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Water-tightness performance of crack-self healing concrete incorporating various types of granule of supplementary cementitious materials and/or Portland cement and other additives

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ABSTRACT

In this study, crack-self healing properties of concrete incorporating various types of granules of supplementary cementitious materials or Portland cement and other additives were investigated. Granules were added to concrete mix by partial sand replacement. A constant water flow through a crack of 0.2-0.4mm wide was conducted for 56 days. The self-healing performance was assessed by monitoring the flow rate, pH value of water passing through a crack and the chemical analysis of precipitated products along the crack surface. The results showed that the inclusion of granules not only had little effect on the compressive strength but also contributes to the crack-closing process. The flow rate was significantly reduced, however the complete healing of cracks were not obtained in concrete incorporating granules of pozzolan and other admixtures. It is thought that the inner of broken granules were hardened by pozzolanic reaction that restrained self-healing additives to dissipate into the crack.

Keywords. Water-tightness, crack, self-healing, granule, supplementary cementitious materials

INTRODUCTION

It is well-known that crack appearance is an intrinsic characteristic of concrete or reinforced concrete structures. In water retaining or underground structures, crack developing and propagating cause a severe water leakage and significantly reduces the serviceability/functionality of these structures. However, based on past research (Edvardsen, 1999; Heide, 2005; Hirozo, 2012; etc) it has been observed that the healing capacity is very limited in normal concrete. It preferably occurs in concrete with low water/binder ratios or using huge amount of cement. Moreover only very small crack, typically whose width is less than 0.1mm, can be healed.

Nowadays the supplementary cementitious materials, such as blast furnace slag, fly ash, silica fume, etc…, have widely been used due to their friendly environmental materials and significant improvement of the mechanical and durability properties of concrete (Hooton, 2011). Besides, according to past research, it was found that the cementitious materials incorporating such pozzolanic substances also contributed to the crack-healing ability (Pipat, 2009; Dechkhachorn).

Recently, one of among several engineered approaches to introduce the healing ability to concrete has been developed by Koide (2011), in which self-healing agents were added to concrete mixture just before mixing in form of granules or capsules to stimulate the chemical reactions after cracking and has a potential to preserve the healing property of concrete in long-term.

In previous research, Kishi, Ahn (2010) and other researchers introduced the self-healing property to concrete by adding some specific mineral and chemical admixtures in form of powder. Even though the self-healing performance was promising, there were some problems with this powder type approach. Typically, the inclusion of self-healing powder into concrete mixture caused a significant reduction in the workability of fresh concrete and possibly reduced the self-healing efficiency of hardened concrete, due to unavoidable further reactions between the embedded powder, water and other products during the mixing and hardening process of concrete. To overcome the above mentioned, the granules were used, instead of powder, by partial sand replacement in this study. Granules were designed to contain self-healing agents inside, coated by an external layer and only activated whenever crack occurs and contacting with water. Moreover, another target is also to successfully produce a crack self-healing concrete at normally used water/cement ratios and has a potential to heal a larger crack width when exposed to water flow in short time.

From the above mentioned, the aim of this research is to investigate the crack healing capability of concrete incorporating granules of pozzolanic materials and/or Portland cement and some specific admixtures and its feasibility in practice. The term "crack self-healing concrete", in this study, is defined as the concrete has an ability to recover the water tightness property after cracking. It is believed that if concrete were designed with crack-self healing capacity, it may contribute to extending the service life and reduction of maintenance and repair for water retaining structures.

EXPERIMENTAL PROGRAM

Hypotheses for crack self-healing phenomenon in concrete incorporating self-healing granules subject to water flow

Self-healing granules, which were fabricated in the laboratory in advance, are added to concrete mix just before casting to minimize the broken possibility of granules during mixing. Once crack penetrates, granules are ruptured. While water flows through a crack, not only the self-healing materials are diffused into crack surface but also the cement hydration products are dissolved in flowing water. Over time, the crack in concrete is healed and the water leakage is stopped mainly due to the formation of new products by the chemical reaction between self-healing materials with external water and the recrystallization of dissolved ions. It is thought that the pozzolanic or hydraulic reactions together with calcite formation reaction are major contributors to the crack self-healing capacity of concrete.

The proposed granules and the granulation process

In this research the granulation technique, introducing the self-healing ability to concrete, was developed by Koide (2011), based on the capsulation technique used in food/medicine industry with special considerations of the cost and required equipment for the process. In case of capsule, the inner material is commonly protected by an organic layer while the inner agent of proposed granule is coated by cement compounds, containing self-healing materials. By this approach, the coating layer may be reacted with water or other products during granulating, mixing and hardening process, however the inner material is expected to be unreacted and only activated when crack penetrates and contacts to water flow.

Granules are manufactured through the granulation process by using a typical mixer in laboratory (i.e. roller mixer or mortar mixer). It changes the initial condition of self-healing powder to granule type of several millimetres. The principle process is as follows: firstly the inner granule manufacture is done, and then a coating layer built-up is followed.

Based on the proposed healing hypothesis, the basic design concept of self-healing material proposed by Ahn (2010) and to investigate the self-healing performance and the feasibility of this approach, various types of granules are designed and fabricated with different ingredients and granulation methods. Typically, the granules used in this study are classified into three groups and the list of granules used in this study can be seen in Table 1.

Table 1. Three groups and list of self-healing granules

Group	Main ingredients	Granule
1	Portland cement and self-healing admixtures	1
2	Pozzolans and self-healing admixtures	2a/2b/2c/2d
3	Portland cement and pozzolans and self-healing admixtures	3

Fabrication and Mix proportions of concretes

After manufacturing in the laboratory, the granules are stored in plastic bags and transported to the ready-mixed concrete plant to make the concrete. There are two mix proportions used in this trial with the attention of the normally used ranges of water/cement ratio and the required workability of fresh concrete (especially the slump). And, self-healing granules are added to concrete mixture just before casting as partial replacement of sand.

Table 2. Mix proportion of design 1 concrete (Mix1)

Type of concrete	W/C [%]	s/a [%]	G_{max} [mm]	Air [%]	Kg/m^3				
					W	C	SH	S	G
Plain	49.6	51.3	20	4.5	175	353	-	900	869
Self-healing concrete (SH-Mix1)	49.6	51.3	20	4.5	175	353	70	830	869

Table 3. Mix proportion of design 2 concrete (Mix2)

Type of concrete	W/C [%]	s/a [%]	G_{max} [mm]	Air [%]	Kg/m^3				
					W	C	SH	S	G
Plain (Plain-Mix2)	49	44.5	20	4.5	171	349	-	802	953
Self-healing concrete (SH-Mix2)	49	41.2	20	4.5	171	349	100	702	953

Techniques to verify the watertight performance and self-healing products

In this study, the water pass test, described by Morita (2011), was used to quantitatively evaluate the self-healing performance as water leakage control.

Before testing, a penetrating crack would be induced to concrete specimen by the tensile splitting test. Initially, the internal crack was controlled to around 0.2mm by the thickness of Teflon sheet, while the surface crack width at the top and bottom of specimen was measured by Microscope at three different spots (Figure 1a). The average value of those spots was adopted as the surface crack width, ranging from 0.2-0.4mm.

After preparation, the specimen was exposed to continuous water flow by attaching a pipe and supplying the water. Then the water pass test, in which a constant water flows through a crack under pressure of approximately 9cm water head, would be performed (Figure 1b and 1c). The expected results were the water leakage, the pH value of water passing through a crack over time.

Furthermore, the optical microscopy and digital image were conducted to assess the degree of self-healing at crack and the X-ray Fluorescence (XRF) and Thermogravimetry-Differential Thermal Analysis (TG-DTA) were also performed to analyse the chemical and mineral components of healing product precipitated at the crack.

(a) (b) (c)

Figure 1. (a) Surface crack width- (b) Exposure condition- (c) Water pass test

EXPERIMENTAL RESULTS AND DISCUSSIONS

Effect of the inclusion of self-healing granules on the workability of fresh concrete

Table 4 showed the workability of fresh concrete just after discharging from the mixer at the ready-mixed concrete plant. It can be seen that the dosage of the super-plasticizer was in the range of 0.6-1.3% (by weight of cement) to achieve the designed slump in design 2 concrete (Mix2). Generally, the input of self-healing granules into concrete mix caused a slightly increased quantity of super-plasticizer compared to that in plain concrete. The possible reason is the water absorption and further reaction between the coating layer of granule and the mixing water. Based on the obtained results, the proposed granule type can be considered as one of the effective approaches to overcome the drawbacks of the reduction of workability in concrete incorporating self-healing materials in form of powder as mentioned above.

Table 4. Workability (slump) of fresh concretes

Concrete	Granule	Designed slump (cm)	Measured slump (cm)	SP (C x %)	AE (C x %)
SH-Mix1-1	1	20.0±1.0	20.0	1.7	
Plain-Mix2	-	12.0±2.5	20.5	0.4	0.03
SH-Mix2-2a	2a		12.5	0.6	0.02
SH-Mix2-2b	2b		11.5	1.3	0.04
SH-Mix2-2c	2c		16.5	0.6	0.02
SH-Mix2-2d	2d		17.0	0.6	0.02
SH-Mix2-3	3		14.5	0.6	0.02

Effect of self-healing granules on the compressive strength of hardened concrete

The compressive strengths of concretes incorporating several types of self-healing granules were measured at the age of 7, 28 and 91 days old after casting in order to investigate the influence of granules on the hardened concrete. From the Figure 2, it can be seen that all self-healing concretes showed a satisfaction of the required strengths for both types of design concretes (dot line for Design 1 concrete, dash line for Design 2 concrete). Moreover, it is found that depending on the ingredients of granules and their usage in granulation process, the differences in strength compared to plain concrete will vary 2-13% (at 28 days old) and 3-19% (at 91 days old). Thus, it may be concluded that up to the level of substitution of 70-100kg/1m^3 concrete, the addition of self-healing granules to concrete mix as partial sand replacement had no detrimental effect on the mechanical property, typically the compressive strength, of matured concrete.

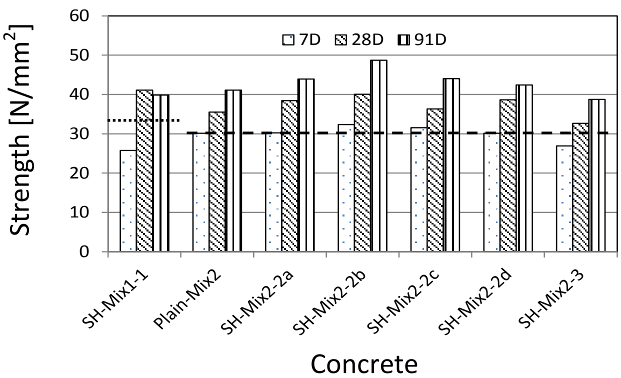

Figure 2. Compressive strengths of concretes

Change of water flow rate with time

Water permeability is of paramount importance on the durability and functionality of concrete structures. In this experiment, the water permeability decreasing with time is one of expected results to achieve the target of self-healing concrete. In order to observe the time dependent permeability of a cracked concrete, the water flow rate was measured and calculated by the formula (1), on the specific days until 56 days subjected to continuous water flow.

$$FR_i = \frac{V_{5\min,i}}{t} \qquad (1)$$

in which: FR_i-water flow rate on i days [cm^3/sec]; $V_{5min,i}$-volume of water flowing through a crack in five minutes on i days [cm^3]; t-period of testing, in this test t=300sec.

Moreover, to assess quantitatively of the decreasing flow rate with time, flow relative to initial flow was introduced as in formula (2):

$$RFR_i = \frac{FR_i}{FR_0} \qquad (2)$$

in which: RFR_i- relative flow rate on i days [%]; FR_i- water flow rate on i days [cm^3/sec]; FR_0- initial water flow rate on starting day of testing [cm^3/sec].

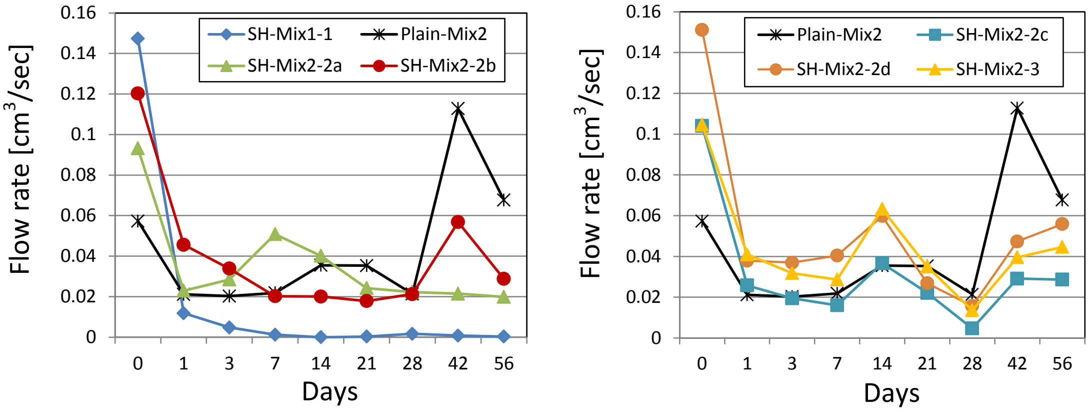

Figure 4. Change of water flow rate with time

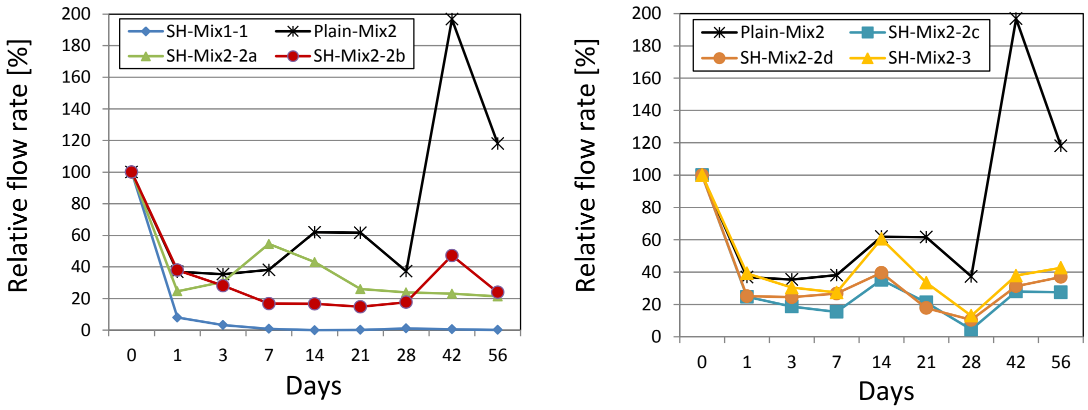

Figure 5. Change of relative flow rate with time

Changes of flow rate with time in all series were shown in Figure 4 and Figure 5 showed the relative flow rate versus duration exposed to continuous water supply for all series.

It can be seen that in case of control design 2 concrete (Mix2), even though the initial flow rate was smallest compared to that of self-healing concrete, the water leakage was maintained at the level of 40% of initial value after the first day subjected to water flow, and then it fluctuated about 40-60% until 28days. After 28 days exposure, due to discontinuous water supply, there was a dramatic increase in flow rate in this series. On the other hand, with the higher initial flow rate, the water flow through a crack after one day in other self-healing concrete series dropped more drastically, typically about 25% of initial value, except for series SH-Mix2-2b and 3 due to the larger average crack width at the beginning. Under a continuous water flow up to 28 days exposure, the self-healing effect in term of reduction of water leakage was promising at about less than 20% of initial flow rate. After that, when the exposure condition was changed, the leakage of water through a crack was slightly increased, in ranges of 20-40% of initial value, depending on the types of granules.

Series SH-Mix1-1 showed the best performance, in which the water leakage after one day was just about 8% of initial water flow and then after one week exposure, it was observed that the water flow was almost stopped during the permeability test as clearly seen in Figure 4 and 5.

Change in pH value of water flowing through a crack

Another parameter is also obtained from the water pass test to evaluate the healing process is the pH value of water flowing through the crack. It is well known that calcite formation, mainly depending on the concentration of Ca^{2+} and CO_3^{2-} ions and pH value of water, is the most important mechanism for sealing of a surface crack. When water flows through a crack, calcium hydroxide is leached and hydration products are dissolved or liberated. Therefore, both the pH value of water passing through a crack and dissolved ions will decrease with time at crack. As a result, the crack-healing process will take longer time if the crack width is small enough to be healed. Above phenomenon can be observed in case of control normal concrete. As seen from Figure 6, after 56 days exposed to water flow, the pH of water in control Mix2 concrete was almost neutral (The break line showed the lower limit value of pH of 7.5 that the calcite formation reaction can occur). It can be inferred that the calcite formation or healing process would be restrained.

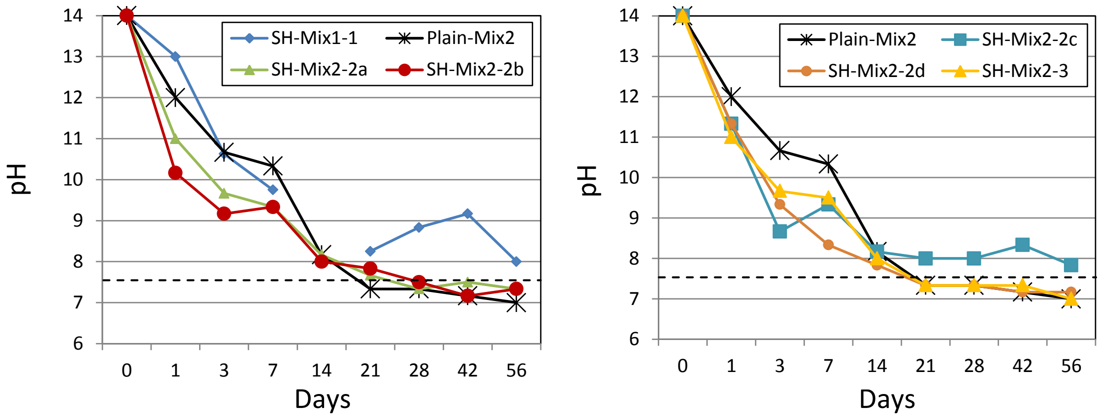

Figure 6. Change of pH value of water flowing through a crack with time

On the other hands, in case of self-healing concrete, there was also a decline in pH value of water and required ions with time, however in such slower rate due to the further supply from the diffusion process of self-healing granules. Under these conditions, the calcite precipitation was facilitated to occur in longer period and provide the crack a higher opportunity to be filled. It is clearly observed that in series of self-healing concretes which showed the best performance, the pH of flowing water was maintained high enough to stimulate the calcite precipitation up to until 56 days subjected to water flow as seen in Figure 6.

Surface crack closing process and chemical analysis of precipitated products at crack

Figure 7 showed the precipitated products on the surface crack at the bottom of concrete specimens after exposed to water flow by using a digital camera. Furthermore, the change of crack width was also observed by a microscope (100X magnification) as seen in Figure 7.

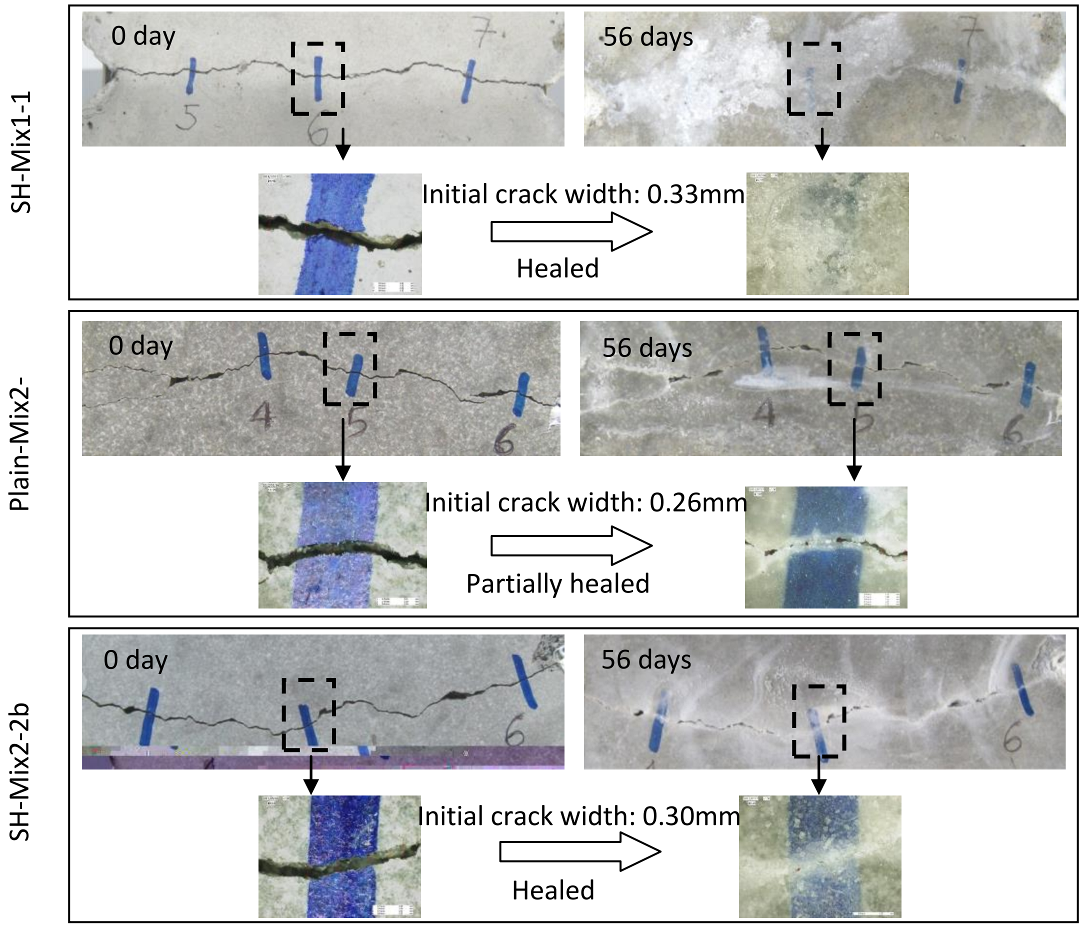

Figure 7. Crack closing process

From Figure 7, it can be seen that with a crack, larger than 0.2mm wide, a partial healing of crack was observed in Series control-Mix2. In other self-healing concrete series, most of cracks which are in ranges of 0.2-0.3mm wide can completely heal after 56 days exposure. However, when crack width is larger than 0.3mm, only partial healing is achieved, except for Series SH-Mix1-1, known as the best performance.

It was found that the decrease of crack width or the healing process was mainly observed at the bottom of specimen. It was thought that due to effect of flowing water, there was a higher concentration of both calcium and carbonate ions at this area that facilitated the precipitation of calcite.

Furthermore, based on the result of XRF test, it is found that the healing products deposited on the surface of aggregate are mainly composed of calcium compounds while calcite is confirmed as the main mineral forming at the surface crack (bottom of specimen) by analysing the product via TG-DTA test.

Discussion on the effectiveness of proposed self-healing granules

In this study, crack self-healing properties were introduced to concrete at normally used water/cement ratio by applying the granulation technology. Ideally, the inner material of granule, coated by an external layer, containing self-healing additives should be diffused spreadingly on the surface of crack when cracking. The more self-healing additives are released, the higher healing capacity is expected. It is necessary to point out that the self-healing capacity of concrete incorporating granules in this approach is mainly influenced by the quality of granulation process, such as selective self-healing additives and granule manufacture process, and the distribution or the size of granules in concrete if the same amount of granules is used. Three groups of self-healing additives are proposed in this experiment. The experimental results show that in case of granule of pozzolanic materials and other additives, the water permeability is significantly improved, especially in series including granules containing higher amount of pozzolanic materials and additional admixtures such as a supply source of calcium or calcium hydroxide. However, the water still seeps through a crack and a fully healed crack is not achieved with width larger than 0.3mm. It can be argued by the author that the following matters influencing the effectiveness of this types of granules. One of the possible reasons is the hardened condition of inner materials in/after the granulation process or the surface of broken granules when contacting to water flow. Given this, the inside self-healing additives are difficult to dissipate into the crack surface that restrain the healing capacity. Another point is the size of granules in this trial, which is thought to be too large, typically in several of millimetres. It is the fact that with the same dosage of self-healing granules used, the smaller the size of granules, the better the distribution of granules in concrete is expected and the higher the self-healing efficiency to be achieved.

Even though there were still several factors and variants affecting the result of the tests, concrete containing granules of Portland cement and specific chemical, mineral admixtures showed the best self-healing performance in this research. The water leakage was almost stopped at 7 days and the complete healing of surface crack was achieved after 28 days exposed to continuous water flow. Based on the results, it might be said that this type of granule may provide a higher possibility of crack healing capacity to normal concrete. This phenomenon can be explained by further stimulating both the hydration process of cement and calcite/other healing products precipitation at crack with the surplus supply of unreacted cement particles and other chemical, mineral admixtures from the embedded granules.

CONCLUSIONS

In this research, the normally used water/cement ratio concretes were designed with the self-healing capacity by incorporating the granules of cement replacement materials and/or Portland cement and other additives, considered as a type of engineered self-healing approach.

Even though the performance of concrete embedding the proposed self-healing granules still has not obtained the satisfactory result or definitely confirmed yet, it showed a promising approach to develop self-healing concrete in the future in terms of significant reduction of water leakage. Not only did the watertight performance of concrete significantly improve, but also the workability of fresh concrete and the compressive strength of hardened concrete were not compromised.

Concretes incorporating self-healing granules of Portland cement and specific admixtures showed the best crack-healing capacity in this study, due to the appearance of broken granules that facilitates further hydration process and calcite precipitation at crack. In case of concretes using granules of pozzolanic materials and other additives, it is observed that the water flow rate through a crack was significantly reduced during the self-healing process, however, completely healed cracks are not obtained, especially at crack with width larger than 0.3mm. It is thought that the inner part of broken granule after cracking were hardened by pozzolanic reactions during water flow through the crack or even in the granulation process. Therefore, self-healing additives were limited to dissipate into the crack that restrained the crack-closing capacity.

Furthermore, this approach showed a high potential to introduce self-healing concrete to the practical construction due to its feasibility in mass production of granules and concrete. However, further research is needed to improve the performance of self-healing concrete by selecting the ingredients of granule and developing suitable method for granulation.

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