallization system interacts with water to provide an effective solution to all water phases associated problems: it minimizes liquid water penetration and consumes available free water in crystal growth; it solves the solid-state water problems by preventing freezing in the pores as it consumes water in a dynamic behavior of crystal growth (and minimizing its penetration), in addition, it assists in heat storage for preventing internal freeze, as confirmed by the experimental results in the following Section (4.2). It also solves the vapor-state water problems, resulting from the re-condensation of vapor which makes it a medium for water associated problems. The system responds dynamically with condensable water in further crystal growth.

4.2. Effect of DCE on concrete resistance to freeze and thaw cycles

Fig. 3 presents results of experiments of repeated freeze and thaw cycles, as the percentage length change as a function of number of freeze and thaw cycles. Fig. 4 presents the loss in weight as a function of number of freeze and thaw cycles. In both figures, the data are presented with error bars of the measurements. After 300 cycles, the average percentage length change of the untreated specimen was -0.09% (see Fig. 3). This value is close to the maximum allowable percentage change in length set by ASTM C 666 (i.e. -0.10%). On the other hand, the percentage length change for the treated specimen was low and acceptable (-0.039%). There was no change in weight of the treated specimen even at the large number of cycles, while the percentage weight loss increased with number of cycles for the untreated specimens beyond 100 cycles (see Fig. 4). This reduction is attributed to the reduction in water penetration through concrete and due to the interaction of the dynamic crystallization system with water. In the control specimen, water penetrates and it is not managed by crystallization. Also, the available water is not consumed in further crystal growth, as in the case of treated specimen, thus it expands upon freezing and causes internal stress that results in concrete damage, which appears as a larger loss of mass and a larger change

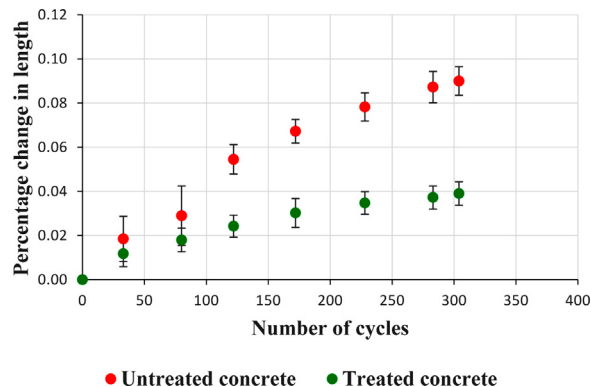


Fig. 3. Percentage change in length as a function of number of freeze and thaw cycles, for treated and untreated concrete samples.

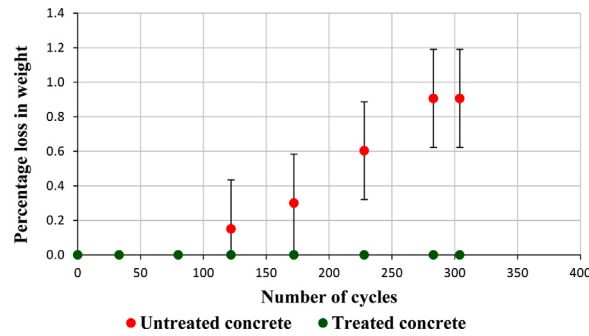


Fig. 4. Percentage loss in weight as a function of number of freeze and thaw cycles, for treated and untreated concrete samples.

in length. On the other hand, the effective blocking mechanism of the DCE results in a major reduction in the available free water in the concrete subsurface, thus eliminate the potential freezing within the pores at freezing temperatures. Thus, concrete treatment with the DCE eliminates the internal stresses resulting from freezing associated with water volume expansion. The positive effect of the presence of the crystals within the pores of concrete appears after 100 cycles, in terms of loss in weight, while it starts to appear at lower number of cycles in terms of change in length.

4.3. Effect of DCE on concrete resistance to chloride ion penetration

The experimental results for chloride ion permeability for both cored and laboratory prepared specimens are presented in Table 3. There is a major difference in electrical current passage between that for treated and that for untreated specimens, for the two cases of concrete specimens. In the two cases, about 55 % improvement in the resistance to the chloride ion penetration was obtained with treated samples. The core specimens seem to be more resistant to chloride ion penetration. Obviously, the average chloride ion penetration for the treated laboratory prepared specimens was equivalent to the passage of 1927 coulombs. This value is less than the acceptable value of chloride penetration less than 2000 coulombs. This reduction occurs because chloride ion penetration requires water medium to occur. However, DCE reduces water penetration through concrete and consumes any free moisture that reaches the pores in further hygroscopic crystallization. Thus, DCE eliminates the medium for the activity of chloride ions. In addition, the relative dryness of the concrete pores assists in preventing the continuity of water medium. It thus hinders chloride ion diffusion, which is derived by the concentration difference between surface regions and internal concrete regions that must be in a continuous water media. However, in the control specimen, much more water was available for chloride ion penetration and it was not managed by crystallization.

In addition, Fig. 5 presents the experimental results for chloride ion content for laboratory prepared specimens, at two different depths. For the same reasons mentioned above, a reduction of about 56 % in the concentration of chloride ion was obtained with the dual crystallization. This mechanism minimizes the possibility of transporting chloride ions from the deicing salts, by penetration through the network, that explains the reduction in the measured concentration of chloride ion in concrete (see Fig. 5).

With this effective reduction in chloride ion penetration, the dual crystallization system solves the associated problems with chloride ion penetration: it assists in preventing the corrosion of steel reinforcing bars, and minimizes chloride ions attack to the concrete matrix, which upon occurring breaks the cement paste bond. This protection mechanism prevents the propagation of cracks, that weakens the structure, and it prevents surface dusting.

In addition, the reactivity of the active ingredients of the system assists in the consumption of calcium hydroxide, produced from cement hydration reactions. Such a reactivity converts the calcium hydroxide into concrete harmless product thus prevents its reactivity through the alkali silica reaction that, when occurs, increases the pores size. Second, the product of such a reaction fills the pores and capillaries and expands with water due to its hydrophilic behavior. Also, the elimination of free water medium through utilizing water by crystals hygroscopic growth eliminates the ability of solvation of any available calcium hydroxide and minimizes its reactivity, and thus minimizes the availability of hydroxide to react with the aggregate and silicates. This mechanism contributes in minimizing the alkali-silica reactions and the alkali-carbonate

Table 3

The experimental results for chloride ion permeability for both cored and laboratory prepared specimens.

Type of Specimen	Chloride ion Permeability (coulombs)				% improvement
	untreated concrete		treated concrete		
	average	STD	average	STD	
Laboratory Prepared Specimens	4140	96.9	1930	194.	53.5 %
Core Specimens	145.0	28.3	62.0	9.9	57.2 %

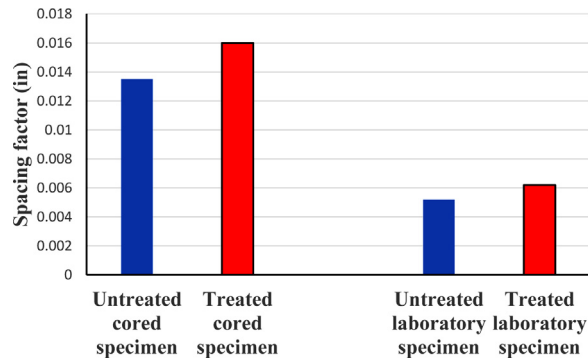


Fig. 5. The experimental results for chloride ion content for laboratory prepared specimens.

reaction (ASR) suspected for the degradation of concrete containing dolomite aggregate. Also, the relative dryness of the concrete pores and the hygroscopic behavior of the DCE minimizes the availability of water as the essential medium for the activity of mold and mildew growth. Preventing moisture build up on the subsurface of the concrete through the created hydrophobicity of capillaries network assists in preventing hosting mold growth.

4.4. Petrographic results for air content

The results of petrographic analysis are presented in Table 4 for both cored and laboratory prepared specimens, comparing investigated parameters for treated and untreated specimens. The test results provide percentage void content, and the specific surface area, which are important in analyzing the resistivity to freeze-thaw cycles. These results show that the percentage air void content for both treated and untreated specimens for laboratory prepared specimens are the same, as expected. No difference is observed between the treated and non-treated specimens since air content is set by the concrete mix design, and these is no active chemicals in the DCE that enhance air entrainment. Such an air void content of 9.67 % is considered to be excellent for the concrete with good freeze and thaw resistance. The air void content for the cored treated and untreated specimens are low, possibly no air entrainment admixture was used in the original concrete mix design of the old concrete. This level of air content is not sufficient for freeze-thaw condition.

The differences in the measured specific surface areas for both treated and untreated specimens, shows a reduction of 7–11 % for the treated specimens. Although this not a major difference, it may provide an indication of crystal growth, since part of internal surfaces of the pores are covered by the system products of reactions causing pore blocking.

Fig. 6 presents the spacing factor through the specimens: The spacing factor gives the average maximum distance from any point in the cement paste to the edge of the nearest void. The values of the spacing factor for laboratory prepared specimens, for both treated and untreated specimens are lower than that for acceptable concrete having good resistance to freeze-thaw damage. Comparing the spacing factor for the treated and untreated specimens indicates an increase by about 19 % for the treated specimens (for both cases of cored and laboratory prepared specimens). This is interpreted by the partial pore blocking, which, when occurs, increases the distance from pore to pore (i.e. the spacing factor). Such a small increase in the spacing factor is compensated with a reduction in water penetration and thus the net effect is a major increase in the concrete resistivity to freeze and thaw damage.

4.5. Impact of DCE treatment on the pull off strength and abrasion resistance

Table 5 lists the results of pull off strength for untreated and treated concrete for laboratory prepared specimens and core Specimens. There are no major differences in the pull off strength for treated and untreated specimens in both cases. In fact, the estimated percentage difference is less than the standard deviations of the measurements. Consequently, the treatment of concrete with DCE has no or little effect on the pull off strength. This indicates that the concrete treated with Chem-Crete

Table 4

The results of petrographic analysis for both cored and laboratory prepared specimens, comparing investigated parameters for treated and untreated specimens.

Parameter	From tests on cored specimens			From tests on laboratory prepared specimens		
	Untreated Specimen	Treated Specimen	% Difference	Untreated Specimen	Treated Specimen	% Difference
Air Void Content (%)	0.67 %	0.37 %	44.8 %	9.67 %	9.67 %	0.0 %
Specific Surface Area (in ² /in ³)	826	734	11.1 %	581	541	6.9 %
Specific Surface Area (m ² /m ³)	32,500	28,900	11.1 %	22,900	21,300	6.9 %

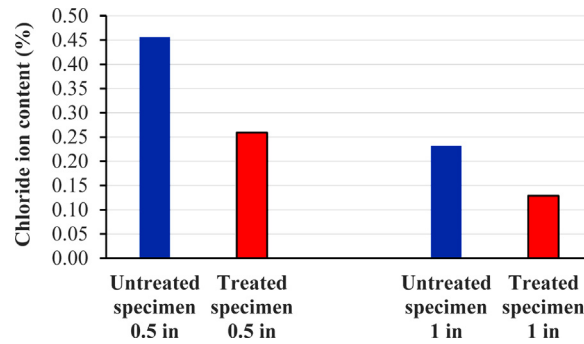


Fig. 6. Spacing factor measured using petrographic analysis, for both cored and laboratory prepared specimens, comparing treated and untreated specimens.

Table 5

The results of pull off strength for untreated and treated concrete for laboratory prepared specimens and core Specimens.

Type of Specimen	pull off strength for untreated concrete (psi)					pull off strength for treated concrete (psi)					% difference
	S1	S2	S3	average	STD	S1	S2	S3	average	STD	
Laboratory Prepared Specimens	371	385	393	383	11.1	318	456	333	369	75.7	-3.7 %
Core Specimens	335	244	327	302	50.4	240	271	229	247	21.8	-18.3%

Pavix CCC100 will experience no measurable loss in bond strength. This is because the mechanical strength of concrete results from its hydration reactions, while there is no involvement of crystallization treatment in this process. However, in these experiments, cored concrete specimens and fully cured concrete specimens were used and thus their pull of strengths were already set before the application of the DCE.

Results of the abrasion resistance test confirm the above findings from the pull off strength tests, the average mass loss on control specimen was 14.4 g, while that for the treated specimen was 13.4 g. This indicates an improvement in the abrasion resistance of the treated concrete. This slight improvement is attributed to the slight increase in the density of the treated concrete as demonstrated in Section 4.1, due to the filling effect of crystallization products. This also enhances concrete integrity and thus makes it harder.

Table 6 lists results of friction tests for untreated and treated concrete. These results show that there is a small reduction in the coefficient of friction (5.3 %), as a result of the application of the DCE to the concrete. However, the measured coefficient of friction for both treated and untreated concrete surfaces was above the minimum static coefficient of friction (0.5) set by the Occupational Safety and Health Administration. Similarly, a minor reduction in the British pendulum number was observed for the treated specimens. This slight reduction in the coefficient of friction, and in the British Pendulum number, are explained by the slight crystal growth and filling part of the surface roughness. However, fast traffic in most weather conditions require concrete pavements to have a British pendulum number above 65. Under these conditions, little to no effect on skid and slip resistance. Consequently, the treatment of concrete pavements with Chem-Crete Pavix CCC100 does not create skid or slip dangerous on roads.

4.6. Results of toxicity of DCE spills

Fig. 7 presents the toxicity results from the static acute 48-h definitive toxicity tests using *Ceriodaphnia dubia* and *Pimephales promelas* indicators, according to EPA-821-R-020012 standards. *Ceriodaphnia dubia* were determined to be the more sensitive species to Pavix CCC100 with an estimated 48 -h LC50 concentration of 7.32 ml/L. The estimated concentration to 48 -h LC50 concentration of *Pimephales promelas* was 20.3 ml/L. Obviously, adjustment to pH 8.5 increased survival for both species.

Based on the recommended application rate (100–200 ft²/gallon) (2.46–4.91 m²/L) and the probable dilution factor associated with a rainfall run-off event or drift during application, indications were that this product would not cause acute toxicity in the receiving body of water.

Table 6

Results of friction tests for untreated and treated concrete.

parameter	untreated concrete	treated concrete	% difference
coefficient of friction	0.76	0.72	-5.3
British Pendulum number	79	73	-7.6

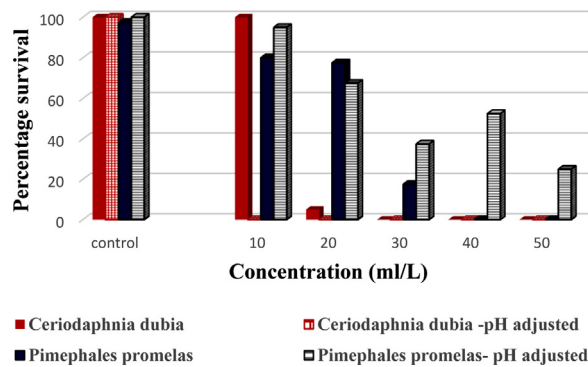


Fig. 7. Toxicity results from the static acute 48-h definitive toxicity tests using Ceriodaphnia dubia and Pimephales promelas indicators.

Table 7

Results of fungal resistance of MCE treated concrete and control specimens tested according to MIL-STD 810G.

Specimen	Inoculation	Fungus growth observation	Rating according MIL-STD 810G
MCE treated concrete	After MCE treatment	None	0
MCE treated concrete	Before MCE treatment	None	0
Untreated	Control	Traces of growth	1

4.7. Effect of DCE on concrete resistance to mold growth

The results of fungal resistance of MCE treated and control concrete specimens tested according to MIL-STD 810 G are summarized in Table 7. For the untreated control specimen, trace growth of fungus was observed. According to MIL-STD 810G, trace growth may be defined as scattered, sparse fungus growth. The growth was seen using 20× magnification or greater. MCE treated specimen did not show any visible growth of fungus for both cases of treatments. This confirms that concrete treated with MCE is resistant to mold growth. This is attributed to the major reduction in available moisture on treated surface and lack of an available carbon source to provide nutrients for growth. Experimental conditions, including temperature and relative humidity in the incubator, were conducive to mold growth as observed on cloth viability sample.

5. Conclusions

Dual crystallization waterproofing technology combines the hygroscopic and hydrophilic functions together with the hydrophobic characteristics in one single aqueous environmentally friend mixture.

The density of DCE treated concrete was increased by about 6–7 %, with a reduction in the volume of permeable pore space by approximately 45–60 % and a reduction in the percentage water absorption of about 60–75 %. The specific surface area of DCE treated concrete was decreased by about 7–11 %, with low values of the spacing factor, but with an increase of about 19 % for the treated specimens as a result of the partial pore blocking.

The DCE improved the resistance to freeze-thaw damage by 57 %. There was no change in weight after complete 300 freeze-thaw cycles for treated specimens, while the percentage weight loss increased with the number of cycles for the untreated specimens beyond 100 cycles. The DCE improved the resistance to the chloride ion penetration by about 55 %, associated with a reduction of about 56 % in the concentration of chloride ion.

The values of the pull off strength of untreated and treated concrete were close. However, results of the abrasion resistance indicated a reduction in the mass loss by 7 % after two minutes abrasion period. There was a small reduction in the coefficient of friction (5.3 %), and in the British pendulum number (7.6) as a result of the application of the DCE to the concrete. However, the measured coefficient of friction was above the minimum static coefficient of friction (0.5) set by the Occupational Safety and Health Administration.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

This research is done for Chem-Crete company to which the authors are related.

The testing was done in independent labs.

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