Construction and Building Materials 220 (2019) 90–101

Contents lists available at ScienceDirect

Construction and Building Materials

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

Laboratory and field study on the performance of microcapsule-based self-healing concrete in tunnel engineering

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HIGHLIGHTS

- Microcapsules had dual effects on the mechanical properties of self-healing concrete.
- With microcapsules incorporation, the chloride migration coefficient decreased obviously.
- The influence on the long-term shrinkage of self-healing concrete was acceptable.
- Microstructure of concrete was modified mainly due to the presence of microcapsules.
- There is a need to balance the amount of microcapsules for the future practical application.

ARTICLE INFO

Article history:
Received 10 October 2018
Received in revised form 1 June 2019
Accepted 3 June 2019
Available online 7 June 2019

Keywords:
Self-healing
Microcapsules
Concrete
Durability
Engineering application

ABSTRACT

Microcapsule-based self-healing concrete was used in a tunnel engineering project in the Qianhai area, Shenzhen and the concrete performance was investigated using laboratory and field tests. The physical properties of the microcapsules and the microstructure of the self-healing concrete were experimentally investigated. The effects of the microcapsules on the strength, permeability, and long-term shrinkage of the self-healing concrete were also investigated. The self-healing efficiency was evaluated using a compressive strength test and a rapid chloride migration (RCM) test. The results indicated that the self-healing functionality of the concrete containing 10% microcapsules gradually increased over time. The microcapsules had both positive and negative effects on the microstructure of the self-healing concrete. The use of the microcapsules resulted in a significant increase in the long-term shrinkage but the amount of shrinkage is acceptable for practical applications. No significant difference of the strain evolution was observed between the experimental and control groups in the field test, indicating that the use of microcapsule-based self-healing concrete is feasible and promising to improve the durability of concrete structures, especially in coastal civil engineering.

1. Introductions

1.1. Microcapsule-based self-healing system

The main cause of deterioration in concrete is the appearance of cracks, which threaten the safety, durability, and the functionality of concrete structures [1–3]. In general, cracking inevitably occurs when the concrete structure is subjected to a variety of mechanical and environmental actions. In order to reduce the repair and maintenance costs of concrete infrastructure, multiple studies [4–7] have been conducted in the past twenty years to develop techniques to prevent concrete from cracking. Over the years, mineral additives have received much attention due to its physicochemical properties, which reduce the cracking risk by minimizing the early cracking susceptibility of concrete [8,9]. However, this enhancement may only improve the durability of the concrete for a limited period rather than lengthen the service life of concrete structures in a smart way from long-term point of view. In addition, due to the fact that micro-cracks formed in the concrete matrix are invisible and their locations are usually difficult to determine, this method cannot be applied to prevent micro-cracks caused by loads or environmental conditions in the later service stages.

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https://doi.org/10.1016/j.conbuildmat.2019.06.017
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Fig. 1. A photograph of the Qianhai Tunnel under construction in Shenzhen.

One way to address this problem is using incorporation of a certain amount of self-healing microcapsules [10], which disperse throughout the cementitious materials and provide mechanical protection for the healing agents that are inside the microcapsules [11,12]. When cracks appear, the embedded microcapsules around the crack surfaces break under the crack-tip stress and the healing agent inside the microcapsule is then pulled into the crack by capillary action to achieve healing [13]. In recent years, researchers and practitioners have carried out extensive research in the application of the microcapsule-based self-healing technique that is capable of reducing permeability and restoring the mechanical properties of the cementitious materials after the damage has occurred [14–19]. For example, Wang et al. showed that the concept of using microcapsules as bacterial carriers to effectively heal cracks is feasible and promising [20] and more recently, Gupta reviewed the current state of autonomous healing in concrete by bio-based healing agents [21]. In addition to the bacterial precipitation, the approach to encapsulate chemical healing agents [22–25] has also been found to be effective in restoring the performance and properties of cementitious materials and this approach is widely considered a versatile strategy for self-healing. However, although a number of studies have been conducted on the use of cement paste or mortar, to date, only a few publications on the use of a microcapsule-based self-healing system in concrete are available. Furthermore, most of the previous studies were mainly focused on lab-scale research rather than on practical applications in construction engineering.

In this study, building on previous research activities and achievements at Shenzhen University [26–31], a microcapsule-based intelligent resilience system was applied to tunnel construction in an attempt to promote further development of the novel microcapsule-based self-healing system for application in concrete structures. Focusing on the control principles of strength and durability of concrete structures, this application study examines the strength and durability-related transport performance of self-healing concrete by using a microencapsulated healing agent in a practical engineering environment and conducting laboratory and field tests. The physical properties of the microcapsules synthesized by an in-situ polymerization method were investigated first. A long-term shrinkage test was used to assess the influence of the embedded microcapsules on the shrinkage behavior of the self-healing concrete. To investigate the performance recovery regarding the strength and permeability of the self-healing concrete, a compressive strength test and rapid chloride migration (RCM) test were conducted respectively. The microcapsule-based self-healing concept was investigated using scanning electron microscopy (SEM) and energy dispersive spectroscopy (EDS). In addition, we briefly address the objectives of the field test and then discuss the results of the strain monitoring and experimental observations. The future prospects of the use of microcapsules in large-scale self-healing concrete applications are also discussed.

1.2. Engineering background: The Qianhai tunnel project

The Qianhai Shenzhen-Hong Kong modern service industry cooperation zone was officially approved by China's State Council in 2010; it has the purpose of serving as an experimental business zone for better interaction between Mainland China and Hong Kong. Located at the edge of Shenzhen, the Qianhai Tunnel is surrounded by many complex engineering projects in infrastructure construction and has drawn increasing attention from researchers and engineers.

Construction began in 2016 (Fig. 1) and the tunnel was planned as the main urban underground road between two major roads of the cooperation zone in the southwest of Shenzhen. In addition to some special components, a cast in situ construction method was adopted for the tunnel in the construction of the reinforced concrete structures. The large-scale use of concrete makes crack control very important even though this is a relatively mature technology, especially for tunnel engineering. The Qianhai Tunnel is located in a coastal area and the moist environment may substantially increase the risk of ingress of aggressive ions that may accelerate the structural degradation. Therefore, it is significant to conduct a study of the self-healing concrete performance in combination with an applied construction project.

2. Materials and methods

2.1. Materials

The urea-formaldehyde/epoxy resin microcapsules used in this study were prepared by means of an in-situ polymerization procedure in the Guangdong Key Laboratory of Durability in Coastal Civil Engineering. Fig. 2 shows the formation process of the microcapsules synthesized by using urea-formaldehyde resin as the shell and epoxy resin E-51 as the healing agent. The details of the synthesis method were presented in our previous study [12]. The mechanical properties of the microcapsules have been investigated and given in [27,29]. The catalyst (MC120D) used for the reaction with the healing agent (epoxy resin E-51) was obtained from Guangzhou Chuanjing Electronic Materials Co., Ltd. A strain gauge with the sensitivity of 1 $\mu\varepsilon$ was used for monitoring the internal strain of the concrete. The commercial fresh concrete used for the tunnel construction and experimental study was supplied by Shenzhen Gaoxinyuan Co., Ltd.

2.2. Mix proportion and specimen preparation

The mix proportions of the concrete are given in Table 1. Two concrete mixtures were used in this study and all the concrete mixtures had the same water to binder ratio (w/b) of 0.32. As described in our previous studies [12,30], the mix proportion for the experimental group was modified based on the control group

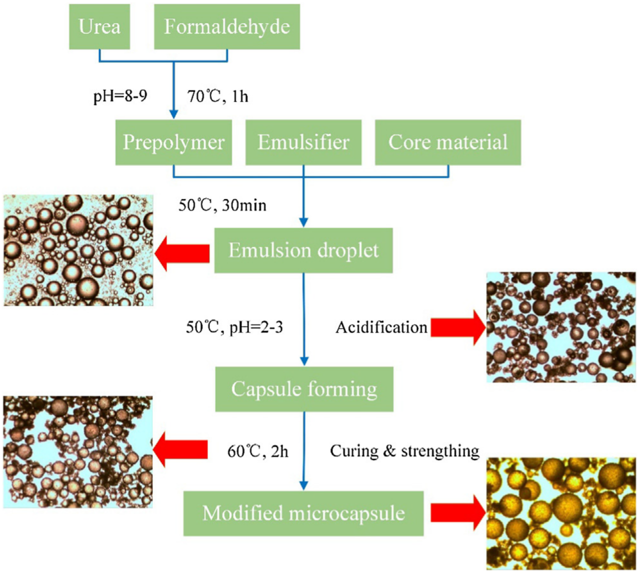


Fig. 2. Synthesis process of the microcapsules prepared by the in-situ polymerization method.

by adding 10% microcapsules and 5% curing agent by weight of the total binders. The results of the experimental group with the microcapsules and the curing agent were compared with those of the control group to evaluate the effect of the urea-formaldehyde/epoxy resin microcapsules on the self-healing efficiency of the concrete. Prior to casting the concrete, the microcapsules, curing agent, and fresh concrete were equably mixed by continuous mechanical stirring.

Table 2 summarizes the test specimens and test methods for each concrete mixture. Four types of specimens of different sizes and functions were used in this study to meet the different test requirements. All the specimens including the precast concrete slabs (Type-IV specimens, Fig. 3) were cast at the construction site at an ambient temperature of about 30 °C. In order to prevent moisture loss, the molded specimens were covered with plastic film. After demolding, the Type-I, Type-II, and Type-III specimens were transferred to the laboratory and were cured until the specified testing age. For the Type-IV specimens, two strain gauges with two different directions were embedded in each precast concrete slab and the slabs were cured in a concrete-curing pool until they were assembled. The schematic illustration of the slab is shown in Fig. 3. The specific test method for each type of specimen will be described further in Section 2.3.

It is noted from Table 2 that for cube compressive strength test, the sample size is 100 mm × 100 mm × 100 mm (cube); while for rapid chloride migration test, the sample size is Φ100 mm × 50 mm (cylinder). Before pre-loading, the difference between two types of sample is taken into consideration. The purpose of the pre-compressive load is to intentionally introduce damage that will increase the number of internal defects and change the pore structure of self-healing concrete. Unlike the compressive strength test, the rapid chloride migration (RCM) test is typically measured on cylindrical specimens to determine the chloride diffusivity. According to the pre-experiment, excessive pre-compressive load would lead to undesirable results, i.e. some through cracks may occurred in the specimens, which may invalidate the RCM test. This unfavorable result should be caused by a mismatch between the preloading level and the size of the specimens. To this end, in order to find a balance between the test requirements and the self-healing effect, we adopt a differentiated treatment strategy to suit different test specimens.

Regarding the treatment of the specimens, as illustrated in Table 2, firstly, all the specimens (Type-I) were cured in a curing room (temperature T = (20 ± 2) °C, humidity RH = 95%) for 60d. Secondly, all the specimens were subjected to a compressive load with 60% of the maximum compressive strength (the control group σ_{max} = 52.66 MPa, the experimental group σ_{max} = 38.05 MPa). Thirdly, after pre-loading, one part of the specimens was retained to test the compressive strength ($\boldsymbol{f}_{\mathbf{initial}}$) immediately while another part was healed at a temperature of 60 °C for 3d, 5d, 7d, 14d, and 28d until they were tested ($\boldsymbol{f}_{\mathbf{healed}}$).

2.3. Test program

2.3.1. SEM, thermal gravimetric analysis, and differential thermal analysis of the microcapsules

After drying the fabricated microcapsules, SEM (FEI, Quanta TM 250 FEG, USA) was used to evaluate the morphology of the microcapsules. In addition, 100 additional microcapsules were analyzed using SEM to investigate the particle size distribution of the microcapsules. The thermal stability properties of the microcapsules were measured using thermal gravimetric analysis (TGA) and differential thermal analysis (DTA) (STA 409PC, Netzsch, Germany) under a dynamic nitrogen atmosphere at a heating rate of 10 °C/min. The morphology of the microcapsules at different temperatures (50 °C, 100 °C, 150 °C, 200 °C, 250 °C) was investigated using a biomicroscope (QianKe, XSP-BCC, China).

2.3.2. Concrete cube compressive strength test

The Type-I specimens with a dimension of 100 mm × 100 mm × 100 mm were cast for the compressive strength test. In order to minimize the effect of the hydration of the cement particles, all specimens were cured in a curing room (temperature T = (20 ± 2) °C, humidity RH = 95%) for 60 d. Damages were then caused to the self-healing concrete by applying a compressive load with 60% of the maximum compressive strength (σ_{max}). After pre-loading, one part of the specimens was retained to test the compressive strength immediately while another part was healed at a temperature of 60 °C for 3 d, 5 d, 7 d, 14 d, and 28 d until they were tested. A universal testing machine (YAW6306 from MTS Systems Co., Ltd) was employed to load the specimens at a uniform speed of 0.5 MPa/s according to the China National Standard (GB/T50081-2002) [32]. The compressive strength was calculated as follows:

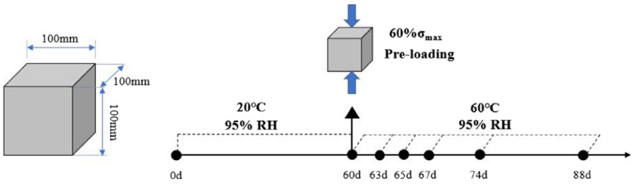
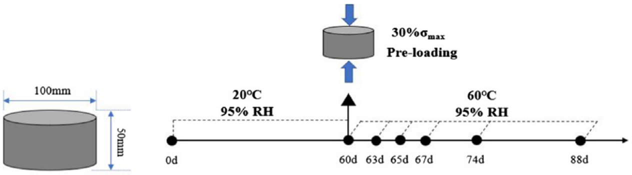
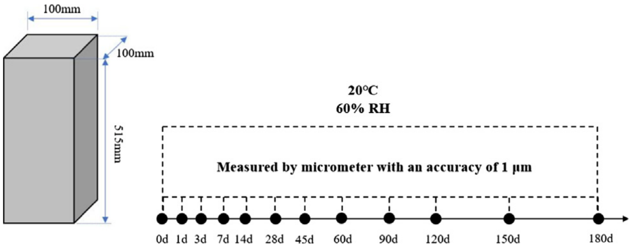
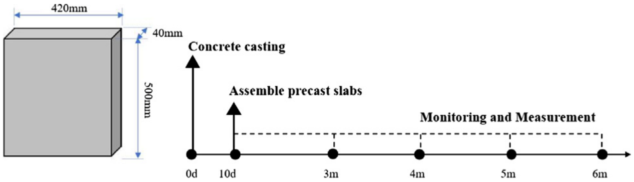
$$f_c = 0.95 \times \frac{P}{A} \tag{1}$$

where fc refers to the compressive strength of the concrete (MPa), 0.95 is the size conversion coefficient, P and A refer to the failure load (N) and area under compression (mm^2), respectively.

Table 1
Mix proportions of the self-healing concrete.

Mix ID	Cement (kg/m^3)	Slag (kg/m^3)	Fly Ash (kg/m^3)	Expansive Agent (kg/m^3)	Sand (kg/m^3)	Stone (kg/m^3)	Superplasticizer (kg/m^3)	Water (kg/m^3)	Microcapsule (kg/m^3)	Curing agent (kg/m^3)
Control	282	94	51	48	671	1049	11.88	153	0	0
Experimental	282	94	51	48	671	1049	11.88	153	47.5*	24

Table 2
Test specimens and test methods for each concrete mixture.

Test type	Sketch of the specimen preparation, curing, and testing	Test method
Type-I		Compressive strength test
Type-II		Rapid chloride migration test (RCM)
Type-III		Long-term shrinkage test
Type-IV		Field test

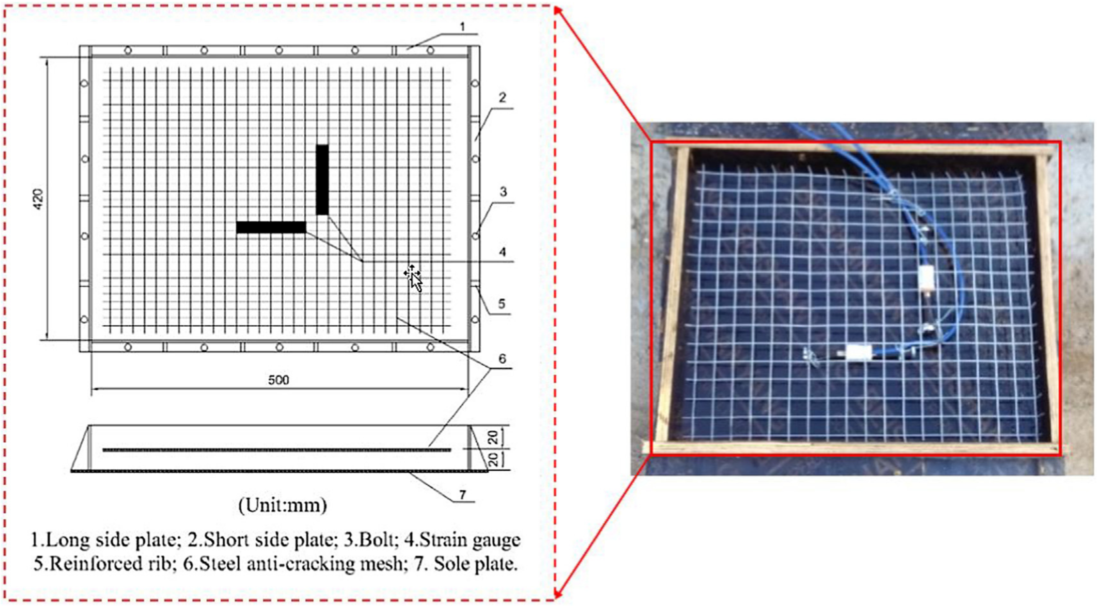


Fig. 3. Schematic illustration of the precast concrete slab used in the tests.

2.3.3. RCM test

The RCM test was conducted for the Type-II specimens (Φ100 mm × 50 mm), which underwent the same pretreatment process as the compressive strength test specimens except for the preloading of the specimens. According to our previous studies on the fracture behavior of microcapsules [13,27], it has been confirmed that microcapsule breakage is tightly associated with micro cracks appeared in the concrete matrix. When a crack approaches a microcapsule and the crack-tip stress exceeds the threshold value of 10 mN to 20 mN, the microcapsule will break due to the stress concentration effects. Specifically, cracks generated in the matrix determine the microcapsule breakage, as these create large stress amplification at cracks tips. In fact, the microcapsule based self-healing system is capable of healing cracks caused by external loads within a load range (about 30%$\sigma_{max} \sim$ 80%σ_{max}) [25,28], rather than a certain load. When a greater load is applied, new cracks appear and propagate in the concrete matrix. In this case, the self-healing properties of microcapsules can be divided into two parts: on the one hand, the healing agent released from broken microcapsules block the propagation of cracks by bonding matrix materials; on the other hand, those microcapsules that did not rupture during the first loading can still provide healing function. Therefore, combining test requirements and the technical feasibility, 30% of σ_{max} was used as the preload to investigate the self-healing effect of microcapsules on the permeability.

Before the test, all the specimens with a thickness (L) of 50 mm were coated with paraffin and then vacuum saturated in a calcium hydroxide solution (saturated CaOH). A sodium hydroxide solution (0.3 mol/L NaOH) acted as the anode and a sodium chloride solution (10 wt% NaCl) acted as the cathode. During the test, an electric field (U = 60 V) was continuously applied through the specimens until the end of the test for a given duration (t). After that, the specimens were cut from the middle portion and a silver nitrate solution (0.1 mol/L) was applied to the freshly split specimens to measure the average chloride penetration depth (X_d). By recording the test temperatures (T), the chloride migration coefficient was determined using Eq. (2) according to the China National Standard (GB/T 50082-2009) [33].

$$D_{RCM} = \frac{0.0239 \times (273 + T)L}{(U - 2)t}\left(X_d - 0.0238\sqrt{\frac{(273 + T)LX_d}{U - 2}}\right) \quad (2)$$

For each mixing proportion, three Type-III specimens with a dimension of 100 mm × 100 mm × 515 mm were cast for the long-term shrinkage test to investigate the effect of the microcapsules on the shrinkage behavior. After demolding, the contact shrinkage measuring method was employed to determine the shrinkage and the test was carried out under the conditions of constant temperature (T = (20 ± 2) °C) and constant humidity (RH = (60 ± 5)%) until the time of testing. The shrinkage of the concrete was measured at 1 d, 2 d, 3 d, 4 d, 5 d, 6 d, 7 d, 14 d, 28 d, 45 d, 60 d, 90 d, 120 d, 150 d, and 180 d according to the China National Standard (GB/T 50082-2009) [33]. The test time began when the specimens were moved into the constant temperature and humidity room and the shrinkage was measured using a dial indicator with an accuracy of 1 μm.

2.3.4. SEM and EDS analyses of the specimens

In addition to evaluating the morphology, SEM and EDS (AMETEK EDAX, EDS, USA) were used to verify the breakage of the microcapsules upon cracking and to identify the crack-healing products. After the compressive strength test, samples were obtained from the fractured surface of the specimens. Prior to the SEM observation, the samples were dried in a vacuum oven and gold-coated for 120 s to achieve better observation results.

For the Type-IV specimens, three precast concrete slabs were cast for the control group (numbered P1, P2, P3) and three precast concrete slabs with 10% microcapsules addition were cast for the experimental group (numbered P4, P5, P6). Both experimental group and control group were prepared by the same specimen size, casting procedure and cured under the same condition. The only difference between these two groups is whether the group incorporated microcapsules or not. The slabs with a thickness of 40 mm were fixed on the stirrups prior to the concrete casting and acted as a concrete cover layer of the structure. In order to strengthen the binding interface between the fresh and old concrete, surface treatments of the slabs were performed. The strain gauges embedded in the precast concrete slabs were measured at specified times (0 d, 10 d, 3 months, etc.) to determine the effect of the evolution of the concrete structures on the strain values. After removing the concrete formwork, the precast concrete slabs assembled in the structures were continuously monitored for 4 months, including strain monitoring and experimental observations.

2.4. Evaluation of the self-healing efficiency

In this research, the healing efficiency of the self-healing system was evaluated by testing the specimens (Type-I and Type-II). $\eta_{STR}$and η_{RCM} were calculated based on the experimental results of the compressive strength test and the RCM test, respectively. The healing efficiency in terms of the mechanical property was determined as:

$$\eta_{STR} = \frac{f_{healed} - f_{initial}}{f_{initial}} \times 100\% \quad (3)$$

where $\eta_{STR}$is defined as the healing ratio of the self-healing concrete in terms of the mechanical property. $f_{initial}$is the compressive strength of the unrepaired specimen after pre-loading (conducted the compressive strength test immediately) and f_{healed} is the compressive strength of the healed specimen (healed at a temperature of 60 °C for 3 d, 5 d, 7 d, 14 d, and 28 d).

The healing efficiency in terms of the impermeability was calculated as follows,

$$\eta_{RCM} = (-1) \times \frac{D_{healed} - D_{initial}}{D_{initial}} \times 100\% \quad (4)$$

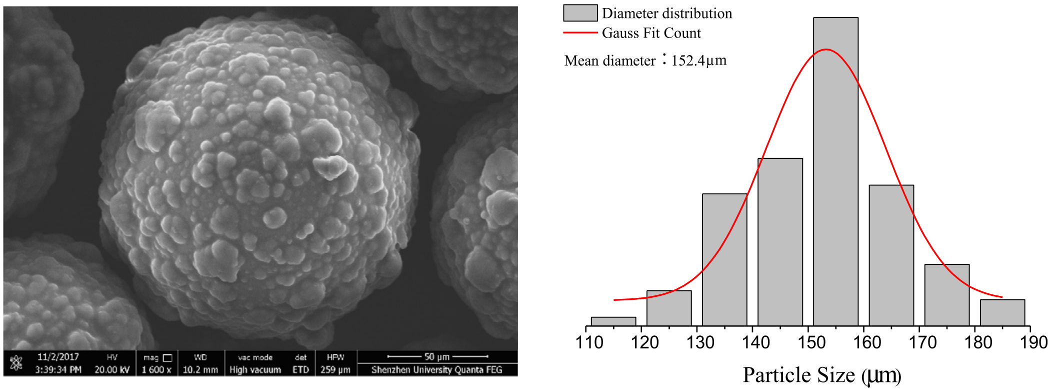
where η_{RCM} is defined as the healing ratio of the self-healing concrete in terms of the impermeability. $D_{initial}$is the chloride diffusion coefficient of the specimen after pre-loading, D_{healed} is the chloride diffusion coefficient of the healed specimen.

3. Results and discussion

3.1. Physical properties of the microcapsules

Since the physical properties of the microcapsules synthesized by in-situ polymerization may significantly affect the suitability of the self-healing system [30], the morphology, size distribution, and thermal stability properties of the microcapsules were investigated as mentioned in Section 2.3. The surface morphology of microcapsules is shown in Fig. 4(a). It was clearly observed that the particles of the urea-formaldehyde resin were uniformly deposited on the shell rather than condensed into a block and the microcapsules exhibited a relatively regular round shape. During the preparation process, the morphology and surface characteristics of the microcapsules can be adjusted by changing the surfactant concentration, the pH of the solution, and the heating rate. However, it should be noted that the process parameters directly affect the roughness of the microcapsule shells, which greatly improves the debonding strength of the interface between the microcapsule and the cementitious matrix [13]. Fig. 4(b) shows the particle size distribution and the mean diameter of the microcapsules. The results indicate that the microcapsule particles conformed to a normal distribution with a mean diameter of 152.4 μm.

Regarding the thermal stability properties of the microcapsules, a clear absorption peak was observed at around 250 °C and the samples showed distinct weight loss events from 250 °C to 600 °C (Fig. 5). The TGA/DTA curves show that the degree of weight loss was quite small and steady until 250 °C, after which the shell of the urea-formaldehyde resin was thermally decomposed and the integrity of the surface of the microcapsules was compromised. The weight loss rate of the microcapsules was low below 250 °C and this was attributed to the evaporation of the moisture trapped



(a) SEM morphology of the dried microcapsules (b) Particle size distribution of the samples

Fig. 4. Urea-formaldehyde/epoxy resin microcapsules used in this study.

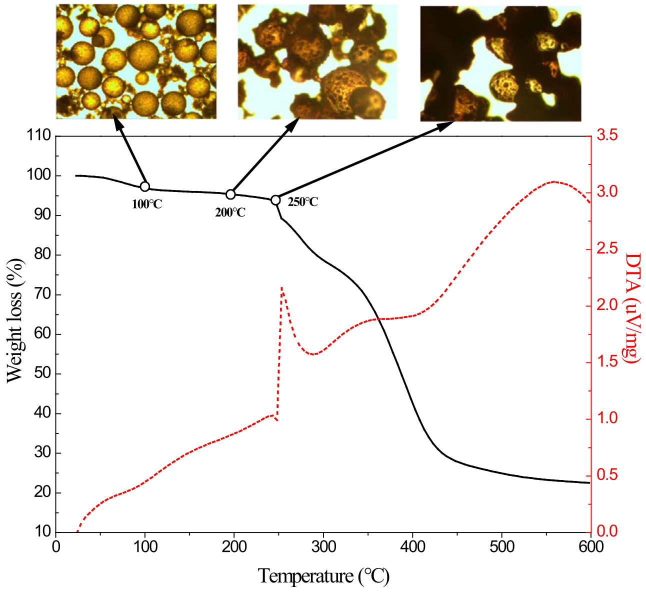


Fig. 5. TGA/DTA curves of the microcapsules.

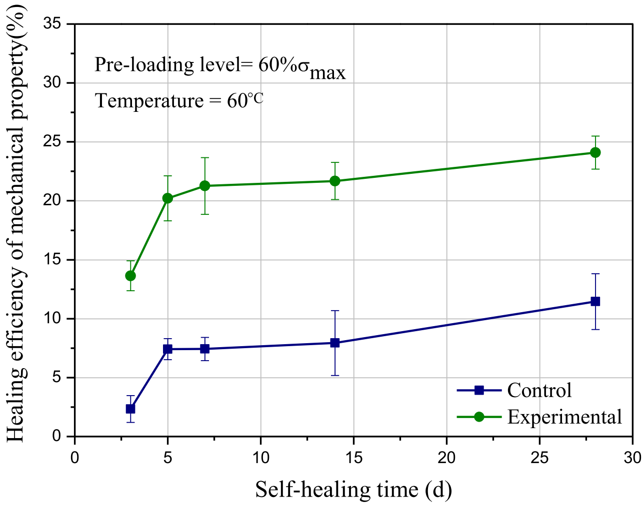


Fig. 6. The self-healing efficiency in terms of the mechanical property of the experimental and control groups over time.

in the particles. The weight loss rate of the samples was about 10% at 250 °C and the shell of the microcapsules maintained the structural integrity and prevented the release of the healing agents until decomposition; this result indicates that the urea-formaldehyde/epoxy resin microcapsules used in this study have good thermal stability. The experimental results confirm that the microcapsules have good physical properties that meet the requirements of the working conditions of the self-healing system.

3.2. Concrete cube compressive strength

The mechanical properties of the experimental and control groups were investigated to determine the healing effect of the microcapsules. In this case, the compressive strength of the Type-I specimens subjected to the preload of 60% σ_{max} at various self-healing times (3 d, 5 d, 7 d, 14 d, 28 d) was tested and the self-healing efficiency was calculated using Eq. (3). The results are shown in Fig. 6. A significant difference was found in the healing efficiency between the experimental and control groups. The healing efficiency of the compressive strength increased markedly by 11% after 3-d healing when the concreted contained 10% microcapsules. The maximum increase of around 14% was observed after 7-d healing. The healing efficiency gradually increased over time for the specimens with and without the microcapsules. In addition, it was noted that the rate of increase in the self-healing efficiency was rapid early on and decreased gradually for the longer aging time. This behavior of the self-healing concrete was similar to that reported for cement mortar [12].

We noted that the addition of 10% microcapsules significantly improved the self-healing efficiency of the concrete but had a negative effect on the compressive strength of the specimens. In the initial stage, the compressive strength of the experimental group was approximately 70% of the control group (see Fig. 7). Although the ratio of the compressive strength between the experimental and control groups showed an increasing trend, the maximum ratio was only about 82%. Furthermore, the ratio was maintained at a relatively constant level (80–82%) after healing, which indicated that the embedded microcapsules were broken and the healing agent inside the microcapsule was released to achieve the healing function.

These results let us conclude that the addition of 10% of urea-formaldehyde/epoxy resin microcapsules had dual effects on the mechanical performance of the self-healing concrete. First, the addition of the microcapsules markedly increased the self-healing efficiency in terms of the mechanical strength. However, it had a negative effect on the compressive strength of the

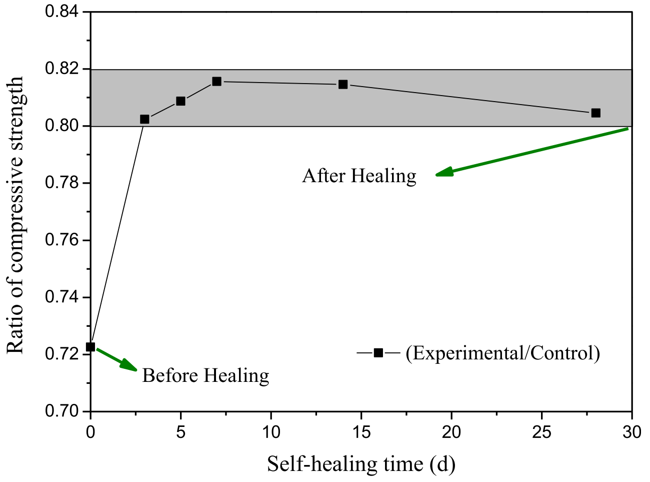


Fig. 7. Changes in the compressive strength ratio with increasing self-healing time.

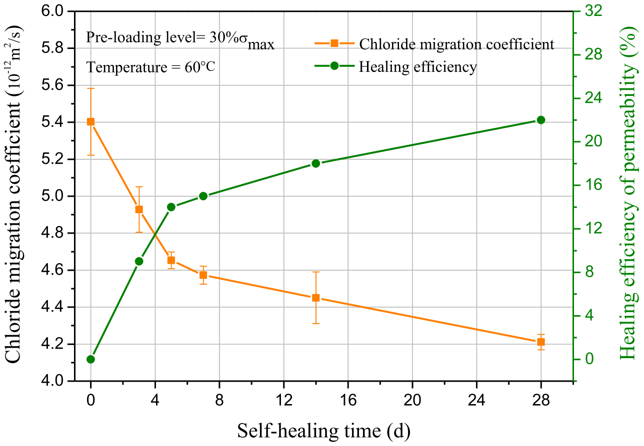


Fig. 8. Development of the chloride migration coefficient and healing efficiency of the specimens with increasing self-healing time.

experimental group. Generally speaking, by adjusting the composition, age, curing condition, and other factors, significant changes in the mechanical strength of the concrete occur because of the influence of the microstructure and the hydration degree [8,34]. The microstructure of the concrete in most cases has a dominant effect on the mechanical strength [35–37]. In this study, all the specimens were created using the same water to cement ratio and the same materials except for the experimental group, which contained the microcapsules and curing agents. The 60-d curing age prior to the test greatly reduced the impact of the secondary hydration on the cementitious materials for both the experimental and control groups. Therefore, the 20–30% decrease in the compressive strength was attributed to the presence of the microcapsules, which changed the microstructure of the self-healing concrete. However, it should be noted that, for many practical applications, this decrease in strength is acceptable and some functional materials such as fiber can be used to increase the mechanical properties in practical situations.

Contrary to the decrease in the compressive strength, a significant increase in the healing efficiency in terms of the mechanical strength was observed in the experimental group. This phenomenon can be explained by the healing effect of the microcapsule-based self-healing system. When cracking triggered the breakage of the microcapsules, the healing agents were released from the broken microcapsules and reacted with the curing agents embedded in the crack zone, resulting in the increased self-healing efficiency of the concrete. To assess the comprehensive effect of the microcapsules on the cementitious materials, some other factors concerning the self-healing functionality should be taken into consideration, in particular, the permeability properties, which will be discussed in Section 3.3.

3.3. Chloride migration coefficient

It is generally believed that a permeability assessment is an important part of the durability assessment of concrete structures. The chloride migration coefficient obtained from the RCM test is the most commonly used method to investigate the permeability of cementitious materials [38,39]. In this study, the chloride migration coefficient and the self-healing efficiency in terms of the permeability of the self-healing concrete was calculated using Eqs. (1) and (4), respectively. As shown in Fig. 8, the chloride migration coefficient decreased significantly with increasing self-healing time, which indicated that the ability of the chloride ions to permeate the self-healing concrete decreased over time as a result of the microcapsules and the resistance of the specimens to the chloride ion penetration gradually improved over time. It was also found that the rate of decrease in the chloride diffusion coefficient was fast in the beginning, i.e., the coefficient decreased from 5.4 to 4.6 (10^{-12} m^2/s) during the first 5 days of healing, and then the rate of decrease slowed. However, the self-healing efficiency in terms of permeability exhibited the opposite trend of the chloride migration coefficient. It was clearly observed that the self-healing efficiency in terms of permeability gradually increased over time. The healing ratio of the self-healing concrete at the age of 28 d was about 22% higher than that without healing. Therefore, the addition of 10% microcapsules has obvious effects on the chloride ion mobility and decreased the chloride migration coefficient resulting in an increased self-healing efficiency in terms of permeability.

As mentioned in the preceding sections, in order to achieve self-healing, damages were caused to the concrete by applying a compressive load prior to healing. With the introduction of damage, the pore structure of the self-healing concrete was changed and there was also an increase in the number of internal defects [11]. Therefore, when the microcracks appeared and propagated, the embedded microcapsules around the crack surfaces would break under the crack-tip stress and then the healing agents was released from the broken microcapsule to achieve healing. The increasing amount of healing agent made the pathways for the chloride ions more tortuous or blocked them partially. This is one reason why the addition of the microcapsules decreased the chloride migration coefficient. In addition, the microcapsules modified the microstructure and the healing products increased the density of the cementitious paste (will be discussed further in Section 3.5), which might have contributed to the decrease in permeability. Consequently, according to the RCM test results, it was demonstrated that the microcapsule-based self-healing system had a significant healing effect on the permeability performance of the self-healing concrete.

3.4. Long-term shrinkage

The long-term shrinkage of the Type-III specimens was determined based on changes in the volume and length using the contact shrinkage measuring method. The time-dependent curves of the shrinkage are shown in Fig. 9. It was clearly observed that the shrinkage gradually increased over time and exhibited a similar trend for the experimental and control groups. However, the difference between the shrinkage strain of the two groups

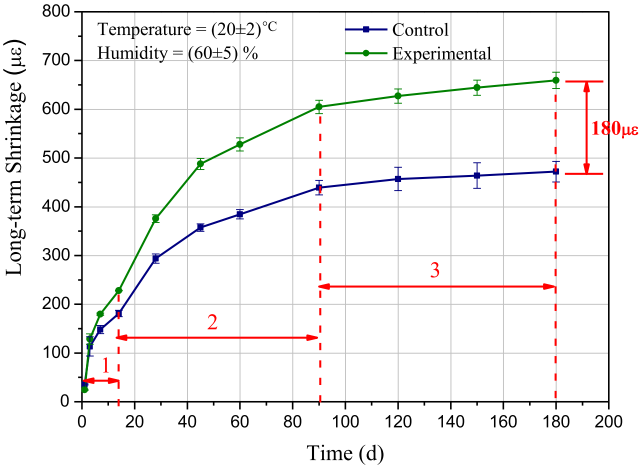


Fig. 9. Long-term shrinkage of the concrete in the experimental and control groups over time.

increased with increasing curing age. The largest difference in the shrinkage strain of 28% between the experimental group and the control group was observed after 180 d. It was also noticed that the rate of change of the shrinkage was very high during early aging and then gradually flattened with increased aging. The relationship between the shrinkage and the curing age can be roughly divided into three stages, namely the acceleration period (stage 1, 0–14 d), the transition period (stage 2, 14 d–90 d), and the stationary period (stage 3, 90 d-180 d). In fact, this shrinkage behavior is found in most cementitious materials [40,41].

The experimental results demonstrated that the 10% microcapsule addition had a significant influence on the long-term shrinkage of the self-healing concrete, especially during the later aging period. In the acceleration period (stage 1), only a slight difference in the shrinkage between the experimental and control groups occurred from 1 to 14 d. The hydration degree of the two groups was similar and the microcapsules had a rather negligible impact on the shrinkage because the autogenous shrinkage accounted for most of the volume change during early aging [42]. The experimental and control groups exhibited a rapidly increasing shrinkage rate, which was mainly due to the increased cement hydration. In the transition period (stage 2), the drying shrinkage was dominant. The low stiffness of the microcapsules largely reduced the constraints on the concrete's shrinkage and resulted in a large volume deformation. Thus, the difference in the shrinkage between the two groups gradually increased because the microcapsules had a lower strength than the cementitious materials [20]. In the stationary period (stage 3), both groups exhibited a low shrinkage rate, which indicated that most of the shrinkage had taken place in the concrete and only little shrinkage was likely to occur in the future. Although the long-term shrinkage test was conducted in an air-conditioned room with no constraints, unlike in practical situations, a significant difference (180 με) was observed at 180 d. Furthermore, in practical engineering applications, functional materials such as a shrinkage-reducing admixture (SRA) can be used to decrease the shrinkage of concrete.

3.5. Microstructure of the specimens

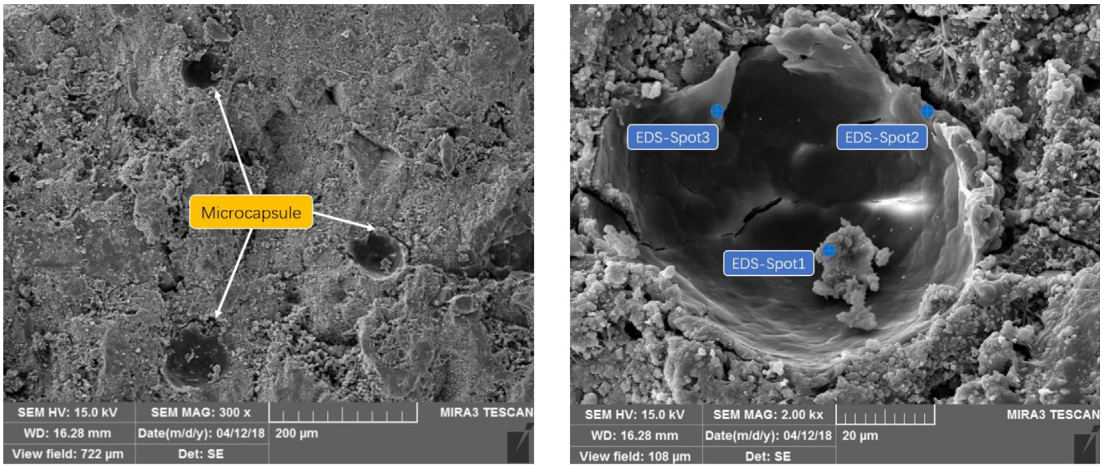
In the field of concrete science, it is generally believed that the microstructure of the concrete has a strong correlation with its mechanical properties and durability. Therefore, a microstructural investigation was conducted on the samples obtained from the Type-I specimens with 10% microcapsule addition to characterize the self-healing concrete. The SEM images of the fracture surface and the EDS analysis results are shown in Fig. 10. The two main purposes of the SEM images are to serve as instruments: (1) to demonstrate the breakage of the microcapsules upon cracking, and (2) to provide a proof-of-concept for self-healing concrete. It was clearly observed that the embedded microcapsules were broken and the shells still remained in the cementitious matrix, indicating good bond strength between the microcapsules and the matrix (Fig. 10(a)). In terms of the compressive strength of the self-healing concrete, the microstructure of the specimens significantly affected the strength because the microcapsules decreased the matrix density. As shown in Fig. 4(b), the mean diameter of the microcapsules used in this study is 152.4 μm. In terms of the concrete strength, the microcapsules can be regarded as harmful pores based on the classification of the pore size [43] because they cannot withstand the compression load during the compressive strength test. Therefore, based on the results of the compressive strength test and the SEM images, it can be concluded that the addition of the microcapsules had a negative effect on the compressive strength of the specimens not only because the microcapsules resulted in a much lower strength than that of the cementitious material but also because the microcapsules took up some pore space and resulted in larger pores in the cementitious matrix.

As shown in Fig. 10(c), high amounts of nitrogen, oxygen, and carbon were observed based on the element identification of the "EDS-Spot" shown in Fig. 10(b). The elements detected by the EDS are rare in cementitious materials and are mainly derived from the organic microcapsules and healing agents. The results indicated that the self-healing behavior can be identified using element detection. From the perspective of the microstructure of microcapsule-based self-healing concrete, the breakage of the microcapsules upon cracking was already confirmed by the SEM images showing the shell of the broken microcapsules was tightly embedded in the matrix (see Fig. 10(b)). Hence, due to a large number of broken microcapsules, the reaction of the released healing agents and curing agents made it possible for the self-healing system to fill the defects and heal the matrix microcracks, which decreased the permeability of the concrete. Furthermore, as shown in Fig. 10(d), it was observed that the self-healing products partly filled the microcracks. The EDS analysis results of the fracture surface demonstrated the feasibility of using microcapsules as carriers for healing agents to heal microcracks in a cementitious matrix. Thus, on the basis of the RCM test and the SEM/EDS analysis results, it can be deduced that the enhancement in impermeability was most likely due to the improvement in the microstructure, which was attributed to the excellent self-healing functionality of the microcapsules.

In summary, the microstructure of the self-healing concrete was modified mainly due to the presence of the microcapsules. The addition of the microcapsules had both positive and negative effects on the microstructure of the specimens. On one hand, the embedded microcapsules reduced the density and homogeneity of the concrete, resulting in a decrease in the compressive strength; on the other hand, the healing agent released from the broken microcapsules blocked the pores and healed the microcracks by reacting with the curing agents and then achieved healing, which decreased the permeability and increased the self-healing efficiency of the concrete in terms of compressive strength as well as permeability.

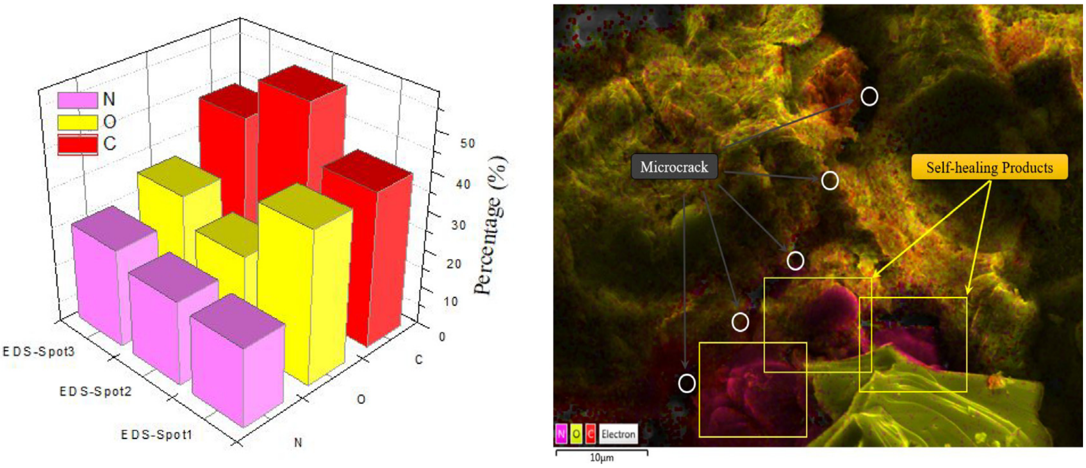
3.6. Field test results: strain monitoring and experimental observations

For concrete structures, a field investigation is essential for providing researchers and engineers with adequate useful information, which is required for monitoring and evaluating the structural performance, particularly in applied research. As an



(a) Microcapsules embedded in cementitious matrix

(b) Broken microcapsule



(c) EDS analysis of a broken microcapsule

(d) EDS analysis of the fracture surface of a specimen

Fig. 10. The microstructural characteristics of the microcapsule-based self-healing concrete.

important part of this study, strain monitoring and experimental observations were carried out with the objective of investigating the performance of the structural members (precast concrete slabs) constructed with microcapsule-based self-healing concrete in tunnel engineering. The strain monitoring was conducted by means of strain gauges, which detected the local structural property variations, whereas the experimental observations included crack tracking and surface observation to verify the suitability of the structural member. Placing the structural members is a key step in an effective field test. The placement of the precast concrete slabs is shown in Fig. 11.

Due to the rapid development of sensor technology, the use of strain measurements is effective for monitoring the performance of a structure and this method is commonly used as an important testing element in structural health monitoring in civil engineering [44]. The strain gauges in this study were placed in two directions perpendicular to one another (see Fig. 3). The results of the strain measurement over time are shown in Fig. 12. Except for the strain gauges that were destroyed during construction, it can be seen that both the horizontal and vertical strains exhibit relatively low shrinkage strain and the values were within the range of the strain gauge (±1500 $\mu\varepsilon$). No significant changes were observed in all strain measurement at regular time intervals, indicating that the local structural property variations were in accordance with the normal state. In addition, a comparison of the results of the experimental and control groups indicated similar development trends in the shrinkage strain in all structural members except for the ones showing very low horizontal strain (Fig. 12(a)). In general, the boundary conditions have a significant influence on the performance of the concrete structure, especially for structures exposed to complex boundary conditions [45]. In this study, the precast concrete slabs were embedded in the concrete structure as a structural member and the boundary of the slabs was restrained instead of having a free end. Thus, this may be one reason for the small difference between the horizontal and vertical strain of the structural members.

After demolding, the precast concrete slabs embedded in the concrete structure were continuously tracked for 4 months prior to coating. The results of the experimental observations are shown in Fig. 13. As described in Section 2.3, the control group (numbered P1, P2, P3) contained no microcapsules, while 10% microcapsules were added in the experimental group (numbered P4, P5, P6). In laboratory test, it has been demonstrated that the presence of microcapsules indeed has significant effect on the performance of self-healing concrete. However, from the experimental observations, it can be found that microcapsules have little effect on the surface characteristics of concrete. Except for the cement slurry covering the slab surfaces, no obvious changes were observed

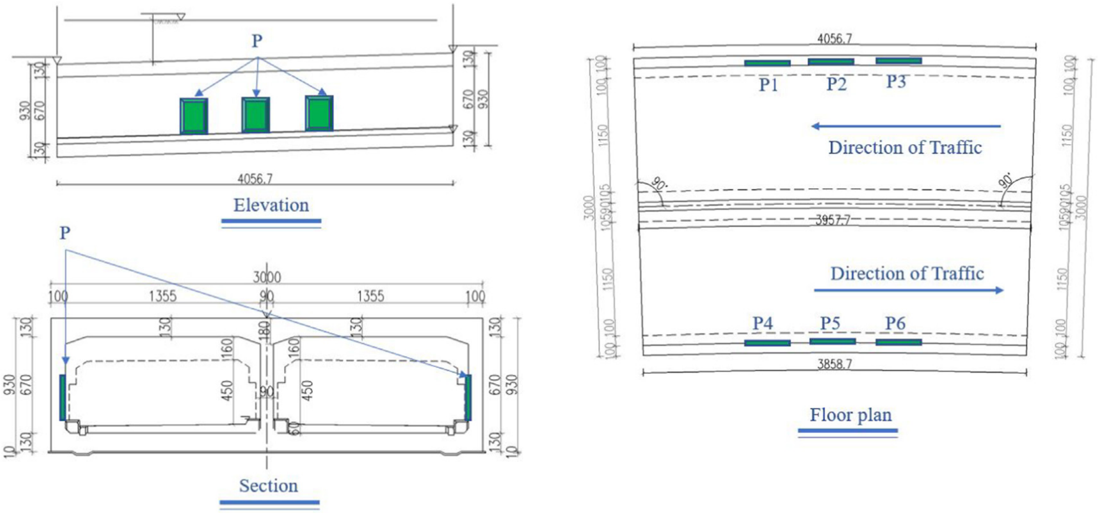
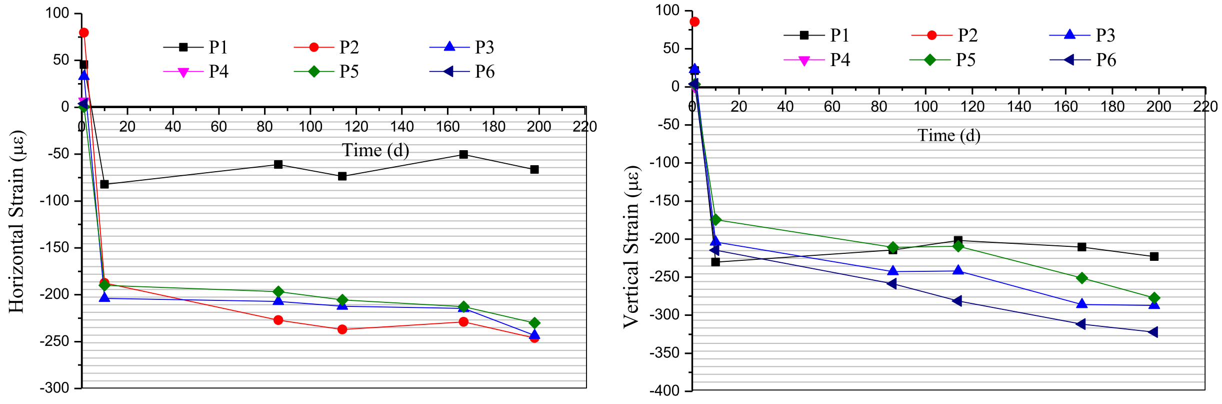


Fig. 11. Schematic drawing of the precast concrete slabs placed in the tunnel.



(a) Horizontal strain

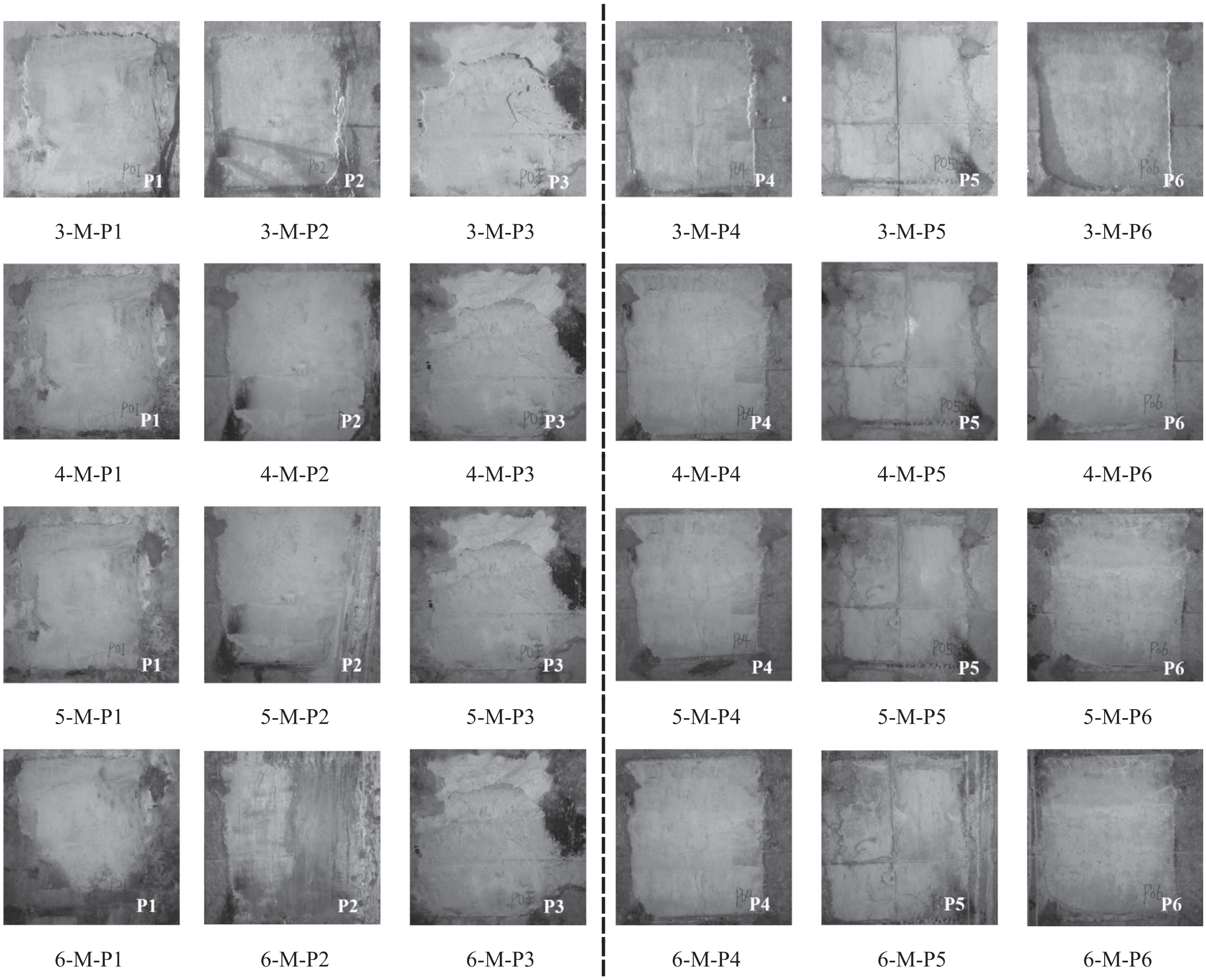
(b) Vertical strain

Fig. 12. Relationship between the internal strain of the concrete in the experimental and control groups over time.

between the experimental group and the control group shown in the horizontal direction in Fig. 13. During the first month (3-M) of experimental observation, no visible cracks were detected in the two groups. In terms of the appearance of the slab surfaces, almost all the precast concrete slabs had similar surface voids and color and those properties remained the same over the 4 months. A comparison of the progression over time (vertical view) indicated also no significant difference in the cracks and the surfaces between the two groups throughout the continuous monitoring of the precast concrete slabs for 4 months. In addition, the experimental observations are an important means of non-destructive testing; in this case, the observation results were in agreement with the results of the morphology analysis of the concrete structure. The results of the experimental observations were also in agreement with the strain measurement results, indicating that the precast concrete slabs had good compatibility with and applicability to the concrete structure.

The field test results let us conclude that the internal strain of the precast concrete slabs exhibited no apparent fluctuations and there were also no significant changes in the experimental observation for the experimental and control groups. Thus, the precast concrete slab embedded structure had no significant negative effects on the performance of the concrete structure. However, as demonstrated in the laboratory test, the experimental groups with a 10% microcapsule content exhibited a remarkable improvement in the mechanical self-healing efficiency, and the impermeability over the control groups. Taking into account the laboratory and field tests, it can be concluded that the experimental results demonstrated the feasibility of using a microcapsule-based self-healing system to improve the durability of concrete in tunnel engineering. Furthermore, due to the importance of the permeability in underground tunnels, especially in a coastal environment, the concept of using self-healing concrete as a construction material in tunnel engineering is very promising. On the other hand, it should be noted that external loads on structures are constantly changing in practical engineering. It is virtually impossible for the load level to keep constant, especially for a concrete structure under dynamic loading. A wide variety of loads may lead to the complexity of stress fields generated by cracks. These conditions expose the limitations of the microcapsule based self-healing concrete, such as, supplement of the healing agent, healing cracks under the mutual coupling of the loads. These are the major challenges of the microcapsule based self-healing system for practical application, and all these aspects deserve further study.

Note that, microcapsules used in the research of self-healing concrete are mainly produced by laboratory-synthesis (such as physical, chemical or physicochemical methods) at present, rather than commercially available. A large fraction of microcapsule's production cost comes from raw materials, which include bisphenol A type epoxy resin (E-51), N-butyl glycidyl ether (BGE), urea, formaldehyde, triethanolamine, sodium dodecylbenzene sulfonate (SDBS) and sulfuric acid. According to the current market price of



3-M-P1 3-M-P2 3-M-P3 3-M-P4 3-M-P5 3-M-P6

4-M-P1 4-M-P2 4-M-P3 4-M-P4 4-M-P5 4-M-P6

5-M-P1 5-M-P2 5-M-P3 5-M-P4 5-M-P5 5-M-P6

6-M-P1 6-M-P2 6-M-P3 6-M-P4 6-M-P5 6-M-P6

Fig. 13. Experimental observations of the precast concrete slabs embedded in the structure. (M: month).

the raw materials in 2019, the cost of preparing microcapsules is about 100 CNY/kg. The cost comparison analysis of self-healing concrete and manual repair has not been accurately evaluated, and we are convinced that this is another major topic which is important and deserves further research. However, it is expected that the developments of self-healing concrete will achieve a major breakthrough driven by societal needs (tunnels, bridges and large infrastructural works, etc.), including improving the self-healing efficiency, optimizing the preparation process and reducing the production cost. On the basis of these developments, the large-scale commercialization of the microcapsule based self-healing concrete in a particular application is feasible and promising.

4. Conclusions

In this paper, we presented an experimental study on the performance and application of microcapsule-based self-healing concrete in practical engineering. Based on the experimental results from laboratory and field tests, the following conclusions can be drawn:

(1) The urea-formaldehyde/epoxy resin microcapsules synthesized by in-situ polymerization exhibited good physical properties that were in agreement with the working condition of a self-healing system.

(2) The experimental results show that microcapsules had dual effects on the mechanical properties of self-healing concrete. The incorporation of 10% microcapsules by weight increased the self-healing efficiency of the concrete in terms of mechanical strength by 14% after 7-d healing and decreased the compressive strength before and after healing by 30% and 20%, respectively in comparison to the control group. On the other hand, significant improvement was observed in the impermeability of self-healing concrete. The addition of microcapsules resulted in a decrease in the chloride migration coefficient under an externally applied electric field and as a result, the self-healing efficiency of the concrete in terms of permeability increased.

(3) Microcapsules had a relatively strong impact on the shrinkage of the self-healing concrete in the stationary period, whereas no difference was observed between the two groups in the acceleration period. Also, the microstructure of the self-healing concrete was modified mainly due to the presence of the microcapsules, which had both positive and negative effects on the microstructure of the specimens. Some supplements may be added to the self-healing concrete to compensate for the negative effects of the microcapsules.

(4) The laboratory and field test results demonstrated that the concept of the microcapsule-based self-healing system is feasible and promising and microcapsules have great

potential for use in self-healing concrete for sustainable infrastructure, especially in coastal civil engineering. However, the microcapsule-based self-healing concrete technology is a double-edged sword. In order to achieve an excellent self-healing effect and avoid the negative effects of the microcapsules, there is a need to balance the amount and particle size of the microcapsules in future practical applications.

Declaration of Competing Interest

None.

Acknowledgments

The authors gratefully acknowledge the support by the National Natural Science Foundation of China (51478272), the joint funds of the National Natural Science Foundation and Guangdong Province of China (U1301241), the International Cooperation and Exchange of the National Natural Science Foundation of China (51520105012), the Science and Technology Foundation for the Basic Research Plan of Shenzhen City (JCYJ20160422095146121), and the Collaborative Innovation Research Centre for Environment-Friendly Materials and Structures in Civil Engineering, Southeast University.

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