

**Performance improvement of self-healing concrete by  
development of semi-capsulation technique for  
functional effective ingredients**

(機能的有効成分の準カプセル化技術の開発による  
自己治癒コンクリートの高性能化)

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# PERFORMANCE IMPROVEMENT OF SELF-HEALING CONCRETE BY DEVELOPMENT OF SEMI-CAPSULATION TECHNIQUE FOR FUNCTIONAL EFFECTIVE INGREDIENTS

## ABSTRACT

In practical civil infrastructures, the crack appearance and water permeation are considered as the main factors facilitating the deterioration process in concrete or reinforced concrete. Especially in case of water-retaining or underground structures, the above problems cause a significant reduction in the functionality, serviceability and also aesthetic appearance.

It has been known for long time that concrete generally has a certain capability of self-healing. According to literature review, the main mechanisms of self-healing in normal concrete at young age are the continued hydration of unreacted cement particles and the calcite formation at cracks. On the other hand, for old (mature) concrete the precipitation of calcium carbonate at crack site is thought to be the dominant process to fill the crack. However, it is necessary to point out that the healing capacity is very limited in the normal concrete and it preferably occurs in a 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. Unfortunately, in practice concretes with moderate or high water-cement ratios are commonly used and real crack width could be larger. Therefore, healing of crack in normal concrete in practice is very rare and insignificant.

Cementitious material incorporating some specific mineral and chemical admixtures, in terms of swelling, expansion and precipitation, as partial cement replacement showed a promising crack-healing effect (*Ahn, 2008*). However, there were some problems when applying these powder to concrete, such as a significant reduction in the workability of fresh concrete and in self-healing efficiency of hardened concrete. This was due to unavoidable further reactions between the embedded powder, water and other products during the mixing and hardening process of concrete.

To overcome mentioned-above obstacles, the concept of granules having

semi-capsulation effect, in which self-healing powder was stored inside by introducing a coating layer of cement compound, was proposed by Koide & Morita 2010. That approach was developed based on a conventional granulation technique used in food/medicine industry. At that moment, granules made by a conventional granulation method did not fulfill the requirements of granules having semi-capsulation effect due to immature of technique development and also the lackness of basic design concepts. Therefore, the performance of concrete incorporating those granules was not so good. Moreover, the manufacturing cost of granules was still high and a special granulator was required that further restrained the possibility to apply this technique to construction industry. However, this approach was believed to be a preferable concept to introduce self-healing properties to concrete. It has a potential for long-term preservation of healing capability as long as a crack ruptures embedded granules and exposed to water. It is necessary to develop a semi-capsulation technique for powder material.

Due to its own advantages and high potential to improve the healing performance and manufacturing cost, self-healing granules having semi-capsulation effect (proposed by Koide & Morita, 2010) were chosen as research approach in this study.

The aim of this research is to improve the self-healing performance of granules in concrete by enhancing the semi-capsulation technique and try to apply this sort of approach to practical construction industry. In order to obtain the above-mentioned target, following objectives should be done:

+ **Objective 1:** To propose the basic design concepts of granule having semi-capsulation effect for powder material.

+ **Objective 2:** To enhance the semi-capsulation technique for functional effective ingredients with considerations of simple & cheap granule fabrication.

Based on proposed requirements and design concepts of granules for powder material, several strategies to select the functional effective ingredients and to improve the granulation technique were proposed.

There was an effort to fabricate various types of granules containing supplementary cementitious materials or Portland cements and other additives with different ratios and techniques. After being manufactured and cured, granules were added to concrete mixture as a partial sand replacement (normally dosage of granules 70kg/m<sup>3</sup> concrete was used). The effects of inclusion of granules on the properties of fresh concrete (such as the workability) and hardened concrete (such as compressive strength) were examined. And the water pass test, in which a constant water head of eight centimeters flowing through a static crack of 0.2-0.4mm, was performed to investigate the healing capability of concrete. And its capability was assessed by observing the reduction of water leakage over time, the closing process of surface crack and chemical analysis of deposit products in the crack.

Based on the obtained experimental results, it was found that concrete incorporating granules of the functional ingredients has no adverse effects on the properties of concrete such as the workability of fresh concrete and the compressive strength of hardened concrete; has a possibility of recovering the water tightness property of cracked concrete, especially in young concrete; and can preserve the healing ability of concrete for a long period of time.

Even though the optimum ratio of effective ingredients of granules still have not been confirmed yet and it was still difficult to verify the self-healing performance in concrete, the requirements and basic design concepts for granules having semi-capsulation effect were established. The basic ideas are embedded granules should be broken by a penetration of crack and inner self-healing material should be released into crack surface. In order to obtain the aboved targets, both requirements of coating layer and inner material should be satisfied simultaneously: coating layer is strong and dense enough; inner material containing reactive agent is weak enough and easy to spread away out of the granule.

In order to achieve the granules having semi-capsulation effect, it was proposed that following materials should be used as self-healing ingredients: rapid hardening material should be used to induce an effective coating layer by providing the watertightness property and enough strength to coating layer; inner material should

be composed of reactive material (typically self-healing materials proposed by Ahn,2008) and a water-soluble/water-reducing agent in order to satisfy both healing effect and spreading effect.

Throughout this study, it can be concluded that it is feasible to fabricate finer granules to improve the distribution of granules in concrete matrix and healing capability of concrete with the considerations of simple & cheap granule fabrication. 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 by using a typical mortar/concrete mixer for granulation process and using fine sand as nuclei for granulation.

Moreover, it is important to bear in mind that both self-healing ingredients of granule (term of chemical effect) and granulation technique (term of physical effect) are paramount importances to ensure the performance of concrete incorporating granules having semi-capsulation effect.

In order to investigate the potential application of this technique to the construction industry, a field trial was also performed. A ready-mixed concrete were prepared at the plant and transported to construction site by a truck with a drum agitator. Then pre-fabricated self-healing granules were added to drum agitator and mixed with ready-mixed concrete just before casting a portion of slab mock-up.

Based on this experimental investigation, it is expected that the designed concrete possesses an excellent crack self-healing performance, in terms of watertight property recovery and long-term preservation of its capacity. Moreover, there is a promising possibility to broadly apply this concept to practice to improve the durability or functionality, especially in the water retaining and underground structures.

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1-INTRODUCTION

# CHAPTER 1

## 1.1 GENERAL

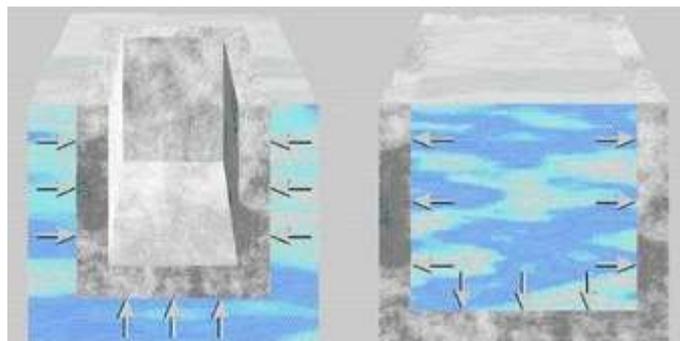
Concrete or reinforced concrete is one of the most widely used construction materials nowadays. Unfortunately, in the practical civil infrastructures the crack appearance and water permeation are common phenomena that facilitate the deterioration of concrete or reinforced concrete structures (*Figure 1.1*). The propagation of penetrating cracks causes a significant reduction in the durability, functionality, serviceability and also the aesthetic aspect of structures.



(a) Bridge slab



(b) Tunnel



(c) Basement/Water retaining structure

**Figure 1.1 Crack problems in civil infrastructures**

In structures subject to contact with water, especially water-retaining/underground structures, the water tightness and durability are paramount importance. In order to achieve watertight properties,

conventional crack repair methods, such as filling the crack by repair materials or surface treatments, can be applied. However, the performances of these methods show not so much efficiency in terms of durability. As a result, the maintenance cost will be increased. One of the main problems is the bonding property between host concrete and new repair materials. Another is its disturbance in structural service or function (for examples, water containers or underground structures should be in dry condition; bridge slab should be free from traffic during period of reparation) and in some specific cases, it is difficult or impossible to access the cracking site to remedy the damage (for instances, very high bridges or containers containing toxic or radioactive wastes).

Due to the features of these structures and drawbacks of conventional methods, it is important to point out that if concrete possessed a sufficient crack-healing ability, in which a crack will be self-healed without human intervention, it was believed that the mechanical (strength, toughness, etc.) or also the water tightness property and the crack closing of concrete would be recovered. Consequently, the service life and functionality of structures will be improved. Such crack-self healing concrete is considered as a smart sustainable construction material nowadays.

It has been known for a long time that concrete has an inherent healing ability. However, based on past researches (*Edvardsen, 1999; Heide, 2005; Hirozo, 2012; etc.*) it has been found that the healing capacity is very limited in normal used concrete. This phenomenon preferably occurs in concrete with low water-cement ratio and only small crack width (typically less than 0.1mm) could be healed (*Reinhardt, 2003; etc.*). In order to obtain the target of sustainable materials, it is necessary to boost the healing properties in concrete. Recently self-healing concrete has become a very hot issue and a great encouragement for a variety of self-healing approaches being developed. Furthermore, 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 nowadays (*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*).

In previous research, *Ahn & Kishi (2008)* and other researchers introduced the self-healing property to concrete by using Portland cement incorporating 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. Further improvements should be investigated.

Recently, one of among several engineered approaches to introduce the healing ability to concrete has been developed by *Koide & Morita (2010)*, in which self-healing agents were added to concrete mixture during mixing in form of granules to stimulate the chemical reactions after cracking and has a potential to preserve the healing property of concrete in long-term. It also is called as self-healing granules having semi-capsulation effect produced by a conventional granulation technique, a common approach to make the capsules for polymer material, or capsulation technique used in food/medicine industry.

Even though there was an improvement on the workability of fresh concrete and compressive strength of hardened concrete by granules, due to its high cost (approximately 10 times) compared with using normal concrete and the insignificant difference in self-healing performance compared with powder type, the application of this approach is constrained in mass construction industry. It is essential to bear in mind that at that moment due to immature technology, granules made by a conventional granulation method did not

fulfill the requirements of granules having semi-capsulation effect. Moreover, there is no doubt that this approach is a preferable concept to introduce self-healing properties to concrete. Therefore, it is necessary to develop a semi-capsulation technique for powder material.

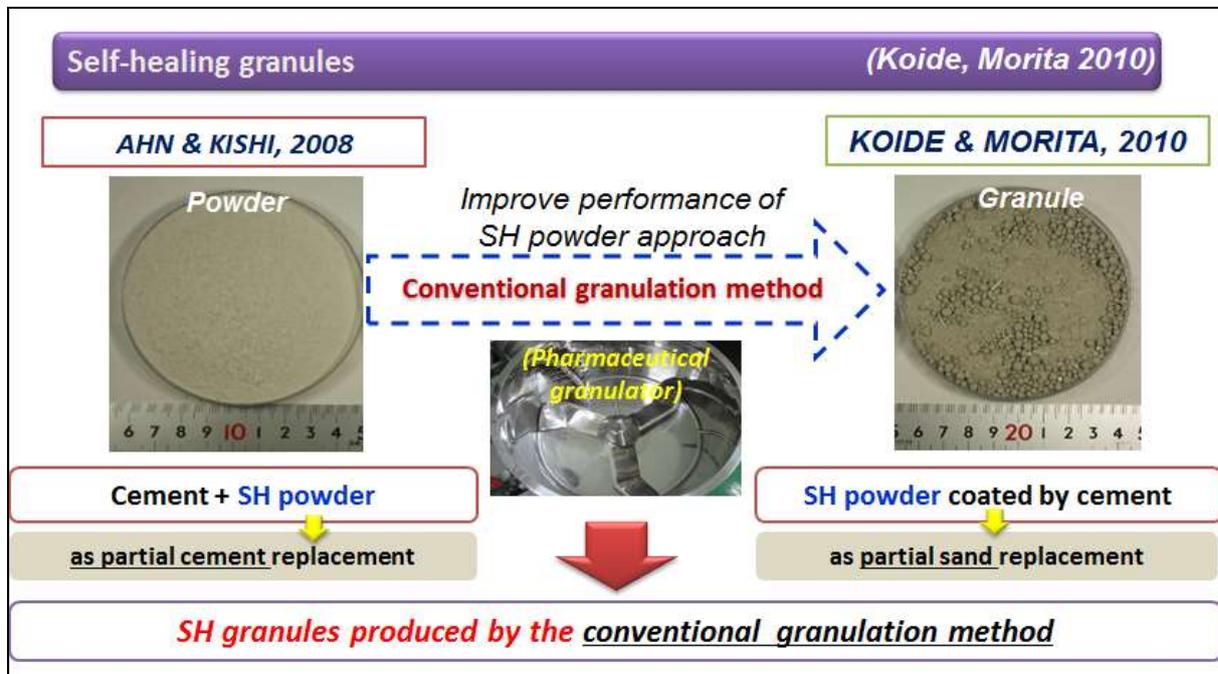


Figure 1.2 Self-healing granules having semi-capsulation effect produced by a conventional granulation technique

## 1.2 RESEARCH OBJECTIVES & SCOPE OF RESEARCH

With its own advantages and the potential to improve the self-healing performance and manufacturing cost (by selecting technologies, ingredients and dosages of granules), self-healing granules having semi-capsulation effect was chosen as a research object in my study. Granules were designed to contain self-healing agents inside, a composition of Pozzolan and/or Portland cement and some specific admixtures based on the basic design concept of self-healing proposed by *Ahn (2008)*, coated by an external layer of cement compound and only activated whenever cracking occurs and contacting with water. The performance of the proposed granules and concretes are enhanced by:

+ Selecting self-healing ingredients and preventing self-healing materials from unexpected reactions during mixing and hardening of concrete (*Improvement compared with Ahn's research, 2008*).

+ Developing suitable granulation methods to make self-healing granules having semi-capsulation with special considerations of the cost and required equipment for the process (*Improvement compared with Koide & Morita's research, 2010*).

The aim of this research is to improve the self-healing performance of granules in concrete by introducing the semi-capsulation technique and try to apply this sort of approach to practical construction industry. In order to obtain the above-mentioned target, following objectives should be done:

+ ***Objective 1:*** To propose the basic design concepts of granule having semi-capsulation effect for powder material.

+ ***Objective 2:*** To enhance the semi-capsulation technique for functional effective ingredients with considerations of simple & cheap granule fabrication.

It is expected that this self-healing granule approach will provide a wide range of applications in practice depending on the required self-healing performance, manufacturing cost, exposure conditions and types of structures, by improvement of the granulation technique, selection of self-healing ingredients and dosage.

The term “crack self-healing concrete”, in this study, is defined as the concrete that has an ability to recover its water tightness properties, typically the reduction of water leakage and crack closing process after cracking. And the scope of this research is focused on the investigation of the recovery of watertight properties in structures where there is an availability of water flow and a static penetrating crack, for examples, the water retaining structures or underground structures. 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.

### 1.3 DISSERTATION OUTLINE

*Chapter 1:* General idea, the objectives and scope of this research are presented.

*Chapter 2:* Literature reviews on natural self-healing and engineered self-healing approaches in cementitious materials are summarized.

*Chapter 3:* Application concept of crack healing concrete using self-healing granules and methodology of this research will be discussed.

*Chapter 4:* Trials to develop granules of cement/pozzolan and self-healing additives is shown and discussed.

*Chapter 5:* Trials to enhance the enhance semi-capsulation technique for cement/pozzolan material.

*Chapter 6:* Investigation on the applicability and healing effect of self-healing concrete in field trial, applied for slab mock-up.

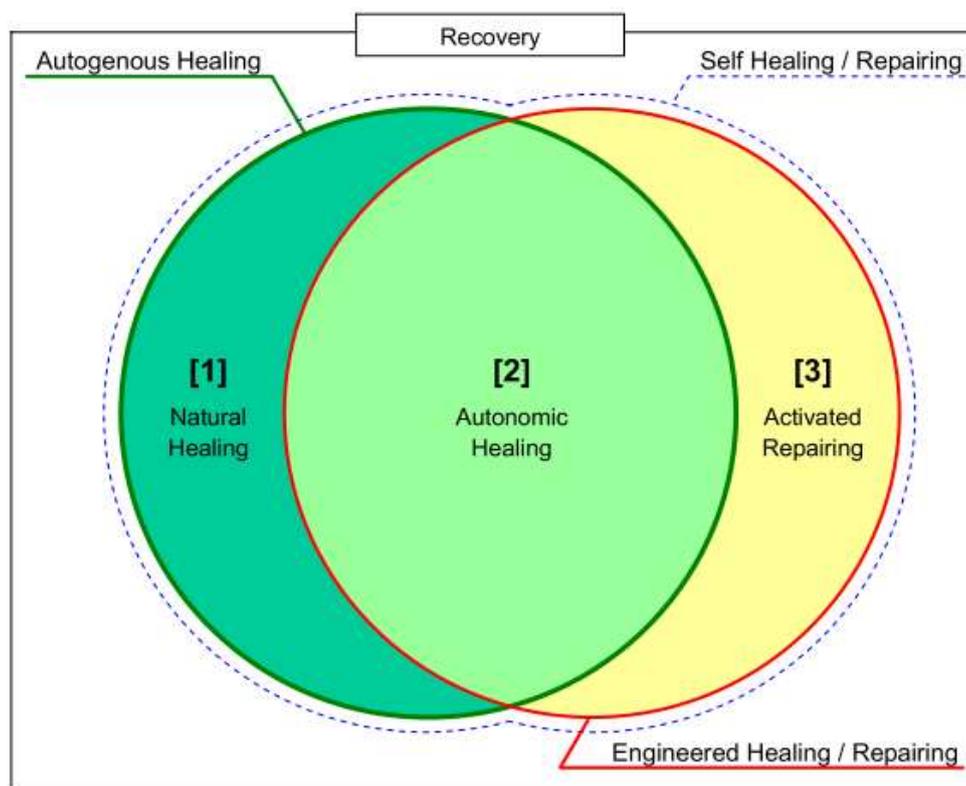
*Chapter 7:* Conclusions , recommendations and further researches will be drawn and proposed.

2-LITERATURE REVIEWS

**CHAPTER 2**

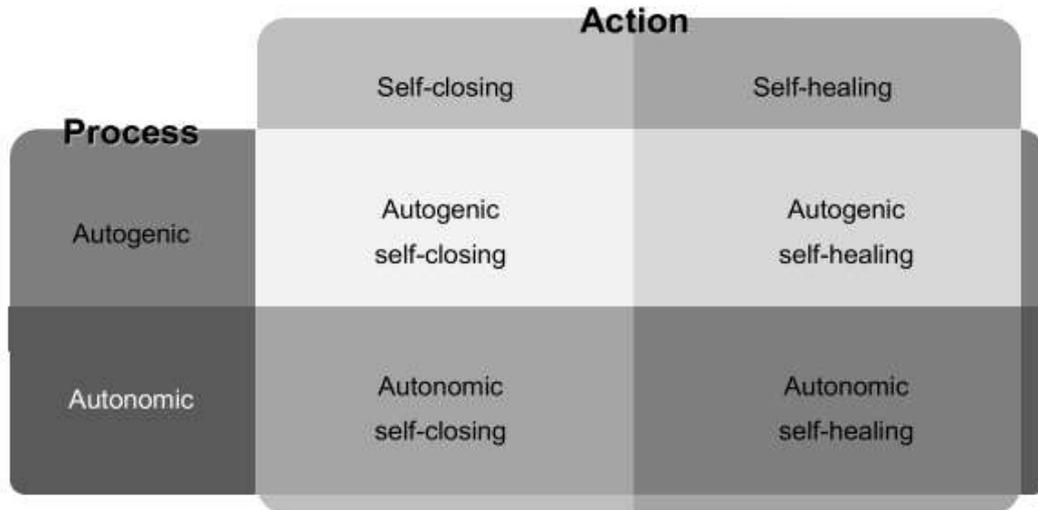
## 2.1 DEFINITION OF SELF-HEALING

Concrete or reinforced concrete is one of the most widely used construction materials nowadays. However with the time being, due to the deterioration process, the quality of structures is gradually degraded and the service life is significantly reduced. With the target of achieving sustainable materials, self-healing/self-repairing concrete is considered as one of the most promising countermeasures and attracts a lot of attention recently. There are many definitions of self-healing/repairing concrete, such as *JCI 2009, Igarashi et al. 2009 (Figure 2.1a)*, *RILEM-TC221 (de Rooij & Schlangen 2011) (Figure 2.1b)*. In this literature reviews, the definitions and categories of self-healing are mainly based on the terms proposed by *H. Mihashi & T. Nishiwaki (2012) (Figure 2.1c)*.



(a) Definite of self-healing/self-repairing concrete

(*JCI 2009, Igarashi et al. 2009*)



(b) Definition of self-healing by RILEM-TC221

*(de Rooij & Schlangen 2011)*(c) Definition of self-healing/self-repairing by *H. Mihashi & T. Nishiwaki (2012)***Figure 2.1 Definitions of self-healing/self-repairing**

According to this, self-healing is divided into two categories: Natural and Engineered self-healing. Natural self-healing is the healing phenomena in concrete without adding specific materials while engineered self-healing occurs in concrete incorporating some special admixtures to promote the healing process.

Another issue is the quality of crack healing. Ideally, the healing of crack in

concrete should recover both the mechanical properties (strength, toughness, etc.) and water tightness properties (stop water leak, crack closing, etc.). However, it is thought that depending on the functionality of structure, the requirement of healing can be rather different. For instance, for the water retaining or underground structures, the tightness property of concrete is of paramount importance (e.g. water leakage should be stopped and crack opening should be closed, but the strength might be unrecovered). While in case of bridge slabs or structures that are subjected to the loading, the recovery of the structural performance is necessary (e.g. complete recovery of original strength is achieved).

Moreover, other self-healing approaches, such as the encapsulation of healing agents in form of chemical solution, epoxy or glues, are categorized as self-repairing concretes that are not mentioned in this review.

## 2.2 NATURAL SELF-HEALING IN CONCRETE

It has been known for a long time that concrete generally has certain capability of self-healing. And its capability is mainly related to the amount of unreacted cements and the availability of  $\text{Ca}^{2+}$  &  $\text{CO}_3^{2-}$  ions. In practical civil infrastructures, it is evident to see white residues depositing at the crack surface of concrete structures, as can be seen in *Figure 2.2*.

In practice, concrete mixtures are often cast at the range W/C ratios of 0.40-0.55. At W/C of 0.40, it was found that there was about 30% amount of un-hydrated cement left in concrete. This amount of un-reacted cement particles is higher if concretes are cast with lower W/C or using coarser cement particles. Due to this fact, concrete generally possesses a self-healing ability due to continued cement hydration. When crack penetrates into concrete matrix and exposed to water, those un-hydrated cements will be ruptured and reacted with water to form the healing products at crack.

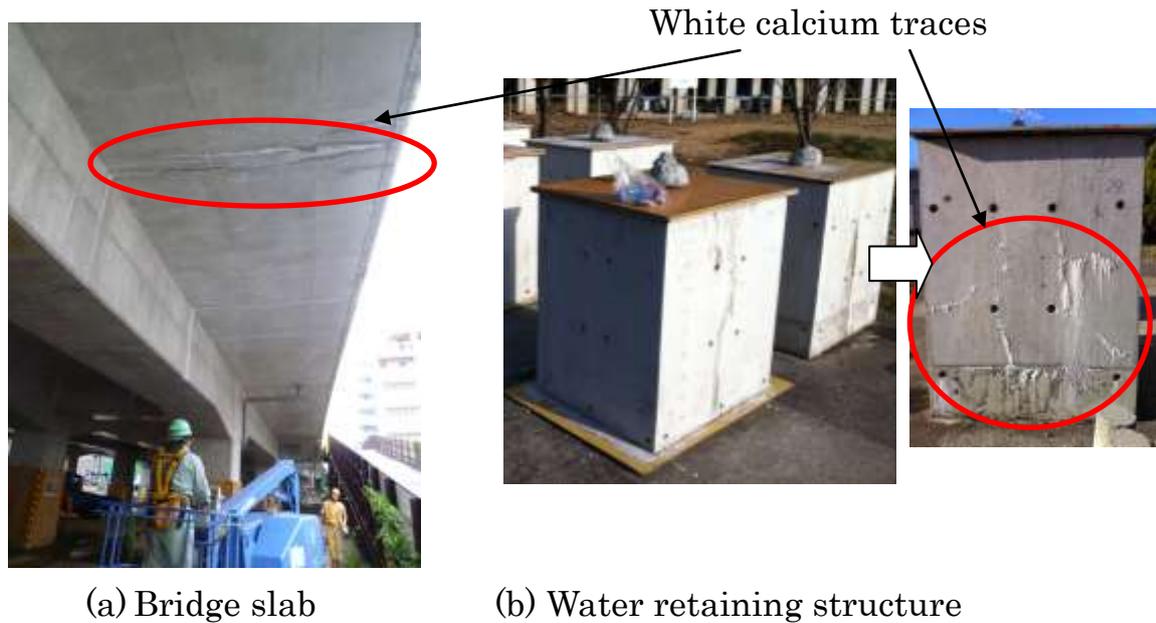


Figure 2.2 White residues depositing at the surface of cracks

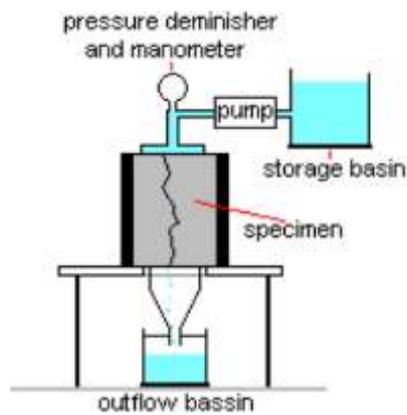
### 2.2.1 Self-healing phenomena in normal strength or low W/C (high strength) concrete:

Most of researches on self-healing were investigated with respect to the water flow through a crack.

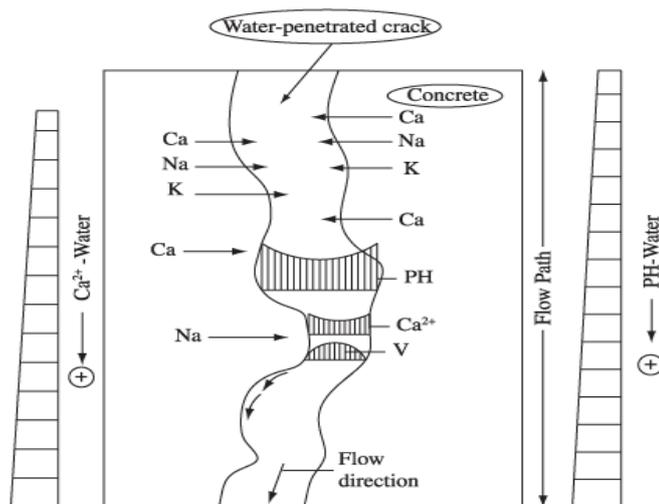
*Clear (1985)* conducted a study on the effects of autogenous healing upon the leakage of water through cracks of 0.1, 0.2 and 0.3mm width in different concretes. It was concluded that there was a significant reduction in water flow depending on the crack width, concrete composition and flow rate. The chemical analysis confirmed the calcite precipitation in the crack. However, during the first days the author thought that the crack blocking due to loose particles in the crack was the main reason accounted for decreasing of flow rate.

*Edvardsen (1999)* conducted a large scale water permeability and autogenous healing of a through crack in concrete (*Figure 2.3*). Based on her research, the healing of cracks was able to be observed in both reduction of flow rate and closing of crack, depending on the crack width and water pressure. However this phenomenon was not affected by concrete composition and the hardness

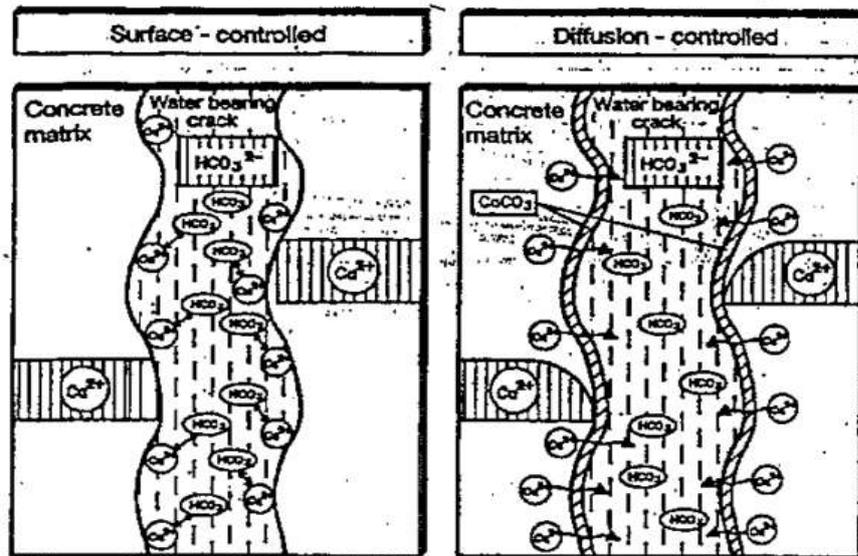
of water. The author concluded that the main mechanism for healing the cracks was calcite formation. Especially, the precipitation of calcium carbonate and the time-dependent flow rate in crack subjected to water flow was also discussed based on two different crystal growth processes. During the first days exposed to water flow, the crystal growth was surface controlled and then it changed to a diffusion controlled process, due to the changes with time in the chemical and physical conditions in the crack (**Figure 2.4**). Another interesting finding was active cracks also showed self-healing effects. However, they needed longer time to obtain the same result of that occurred in static cracks.



**Figure 2.3** Permeability test set up (*Heide 2005*)



(a) Conditions in water-penetrated concrete crack



(b) Surface and diffusion controlled process of calcite growth

Figure 2.4 Conditions and crystal growth of calcite processes in crack subjected to water flow (Edvardsen 1999; J.H. Yu 2010)

Reinhardt (2003) had tested the self-healing capability of high strength concrete under influences of temperatures of flowing water (20, 50 and 80°C) and ranges of crack width (0.05-0.20mm) (Figure 2.5). It was found that the crack healing effect with respect to reduction in flow rate is faster if crack width is smaller and the temperature is higher (Figure 2.6).

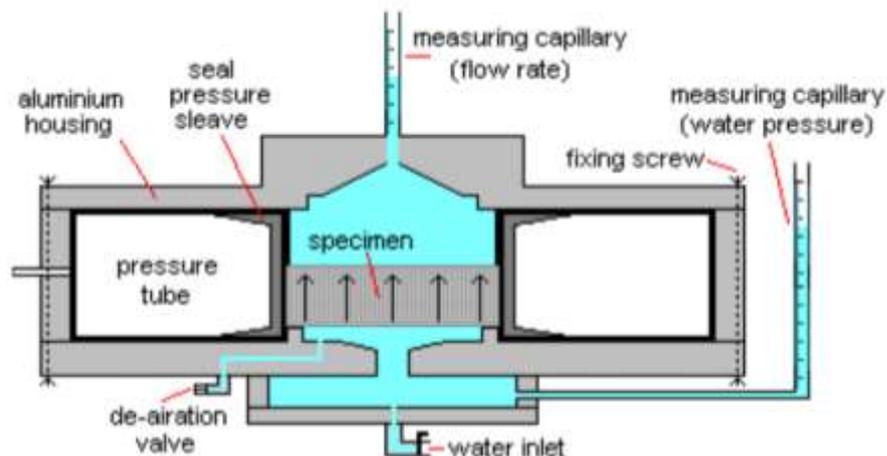
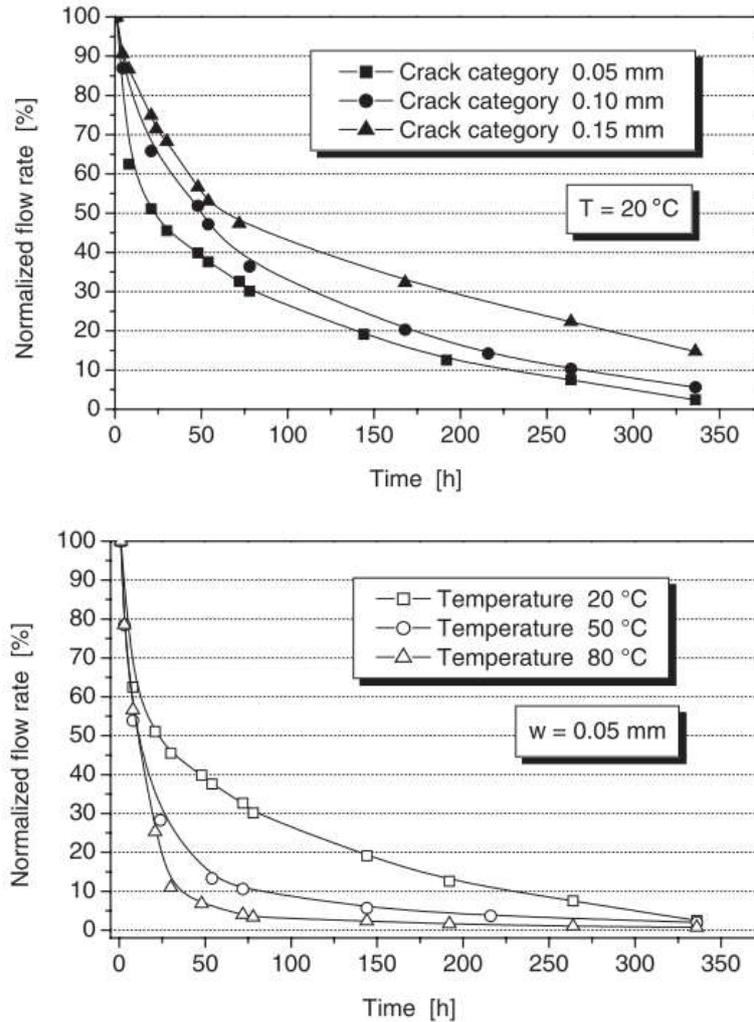
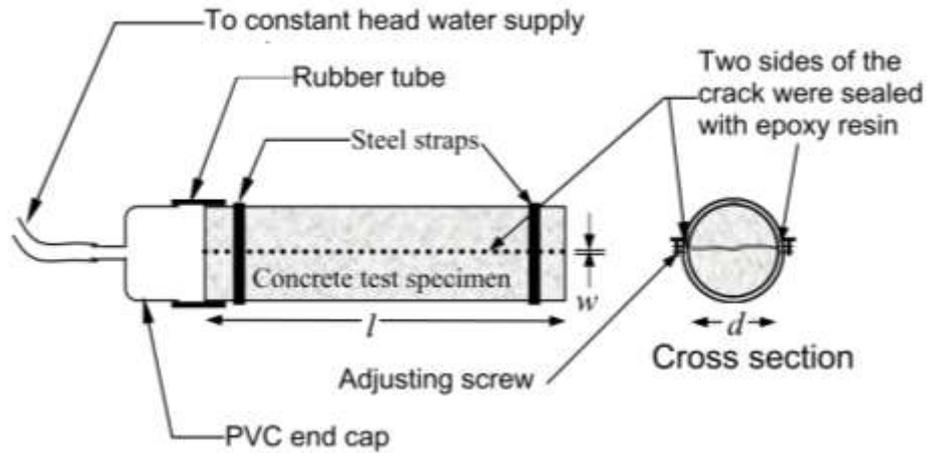


Figure 2.5 Permeability test set up (Heide 2005)

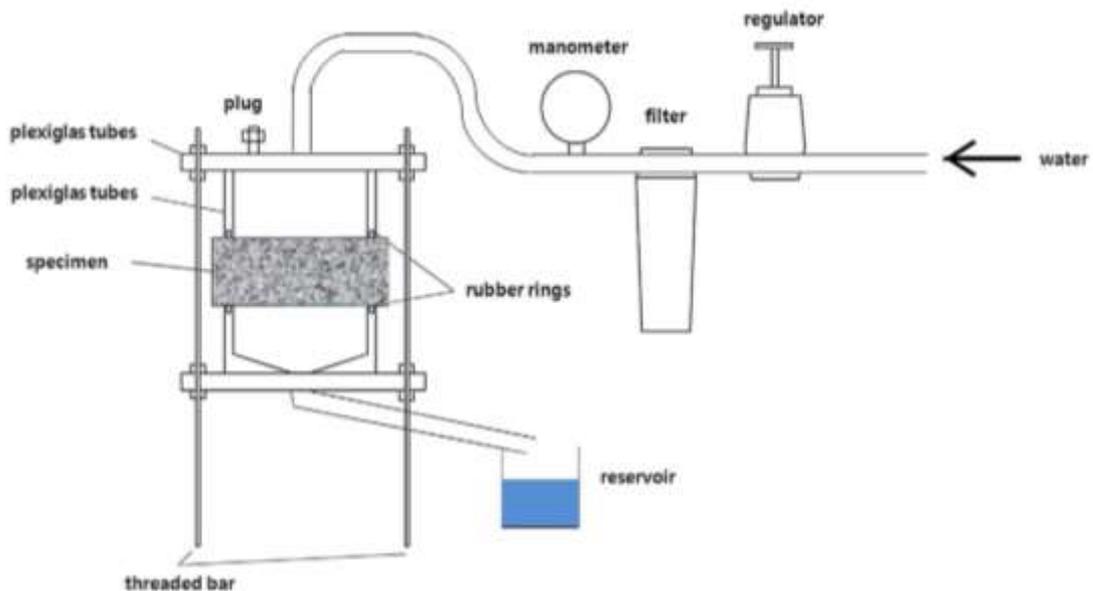


**Figure 2.6 Influence of crack widths and temperatures on water flow rate**

The effects of water pressures and crack widths on healing properties of concrete were also studied by *Anura (2003) (Figure 2.7a)* and *S.T. Yi (2010) (Figure 2.7b)*. According to the first author, there was a relationship between the healing time and crack width under the same pressure gradient (liquid height/crack width). Furthermore it was verified that under specific hydraulic gradient or water pressure, the crack width should not be larger than the critical value so that the healing process would occur (*Table 2.1 –Breugel,1984*). When the crack width is small, typically less than  $50 \mu\text{m}$ , the water permeability of concrete is almost negligible. However, with a larger crack width and a higher water pressure, the permeability of concrete increases significantly.



(a) Water permeability test (Anura 2003)



(b) Water permeability test S.T. Yi (2010)

**Figure 2.7 Tests set up for evaluating the healing properties****Table 2.1 Relationship between hydraulic gradient & critical crack width**

Hydraulic gradient (Liquid height/crack length)	Critical Crack width (mm)
< 2.5	<0.2
<5	<0.15
<10	<0.10
<20	<0.05

*J. Parks (2010)* investigated the effects of bulk water chemistry on the

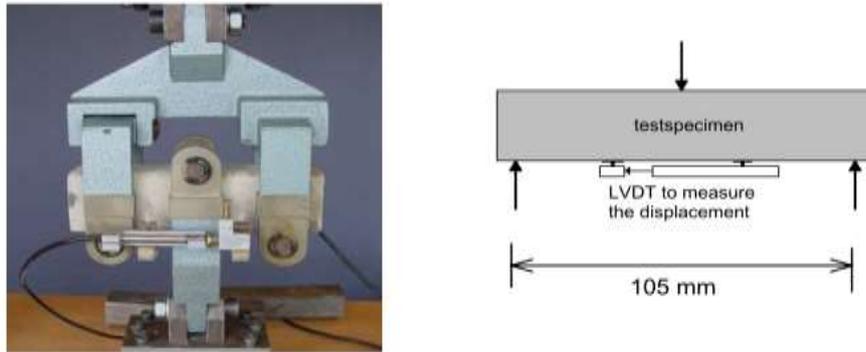
autogenous healing of concrete. Precipitation of calcite was thought as the main mechanism that healing the crack, therefore it was expected that water containing high amount of calcium could contribute to the healing ability. However, there was no significant strength gain in this experiment. It is also found that water with high concentrations of magnesium and silicate near pH of 9.5, both the crack closing and shear strength of concrete were improved. Based on the result, this study was considered as one of the important approaches to stimulate the healing process in concrete. *Fagerlund & Hassanzadeh (2010)* also made a long-term test on self-healing of cracks in concrete exposed to different types of water: salt, brackish and pure. In their test, two types of water exposure have been chosen as: permanent immersion and cyclic wet and dry in lab conditions. It was found that some certain healing of cracks could be observed, especially in the case exposed to salt water. The evidences of crack healing were verified by photography, measurement of chloride content on the crack wall and chemical analysis.

Contrary to number of research on self-healing in term of water flow, there were very few experiments or researches which were investigated on the recovery of mechanical properties of concrete after cracking.

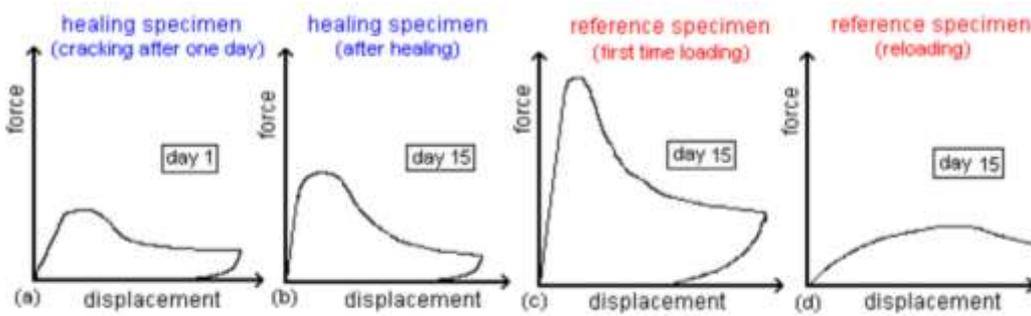
Self-healing of high strength concrete after deterioration by rapid freeze/thaw cycles was conducted by *Jacobsen (1996)*. After exposure to freeze/thaw test, the specimens with micro cracks were cured in water for three months and then healing effects were verified by the recovery of resonance frequency & compressive strength; and the reduction in migration of chloride. It was found that the loss of resonance frequency could be fully recovered, but only small percentage of compressive strength recovery was obtained. Furthermore, due to hydration products filling the micro cracks, it was thought that the rate of chloride migration into concrete was reduced.

*Heide (2005, 2007)* investigated the self-healing of early age cracks in concrete in term of recovery of the mechanical properties (such as the 3 point bending strength and the stiffness – *Figure 2.8*), under various influences: effect of

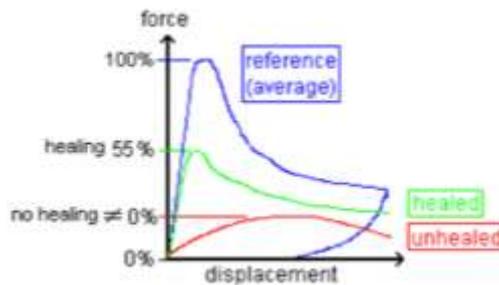
compressive strength, type of cement, age of concrete when introducing a crack, crack opening and effect of relative humidity.



(a) Three point bending test



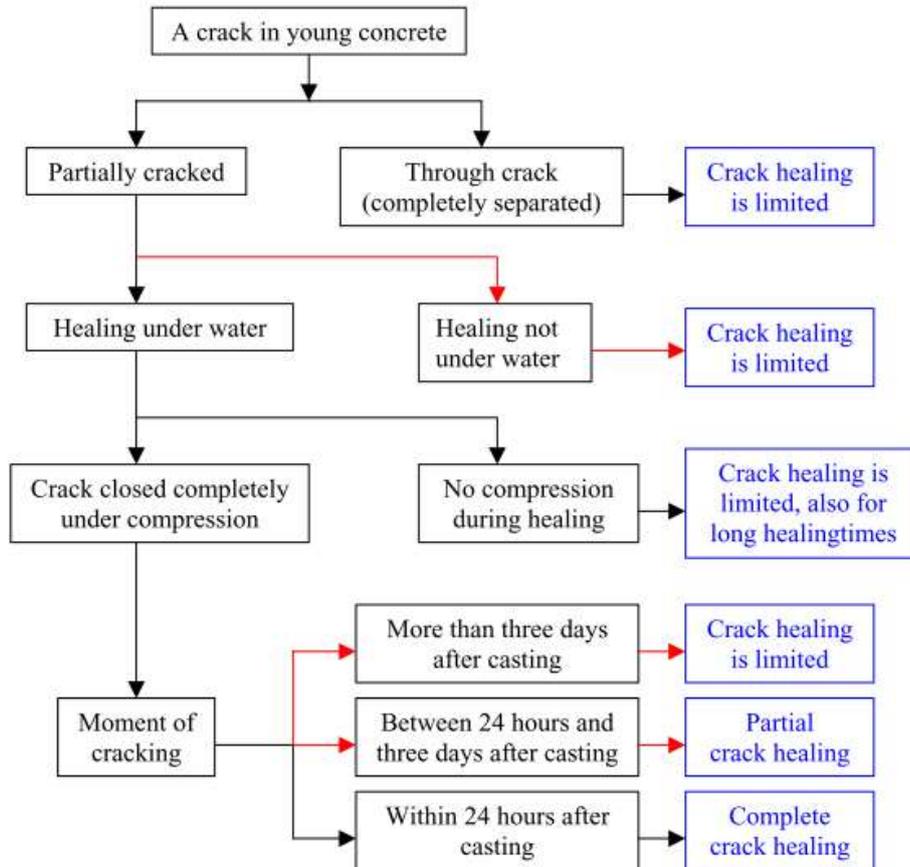
(b) Force-displacement curves for healing specimen (a&b) and for reference specimen (c&d)



(c) Healing relative to the reference specimen

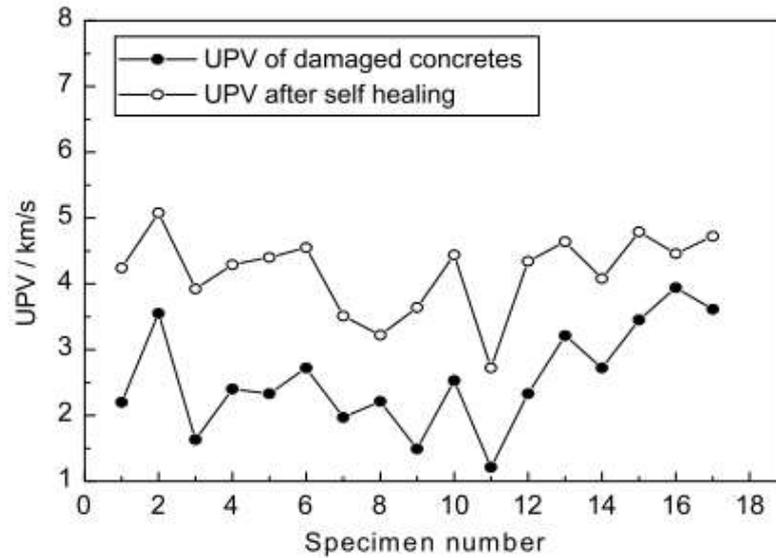
**Figure 2.8 Three point bending test and healing experiment**

The results of these experiments were summarized as seen in *Figure 2.9*.



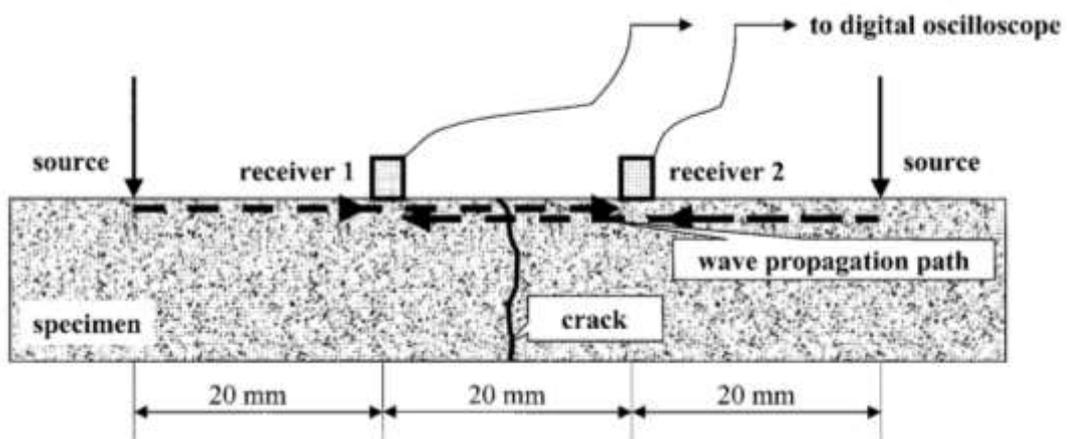
**Figure 2.9 Summary of experimental results**

*Zhong (2008)* studied the influence of damage degrees on self-healing of concrete by measuring ultrasonic pulse velocity UPV (*Figure 2.10*). After inducing micro cracks in normal strength and high strength concrete via compression load, all the specimens were cured in standard room. By comparison the pulse velocity before and after curing, the healing of crack can be detected. Based on the experimental results, it was concluded that there was a “damage threshold” for normal strength and high strength concrete. If applied damage degree is smaller than threshold value,” healing ratio of concrete” will be higher when higher damage is obtained. On the other hand,” healing ratio of concrete” will decrease with higher damage if the damage degree is larger than threshold value.



**Figure 2.10 Changes of UPV in damaged & healed concrete**

Even though UPV measurement can detect the occurrence of crack healing, this technique cannot determine the extent of healing (*Aldea 2000*). To overcome this issue, *Aldea* suggested to apply stress wave transmission measurement (*Figure 2.11*). The results showed that under water flow, both the water permeability and transmission will be recovered with time, however the rate of recovery of “signal transmission” is less significant than that of water tightness.

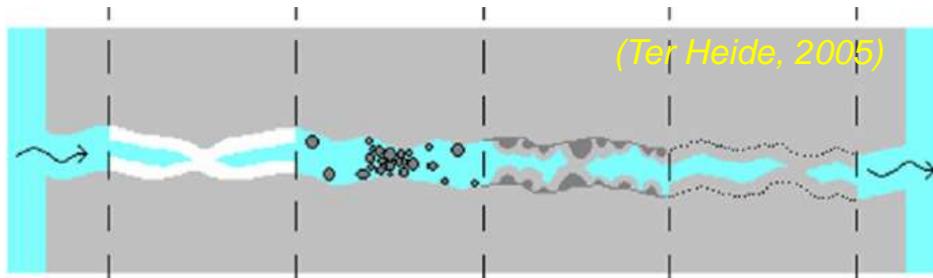


**Figure 2.11 Stress wave transmission test set up**

Based on the literatures, small cracks in cementitious materials exposed to

water flow can be self-healed by following mechanisms, as can be seen in *Figure 2.12* (summarized by T. Heide, 2005):

- Precipitation of calcium carbonate (calcite) or calcium hydroxide.
- Crack blocking by the impurities in water flow and broken particles from cracking.
- Continued hydration of un-reacted cement particles.
- Swelling or expansion effect of cementitious matrix.



**Figure 2.12 Possible mechanisms for natural self-healing in cementitious materials**

In young concretes, there is a huge amount of un-reacted cements in concrete. Therefore the continuing hydration of un-reacted cements when crack penetration and contacting with water is believed as the main mechanism for crack filling. With the elapse of time, it is thought that almost all cement in concrete is hydrated and the crack healing in old or mature concrete is occurred mainly due to the dissolution or precipitation of calcium carbonates.

Unfortunately, this phenomenon preferably occurs in concrete using large amount of cement or low W/C ratio, and the crack width should not be too large, typically less than 0.1mm. Moreover, it is difficult to control or determine the healing capacity in these cases.

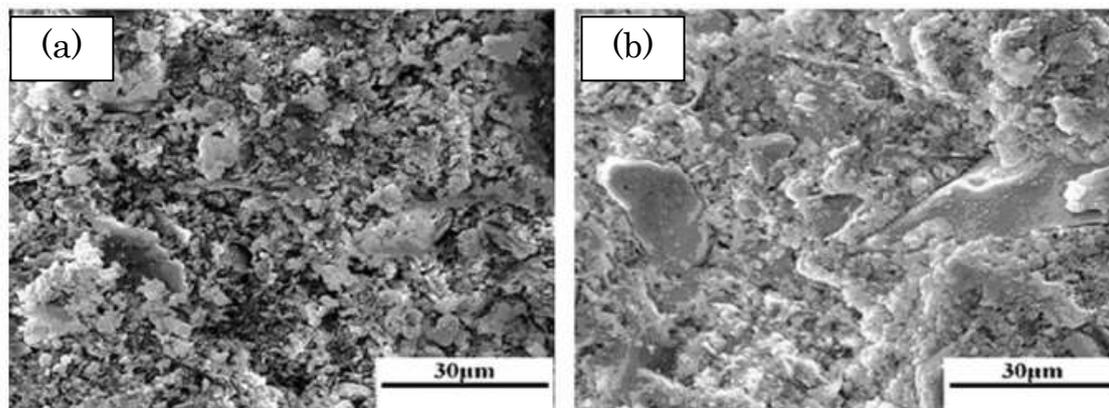
### **2.2.2 Self-healing in concrete using cement coarse particle:**

Due to coarser particles of cements, only the surface of cement is hydrated as can be seen in *Figure 2.13* (T. Zhang 2011). Based on this fact, it inspires the idea to promote the healing effect in concrete by using coarser cement. Lin

(2005) made an experimental study on the healing ability of concrete with different particle size distribution of cement particles. *Li Zhiqiang (2011, 2012)* studied on the self-healing ability of concrete influenced by the particle size (*Table 2.2*) and content of coarse cement. In his research, based on the ultrasonic method it was interesting to find that coarse particles of cement may contribute to self-healing effect of concrete.

**Table 2.2 Physical properties of cement (*Li Zhiqiang, 2011*)**

Cement type	Grinding time/min	Size / $\mu\text{m}$	Median / $\mu\text{m}$	Density / $\text{gcm}^{-3}$	Specific surface area / $\text{m}^2\text{kg}^{-1}$
Normal cement	-	20.14	14.54	3.03	393.0
Coarse cement 1	5	42.97	27.15	3.16	244.5
Coarse cement 2	10	40.64	22.99	3.19	247.2
Coarse cement 3	15	39.25	21.42	3.14	263.9



**Figure 2.13 SEM images of hardened cement pastes: (a) Portland cement (fine particles) paste cured for 24h (b) Portland cement (coarser than  $30\ \mu\text{m}$ ) paste cured for 3d (*T. Zhang, 2011*)**

Even though there is a potential to use coarse cement to promote self-healing properties in concrete, this application causes bleeding when mixing concrete and also reduction in early strength of concrete. Further improvements should be necessarily investigated.

### 2.3 ENGINEERED SELF-HEALING IN CONCRETE

In order to achieve the target of a smart sustainable construction material,

concrete should be designed with higher crack healing capability. Recently with the high development of applied technologies and sciences, several engineered SH approaches are being developed by various group researchers to introduce or improve significantly the healing ability in cementitious materials.

### 2.3.1 Mineral/chemical (powder type) approach:

There are various types of mineral and chemical admixtures being used to improve the healing capability of concrete. Generally, this approach can be divided into three groups as following:

#### a) Supplementary cementitious materials (SCMs)

Nowadays the supplementary cementitious materials (*Figure 2.14*), such as blast furnace slag, fly ash, silica fume, etc..., have widely been used due to their friendly environmental materials (most of them are by-products from industry; partial cement replacement); significant improvement of the mechanical & durability properties of concrete (*Barbara L. 2011*) and also cost benefit. 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; Y. Ishikawa, ICSHM2013*).

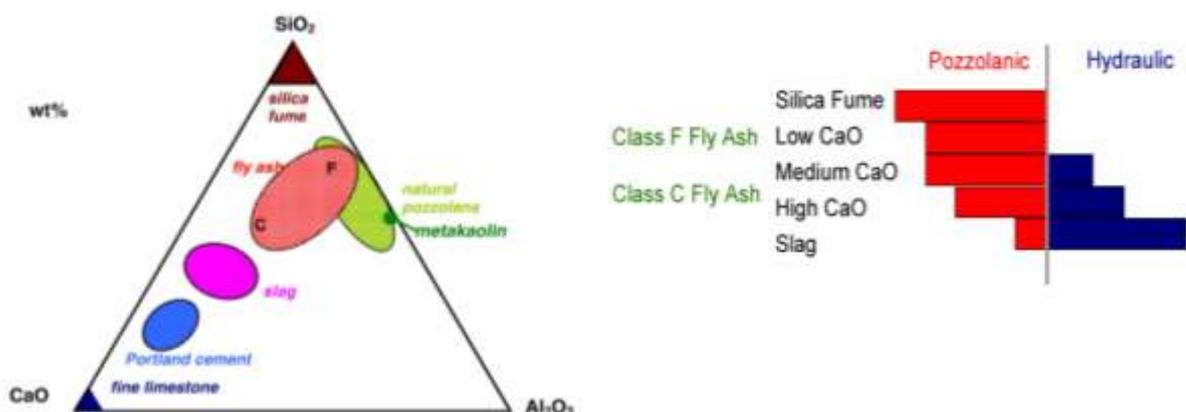
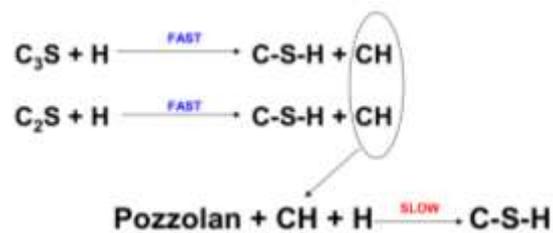


Figure 2.14 (a) SCMs chemical composition (*Barbara L.2011*);  
 (b) Pozzolanic/Hydraulic property of different SCMs (*Kimberly*)

The mechanism is due to pozzolanic reaction between pozzolan and calcium hydroxide, one of cement hydration products, a new product similar to hydration product is formed that filling the crack (*Figure 2.15*). Another reason contributing to the healing capacity is the low reactivity of supplementary cementitious materials. As a result, there is still a huge amount of those materials remaining unreacted over a long time period (*Baert 2009; Gruyaert 2011*). Once the crack occurs, together with moisture, pozzolanic reaction will be triggered and crack will be healed by newly formed products. However, due to its assumption of  $\text{Ca}(\text{OH})_2$ , there is a possibility that the phenomenon of crack healing by the precipitation of calcite will be restrained (*K. Van Tittelboom, ICSHM2013*).



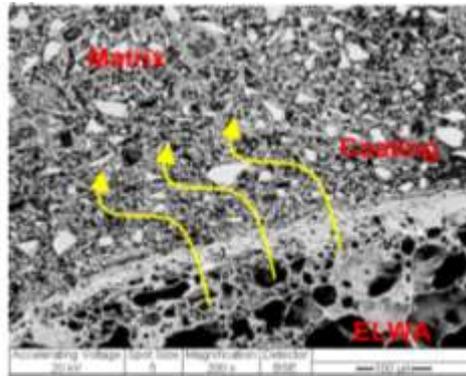
**Figure 2.15** Pozzolanic reaction (*Kimberly*)

Most of researches in this field are focused on the healing of micro cracks in concrete, in terms of the recovery of mechanical properties, such as the compressive strength, ultrasonic pulse velocity; or recovery of impermeation properties, such as chloride penetration reduction or water sorptivity (*Mustafa S. 2008*).

Due to its low reactive material, especially if huge dosage of SCM is used, many researchers try to improve the reactivity by choosing the appropriate activators. *E. Gruyaert (2012)* tried various types of activators for slag and fly ash in mortar. Based on his results,  $\text{Ca}(\text{OH})_2/\text{Na}_2\text{CO}_3$  solution showed best performance, but further experiments should be done to confirm the results.

Recently Sodium monofluorophosphate (Na-MFP) has been proposed as one of countermeasures to improve self-healing in slag-rich concrete by surface

coating (*Bilal 2011*). The mechanism is that the healing agent will react with carbonated matrix to form a stable product that improves the concrete surface against the frost salt attack. Based on this research, *K. Sisomphon* tried to investigate self-healing of slag mortar subjected to carbonation and frost salt scaling by embedding light-weight aggregates, which are impregnated by Na-MFP, coated by cement. In this case the carbonation process triggers the releasing of encapsulated agent into the matrix by coarsening the coating layer (*Figure 2.16*).



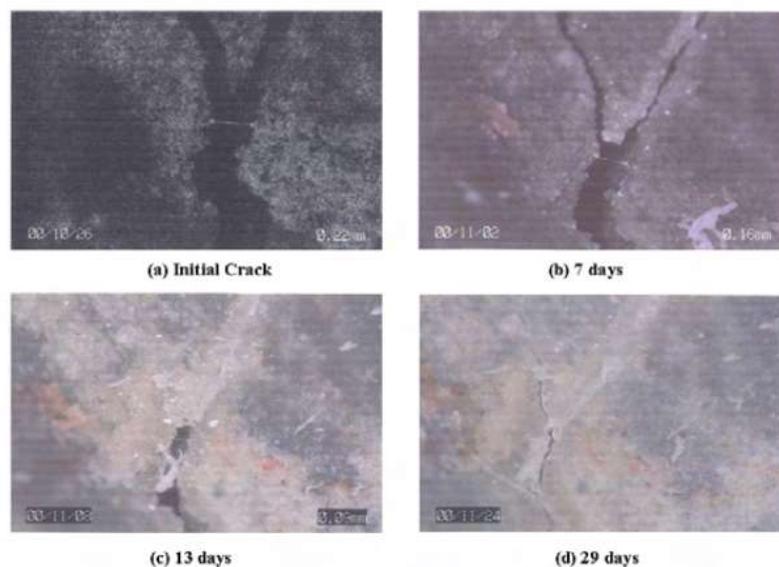
**Figure 2.16 Photomicrograph of ITZ after carbonation attack  
(*K. Sisomphon*)**

*b) Expansive agents*

Initially, expansive agents were added to concrete for shrinkage compensation to prevent an early cracking. *Hosoda A. and Kishi T. (2007)* investigated the self-healing of crack and water permeability of expansive concrete with low W/B ratio and high dosage of expansive agent. It was found that crack width of 0.22mm could be healed after one month cured in water (*Figure 2.17*). The healing of crack was due to the additional expansion and the precipitation of new products. However, the expansion effect was significant when using huge amount of agents that would lead to high initial cost and specimens should be kept under sufficient constrain conditions.

To improve problems above, some special additives were introduced to expansive concrete with higher W/B ratios and lower dosage of expansive agent. In these cases, the crack sealing effect is mainly due to the

precipitation of new products ( $\text{CaCO}_3$ ). For examples, *Kishi T.(2007)* used selective carbonate compounds incorporating with expansive agent (Calcium sulphoaluminate-CSA) in normal concrete to promote the recrystallization and precipitation; *Yamada K. (2007)* incorporated cement crystal breeder material or bicarbonate of soda and expansive agent with various Portland cements to facilitate the precipitation of  $\text{CaCO}_3$ ; *Hosoda A. (2009)* and *K. Sisomphon (2011)* studied the effects of different crystalline additives and expansive agent on crack self-healing properties of concrete and mortar. Recently, group research from Delft University of Technology also has tried to use tri-calcium aluminate – one of cement minerals- as the healing agent to trigger an expansive reaction for crack healing.

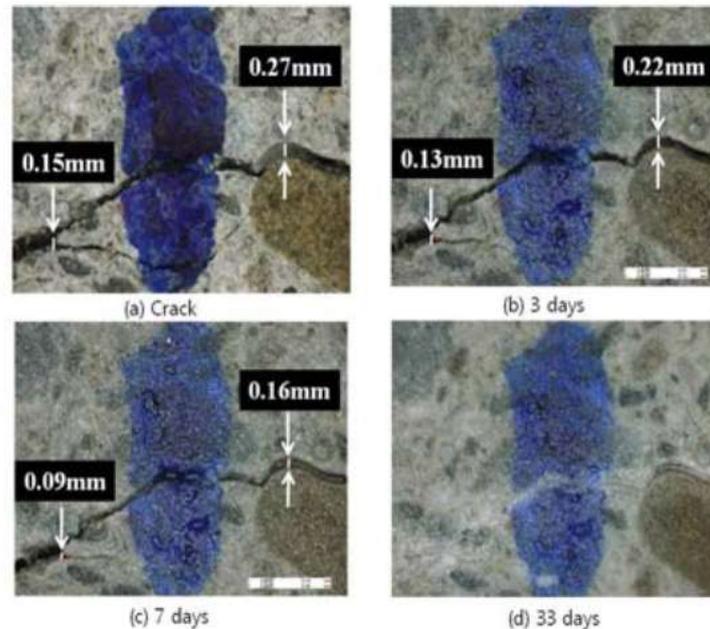


**Figure 2.17 Crack closing process in expansive concrete of low W/B ratio**  
(*Hosoda & Kishi, 2007*)

*c) Swelling + expansive + precipitated agents*

*Ahn & Kishi (2008)* developed cement compounds incorporating expansive agent and geo-materials in form of powder to improve the healing ability of normal concrete by further combination effects of expansion, swelling and carbonate precipitation. A crack width of 0.27mm could be fully healed after 33 days exposed to water (**Figure 2.18**). Based on the obtained results, it was

found that an appropriate usage of alumino-silicate materials and various types of calcium compounds may provide a significant contribution to crack healing capacity of concrete.



**Figure 2.18 Crack closing process of self-healing concrete at W/B of 0.47**  
(Ahn, 2010)

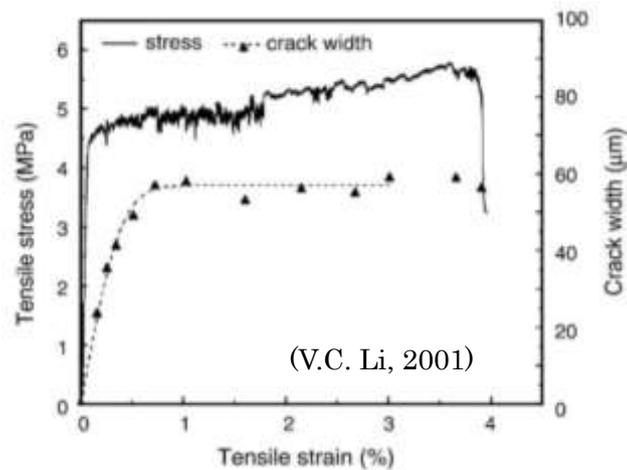
Even though this powder type approach showed promising effects, there was still a need for further improvements to overcome the following problems, such as a reduction in the workability and also self-healing capacity due to further reactions between self-healing powders and mixing water during casting and hardening process of concrete.

### **2.3.2 Fiber reinforcement and low water binder ratio (W/B) approach:**

The idea is small crack width will be controlled and promote a healing phenomenon to occur by continued pozzolanic or hydration reactions. Due to tight crack widths and huge amount of unreacted materials left in cementitious materials, the higher reliability for healing of crack is expected.

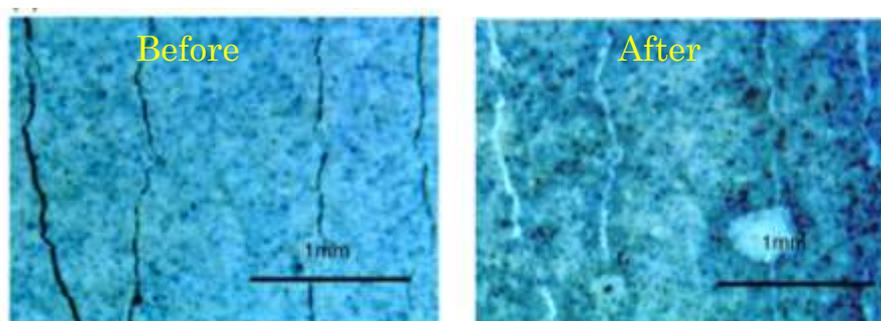
Engineered Cementitious Composite (ECC) developed by *V.C. Li* was considered as one of the best representatives for this approach. ECC, kind of high performance fiber reinforced cementitious composites, is designed from

micromechanical concepts. The tight crack widths are the result of the combination of fiber and cementitious matrix, together with the interfacial properties between these components (*M. Lepech & V.C.Li*) (**Figure 2.19**). Due to its tight crack, it was verified that both the recovery of mechanical and transport properties were obtained by this approach. Based on chemical analysis, it was found that healing products that depositing on the crack surface were mainly C-S-H and  $\text{CaCO}_3$  (*Kan, Mustafa, Yang*).



**Figure 2. 19 Typical tensile stress-strain-crack width curve of ECC**

*Yang (2009)* studied the autogenous healing of ECC under wet-dry cycles. It was observed that the resonant frequency, uniaxial tensile strength and water permeability were recovered after exposure as can be seen in **Figure 2.20 & 2.21** . Based on this result, the author found that if the crack width is smaller than  $50\ \mu\text{m}$ , both the mechanical and transport properties could be fully recovered. However, with crack width in range of  $50\text{-}150\ \mu\text{m}$ , the rate of recovery would be less or just partially healed (**Figure 2.22 a& b**).



**Figure 2.20 Crack closing in ECC (*Y. Yang 2009*)**

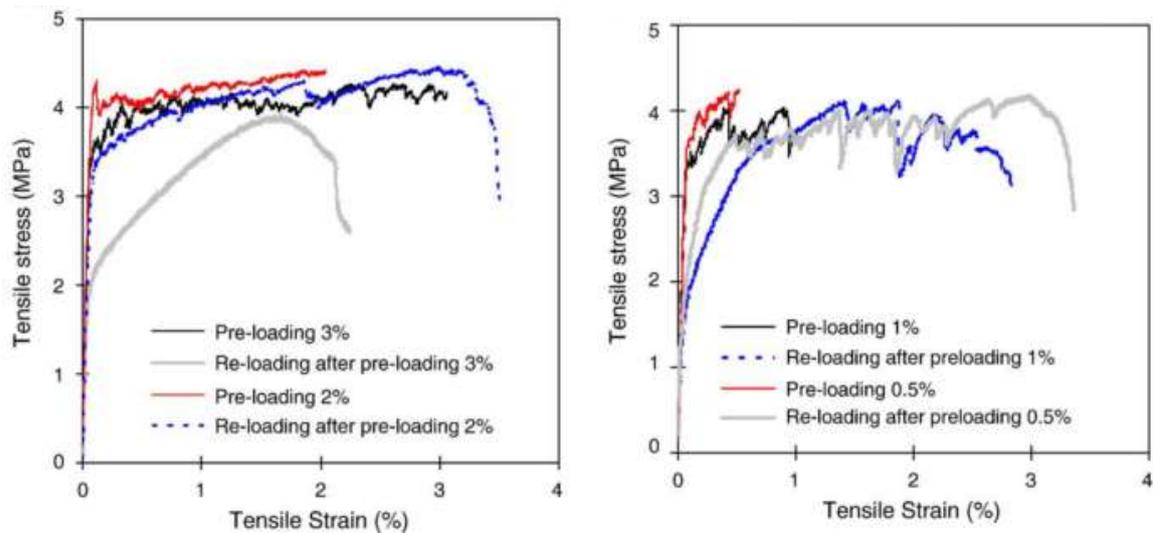


Figure 2.21 Preloading and reloading after wet/dry cycles tensile stress-strain relations of ECC (a) Preloading to above 1% (b) Preloading to 1% or below (*Y. Yang 2009*)

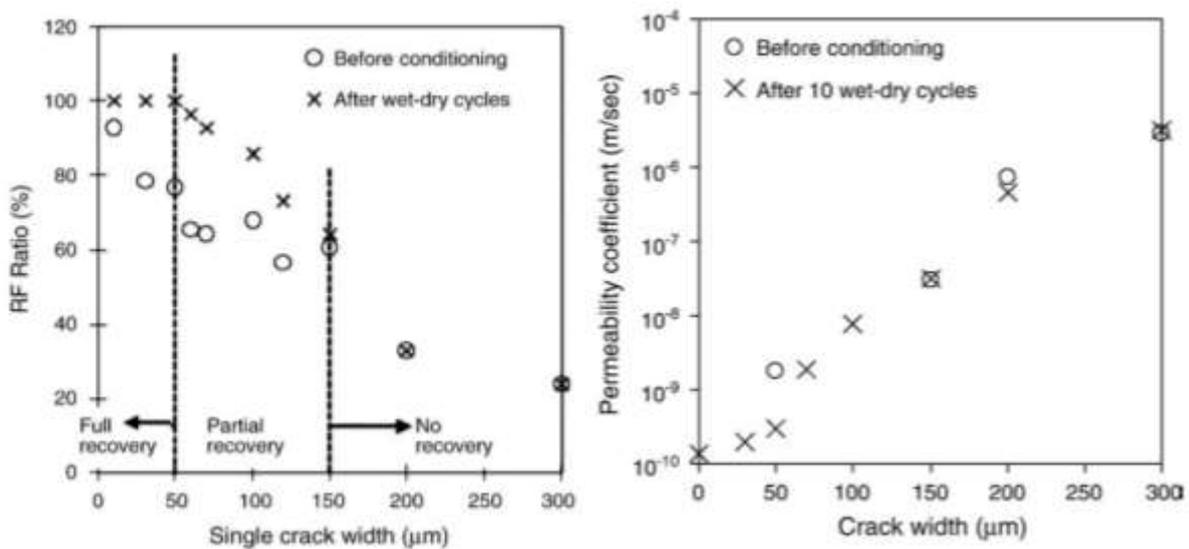
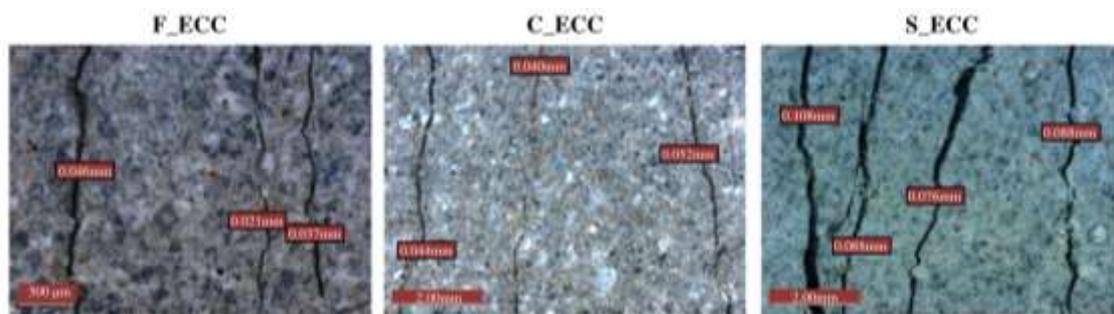


Figure 2.22a (left). Resonant frequency as a function of crack width; b (right) Permeability as a function of crack width (*Y. Yang 2009*)

*Kan (2010, 2012)* also observed the recovery of resonant frequency, ultimate tensile strength and tensile strain capacity of the re-healed ECC under wet-dry conditions.

Moreover, self-healing capability of ECC incorporating different types of supplementary cementitious materials (SCMs) was investigated by *Mustafa S. (2012)*. SCMs, such as fly ash, slag, were used as partial replacement of

cement to reduce the cost and also to be achieved as a “green” material. The healing capability of ECC was examined by measuring the amount of chloride penetration under various exposure conditions: air, water and freeze/thaw cycles. It was thought that due to healing effect, healing products close the crack mouth and prevent the chloride ions penetration. From the experimental results, the slag-ECC showed the best healing effect under continuous water curing due to its “higher pH value of the pore solution and CaO content” (Mustafa) (Figure 2.23).



(a) Before self-healing (60 days of age)



(b) After 60 days of water curing

**Figure 2.23 Crack closing process in ECC**

In general, the healing of crack in ECC or concrete needs the availability of water for triggering the reactions to form new products. However, in practice bridge slabs or other civil structures are not always exposed to water. In that case the healing process will be disturbed. It can be observed from results of the self-healing effect in concrete or ECC under air curing condition. The healing effect of crack is not achieved. In order to overcome this issue, group research from *Delft University of Technology* studied on the use of Super

Absorbent Polymers (SAP) in ECC. SAP will absorb the water during mixing and store them as water pockets in concrete mix. When crack occurs, water will be released from these water pockets and promote the hydration reactions to fill the crack. When these pockets are empty due to rupture, they can be filled again if raining occurs. However, this approach is still under investigation.

Even though ECC possessed high self-healing capability in terms of recovery of both mechanical and transport properties under various environmental exposures, the high initial material cost is one of the main reasons that restrained the wide application of this approach in construction industry.

### 2.3.3 Bacteria-based approach (Bio-chemical):

It is broadly accepted that calcite precipitation is one of the most common mechanisms contributing to crack-healing capability of concrete. Based on this fact, it inspires the idea of using bacteria to produce calcium compounds that fill the crack. Group research from *Delft University of Technology* tried to develop “bio-chemical” self-healing approach by using encapsulation light weight aggregates (LWA) (*Figure 2.24*). Bacterial spores and food (calcium lactate solution) are impregnated into expanded clay particles to prolong the life of bacteria in concrete.

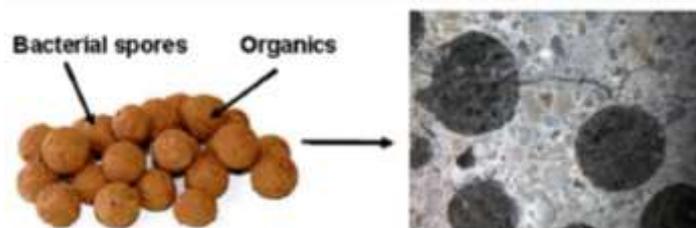
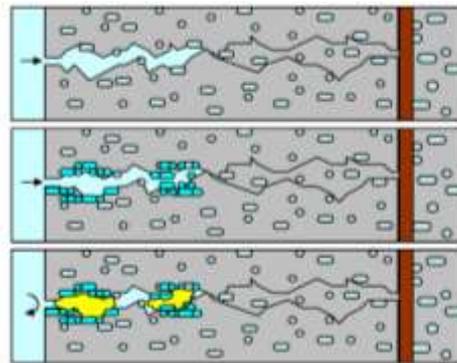


Figure 2.24 Encapsulation LWA (*Jonkers*)

The scenario for this approach can be seen in *Figure 2.25*. Firstly, concrete was cast incorporated with encapsulation LWA. Once the crack occurs, it penetrates the LWA. Under the water supply, the bacteria are activated and convert the calcium lactate to calcium carbonate that fills the crack (*Figure 2.26*).

According to *Wiktor and Jonkers (2011)*, the crack-healing capacity of this approach is more significant than that of normal concrete. Crack with width less than 0.46mm can be fully healed.

However, further improvements and considerations should be done before applying in real structures. Firstly is a reduction in the compressive strength of concrete incorporating large amount of encapsulation LWA. Secondly is the short service life of bacteria in concrete.



**Figure 2.25** Scenario of crack-healing by bacteria based approach  
(*Mors & Jonkers 2012*)



**Figure 2.26** Crack closing process (*Jonkers*)

In summary, different self-healing approaches have their own features, advantages and disadvantages. *Table 2.3* showed the brief comparison of those approaches mentioned above.

Table 2.3 Comparison of different self-healing approaches

SH approach	Fiber reinforced	Bacteria	Mineral/chemical
Features	<b>Small crack width</b> + Pozzolanic + Continued hydration + Calcite formation	Bacteria produce <b>calcite</b> to fill the crack	<b>Swelling + Expansion + Precipitation</b>
Advantages	Recover <b>mechanical/transport properties</b> ; Small crack width (~0.05mm)	Recover <b>transport properties</b> ; Larger crack width (~0.46mm)	Recover <b>transport properties</b> ; Crack width of 0.20mm; Cheaper cost (~2times)
Disadvantages	<b>Very high cost</b> ~-(16-17)times ( <i>Japan</i> )	<b>Short-term service</b> life of bacteria; Reduction in compressive strength; <b>Cost???</b>	<b>Affect the workability &amp; strength of concrete</b> ; SH efficiency

#### 2.3.4 Cement particle approach

In this approach, crack self-healing capability of concrete is mainly contributed by the cement hydration and calcite precipitation as can be seen in *Figure 2.27*. One of the advantages of this approach is “No “strange” material is added to concrete” (*Breugel, 2007*).

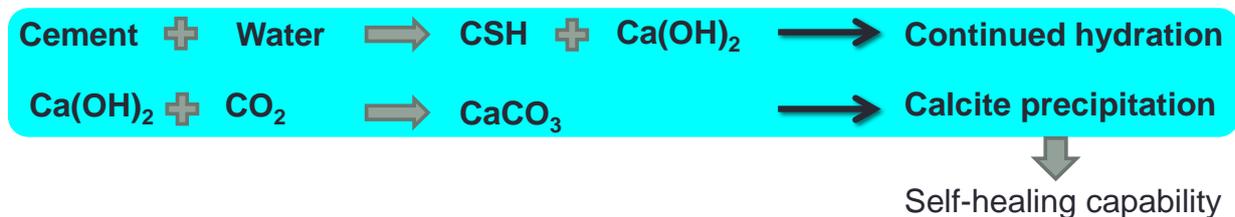


Figure 2.27 Self-healing capability of concrete using cement particle approach

(a) Dissoluble encapsulated cement (or special additives) particles (Koenders, etc.)

This project is still now under development led by *Koenders* at the Delft University of Technology. According to author, “The basic idea is to blend a regular cement with a predefined fraction of dissoluble encapsulated cement particles (DEP)” as seen in *Figure 2.28* (source: <http://selfhealingconcrete.blogspot.jp/p/encapsulated-cement.html>). In that

research, regular cement is coated by a viscous agent acting as a membrane during mixing concrete (high pH environment). Then the coating membrane will be dissolved when contacting with water through the concrete's crack that activating the reaction of the encapsulated cement or other additives.

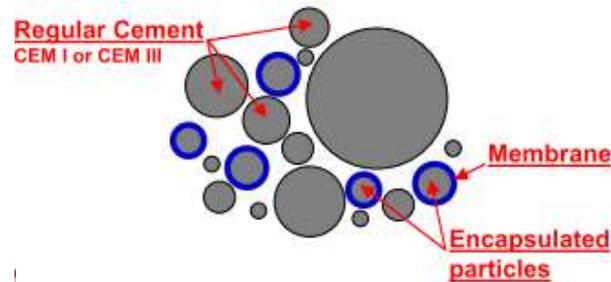


Figure 2.27 Self-healing approach using dissoluble encapsulated particle

*(b) Self-healing granules produced by a conventional granulation technique (Koide & Morita, 2010)*

*Koide & Morita (2010)* proposed to use self-healing granules as partial sand replacement to overcome the obstacles caused by self-healing powder type approach (proposed by Ahn, 2008). The granules ranging from 0.3-5.0mm were produced by applying a conventional granulation technique, a common approach for polymer material or used in food and pharmaceutical products (*Figure 2.28*).

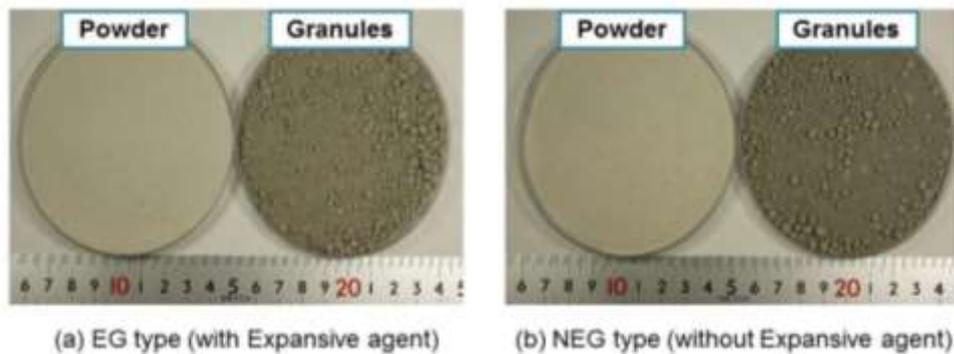


Figure 2.28 Pharmaceutical granulator & technique

And self-healing materials in their study were mainly prepared based on the basic design concept of self-healing material proposed by *Ahn, 2008*. *Table 2.4* shows the material components of granules and the samples can be seen in

*Figure 2.29.***Table 2.4 Self-healing components in percentage**

Sample	Self-healing agent			Binder		*PVA fiber (kg/m <sup>3</sup> )
	Expansive agent	Geo-materials (A, B type)	Chemical additives	Low-heat portland cement	Water + Ethanol	
EG	O	O	O	42	18	0.08
	42					
NEG	-	O	O	49	18	0.08
	33					

**Figure 2.29 Samples of two types of powders and granules**

Even though this approach is a preferable concept to introduce self-healing property to the concrete, further improvement of self-healing performance should be done and due to its high cost that restrains the commercial-based application.

In summary, there is a variety of self-healing approaches nowadays. However, most of them are under development in the laboratory. Moreover, most of researches are focused on the time-dependent property of water flowing through a crack in concrete. It is widely accepted that the flow rate of water through a crack in concrete is a very complicated phenomenon, especially in self-healing concrete, such as various types of materials used, exposure conditions, experiment set-ups, etc. Further knowledge on water flow in cracked self-healing concrete should be obtained to improve the healing performance of self-healing concrete.

Among out of the most important issues are the different performances and exposure conditions between laboratory test and real life; another problem is the feasibility to apply these approaches to the construction industry with the consideration of the manufacturing cost and the complication.

With its advantages and potential to improve the performance through selecting technique, ingredient and dosage, self-healing granules of cement compound and some specific self-healing additives (powder) shows a promising approach to introduce a crack-healing capability to concrete that satisfies the requirements of construction industry.

# CHAPTER 3

## 3-APPLICATION

### CONCEPT OF CRACK

### SELF-HEALING

### CONCRETE USING

### GRANULES & METHODOLOGY

### 3.1 APPLICATION CONCEPT OF CRACK SELF-HEALING CONCRETE USING SELF-HEALING GRANULES SUBJECTED TO WATER FLOW

The basic application concept for crack healing phenomenon in concrete using granules is developed from the autonomic healing concept in polymer composites as seen in *Figure 3.1* (White et al. 2001). Cracking is used as a trigger for releasing the healing agents into crack surface.

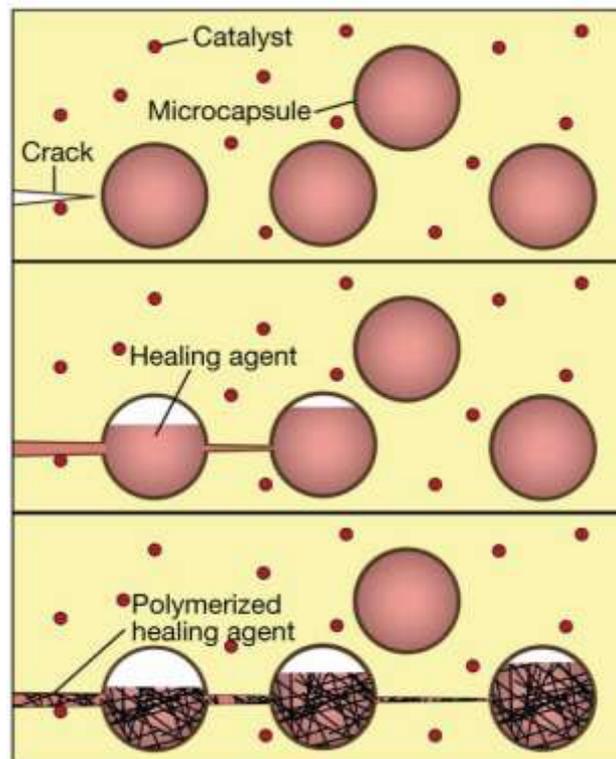


Figure 3.1 Concept of autonomic healing in polymer composites

(White 2001)

In this study, self-healing granules are manufactured in advance and cured in plastic boxes or bags for a period of time. And then granules are added to concrete mixture just before casting to reduce the possibility of being broken under the impact of aggregates (*Figure 3.2a*). When crack occurs in concrete and penetrates the embedded granules, which are distributed uniformly in the matrix, the inner materials will be dissipated into crack surface under the effect of water flow (*Figure 3.2b*).

Gradually, the crack will be healed and the water leakage is stopped by the formation of new products from reactions between self-healing materials and flowing water or other hydration products (*Figure 3.2c*). It is thought that crack-healing capability of concrete is contributed by the combined effects of hydration/pozzolanic reactions; calcite precipitation; and the effect of swelling/expansion of cementitious matrix

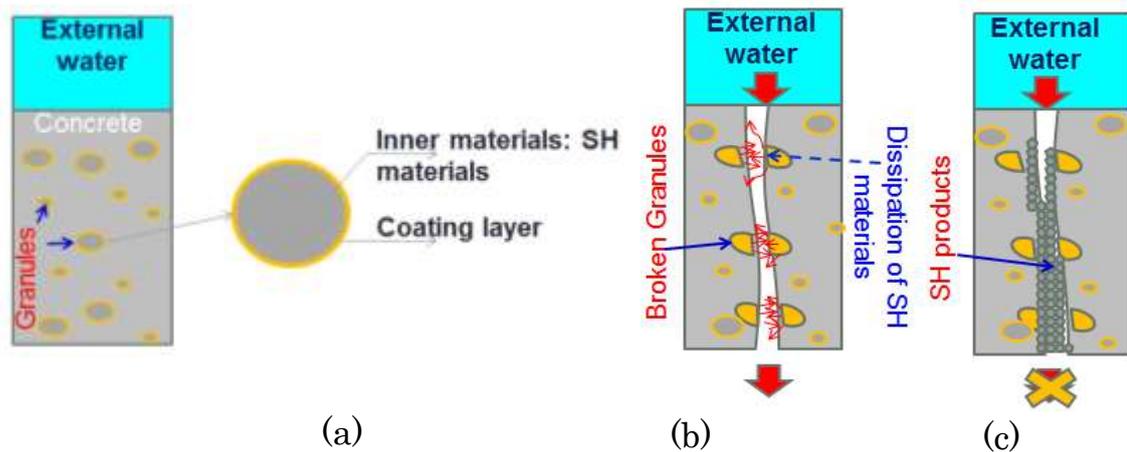


Figure 3.2 Crack self-healing concept in concrete using granules

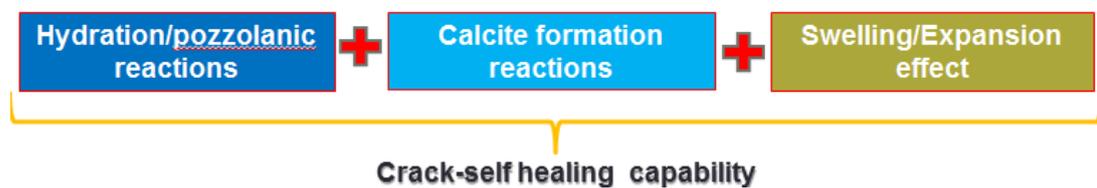


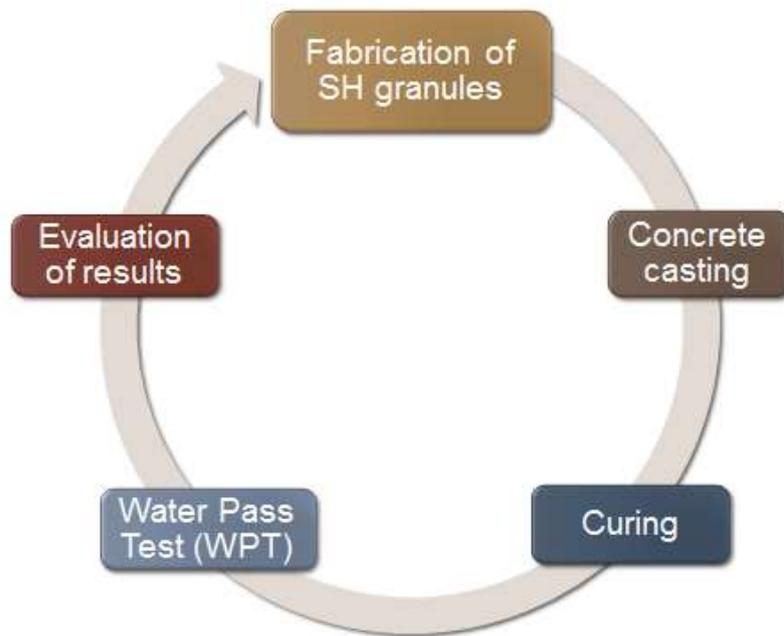
Figure 3.3 Crack self-healing capability

### 3.2 METHODOLOGY & EXPERIMENTAL PROCESS

Based on the basic concept mentioned above, several trials of concretes incorporating various types of granules in self-healing ingredients were cast and investigated for crack-healing capability. Experimental process and methods in this study can be seen in *Figure 3.4*. Briefly, the process can be divided into five steps and details of each step are presented as following:

- Step 1: Fabrication of self-healing granules
- Step 2: Casting concrete incorporating granules

- Step 3: Curing concrete specimens
- Step 4: Conducting water pass test through a penetrated crack
- Step 5: Analyzing/ evaluating the obtained results and giving feedbacks for next trials



**Figure 3.4 Experimental process**

### **3.2.1 Step 1- Fabrication of self-healing granules**

#### **a) Self-healing materials**

The basic idea is self-healing materials and self-healing products should be compatible with concrete matrix. In this research, the designed inner materials of granules are composed of Portland cements/Pozzolans, self-healing additives and liquid, in which Cement/Pozzolan is the base, further improved by self-healing additives and liquid is used to agglomerate the inner materials (*Figure 3.5*). And coating layer is mainly composed of Portland cement compound.

Cement/Pozzolan compounds are mainly contributed to crack self-healing

capability via hydration/pozzolanic reactions. Various types and different ratios of Portland cements and supplementary cementitious materials (SCM), such as fly ash, blast furnace slag, etc., were used.

Self-healing additives used in this study were mainly based on the basic design concept of self-healing materials proposed by *Ahn (2008)*. Various types of granules with different combination of additives were prepared. Depends on its compositions and combination, it is expected that the individual or combined effects of swelling, expansion and precipitation agents may bring a further improvement in healing ability of concrete.

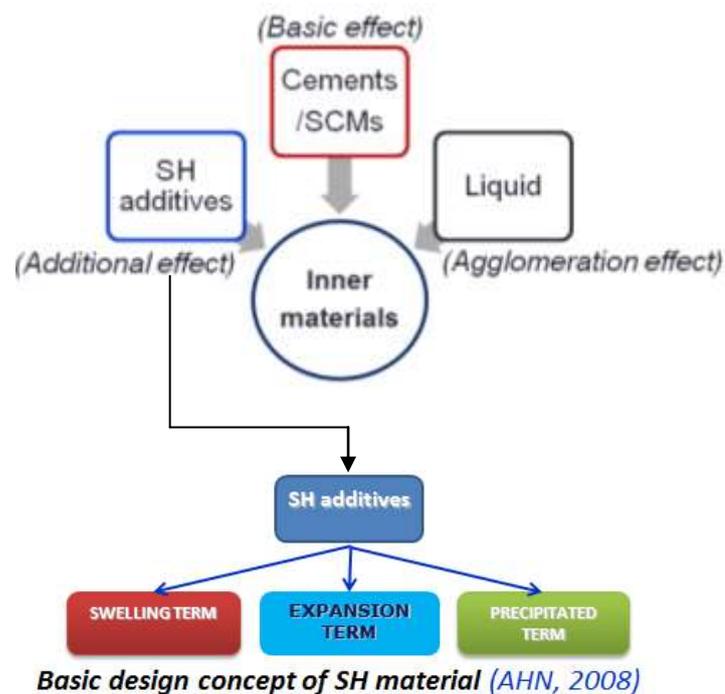


Figure 3.5 Composition of inner materials

b) Granulation process

In this research the granulation technique to make self-healing granules having semi-capsulation effect was developed based on the conventional granulation technique used by *Koide & Morita (2010)*. As mentioned above, this capsulation method commonly uses in food/medicine industry or polymer material (*Figure 3.6*). In case of capsule, the inner materials

## APPLICATION CONCEPT OF CRACK SELF-HEALING CONCRETE USING GRANULES & METHODOLOGY

are commonly protected by an organic layer while the inner of proposed granule is coated by cement compounds, containing self-healing materials (*Figure 3.7*). 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. Due to this feature, it was termed as self-healing granules having “semi-capsulation” effect.

Even though the granulation technique developed by Koide showed promise in the manufacture of granules, its wide application in practice was constrained by a very high initial cost (approximately 10 times higher than using normal concrete) and further improvement in self-healing performance should be done.



Figure 3.6 Conventional granulation technique developed by Koide & Morita

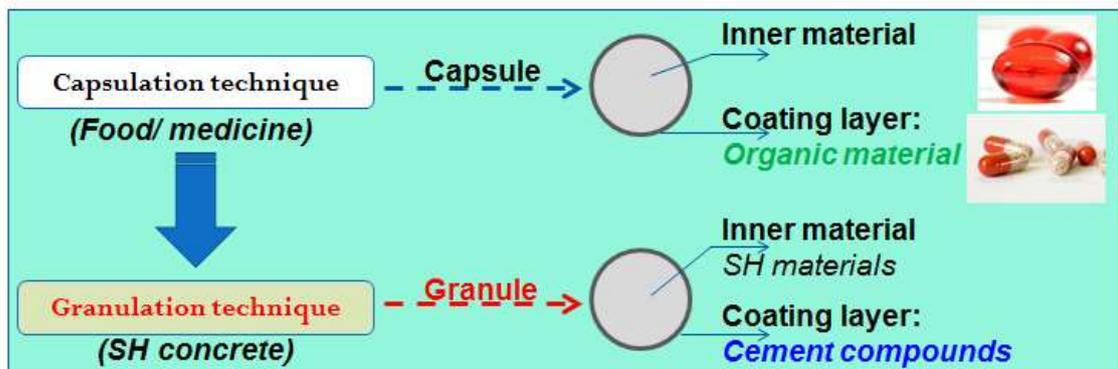


Figure 3.7 Capsules vs Granules having semi-capsulation effect

With the purpose of broadly applying this approach in construction

industry, special considerations of the cost and required equipment for the process should be concerned.

To reduce the cost of manufacture, granules were made by trying to use some typical mixers in laboratory (i.e. roller mixer or mortar mixer). Through this process, the initial condition of self-healing materials, mainly in form of powder, was changed to granule type of several millimeters. The principle process is as follows: firstly the inner granule manufacture was done, and then a coating layer built-up is applied (*Figure 3.8*).

After granulation, the granules were cured in plastic bags or boxes to prevent the loss of moisture so that the formation of coating layer was ensured.



**Figure 3.8 Principle of proposed granulation technique**

### **3.2.2 Step 2- Concrete casting**

After curing for a period of time, granules were added to concrete mixer to cast the concrete specimens (*Figure 3.9*). In this experiment, self-healing granules were used as partial sand replacement at the dosage of 70 kg/m<sup>3</sup> concrete.

Typically, each trial has:

- + 9 cylindrical specimens of D100xH200 mm for the compression

## APPLICATION CONCEPT OF CRACK SELF-HEALING CONCRETE USING GRANULES & METHODOLOGY

tests at 7, 28 & 91 days after casting.

+ 10 cylindrical specimens of D100xH200 mm with notches for the water pass tests (*Figure 3.10*).

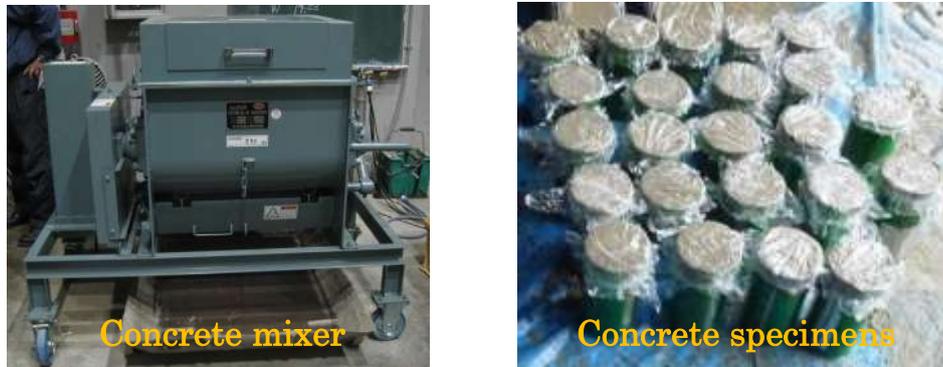


Figure 3.9 Casting self-healing concrete specimens

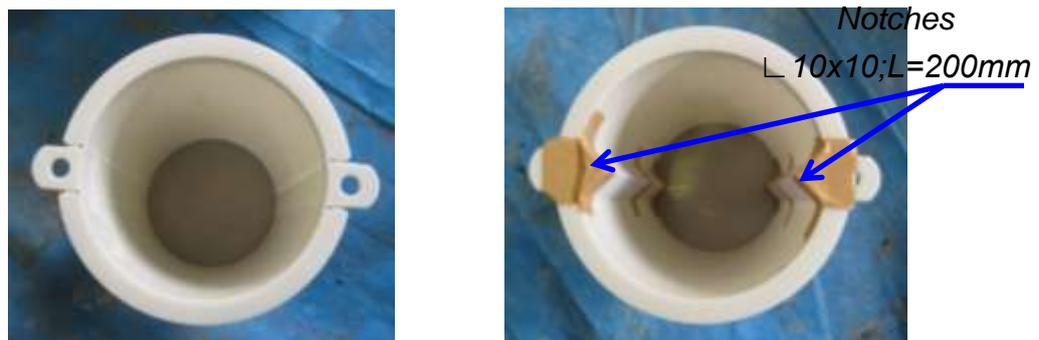


Figure 3.10 Cylindrical mold for compression test (left)  
& water pass test (right)

### 3.2.3 Step 3- Concrete curing conditions

After casting, all concrete specimens were cured under three different curing regimes to investigate the healing capability of concrete at young age, in mature concrete (long-term) (*Figure 3.11a*), and under accelerated hydration of cement base (*Figure 3.11b*), as can be seen in *Table 3.1*.

**Table 3.1 Curing regimes for concrete specimens**

Curing regime	Temperature [°C]	RH [%]	Duration [days]	Condition
1	20	60	14-21	Young concrete
2	20	60	90-180	Long-term
3	40	Water	28	Accelerated hydration of cement base



**Figure 3.11 Sealed curing in room condition and hot water of 40°C curing**

#### **3.2.4 Step 4- Water pass test**

In this study, the water pass test, described by *Morita (2010)*, was used to quantitatively evaluate the self-healing performance in terms of reducing water leakage and sealing cracks with time.

After curing, a penetrating crack would be induced to concrete specimen by applying a splitting load (*Figure 3.12a*). Broken particles by the splitting test would be removed by using a brush. A crack with target width would be formed by inserting Teflon sheet (*Figure 3.12b*) and binding two halves of specimens by stainless clamps (*Figure 3.12c*). 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 3.12d*). 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

APPLICATION CONCEPT OF CRACK SELF-HEALING CONCRETE USING GRANULES & METHODOLOGY

attaching a PVC pipe at the top of specimen and supplying tap water (Figure 3.12e). Then the water pass test, in which a constant water flow through a crack under pressure of approximately 8cm water head, would be performed periodically until 42 or 56 days exposure (Figure 3.12f).

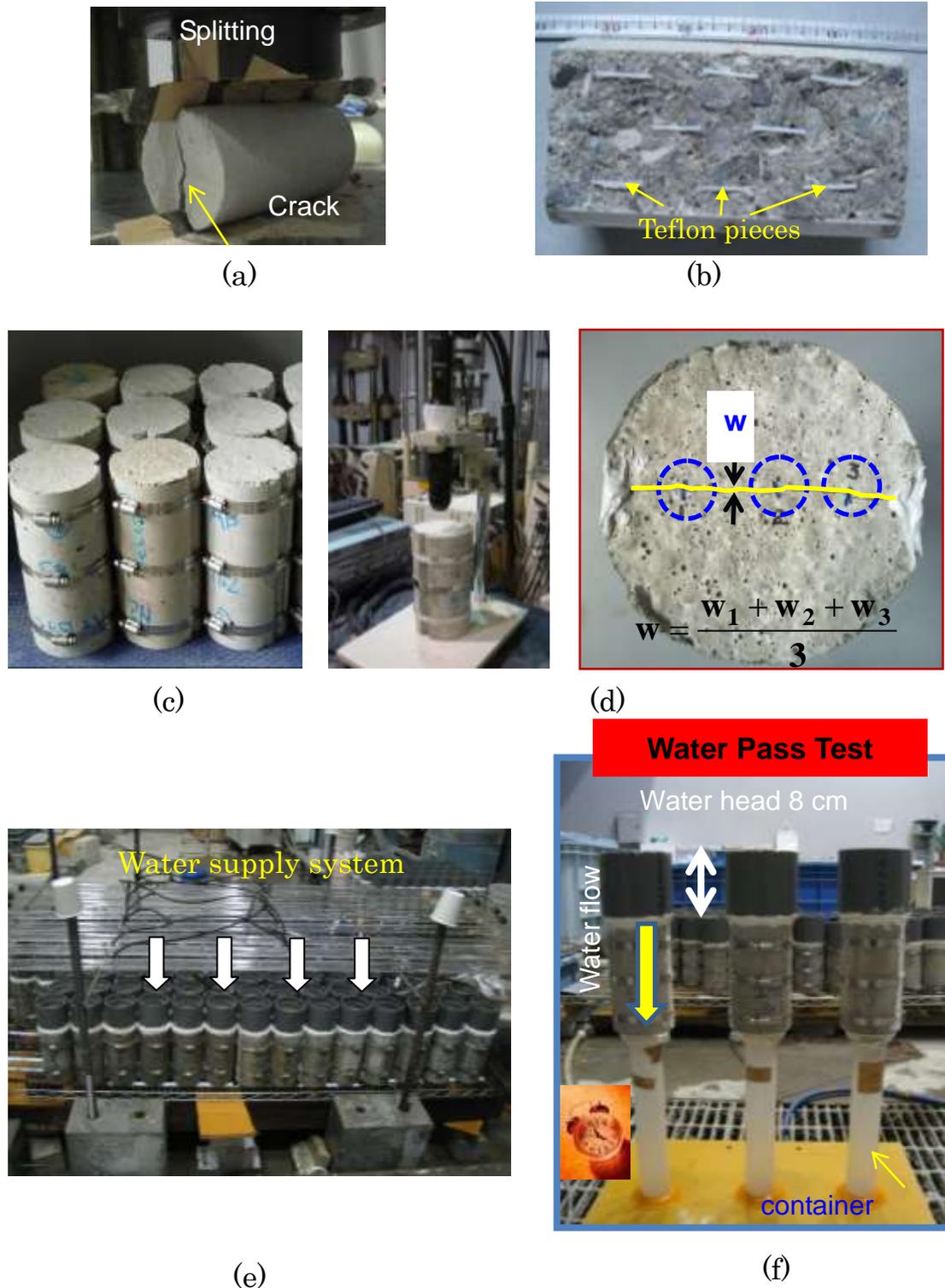


Figure 3.12 (a) Inducing a penetrating crack; (b) Arrangement of

**Teflon sheet on the crack surface; (c) Binding specimen by clamps;  
(d) Measure crack width by Microscope; (e) Continuous water  
supply; (f) Water Pass Test**

The expected results were the water leakage, the pH value of water passing through the crack over time. Furthermore, optical microscopy and digital image were conducted to assess the degree of self-healing at crack and the X-ray Fluorescence (XRF) and Thermo Gravimetry - Differential Thermal Analysis (TG-DTA) were also performed to analyze the chemical and mineral components of healing product precipitated at the crack.

### **3.2.5 Step 5- Evaluation of obtained results and feedback**

Based on the results obtained from the water pass test, such as reduction rate of water flow, crack-closing effect and chemical analysis, the self-healing capability of trial concrete will be evaluated and feedback also will be drawn for improving performance in next trials.

Water permeability is of paramount importance on the durability and functionality of concrete structures. In this study, 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 cracked concrete, the water flow rate was measured and calculated by formula (1), on the specific days until 42 or 56 days subjected to continuous water flow.

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

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

in this test  $t=300\text{sec}$ .

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

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

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

In general, the whole experimental process in this research is tentatively divided into three stages as seen in *Figure 3.13*.

In the first stage, many trials of granules of cement/pozzolan & self-healing additives and self-healing concretes were fabricated and tested in the laboratory. There was an effort to manufacture granules of powder material by using a typical mortar/concrete mixer in the lab. The following tests will be done, such as the workability of fresh concrete, the compressive strength and crack healing capability. At the end of this stage, the requirements and basic design concepts of granules having semi-capsulation effect for powder material will be proposed.

Based on the requirements and basic concepts of granules having semi-capsulation effect, several trials to enhance the granulation technique with cement/pozzolan material were performed in the second stage. Strategies to select the functional effective ingredients and to improve the granulation technique were also proposed.

The third stage is the field trial. The purpose of this stage is to investigate the applicability and the real healing performance of self-healing concrete incorporating granules, which were designed based on the basic design concepts, in real structures- a portion of slab mock-up. Concrete will be produced at the ready-mixed concrete plant and transported to construction site by a truck. Prefabricated self-healing

## APPLICATION CONCEPT OF CRACK SELF-HEALING CONCRETE USING GRANULES & METHODOLOGY

granules will be added to the ready-mixed concrete truck just before casting.

### **Stage 1:**

#### **Trials to develop SH granules with cement/pozzolan & swelling/expansion/precipitation materials**

- 1) TRY TO MAKE GRANULES OF POWDER MATERIAL BY USING A TYPICAL MORTAR/CONCRETE MIXER IN THE LABORATORY
- 2) PROPOSE REQUIREMENTS & BASIC DESIGN CONCEPTS OF GRANULE HAVING SEMI-CAPSULATION EFFECT FOR POWDER MATERIAL

### **Stage 2:**

#### **Trials to enhance the granulation technique with cement/pozzolan**

- 1) STRATEGY TO SELECT THE FUNCTIONAL EFFECTIVE INGREDIENTS
- 2) STRATEGY TO IMPROVE THE GRANULATION TECHNIQUE

### **Stage 3:**

#### **Field trial-application in slab mock-up**

**Figure 3.13 Experimental plan**

# CHAPTER 4

## 4 TRIALS TO DEVELOP SH GRANULES OF CEMENT/POZZOLAN AND SELF-HEALING ADDITIVES

This chapter showed the study on the feasibility of manufacturing of granules having semi-capsulation effect by a typical mortar/concrete mixer in laboratory (*Figure 4.1-above*) and their influences of adding granules to concrete mixture as partial sand replacement on concrete properties, such as the workability of fresh concrete, compressive strength of hardened concrete and crack healing capability (*Figure 4.1-below*).

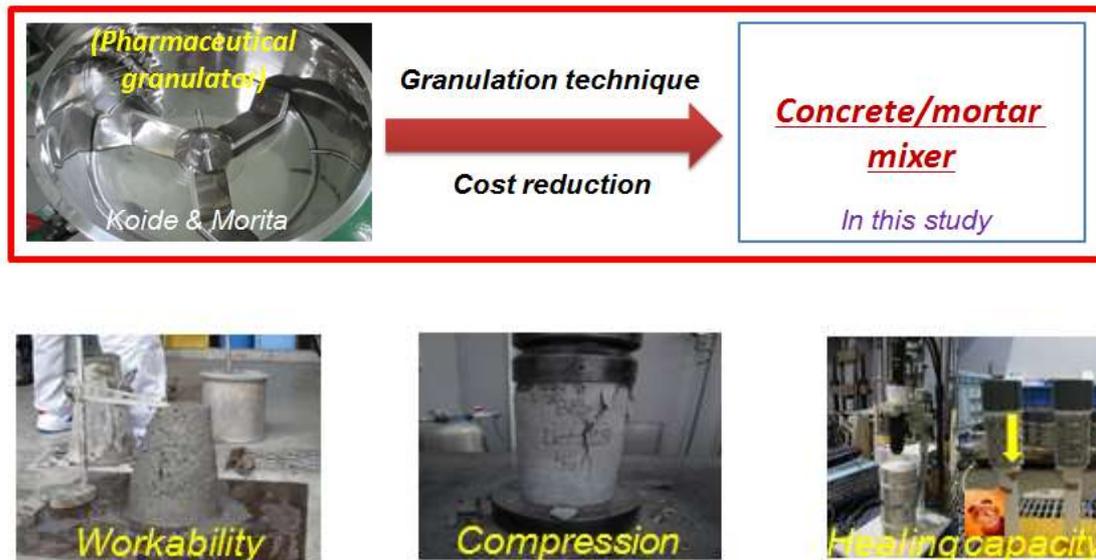


Figure 4.1 Objectives of this chapter

#### 4.1 INGREDIENTS OF GRANULES

Based on the design concept of crack-healing capability (*see in 3.1*), several trials of granules were manufactured with different ingredients of self-healing materials and also different granulation techniques. Typically, granules used in this trial test were classified into three groups and the list of number of trials in this stage can be seen in *Table 4.1*.

Table 4.1 Three groups and list of number of trial self-healing granules

Group	Main inner ingredients	No. of trials
1	Portland cement(s) + self-healing additives	1
2	Pozzolan(s)+ self-healing additives	8
3	Portland cement +Pozzolan(s) + self-healing additives	7

## 4.2 GRANULATION TECHNIQUE (see 3.2.1b)

In general, granulation process were often divided into two steps.

Firstly, main inner ingredients in powder type were mixed together in a mixer (several typical mortar/concrete mixers were tried) and then a certain amount of liquid was supplied to make the powder agglomerate by spraying or pouring gradually.

Secondly, a coating layer of cement compounds would be introduced to prevent further reactions between inner materials and external water. Depending on the condition of inner materials, additional liquid was determined.

After manufacture, granules were cured in a plastic bag (or box) for a few days to maintain sufficient moisture for hardening of coating layer (*Figure 4.2*).



**Figure 4.2 Curing condition of pre-fabricated granules**

## 4.3 MIX PROPORTIONS OF CONCRETES

With an aim to introduce self-healing capability to concrete with normally used ranges of water-to-cement ratio, two mix proportions of concrete were designed with different required workability property of fresh concrete (especially the slump) and different strength design as seen in *Table 4.2a & b*. And, in this trial self-healing granules (SH) were added to concrete mixture during casting concrete as a partial replacement of sand (S) at two different dosages: 70 kg/m<sup>3</sup> concrete for Mix 1 and 100kg/m<sup>3</sup> concrete for Mix 2. In the case of design 1 concrete (Mix1), only self-healing trial was cast (without casting the normal concrete as a controlled one for Mix1); in addition, concrete were produced at a ready-mixed concrete plant and then transported to construction site. Then self-healing granules were added to ready-mixed

concrete truck just before casting to reduce the possibility of broken granules.

**Table 4.2a Mix proportion of design 1 concrete (Mix1)**

Type of concretes	W/C [%]	s/a [%]	G <sub>max</sub> [mm]	Air [%]	kg/m <sup>3</sup>				
					W	C	SH	S	G
Plain	49.6	51.3	20	4.5	175	353	-	900	869
SH concrete (SH-Mix1)	49.6		20	4.5	175	353	70	830	869

**Table 4.2b Mix proportion of design 2 concrete (Mix2)**

Type of concretes	W/C [%]	s/a [%]	G <sub>max</sub> [mm]	Air [%]	kg/m <sup>3</sup>				
					W	C	SH	S	G
Plain (Plain-Mix2)	49	44.5	20	4.5	171	349	-	802	953
SH concrete (SH-Mix2)	49	-	20	4.5	171	349	100	702	953

#### 4.4 CONCRETE CURING CONDITIONS & PREPARATION OF SPECIMEN FOR THE WATER PASS TEST

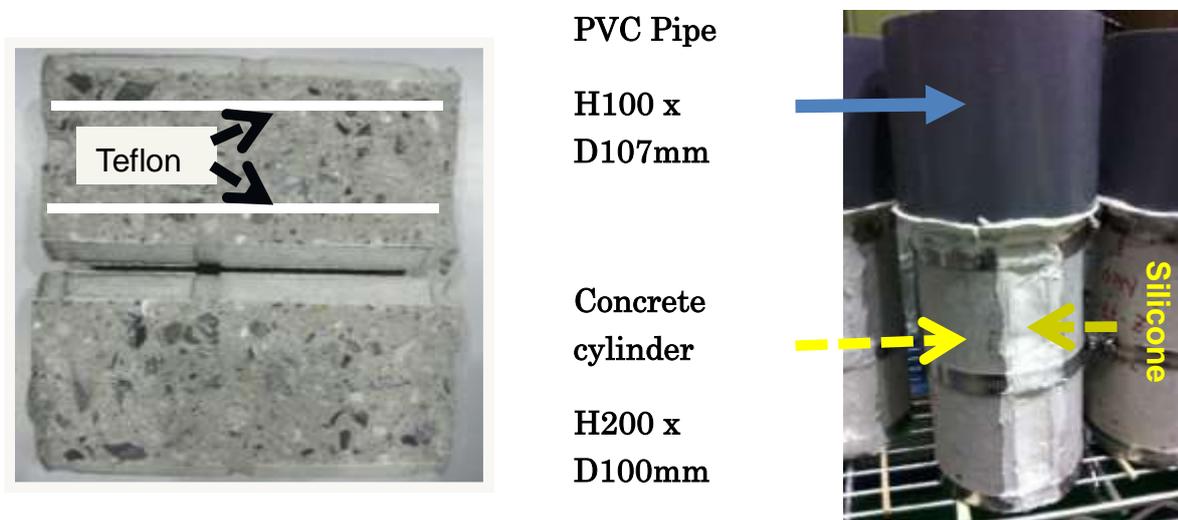
After casting, concrete specimens were cured under different conditions for two mix proportions of concrete. The curing condition applied for Mix 1 concretes was considered as an accelerated hydration condition of cement base in concrete whereas Mix 2 concrete specimens were cured in sealed condition as normally used in practice. The period of curing was about one month, and then a single penetrated crack would be induced (*Table 4.3*).

In this stage, the crack width was also controlled in range of 0.2-0.4 mm by

inserting Teflon sheets and combining two halves of specimen by steel clamps. However, the arrangement of Teflon was different from the one shown in *Figure 3.12b*. Two Teflon strips were set along the height of specimen as seen in *Figure 4.3a*. *Figure 4.3b* showed a typical concrete specimen that was ready for the water pass test.

**Table 4.3 Curing & cracking history**

<b>Trials</b>	<b>Curing conditions</b>	<b>Curing period</b>	<b>Cracking</b>	<b>Start water pass test</b>
SH-Mix1	Sealed in air T20°C,RH60%	7 days	30 <sup>th</sup> day	31 <sup>st</sup> day
	Water T40°C	21 days		
SH-Mix2	Sealed in air T20°C,RH60%	35 days	35 <sup>th</sup> day	42 <sup>nd</sup> day



**Figure 4.3 (a) Arrangement of Teflon strips (b) Typical concrete specimen**

#### 4.5 EXPERIMENTAL RESULTS

Out of several attempts, the best performance trials were chosen and discussed in this part based on the rate of reduction in water flow rate and crack-closing process (*Table 4.4*). The detailed information of those granules

can be seen in *Table 4.5* and *Table 4.6* showed the mixer and sample for each trial.

**Table 4.4 Chosen trials for discussion**

Concrete designation	Granule	Mix proportion
SH-Mix1-1	Group 1	Mix1
Plain-Mix2	Control	Mix2
SH-Mix2-2a/2b/2c/2d	Group 2	
SH-Mix2-3	Group 3	

**Table 4.5 Ingredients of granules**

Designation	Ingredients of granule	Percentage [%]	
		<i>Inner</i>	<i>Coating</i>
SH-Mix1-1 (Cement+SHA)	Portland cement (LC+HC)	37.63	16.13
	SH additives (Water-soluble agent+SHA)	37.63	-
	Liquid (W)	5.38	3.23
SH-Mix2-2a (Pozzolan+ Swelling+additives)	Pozzolans (FA.BFS.SF)	50	-
	SH additives (Swelling)	5.56	2.78 (CaO)
	Liquid (UW+SP)	16.67	-
	Cement compounds	-	25

Designation	Ingredients of granule	Percentage [%]	
		<i>Inner</i>	<i>Coating</i>
SH-Mix2-2b (Pozzolan+ Swelling+ additives)	<b>Pozzolans (FA.BFS.SF')</b>	24.1	-
	SH additives (Swelling.Carbonate. Dolomite)	24.1	4.82 (CaO.Slight acidic powder)
	Liquid (Fatty acid saturated Ethanol solution))	19.28	8.43(UW.SP)
	Cement compounds	-	19.28
SH-Mix2-2c (Pozzolan+ Swelling+additives)	<b>Pozzolans (FA.BFS)</b>	44.19	-
	SH additives (Swelling)	8.18	-
	Liquid (UW.SP)	13.26	9.82
	Cement compounds	-	24.55
SH-Mix2-2d (Pozzolan+ Swelling+additives)	<b>Pozzolan (BFS)</b>	34.63	-
	SH additives (Carbonate.Swelling)	12.99	2.53(Light acidic powder)
	Liquid (UW)	19.05	5.54 (W)
	Cement compounds	-	25.26
SH-Mix2-3 (Pozzolan+ Cement+Swelling +additives)	<b>Pozzolans (FA.BFS.SF')</b>	20.66	-
	<b>Portland cement (cement compounds)</b>	5.97	22.96
	SH additives	23.41	2.3
	Liquid (W+SP+UW)	20.48	4.23(UW)

Table 4.6 Typical mixers & samples of granules

Designation	Mixer	Samples
SH-Mix1-1		
SH-Mix2-2a		
SH-Mix2-2b		
SH-Mix2-2c		
SH-Mix2-2d		

Designation	Mixer	Samples
SH-Mix2-3		

Note: W-tap water; UW: Water-soluble agent solution (Water-soluble agent: Water=1:1); SP-Super-plasticizer; SF-Silica fume; FA: Fly ash type II; BFS-Blast furnace slag; HC-High early strength cement; LC-Low heat cement

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

*Table 4.7* showed the workability of fresh concretes just after discharging from the mixer. It can be seen that the dosage of the super-plasticizer was in the range of (0.6-1.3) % & 1.7 % (by weight of cement) to achieve the designed slump in Mix2 & Mix1 concretes, respectively. Generally, the input of self-healing granules into concrete mix caused a slightly increasing quantity of super-plasticizer compared to that in plain concrete and slump values of self-healing concretes were smaller than that of normal concrete. The possible reasons were the water absorption and further reactions between the coating layer of granules and the mixing water. Based on the obtained results, the proposed granule type can be considered as one of the promising approaches to overcome the drawback of the reduction of workability property in concrete

incorporating self-healing materials in form of powder as mentioned above (*in chapter 2&3*).

**Table 4.7 Workability (slump) of fresh concretes**

Concrete designation	Granule	Designed slump (cm)	Measured slump (cm)	SP (C x %)	AE (C x %)
SH-Mix1-1	Group 1	20.0±3.0	17.0	1.7	
Plain-Mix2	-	12.0±3.0	20.5	0.4	0.03
SH-Mix2-2a	Group 2		12.5	0.6	0.02
SH-Mix2-2b			11.5	1.3	0.04
SH-Mix2-2c			16.5	0.6	0.02
SH-Mix2-2d			17.0	0.6	0.02
SH-Mix2-3			Group 3	14.5	0.6

**4.5.2 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 embedding granules on the hardened concrete’s property. From the *Figure 4.4*, it can be seen that all self-healing concretes showed a satisfaction of the required strengths for both types of design concretes (*the dot line for Mix1 concrete-33N/mm<sup>2</sup>, and the dash line for Mix 2 concrete-30N/mm<sup>2</sup>*). Moreover, it was found that depending on the ingredients of granules and their usages in the granulation process, the differences in strength compared to plain concrete varied 2-13% (at 28 days old) and 3-19% (at 91 days old). However, in general most of differences were less than 10%. Thus, it may be concluded that up to the level of substitution of 70-100kg/1m<sup>3</sup> concrete, the addition of self-healing granules to concrete mix as partial sand replacement had no detrimental effects on the mechanical

property, typically the compressive strength, of matured concrete. However, further improvement in control of strength increase should be done by introducing more effective coating layer.

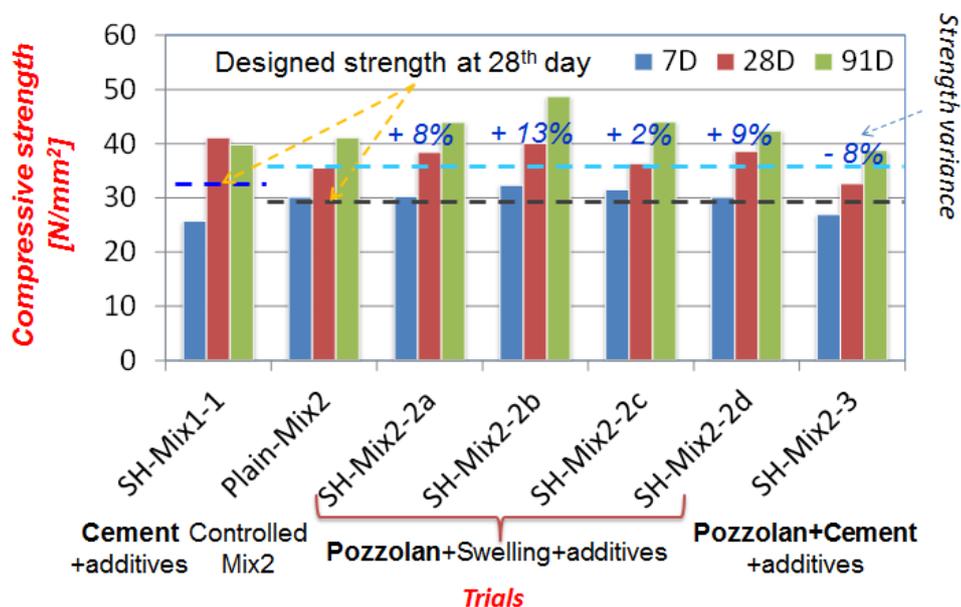


Figure 4.4 Compressive strengths of hardened concretes at 7/28/91 days after casting

### 4.5.3 Change of water flow with time

Changes of flow rate with time in all trials were shown in *Figure 4.5* and *Figure 4.6* showed the relative flow rate versus duration exposed to continuous water supply. The graphs were drawn based on the average values of three specimens for each trial. (*Graphs of water flow rate & crack width in each specimen for each trial can be seen in Appendix 1*).

From these figures, it can be seen that there was similar decreasing tendency of water flow through a crack in the first day with regardless of the types of concrete. The possible reasons were the achievement in complete saturation of the vicinity of crack and further densification of the mortar (*M.D. Lepech at al.*).

It can be seen that in case of control Mix2 concrete, even though the initial flow rate was smallest compared to those of self-healing concretes, 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 controlled trial. On the other hand, with the higher initial flow rate, the water flow through a crack after one day in other self-healing concrete trials 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 of the test.

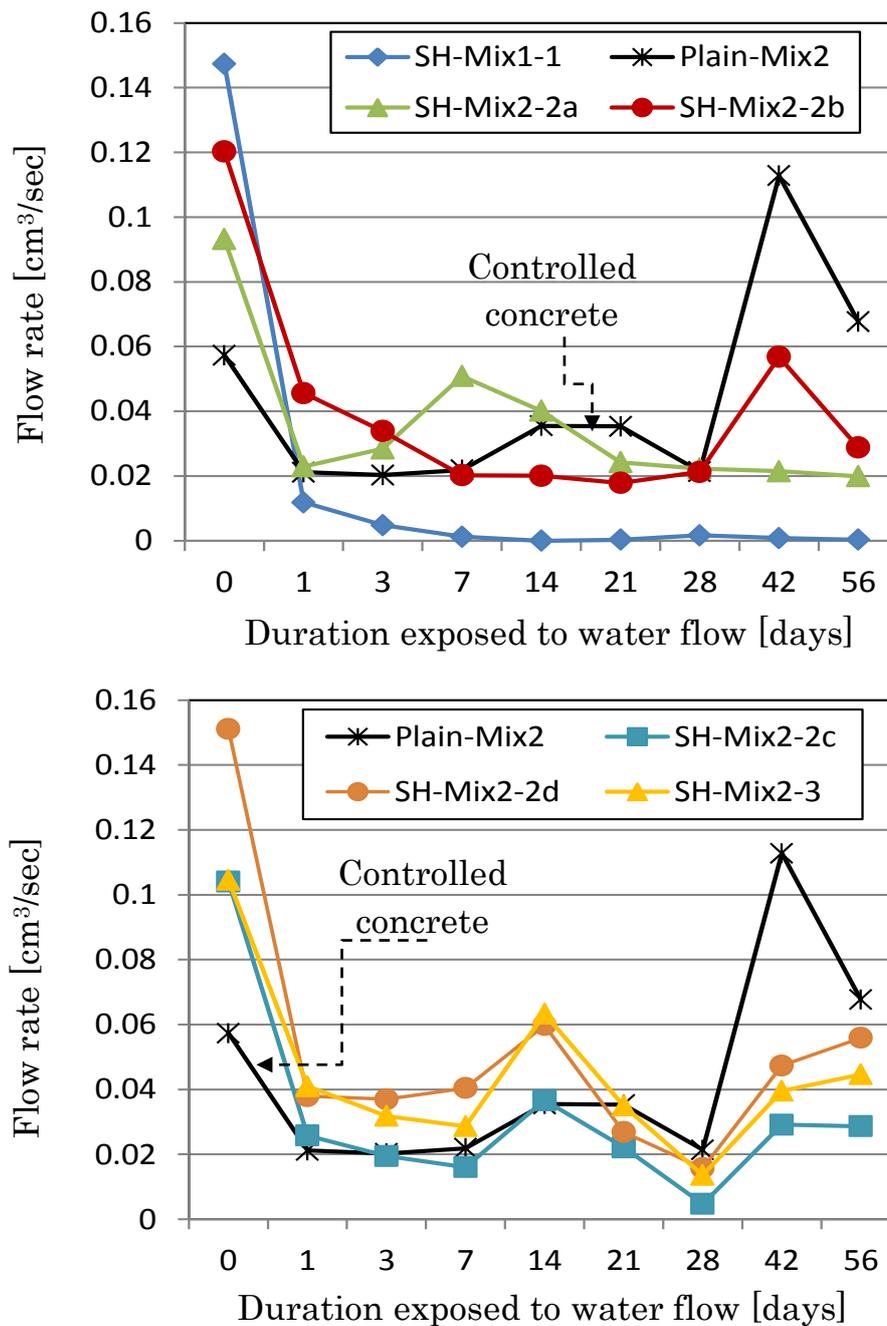


Figure 4.5 Time-dependent water flow rate

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 (just pour water full of the pipe on the top of specimen once a day), the leakage of water through a crack was slightly increased, in ranges of 20-40% of initial value, depending on the types of granules, the initial crack width and flow rate.

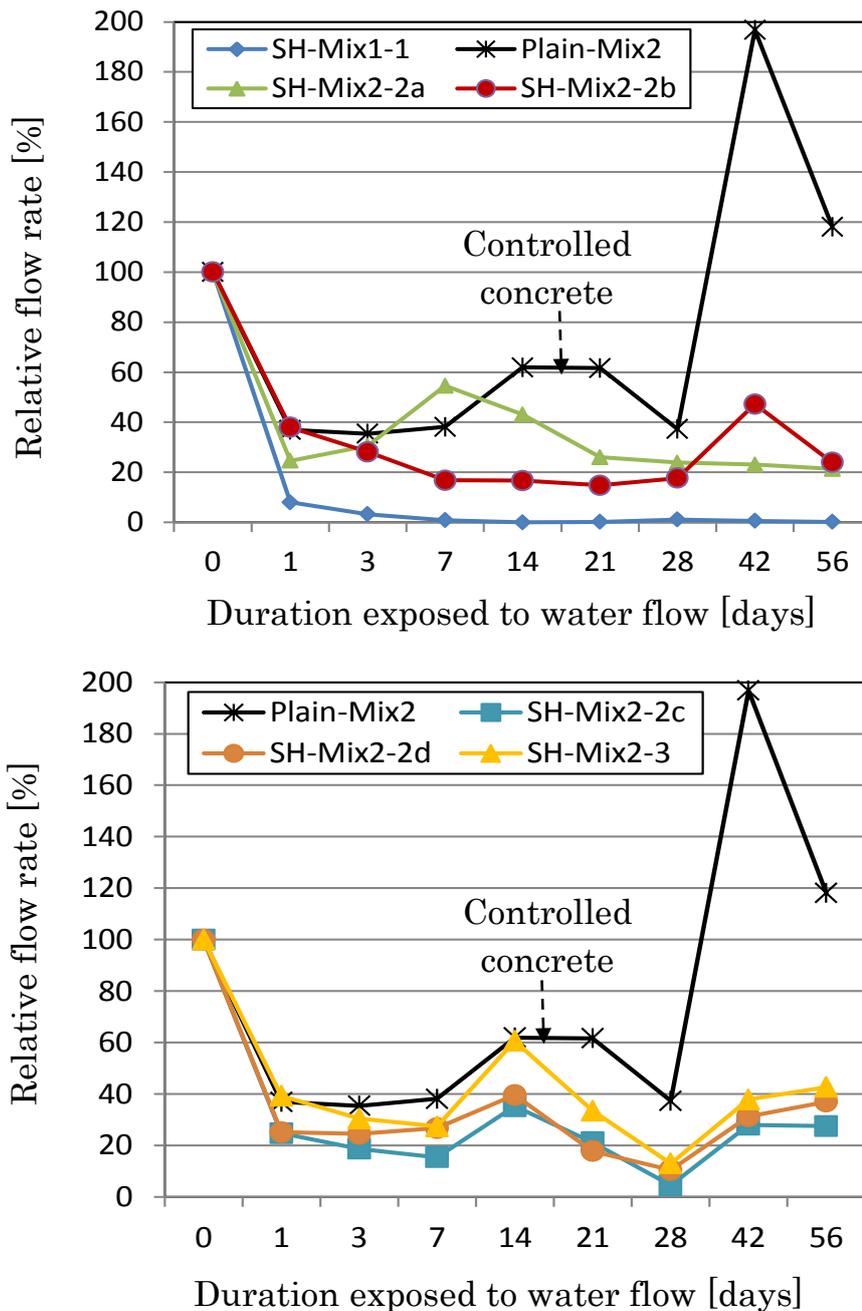


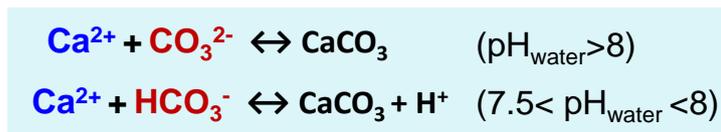
Figure 4.6 Change of relative water flow rate with time

For example, in case of a specimen with higher initial flow rate and crack width, it was observed that water flow rate was more complicated and fluctuated with time. While in case of smaller initial flow rate and crack width, water flow through a crack was more stable.

Out of these self-healing concretes, trial SH-Mix1-1, containing granules of Portland cements and self-healing additives (swelling & expansion & precipitation & water-soluble materials), showed the best performance in term of reduction of water leakage. It can be seen that the water leakage after one day was just about 8% of initial water flow. Then after one week exposure, it was observed that the water flow was almost stopped during the permeability test as clearly seen in *Figure 4.5 and 4.6*

#### 4.5.4 Change in pH value of water flowing through a crack

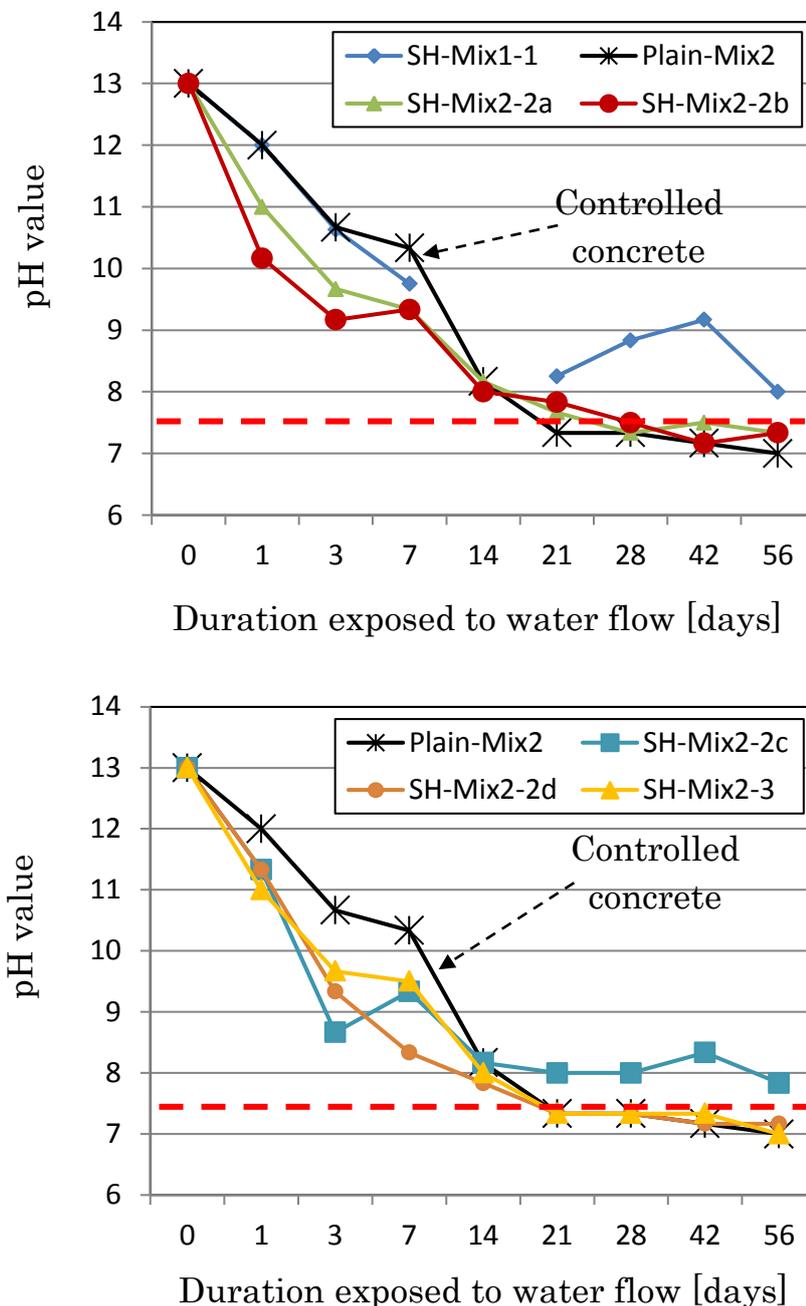
Another parameter also obtained from the water pass test to evaluate the healing process was the pH value of water flowing through the crack. It is well known that calcite formation, mainly depending on the concentration of  $\text{Ca}^{2+}$  and  $\text{CO}_3^{2-}$  ions and pH value of water, is the most important and dominant mechanism for sealing a surface crack (*Figure 4.7*).



**Figure 4.7 Calcite formation reactions**

When water flows through a crack, calcium hydroxide and other hydration products are dissolved or liberated. As a result, both the pH value of water passing through a crack and concentration of dissolved ions will decrease with time at crack. Moreover, with the elapse of time calcite formation will be changed to diffusion-controlled process. As a result, the crack-healing process will take longer time if the crack width is small enough to be healed. The above phenomenon can be observed in case of controlled normal concrete. As seen from *Figure 4.8*, after 21 days exposure 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 4.8 pH level of water flowing through a crack change with time**

On the other hands, in case of self-healing concretes, 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 sources from the diffusion process of self-healing

granules on the crack surface. Under these conditions, the calcite precipitation was facilitated to occur in longer period and provided the crack a higher possibility to be filled. It was clearly observed that in the trials of self-healing concretes which showed the best performance (SH-Mix1-1), the pH of flowing water were maintained high enough to stimulate the calcite precipitation up to until 56 days subjected to water flow as seen in *Figure 4.8*.

#### 4.5.5 Surface crack closing process

*Table 4.8* showed the time-dependent precipitation process of healing products on the surface crack at the bottom of some typical trials after exposed to water flow by digital images analysis. Furthermore, the change of crack width during the testing period was also observed by a microscope (100X magnification) at specific positions with typical crack widths, such as 1<sup>st</sup> mark, 2<sup>nd</sup> mark and 3<sup>rd</sup> mark on the bottom surface of each concrete specimen.

From *Table 4.8*, it can be seen that with a crack, larger than 0.2mm wide, a partial healing of crack was observed in Plain-Mix2 (N2). It was interesting to observe that there was also significant amount of healing products deposited on surface crack in Plain-Mix2 (N1). It was thought that due to small initial flow rate, it promoted the precipitation of calcite or recrystallization of hydration products at the crack mouth. Consequently, larger crack width would be healed and also there was a higher possibility to heal a crack. However, the crack healing process in controlled normal concretes was still uncertain in all circumstances. In other self-healing concrete trials, most of cracks which were in ranges of 0.2-0.3mm wide could completely heal after 56 days exposure. However, when crack width was larger than 0.3mm, only partial healing was achieved, except for Trial SH-Mix1-1, known as the best performance in both reduction of water leakage and crack closing.

It was found that the decrease of crack width or the healing process was mainly observed at the bottom of concrete specimen. The possible reasons were that; firstly when water flowing through a crack, calcium hydroxide or

other hydration products were dissolved and flew along the flow path. Due to this fact, it was believed that the lower part of concrete specimen, the higher concentration of  $\text{Ca}^{2+}$  it was. Secondly, at the crack mouth, especially in this experiment when the bottom of specimen was exposed to the air, there was a possibility of high concentration of dissolved carbonate ions in this area. Finally, the water flow at the bottom part is slower compared to that of upper zone due to the loss of energy along the flow path. As a result, the precipitation of calcite or recrystallization of hydration products at the crack mouth were facilitated or promoted (*Figure 4.9*).

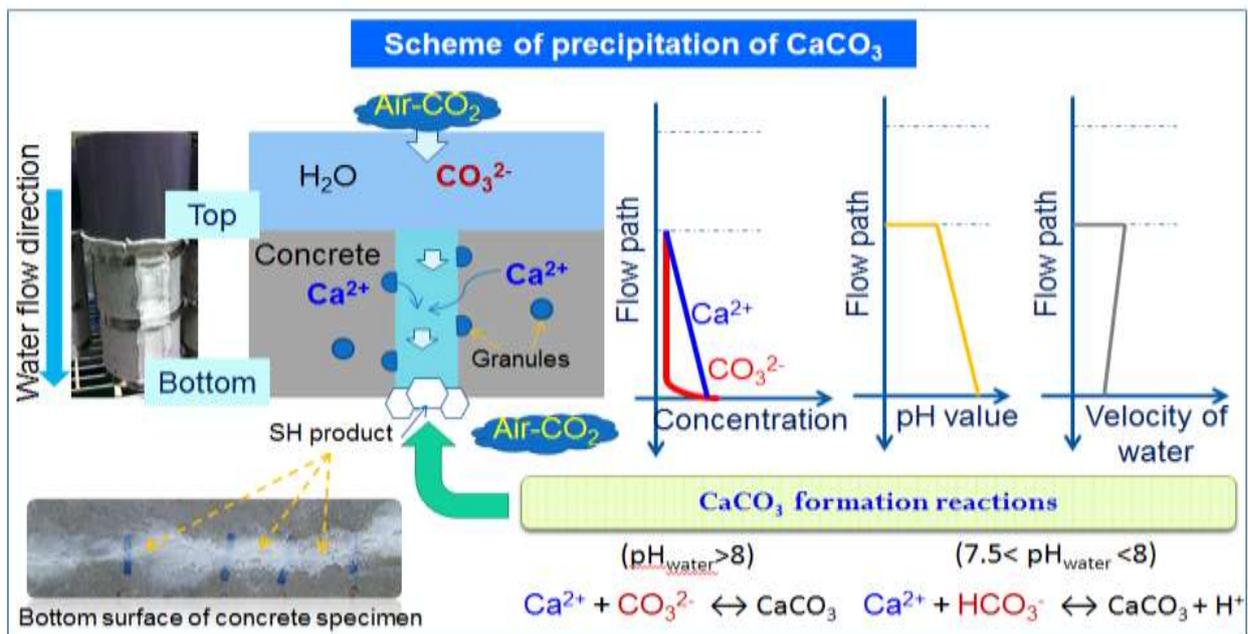
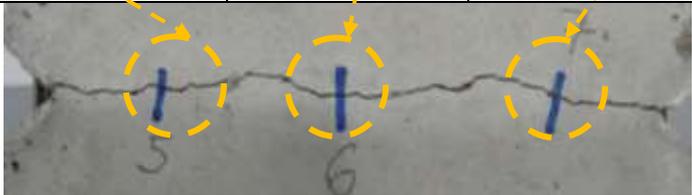
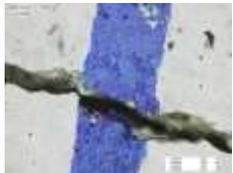
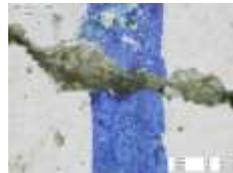


Figure 4.9 Scheme of calcite precipitation at the crack surface.

Table 4.8a Crack closing process in SH-Mix1-1

Concrete designation	Day	Bottom surface			1 <sup>st</sup> Mark	2 <sup>nd</sup> Mark	3 <sup>rd</sup> Mark
		1 <sup>st</sup> Mark	2 <sup>nd</sup> Mark	3 <sup>rd</sup> Mark	CW=0.22mm	CW=0.25mm	CW=0.25mm
SH-Mix1-1 (N1)	0						
	1						
	7						
	28						

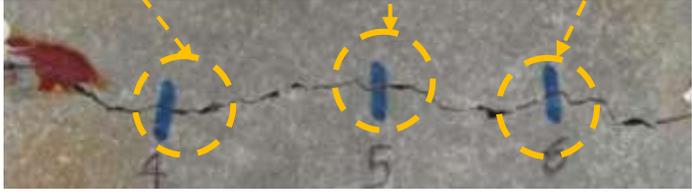
TRIALS TO DEVELOP SH GRANULES OF CEMENT/POZZOLAN & SH ADDITIVES

	56						
Concrete designation	Day	Bottom surface			1 <sup>st</sup> Mark	2 <sup>nd</sup> Mark	3 <sup>rd</sup> Mark
		1 <sup>st</sup> Mark	2 <sup>nd</sup> Mark	3 <sup>rd</sup> Mark	CW=0.37mm	CW=0.33mm	CW=0.44mm
SH-Mix1-1(N2)	0						
	1						
	7						

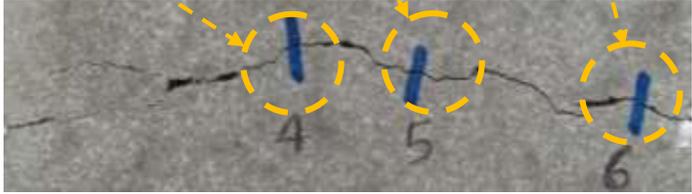
TRIALS TO DEVELOP SH GRANULES OF CEMENT/POZZOLAN & SH ADDITIVES

	28				
	56		-		

Table 4.8b Crack closing process in Plain-Mix2

Concrete designation	Day	Bottom surface			1 <sup>st</sup> Mark	2 <sup>nd</sup> Mark	3 <sup>rd</sup> Mark
		1 <sup>st</sup> Mark	2 <sup>nd</sup> Mark	3 <sup>rd</sup> Mark	CW=0.31mm	CW=0.27mm	CW=0.26mm
Plain-Mix2-(N1)	0						
	1						

TRIALS TO DEVELOP SH GRANULES OF CEMENT/POZZOLAN & SH ADDITIVES

	7						
	28						
	56						
Concrete designation	Day	Bottom surface			1 <sup>st</sup> Mark	2 <sup>nd</sup> Mark	3 <sup>rd</sup> Mark
		1 <sup>st</sup> Mark	2 <sup>nd</sup> Mark	3 <sup>rd</sup> Mark	CW=0.23mm	CW=0.26mm	CW=0.23mm
Plain-Mix2 -(N2)	0						

TRIALS TO DEVELOP SH GRANULES OF CEMENT/POZZOLAN & SH ADDITIVES

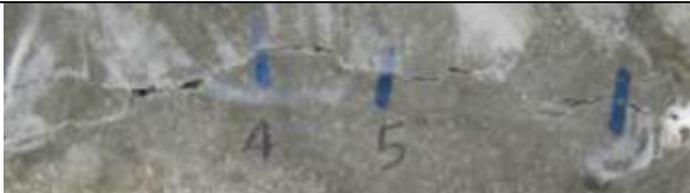
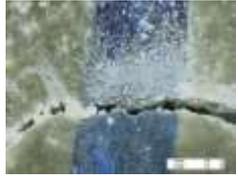
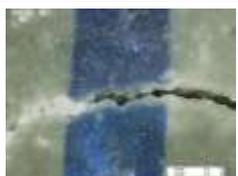
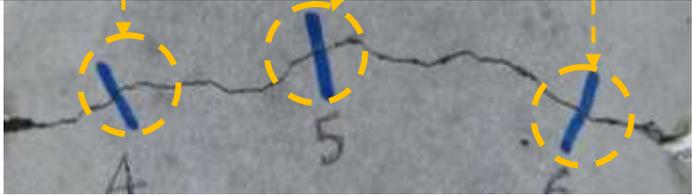
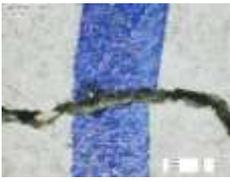
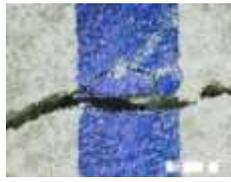
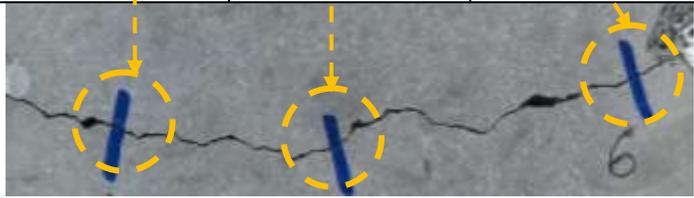
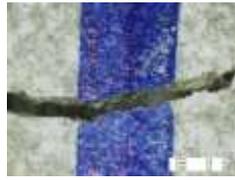
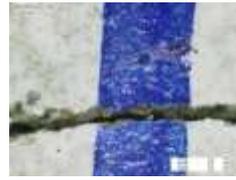
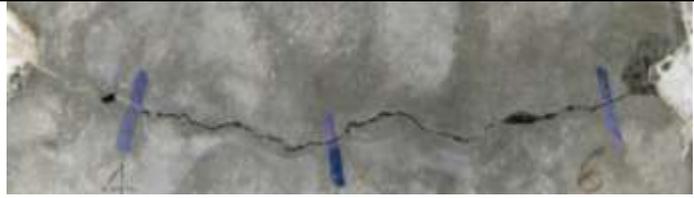
1				
7				
28				
56				

Table 4.8c Crack closing process in SH-Mix2-2b

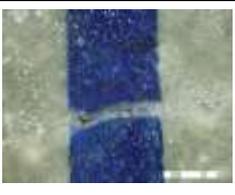
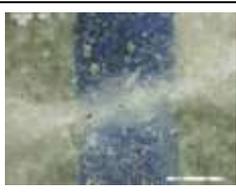
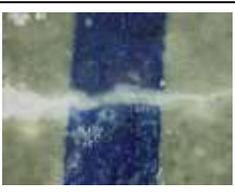
Concrete designation	Day	Bottom surface			1 <sup>st</sup> Mark	2 <sup>nd</sup> Mark	3 <sup>rd</sup> Mark
		1 <sup>st</sup> Mark	2 <sup>nd</sup> Mark	3 <sup>rd</sup> Mark	CW=0.25mm	CW=0.25mm	CW=0.29mm
SH-Mix2-2b(N1)	0						
	1						
	7						
	28						

TRIALS TO DEVELOP SH GRANULES OF CEMENT/POZZOLAN & SH ADDITIVES

	56				
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Concrete designation	Day	Bottom surface			1 <sup>st</sup> Mark	2 <sup>nd</sup> Mark	3 <sup>rd</sup> Mark
		1 <sup>st</sup> Mark	2 <sup>nd</sup> Mark	3 <sup>rd</sup> Mark	CW=0.39mm	CW=0.30mm	CW=0.27mm
SH-Mix2-2b(N2)	0						
	1						
	7						

TRIALS TO DEVELOP SH GRANULES OF CEMENT/POZZOLAN & SH ADDITIVES

	28				
	56				

4.5.6 Chemical/mineral analysis of precipitated products at crack

Table 4.9 Analysis of precipitated products on crack wall along the flow path and bottom surface-SH-Mix1-1

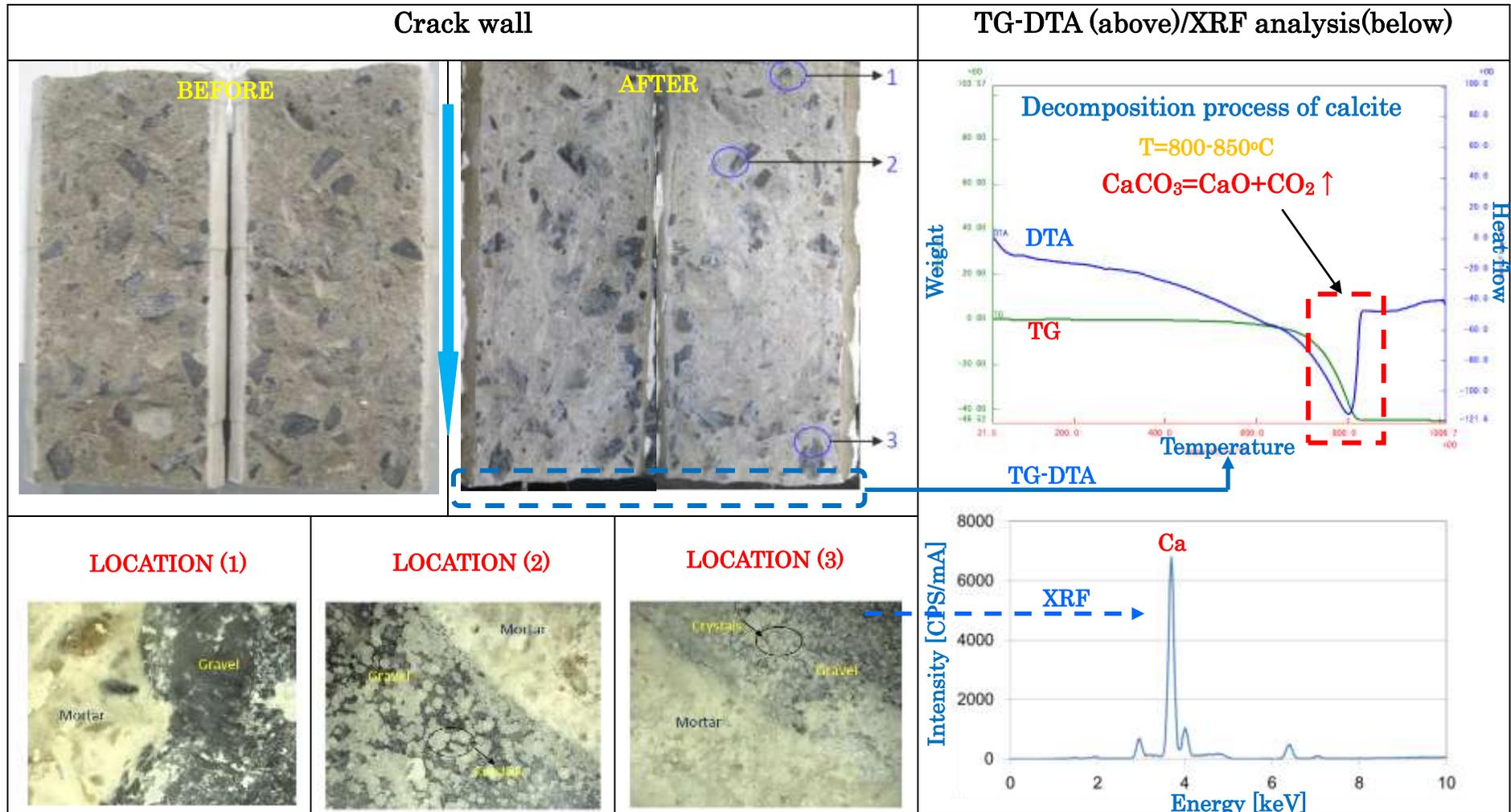


Table 4.10 Analysis of precipitated products on crack wall along the flow path and bottom surface-Plain-Mix2

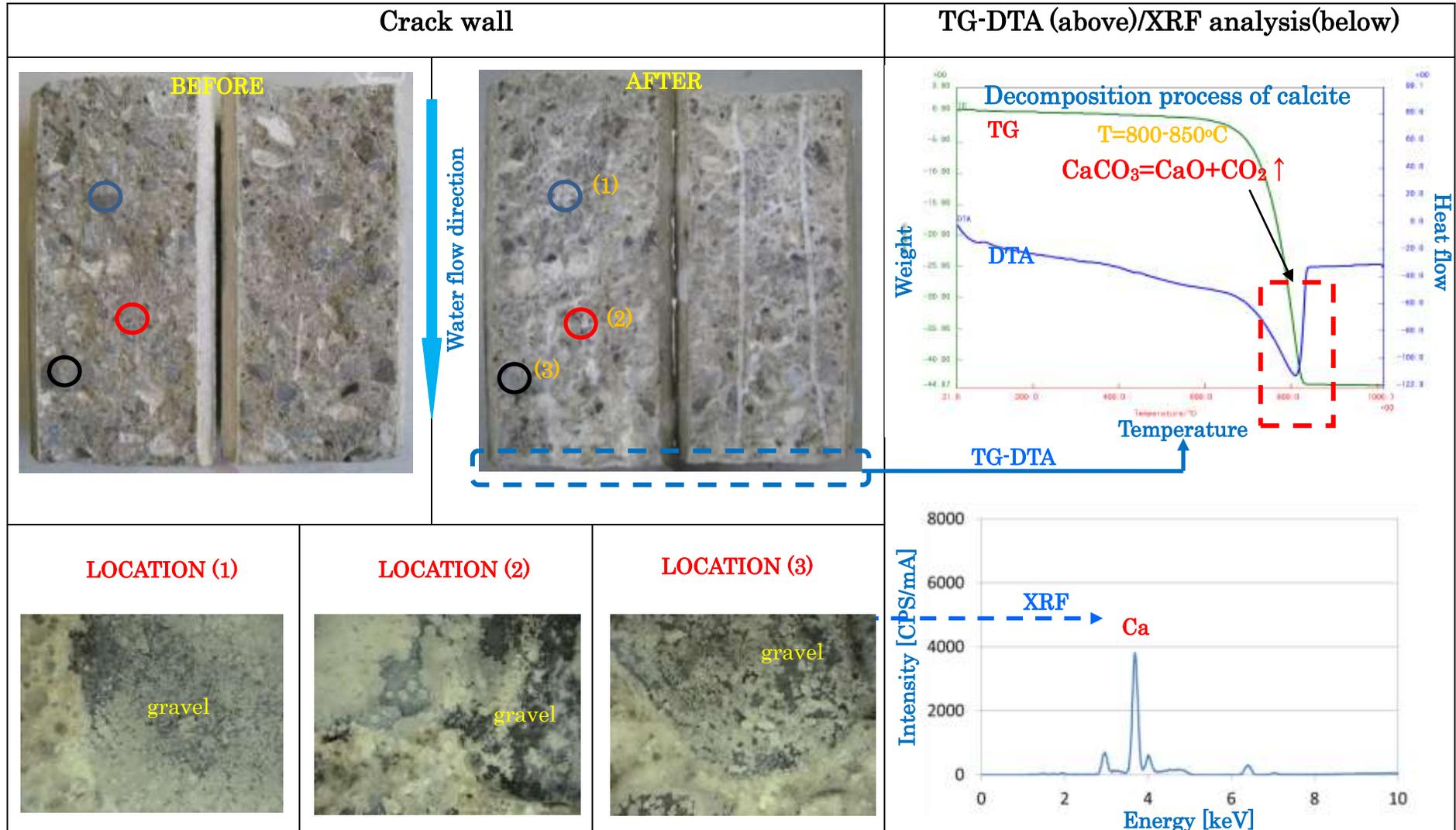
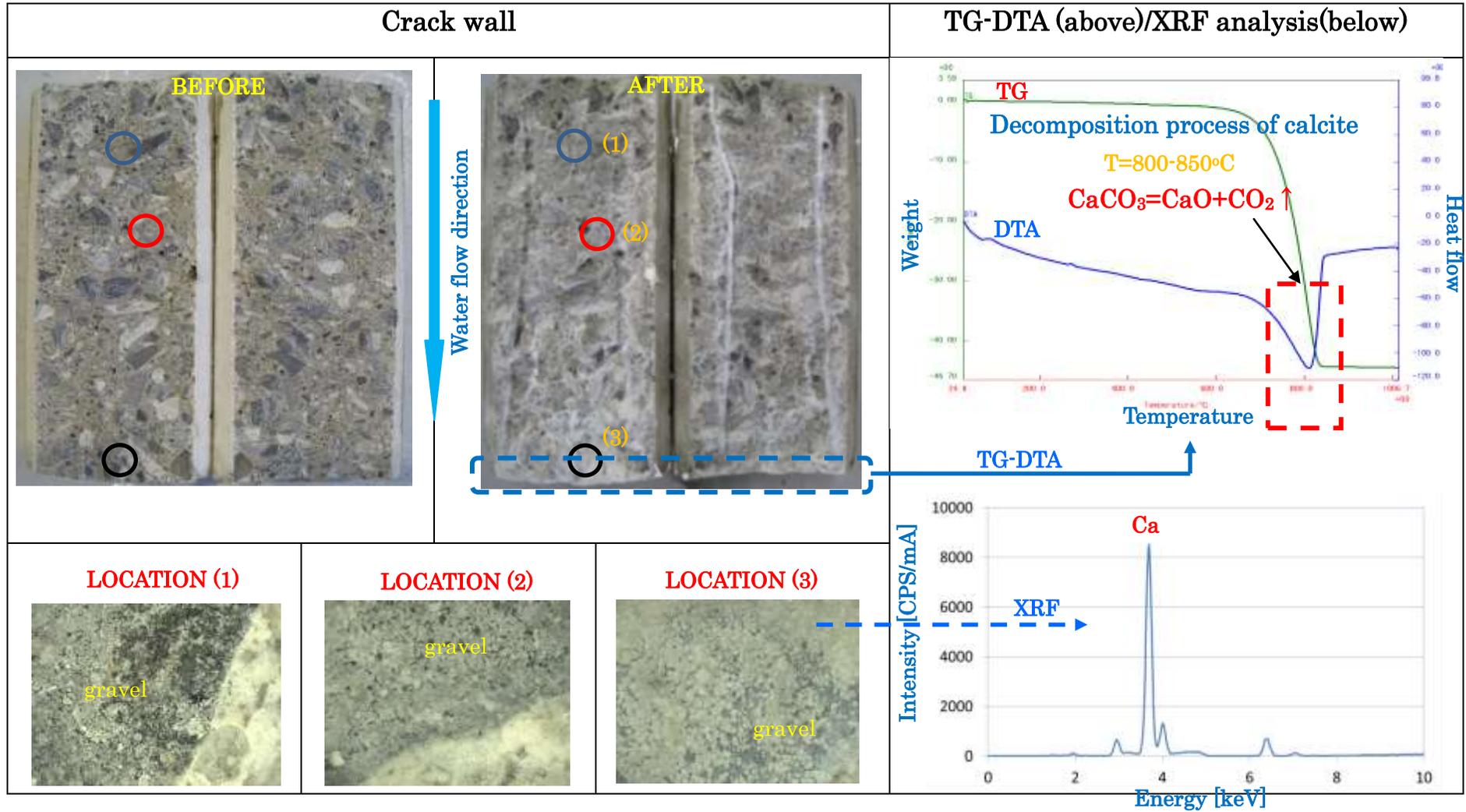


Table 4.11 Analysis of precipitated products on crack wall along the flow path and bottom surface-SH-Mix2-2b



After finishing the water pass test, specimens were kept dry under room conditions of 20°C and RH60%. Then unbinding two halves of specimen was done to observe the crack surface along the flow path.

It could be seen from *Table 4.9, 4.10 & 4.11*, that there was different color on the crack surfaces before and after the test due to the appearance of precipitated products. Moreover, many newly formed crystals were also clearly found on the surfaces of coarse aggregates, especially at the downstream direction of water flow path.

Based on the results of XRF test, it was found that the healing products deposited on the surface of aggregates were mainly composed of calcium compounds while calcite was confirmed as the main mineral forming at the surface crack (bottom of specimen) by analyzing the products via TG-DTA test.

#### 4.5.7 Long-term performance of self-healing concrete using granules

Based on the results above, it can be seen that trial SH-Mix1-1 showed the best performance for short-term test (a crack was introduced to concrete specimen at the age of 30 days after casting). In order to investigate the capability of preserving the healing property of concrete incorporating granules with the time, some specimens were cured continually for 12 months under two different curing regimes as can be found in *Figure 4.10*.



**Figure 4.10 Investigation of long-term preservation of healing capability on concrete using granules SH-Mix1-1**

In this test, the average crack width in the bottom surface was controlled about (0.20-0.26) mm while the crack width on the top ranged from 0.24 to 0.31mm. The results showed that there was no significant difference of healing capacity between two curing regimes. The same tendency of reduction in water leakage and healing products deposit on the surface crack was observed with time even though the initial flow rate was relatively different

as seen in *Figure 4.11 & 4.12*. This indicated that crack healing capability of concrete using granules (in this case granule SH-Mix1-1) could be maintained for long term until it was activated by a penetrated crack and exposed to water. Cracks with width of (0.2-0.3)mm can be fully healed.

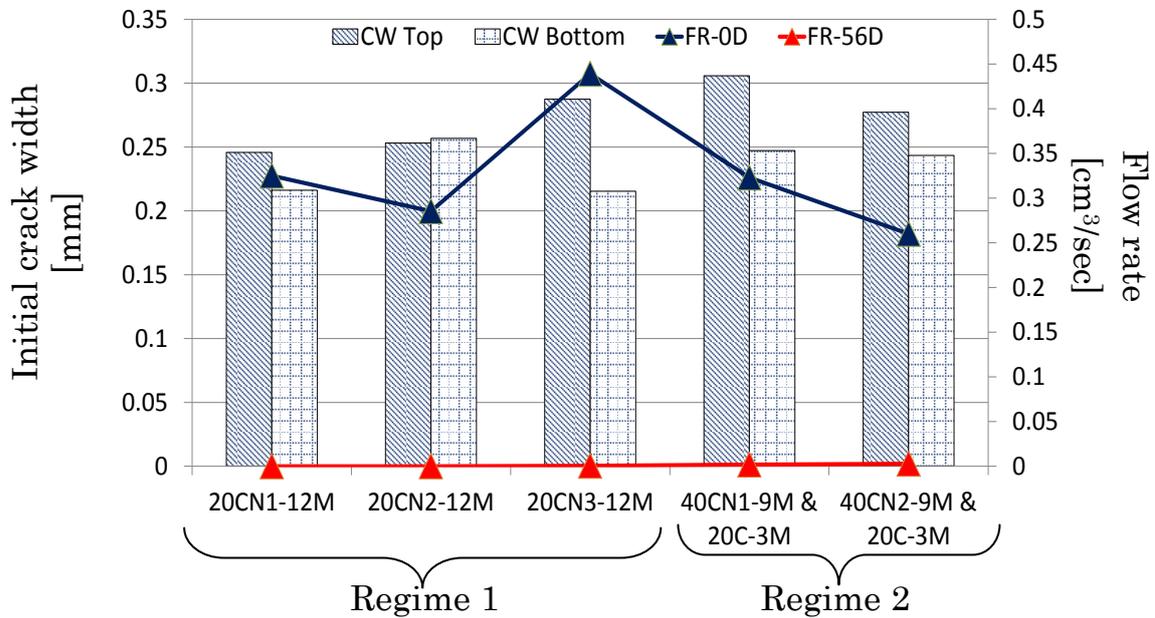


Figure 4.11 Initial crack width (vertical left axis); and water flow rate with time (vertical right axis)

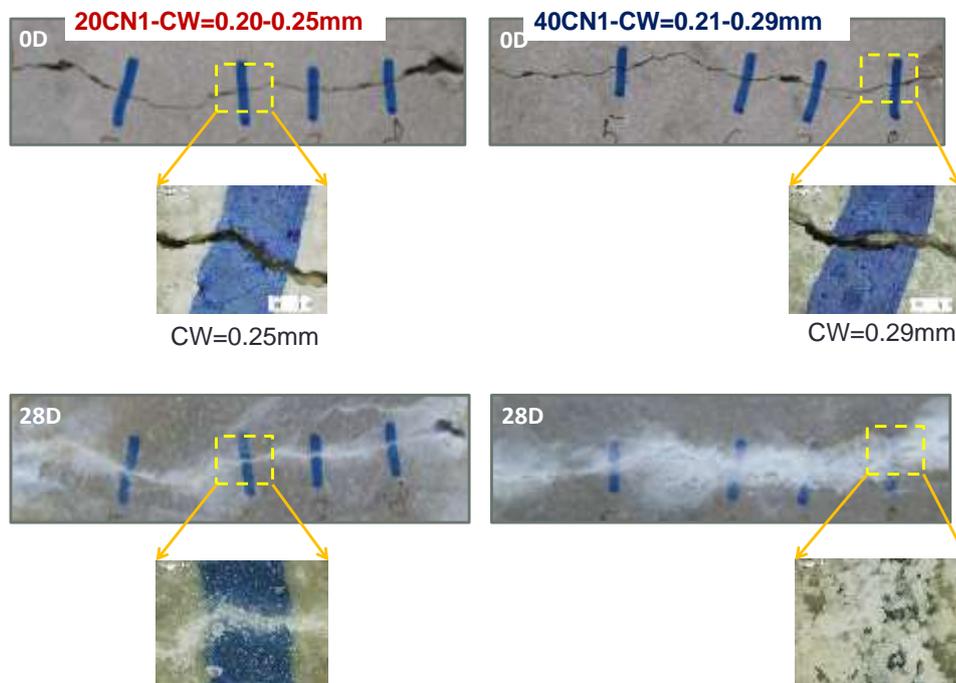


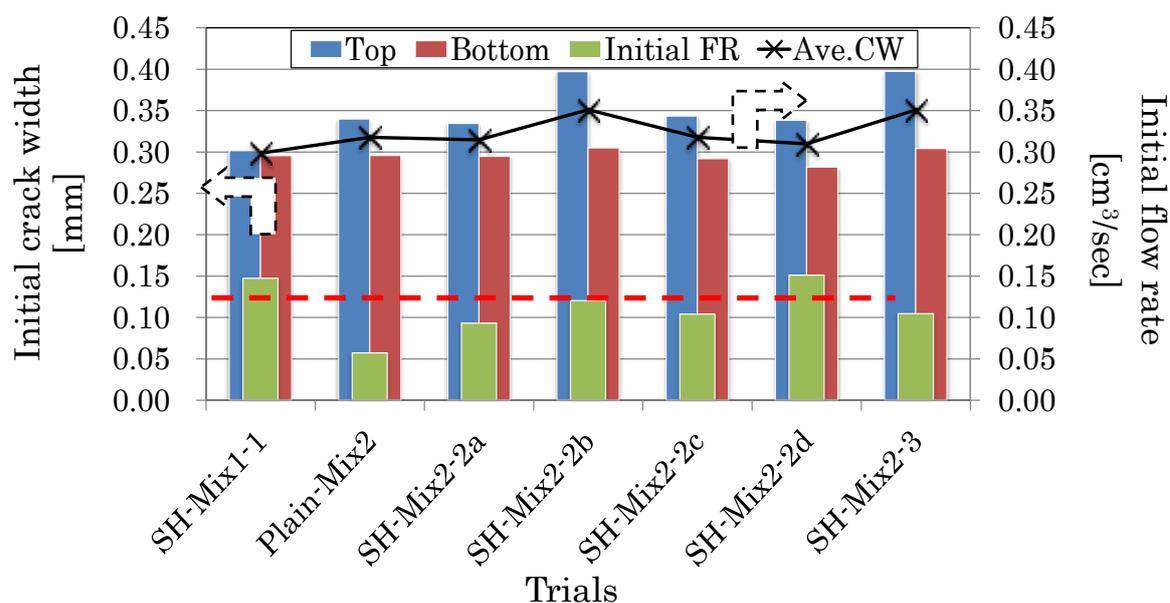
Figure 4.12 Typical crack closing process (above-initial crack; below-after 28days continuously exposed to water flow)

Unfortunately there was no controlled trial for Mix 1; therefore it is necessary to examine the crack healing capability of normal concrete in laboratory condition after long term exposure.

## 4.6 FURTHER IMPROVEMENTS IN WATER PASS TEST

### 4.6.1 Control of initial crack width and initial flow rate

It is well known that crack width is one of the most important parameters affecting the water flow rate. However, in this experiment crack width was not strictly controlled as can be seen in *Figure 4.13*.



**Figure 4.13 Relationship between initial crack width and initial flow rate**

There was an unclear relationship between crack width and flow rate due to the difficulty in controlling the same crack width for all specimens and also in one specimen (surface crack or the internal crack width along the flow path). Even if the crack width was the same, there was also no guarantee that the initial flow rate was the same for different trials. Water flowing through a concrete crack, especially in self-healing concrete is a very complicated phenomenon. It depends on a variety of influencing factors, such as: material used, crack width, roughness of crack surface, water saturation; pressure of water head, temperature, etc. Moreover, it is preferable to restrain the water flow rate as small as possible to accelerate the crack healing process in self-healing concrete.

In order to obtain the reliability of this approach, the performance of each trial was evaluated by observing the average result of three specimens.

Further improvements should be done such as controlling same ranges of initial crack width and flow rate in all trials so that it is effective to compare the performances among these trials.

#### **4.6.2 Change in arrangement of Teflon sheet to control internal crack width**

In this chapter, the internal crack width was controlled by setting two strips of Teflon on the crack surface as seen in *Figure 4.3a*, and then binding by two stainless clamps. To simulate the crack condition in practice and also to better control of crack width, from now on several pieces of Teflon were arranged on the crack surface (*Figure 3.12b*).

### **4.7 SUMMARY**

In this chapter, crack self-healing properties were introduced to concretes at normally used water-cement ratios by improving the granulation technology proposed by *Koide & Morita, 2010*.

#### **4.7.1 Granulation technique**

In this stage, there was an effort to store self-healing material by apply capsulation technique. It was considered as a physical treatment of powder material. Several types of mixer (roller mixer, Omni mixer, small & large mortar mixer) and granulation process (procedures or sequences to add in or to treat the powder) were investigated. There was no doubt that it was feasible to fabricate self-healing granules, typically in ranges of several millimeters, in order to reduce the manufacturing cost of granules by using a typical mortar/concrete mixer in laboratory. In general, even though the size of granules could be controlled by sieving, it was still relatively large that might affect the distribution of granules in concrete matrix. Therefore, a better control in the size of granules should be done with considerations of simple and cheap granule fabrication.

It is important to point out that physical semi-capsulation technique was occasionally realized in this stage (especially in case of manufacturing granules SH-Mix1-1). However, it was still unknown the key points of this technique to implement the semi-capsulation effect. Therefore, it is necessary to further improve the performances of concrete containing self-healing granules by enhancing the concepts and granulation techniques.

#### 4.7.2 Self-healing ingredients & performances of fabricated granules

Ideally, the inner material of granules, coated by an external layer, containing self-healing additives should be spread easily on the surface of crack once rupture and exposure to water flow. The more self-healing additives are released, the higher healing capability 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, selection of self-healing additives and the distribution or the size of granules in concrete matrix, if the same amount of granules is used.

Three groups of inner material were investigated in this experiment. The affects of inclusion of self-healing granules on the properties of concrete were summarized as follows.

In general, it was found that there was a decrease in the slump and an increase in compressive strength of concrete incorporating granules. However, it should be controlled as small as possible. Unfortunately, a reduction of the workability and a variance of compressive strength compared with normal concrete were still high at this stage. That indicated that the coating layer of granule was not so effective because its protection or preservation function still has not successfully obtained yet. As a result, when mixing with water and other components of concrete, the inner material might be activated and became hardened due to further reactions occurred.

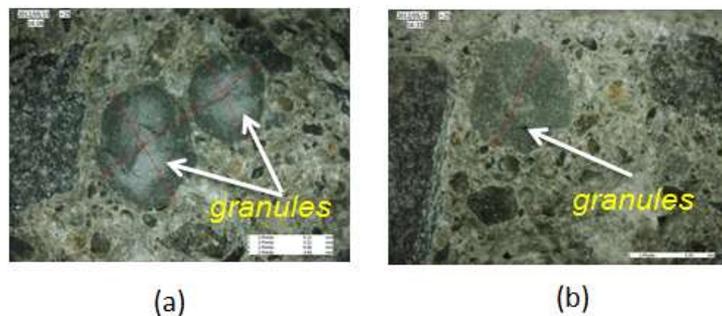
The experimental results obtained from water pass test showed that in case of granules containing cement, pozzolan materials and other additives, the water permeability through a penetrated crack was significantly reduced, especially in trial SH-Mix1-1 embedding granules of cement compound, admixtures proposed by AHN 2008, and a water-soluble agent. It can be inferred that compounds of cement/pozzolan & additives used in this study possess a certain capability of crack healing. However, the water still seeped through a crack and a fully healed crack has not achieved yet with width larger than 0.3mm (except for Trial SH-Mix1-1).

It can be argued by the author that the following matters influence the effectiveness of this type of granules. One of the possible reasons was the size of granules. In most of these trials, the size of granules was thought to be relatively large, typically in several of millimeters as can be seen in *Figure*

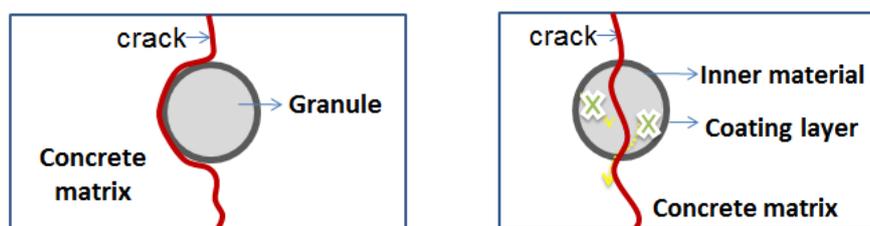
**4.14a.** The coarse size of fabricated granules caused a bad effect on the distribution of granules in the concrete matrix. It is the fact that with the same dosage of self-healing granules used, the smaller the size of granules, the better the distribution of granules in concrete is expected and the higher the self-healing efficiency to be achieved.

Another matter was the hardened condition of inner materials in/after the granulation process (*Figure 4.14b*) or the surface of broken granules when contacting to water flow. Given this, the inside self-healing additives were thought to be difficult to dissipate into the crack surface that restrained the healing capability. The hardened state of inner material might occur:

+ during the curing period of granules due to further reactions between cement compound or supplementary cementitious material (main component of inner material) and water used to prepare granules. In these trials test, the quantity of liquid used to make granule ranged 8.6-27.7%, but most of them were over 20%. Because this fact, the strength of granules became high and there was a possibility that granules cannot be ruptured once a crack penetrates into concrete matrix (*Figure 4.15-left*).



**Figure 4.14 (a) coarse granules; (b) Hardened state of inner material**



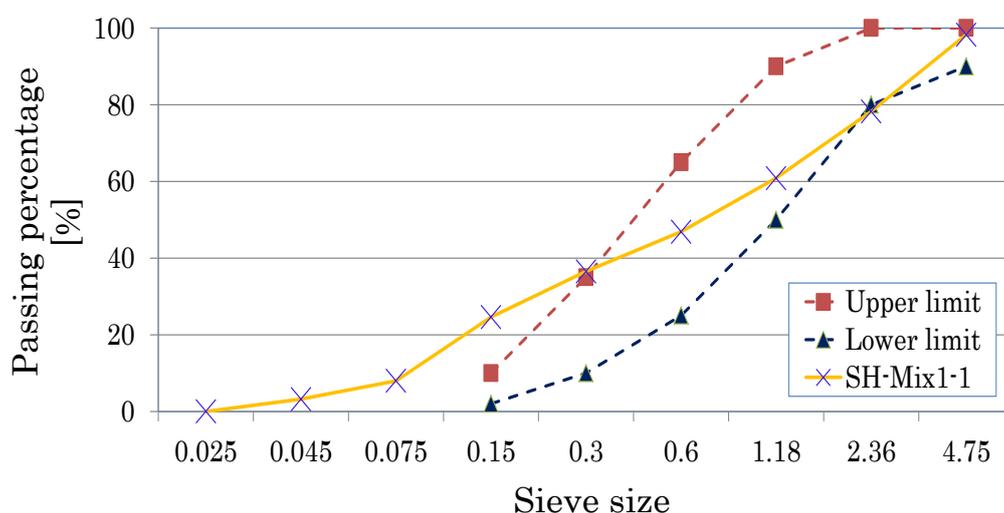
**Figure 4.15 (left) Problem caused by high strength granule  
(right) Hardened state of inner material**

+ just after or during casting concrete due to an ineffective coating

layer (mentioned above). In that case, even a crack can penetrate into the granule, self-healing additives are still difficult to release (*Figure 4.15-right*).

+ when ruptured by a crack and exposed to water flow, but inner material cannot be released into crack surface. The chemical reactions between inner material and flowing water occur inside granule.

Even though there were still several factors and variants affecting the result of the tests, designed concretes SH-Mix1-1 containing granules of Portland cements and specific chemical, mineral admixtures (Water reducing agent, dissolved agents, low water content) showed the best self-healing performance at this stage. The water leakage was almost stopped after 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 for crack in normally used concrete to be self-healed. It was assumed that due to 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, the best self-healing performance was achieved in this trial. In addition, it was thought that the particle size distributions of this type of granules also contributed to the better healing capability (*Figure 4.16*).



**Figure 4.16 Particle size distributions of granules used in SH-Mix1-1**

However, there was no clear concepts to explain why concrete incorporating granules SH-Mix1-1 showed the best performance. Further investigations should be done.

# CHAPTER 5

5- TRIALS TO ENHANCE  
THE SEMI-CAPSULATION  
TECHNIQUE FOR  
CEMENT/POZZOLAN MATERIALS

## 5.1 GENERAL

In this research, a crack self-healing capability was introduced to concrete by developing the semi-capsulation technology for powder material with the considerations of manufacturing cost. The designed granules, in which inner materials containing self-healing agents were coated by a layer of cement compound, were described as self-healing granules having semi-capsulation effect. Due to its features, granules of cement/pozzolan and self-healing additives and their reaction products are thought to be compatible with concrete components. Moreover, it is believed that they are more convenient for the recycling process in the future. However further reactions between granules and water (moisture) during fabrication, mixing and hardening of concrete also bring side effects on the healing capability of concrete using granules, depending on the quality of applied granulation techniques.

In previous chapter, concretes incorporating granules of cements/pozzolans and self-healing additives showed promising improvements in the workability, compressive strength and also crack-healing performance. However, the target of crack-healing capability of concrete has still not yet been achieved. The possible reasons for this constraint were discussed in section 4.7. In order to improve the healing capability of concrete by using this approach, two basic ideas were proposed (*Figure 5.1*). First, fine granules, whose typical sizes (or diameters) were less than 2mm, should be used for better distribution in concrete matrix. If the same dosage of granules used, the finer granule it is, the higher possibility cracks rupture the granules, the better healing effect is obtained. Second, inner material of granule should be designed so that self-healing agents were easily released upon cracking. If inner compound is not released, the healing effect will be limited due to just the surface of broken granules will react with water flowing. Once a layer of newly formed product deposits on the surface of ruptured granule, the diffusion process of self-healing material into crack surface will be further restrained.

Furthermore, “the market needs low cost, cement paste compatible self-healing agents for sustainable material use” (*Source: <http://selfhealingconcrete.blogspot.jp/p/expansive-cementitious-agents.html>*).

The following challenges should be faced to solve above-mentioned issues: reduce the cost of making finer granules and find out the functional effective ingredients for granule having semi-capsulation effect.

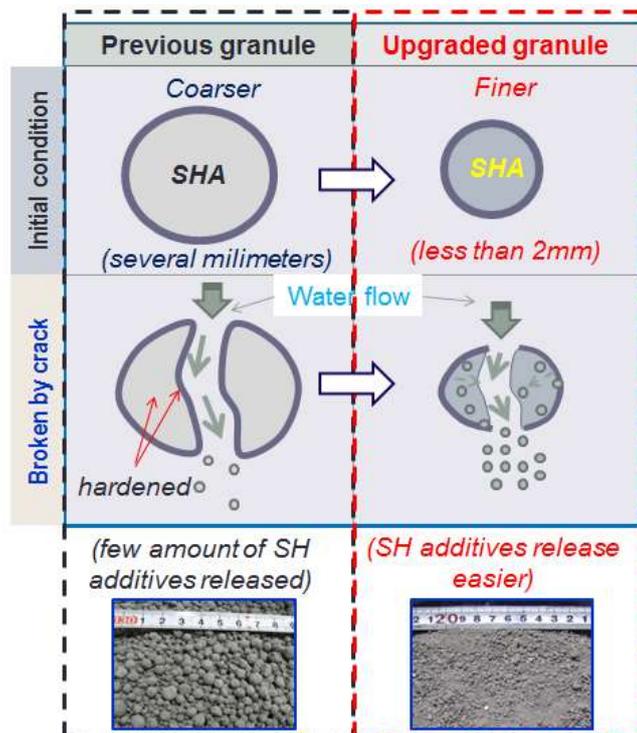


Figure 5.1 Two basic ideas to robust crack-healing capability

## 5.2 REQUIREMENTS AND BASIC DESIGN CONCEPTS OF GRANULE HAVING SEMI-CAPSULATION EFFECT FOR POWDER MATERIAL

Based on the obtained results in research of Koide & Morita (2010) and data achieved from the study in stage 1, the requirements for granule having semi-capsulation effect were proposed as follows.

○ **Requirement 1:** The addition of granules to concrete mixture should not bring bad effects on the properties of fresh/hardened concrete (*Research of Koide & Morita*).

In this study, the coating layer of granule is made by cement material.

Therefore, further reactions between outer layer and mixing water during mixing and hardening process of concrete could not be perfectly excluded. Because of this, even though a better bonding strength between granules and concrete matrix can be obtained, the workability of fresh concrete and the strength of matured concrete will be affected. It is necessary to control of slump loss and strength increase as much as possible.

○ **Requirement 2:** Granules have a capability of preserving the healing property of concrete with the elapse of time (*Research of Koide & Morita*).

One of the most important purposes to use granules in concrete is to provide a sufficient healing capability and to maintain this property for long term, due to the long service life of structures and the uncertainty of crack appearance in concrete during its service life.

○ **Requirement 3:** Self-healing granules should be distributed uniformly in concrete matrix (*Additional requirement*).

This requirement is derived from the fact that it is difficult or complicated to know in advance the position at which a crack will occur and further penetrate in concrete matrix. Moreover, the efficiency of crack healing is also significantly influenced by the distribution of granules as discussed in section 5.1.

○ **Requirement 4:** Embedded granules should be ruptured by the penetration of a crack (*Additional requirement*).

It is well-known that the healing process in concrete incorporating self-healing granules is only activated when a crack breaks through the granules and water is available on the crack surface. Once a crack penetrates, the more broken granules there are, the higher healing capacity is expected.

○ **Requirement 5:** The inner material of granule should be released into the crack surface (*Additional requirement*).

Even the granule is broken by a crack, if the inner material cannot easily

dissipate into the crack surface, the effective healing area will be limited. It can be assumed that when water flows through a crack and pass a broken granule, some of water will penetrate inside the inner part of broken granule to activate chemical reactions at that position. As a result, inner material becomes hardened so that its healing capacity is reduced.

In order to fulfill the semi-capsulation effect of proposed granule, the basic design concepts of granule for powder material were established in this study.

☆ **Concept 1:** Fabricated granule should have a waterproofing property.

In this research, one of the main roles of granule is to contain and protect the self-healing or reactive material inside, which is often considered as a water loving agent. Therefore, a coating layer will be introduced to granule to render the surface impermeable to water. Even though the coating layer is made by cement compound, it is possible to control the water absorption of granule and further reactions between ingredients of granule and mixing water by producing an effective outer layer, which has both good bonding property with concrete matrix and watertightness property to protect inner material. If granule does not possess a waterproof function, there is a possibility for the mixing water penetrating to inner part that facilitates further reactions between water and self-healing component. As a result, the strength of granule becomes high (very thick and strong coating layer; or the hardened state of inner material) that restrains the healing capacity (*Figure 5.2*).



**Figure 5.2** Expected problems when coating layer does not possess a waterproofing property

Another problem is due to further reactions, the slump loss of fresh concrete is also observed.

☆ **Concept 2: Granule containing inner self-healing material should be strong enough to preserve the healing capability from unexpected events during concrete mixing process and also should be weak enough to be cracked when necessary.**

Coating layer of proposed granule has a possibility to further react with water when adding to concrete mixture. Due to this fact, this approach brings to better bonding property with concrete matrix (to ensure that a crack can break through the granule), but also induces bad effect on the healing capacity of concrete if its strength is too high and its thickness is too big (*Figure 4.12*). Therefore, be broken and released when needed is a very important issue for granule to fulfill the semi-capsulation effect. The strength of granule, mainly contributed by the strength and thickness of the coating layer, should be strong enough to withstand the compaction/vibration effect while mixing and casting concrete. However, the outer layer also should not be too strong and thick as well. In addition, the inner material should be weak enough and easily released once rupture to ensure the spreading effect. Besides the quality and techniques of granulation process, the sequence of adding granules to concrete mixture and mixing duration together with other concrete components also affect the performance of granules. To reduce the possibility of broken granules during concrete mixing, self-healing granules should be added to mixture just before casting and the mixing duration with components of concrete should be as short as possible to prevent embedded granules from breaking, but also satisfies the requirement of uniform distribution of granules in concrete matrix.

### **5.3 STRATEGIES TO IMPROVE SELF-HEALING PERFORMANCE VIA SELECTING FUNCTIONAL EFFECTIVE INGREDIENTS**

In general, self-healing granule has two basic components: inner material and

coating layer as seen in *Figure 5.3*. Each component has its own features and effects.



**Figure 5.3** Two basic components of self-healing granule

Based on the requirements and basic design concepts of granule having semi-capsulation effect, following strategies to upgrade the performance of self-healing granules in concrete were proposed and investigated.

### 5.3.1 Inner material

*Figure 5.4* showed the strategies to enhance the healing capability of concrete incorporating granules by selecting functional ingredients.

The inner material should have both the self-healing effect and the spreading effect. Reactive material should be released when the granule is ruptured.

In order to possess the healing property, the compounds of cement and pozzolan were chosen as the main components of inner material. In this investigation, various types and ratios of Portland cements, commercial or special supplementary cementitious materials, such as fly ash, blast furnace slag and silica fume were used. In order to further stimulate healing properties, several types of self-healing additives, in terms of swelling/expansion/precipitation effect, were preferably added to inner material in single or combination form. In this study, one of the most interesting things in improvement of performance of granules via selecting ingredients was the application of several types of materials to provide sufficient healing capability of concrete with the elapse of time.

In order to make the inner material easily to release once rupture, it should not be in hardened state. One of solutions is the usage of water-soluble agent,

such as calcium hydroxide or other kinds of water-soluble materials. Those materials are easily dissolved when contacting to water, that helps to dissipate the healing material into the crack surface. Another approach is to restrain the hydration ratio of cement compound (one of the main components) or chemical reactions of other self-healing additives by using water reducing agents or special solutions. The more un-hydrated cement particles or un-reactive self-healing additives remain, the higher healing capacity of concrete is expected. In this test, the applied agent not only reduces the required quantity of water but also gives a retarding effect. Furthermore, another important effect of water reducing agents was to control the flocculation process of cementitious materials when contacting to water. That helps to reduce the necessary amount of water needed for granulation and also reduce the average size of granules (*Section 5.43*). By using fine sand as one of components of inner material, such as silica sand and slag sand, etc, it was believed that besides the effect of healing effect (due to pozzolanic reactions), the spreading effect would be improved due to the round shape of fine sand.

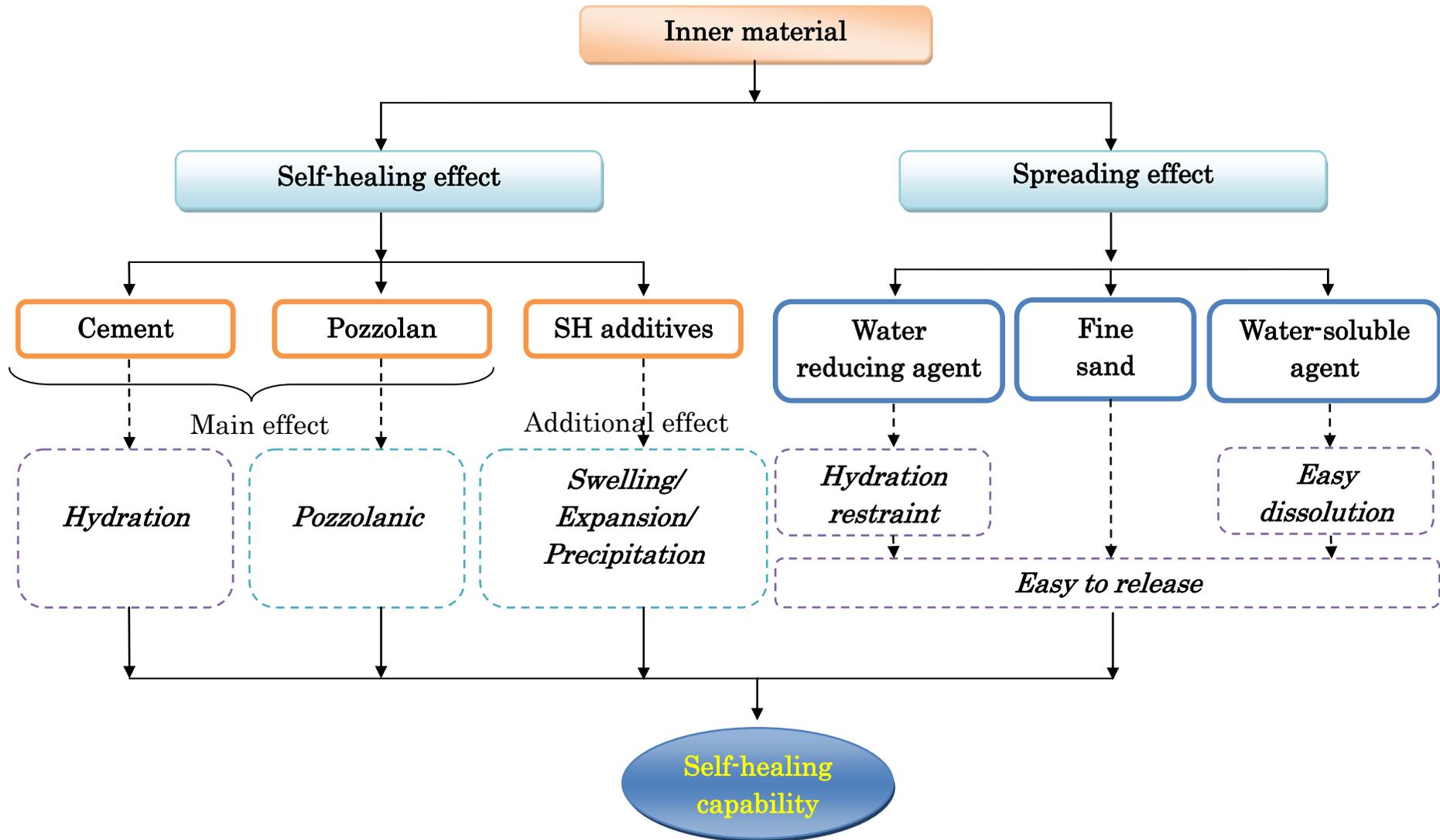
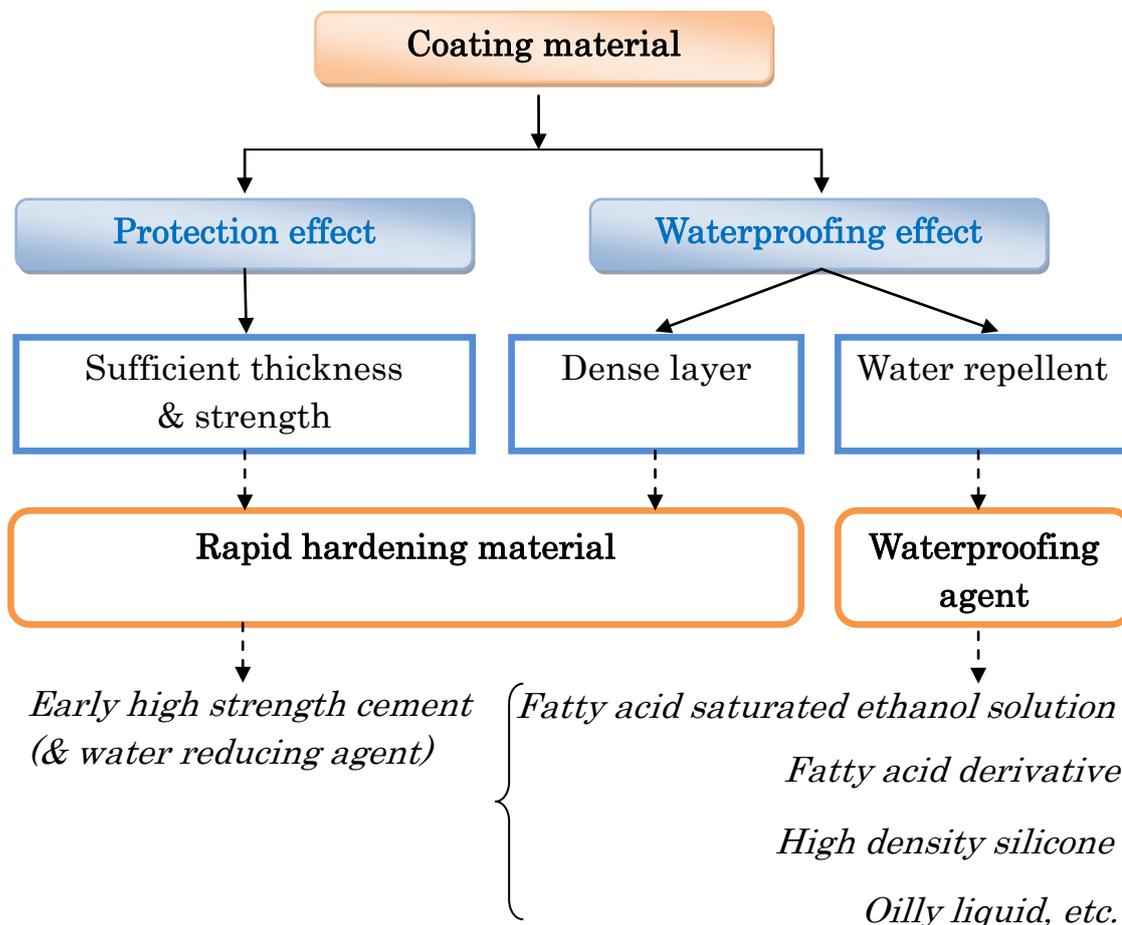


Figure 5.4 Strategies to improve self-healing performance via selecting functional ingredients

### 5.3.2 Coating material

Based on the role of coating layer on the performance of granules, it was thought that the layer should be strong enough to withstand the impact during mixing concrete and has a good bonding property with concrete matrix. On the other hand, it should be thin or weak enough for the crack to break through. Therefore ideally, the coating layer should possess both the protection effect and waterproofing effect. In this research, cement compound was used to form the coating layer.

The strategies were proposed to introduce an effective coating layer to granule, as can be seen in *Figure 5.5*.



**Figure 5.5 Strategies to choose selective ingredients for coating layer**

In order to achieve a protective outer layer, a rapid hardening process would be applied by using highly reactive cements or water reducing agents. Those materials have a potential to limit the long-term reaction between coating

material and water to produce the outer layer sufficient thickness and density (This effect also contributes to the waterproofing property of coating layer).

With respect to preservation of healing capacity and bring no adverse effects to concrete properties, coating layer should act as a barrier to prevent water absorption during mixing concrete. Several types of waterproofing agents, for instance fatty acid saturated ethanol solution, fatty acid derivative in form of powder, high density silicones, oily substances, etc. were tried to investigate its hydrophobic effect.

#### **5.4 STRATEGIES TO IMPROVE SELF-HEALING PERFORMANCE VIA SEMI-CAPSULATION TECHNIQUES**

In the 1<sup>st</sup> stage, the resultant granules were rather coarse due to the effects of agglomeration of powder & water and mixing process. Consequently, the distribution of granules in concrete matrix and the efficiency of self-healing capability of concrete were affected. This inspires an idea of using finer granules for improving the healing performance of concrete (*Figure 5.5*).

Two strategies were proposed to achieve fine granules with special considerations of simple and cheap granule fabrication: (1) to prevent self-healing powder from agglomerating in large size by using steel balls and sieving (*Figure 5.6*), (2) to provide nuclei for granulation process by using various types of fine sand as one of components of inner material (*Figure 5.7*).

It was clear to find that under the impact of steel balls during granulation process, the sizes of granules were controlled better, especially with the help of sieving equipment. By applying this work, large amount of steel balls with a diameter of 11mm were added to the mixer while mixing the materials and liquid together as could be seen in *Figure 5.6*.

On the other hand, another approach to provide nucleus for easier granulation process also showed very promising effectiveness. In this case, fine sands, especially which have a self-healing potential such as silica fine sand, slag

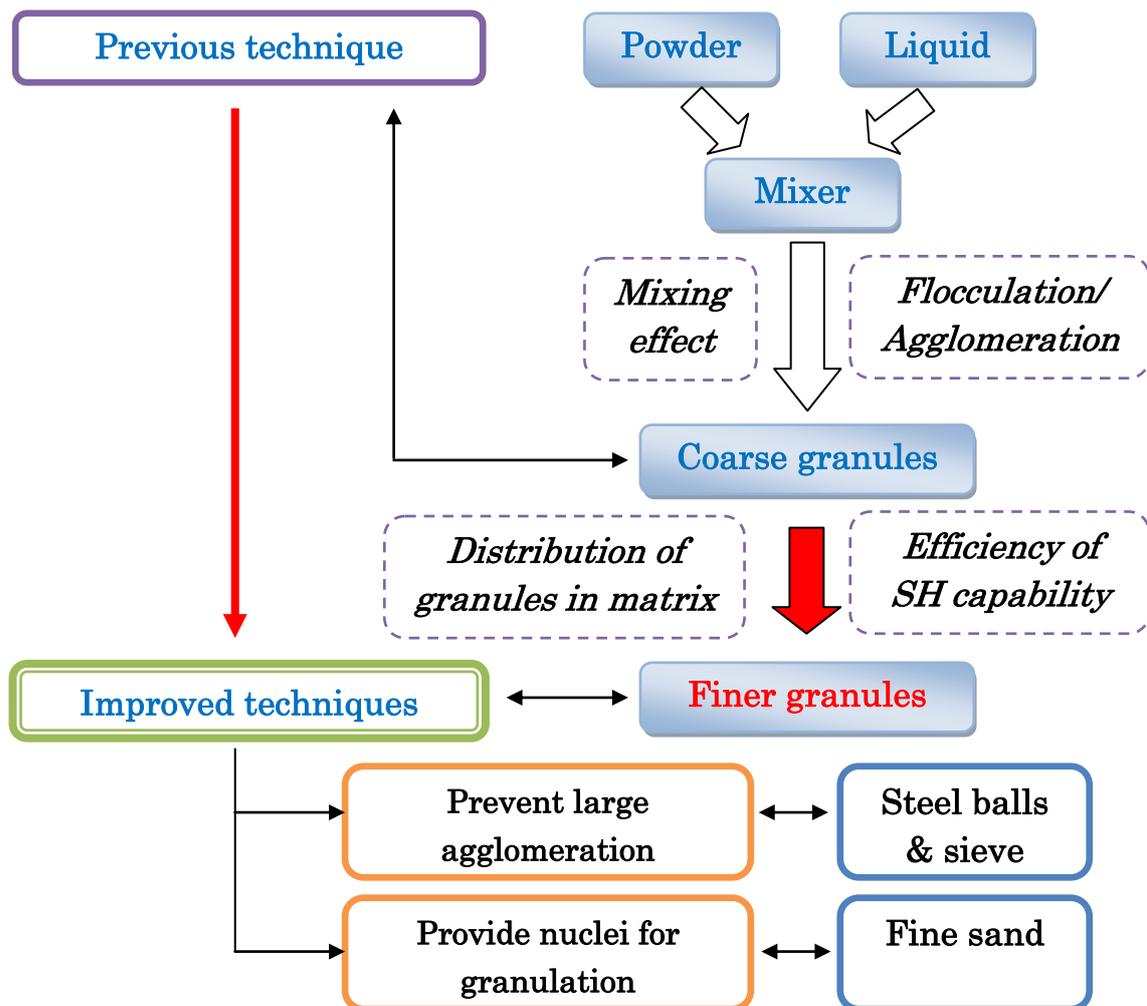


Figure 5.5 Strategies to improve granulation techniques

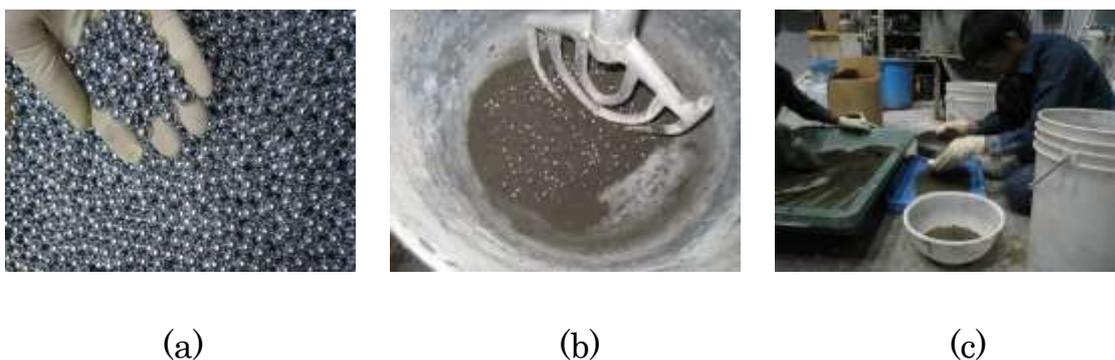


Figure 5.6 (a) Steel balls; (b) Mixing self-healing materials with steel balls; (c) Sieving process

sand or OPC clinker, were preferably used in granulation process to control the sizes of granules (self-healing powder will be accumulated gradually around the core of sand) and also further contribute to the self-healing

capability of concrete (*Figure 5.7*).

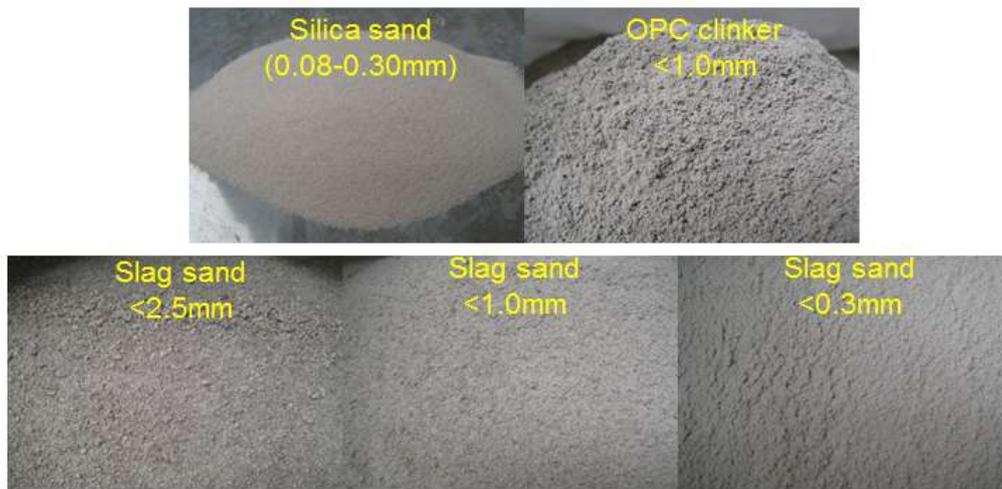


Figure 5.7 Different types of fine sand having certain healing capability

## 5.5 PERFORMANCE OF CONCRETES INCORPORATING ENHANCED GRANULES

### 5.5.1 Experimental timelines

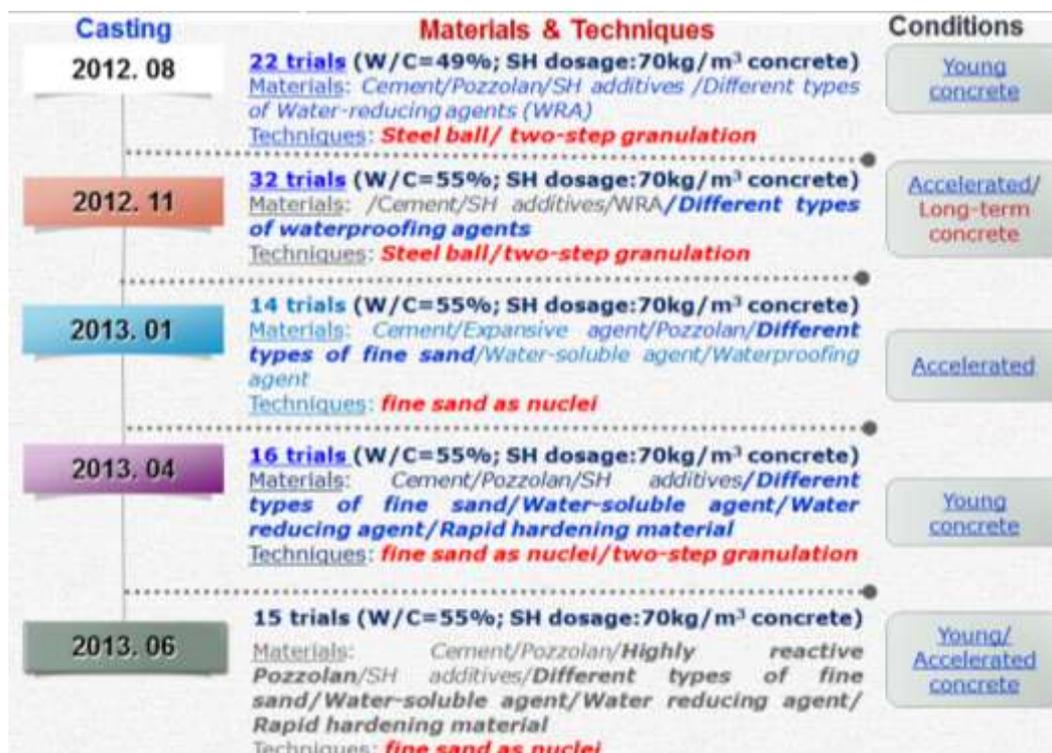


Figure 5.8 Experimental timelines

Based on the strategies proposed above, various trials of self-healing concrete incorporating different types of granules were cast and investigated as can be seen in *Figure 5.8*.

### **5.5.2 Trials using steel balls & two-step granulation technique (self-healing concrete trials cast in August 2012)**

There were totally 22 trials including controlled normal concrete one. In this case, initial crack width at the bottom of specimen was controlled about (0.25-0.30)mm and concrete specimens were induced a crack at the age of 21 days after casting (cured in sealed condition under experimental room conditions).

Two methods of granulation process were tried, as follows:

#### **(a) Using steel balls to control the size of granules**

In this method, steel balls were added to mortar mixer during mixing the self-healing powder and water or liquid (*Figure 5.6*). The quantity of liquid was about 15% of total weight of all ingredients making granule. There was a reduction in the amount of liquid used to make granules with the purpose of restraint of further reaction due to surplus water.

Under the impact of steel balls, smaller granules could be achieved as could be seen in *Figure 5.10 (left)*. However, there was also a high possibility that granules or coating layer of granules would be broken during granulation process. That might bring to a decrease in the performance of self-healing concrete using granules. Another obstacle was that a large amount of steel balls needed for a better control of the size of granules. The more steel balls used, the tougher and heavier work it was.

Furthermore, also in this trial, there was an effort to make granules that had similar self-healing ingredients as used in granules SH-Mix1-1 in the 1<sup>st</sup> stage (which was composed of cement compounds, some specific self-healing materials in terms of swelling, expansion, precipitation (proposed by AHN,

2008) and a special kind of water-reducing agent and also water-soluble agent in form of powder). In this trial, at first fine granules were manufactured by mixing all powders with water and steel balls in mortar mixer. And then, fine granules were steamed to induce coating layers. However, the crack self-healing performance of concrete using these granules was not as good as that of in the 1<sup>st</sup> stage due to using different granulation techniques. Based on this result, it can be found that both self-healing ingredients and granulation technique are important to fabricate effective granules. In another word, if only good self-healing ingredients were used or only good granulation technique was applied to manufacture granules, it was thought that the self-healing performance of concrete using those granules would be not good.

**(b) Two-step granulation technique**

In this method, the granulation process was divide into two steps with an interval of time between them (to wait for the inner material hardening).

+ Step 1: Make fine granules by using steel balls as in section 5.5.1a.

+ Step 2: After curing in plastic bag for a few days, a coating layer was introduced by gradually adding the material to the roller mixer (drum mixer) and spraying the liquid (*Figure 5.9b*).



(a) Mortar mixer + steel balls



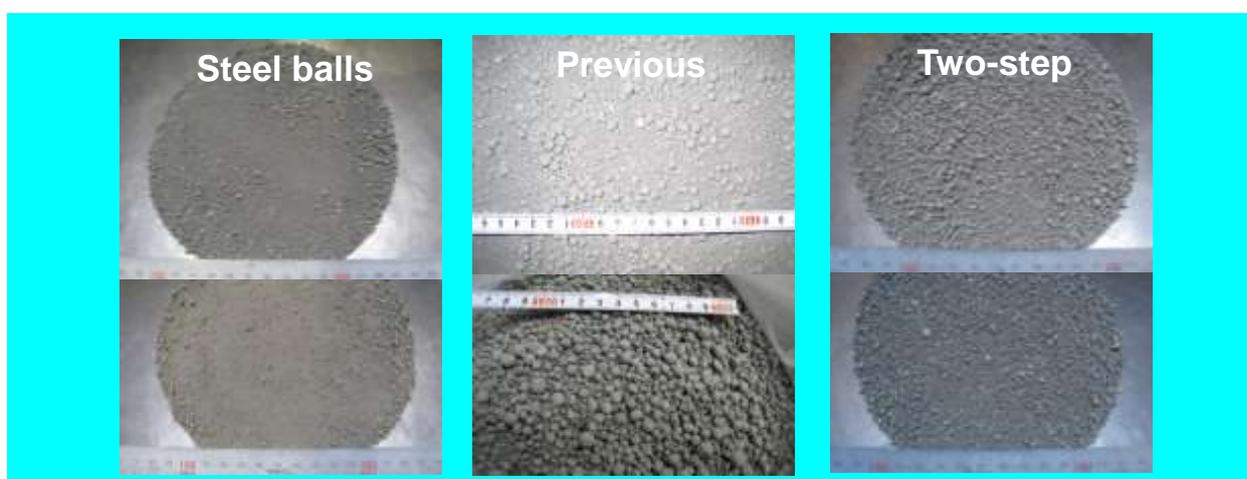
(b) Roller mixer

**Figure 5.9 Mixers used in two-step granulation technique**

It was found that it was easier to make the shape of granules by two-step granulation process. However, it will take longer time to fabricate the

granules and it needs better control in the size of granules by selecting effective coating materials and improving the granulation technique. At that time, due to this technique was still being developed, the resultant granules were relatively large in size as shown in *Figure 5.10 (right)*. Furthermore, based on this technique, it was inspired to use fine sand as nuclei for granulation process that will be discussed later.

The typical samples of granules manufactured by using steel balls, two-step granulation and previous ones were shown in *Figure 5.10*.



**Figure 5.10** Samples of granules made by (left)-using steel balls; (middle)-previous technique; (right)-two-step granulation process

### 5.5.3 Trials introducing the waterproof effect to granules by using waterproofing agent (self-healing concrete trials cast in November 2012)

In these trials, the waterproofing effect was introduced to granules by using various types of waterproofing agents that are commercially available. Firstly, fine granules were made by using steel balls technique (as seen in Section 5.5.2) and trying to reduce the required amount of liquid to around 13% of total weight of all ingredients making granule. And then granules were cured in plastic bag for a period of time to wait for the coating layer hardening. Finally, waterproofing agents in form of powder (dosage: 3% of total weight of granules) or liquid (dosage: 10% of total weight of granules) will be mixed together with pre-fabricated granules in small mortar mixer to provide the

hydrophobic, water repellent surface. *Figure 5.11b* showed the typical samples of granules just after mixed with the waterproofing material.



(a) Small mortar mixer



(b) Conditions of granules just before  
& after mixed with waterproofing agent

**Figure 5.11 Granules before and after applied the waterproofing layer**

Even though waterproofing agents could contribute to the preservation of healing capability of granules for long time or prevention of water penetration into inner part of granules, due to its high cost it is thought that this technique is difficult to reach the commercial-based application. Furthermore, if only the coating layer of granules has the waterproofing property, it is still unsure that the healing performance of granules will be improved if the inner material does not work well its role.

Up to that period, based on the performance of self-healing concretes embedding granules manufactured by using steel-ball granulation technique and waterproofing agents, it could be concluded that those approaches were not promising to achieve the target for simple & cheap granule fabrication

#### **5.5.4 Trials using fine sand as nuclei for granulation and different types of functional powder (self-healing concrete trials cast in April 2013)**

Based on the results obtained from previous trials, it was found that it was feasible to fabricate finer granules for better distribution in concrete matrix by using steel balls. However, this work was too tough to prepare a huge

amount of self-healing granules (sometimes it also further needed to use the seive to control the size of granules).

In this time, various types of fine sand with different sizes and chemical components were used with respect to achieve of a simple & cheap granulation technique (*Figure 5.7*). Besides acting as initial seeds for the granulation process, these sand also contribute to self-healing capacity of granules due to its properties of pozzolanic or cementitious materials. Furthermore there was an effort to identify the simple compound of inner material possessing sufficient healing capability. The main component of granule was also the Portland cement compound. In these trials, self-healing performance of normal concretes with different water cement ratios (*trial 1 & 2*), self-healing concretes using granules of different types of fine sand and Portland cement (*trial 6-9*), self-healing concretes using granules of fine sand (silica sand), Portland cement and different types of functional powders (such as water-soluble agent, expansive agent, carbonate material, pozzolans, and hydrated or unhydrated calcium sulfate) (*trial 10-16*). In addition, the effect of fine sand (silica sand) quantity used to make granules was also investigated (*trial 3-6*).

Two-step granulation process performed by using a small mixer (*Figure 5.11*) was applied in this case, as follows:

+ **Step 1:** Mix all the powders with fine sand & water (amount of water used ranges from (12.5-16.7)% the total weight of powder material). Then curing the resultant material in plastic bag for a few days.

+ **Step 2:** Mix pre-fabricated material with coating powder and water reducing agent to induce a outer layer of granules. In this case, coating layer was made by using high early strength cement and water reducing agent in form of liquid (superplasticizer).

*Table 5.1* showed the types of concretes and self-healing ingredients in percentage used in this investigation.

TRIALS TO ENHANCE SEMI-CAPSULATION TECHNIQUE FOR CEMENT/POZZOLAN

**Table 5.1 Types of concrete and material used**

Trial	W/C	Notification	Self-healing ingredient in percentage				Type of sand	Type of function powder
			W+SP	Cement	Function powder	Fine sand		
1	0.55	Normal concrete as control						
2	0.45	Powder of NC (Ordinary Portland Cement) as SH material						
3	0.55	SHC, dosage of granule: 70kg/m <sup>3</sup> concrete, different types of granules	16.9	77.9	0.0	5.3	Silica sand	
4			19.9	80.1	0.0	0.0		
5			14.0	58.0	0.0	28.1	Silica sand	
6			14.6	63.1	0.0	22.3	Silica sand	
7			14.6	63.1	0.0	22.3	Slag sand <0.3mm	
8			14.6	63.1	0.0	22.3	Slag sand <1.0mm	
9			14.6	63.1	0.0	22.3	Clinker sand <1.0mm	
10			15.9	51.6	10.9	21.7	Silica sand	Calcium hydroxide
11			15.9	51.6	10.9	21.7		Expansive agent
12			15.9	51.6	10.9	21.7		Carbonate
13			15.3	43.0	22.7	19.0		Blast furnace slag
14			15.3	56.4	10.6	17.7		Fly ash type II
15			13.6	34.0	30.1	22.3		Blast furnace slag + Anhydrous calcium sulfate
16			15.6	46.7	21.7	16.1		Blast furnace slag+Hydrated calcium sulfate

**Table 5.2 Mix proportions of normal concrete and self-healing concretes**

Concrete	W/C [%]	s/a [%]	kg/m <sup>3</sup>					
			W	C	S	SH	G	AD(Bx%)
Controlled	55	46	168	306	848	-	949	1
SHC	55		168	306	778	70	949	1
OPC as SH material	45		168	306	778	70	949	1

**(a) Result of the properties of fresh concretes**

It was observed that the addition of self-healing granules into concrete mixture often caused a decrease in the workability of fresh concrete. However, depending on the quality of coating layer, typically the density of the outer layer, the changes in slump values would be different.

*Table 5.3* showed the values of the slump and air contents in all trials when casting concrete specimens. From this figure, it can be seen that there was a significant improvement on the workability of fresh concretes by using upgraded granules. With the same dosage of superplasticizer and air-entraining admixture, it was found that the slump values of all self-healing concrete trials were satisfied with the designed value.

**Table 5.3 Properties of fresh concretes**

Trial	Properties of fresh concrete				AD (Bx%)	AE (Bx%)
	Slump [cm]		Air content [%]			
	Design	Testing	Design	Testing		
1	12±2.5	14.0	4.5±1.5	4.2	1	0.015
2		14.0		4.0		
3		13.0		5.4		
4		20.0		6.0		
5		17.0		6.7		
6		14.0		6.7		
7		11.5		6.3		
8		14.5		6.3		
9		13.5		6.0		
10		11.0		5.9		
11		13.0		5.8		
12		10.5		5.6		
13		15.5		5.4		
14		15.0		5.4		
15		13.5		4.7		
16		14.5		5.0		

**(b) Result of the compressive strength at 28<sup>th</sup> day curing in experimental room conditions (sealed condition, temperature of 20°C and relative humidity of 65%):**

*Figure 5.12* showed the results of 28<sup>th</sup> day compressive strength of all trials which were cured in sealed condition at the temperature of 20°C and relative humidity of 65%.

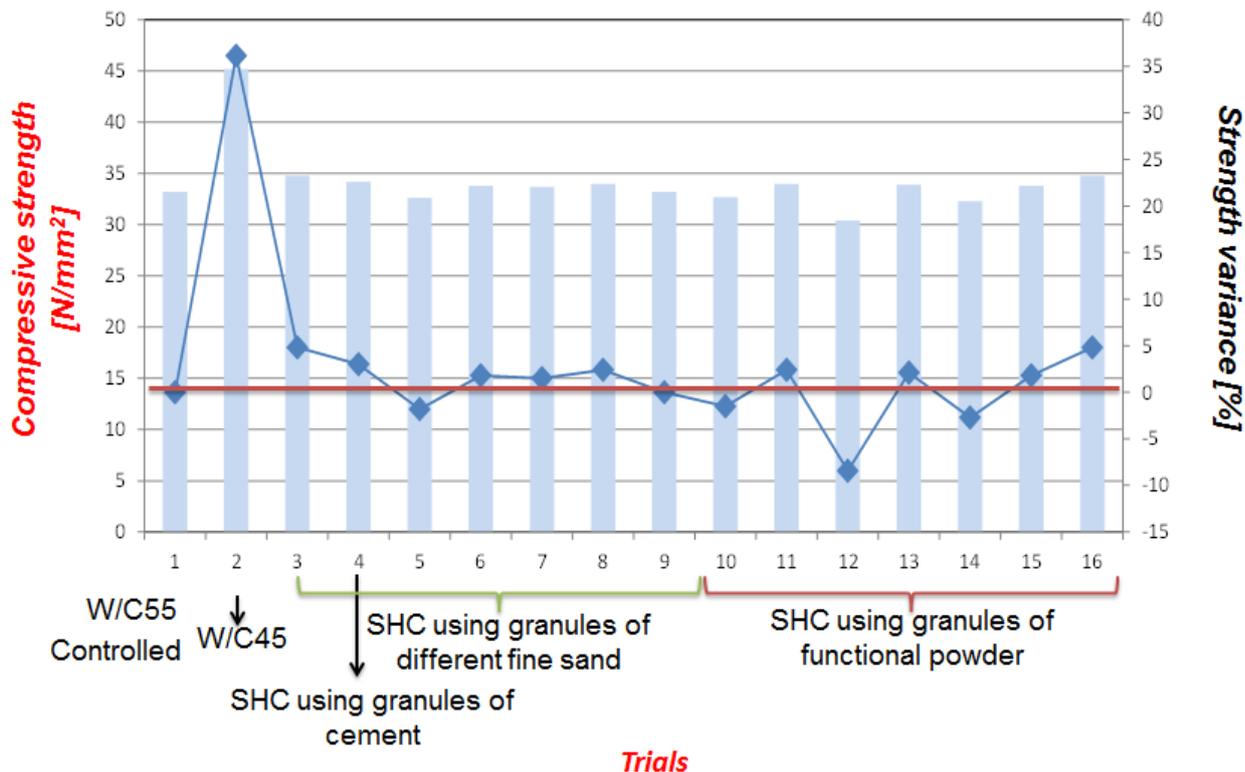


Figure 5.12 Compressive strength of all trials at 28<sup>th</sup> day

There was a improvement of strength variances between the normal and self-healing concretes as shown in *Figure 5.12*. In most cases, the variance was smaller than 5%, except for trial 12. This indicated that early hardening Portland cement & water reducing agent (superplasticizer) can provide a dense and effective waterproofing layer to granule. Due to this property, the mixing water just reacts with outer material and difficult to penetrate into the inner part of granules that may bring to a good bonding effect between the embedded granules with concrete matrix and a better control of slump loss and strength increase. As a result, it was believed that the healing capacity of concrete incorporating granlues should be maintained for a long period of time.

Moreover, it also can find clearly that the effect of different approaches to add powder material to concrete mixture on the properties of fresh and hardened concretes, such as in form of powder (trial 2) and in form of granules (trial 4). In case of trial 2 using powder of Ordinary Portland Cement as self-healing

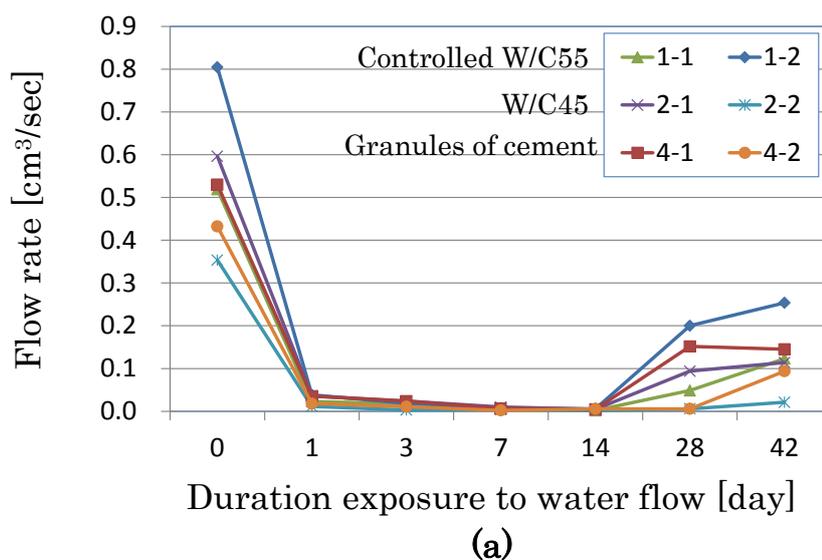
material (in this case cement was added to concrete by partial replacement of sand with the same dosage), the strength of concrete was significantly increased (about 35%) due to the reduction of water cement ratio. While in case of trial 4, in which powder was added to concrete in form of granules, the strength of concrete was insignificant different with that of controlled one.

However, a further investigation should be done to verify the effect of coating layer with the elapse of time and under different exposure conditions, such as water submersion, relatively high humidity and temperature, etc.

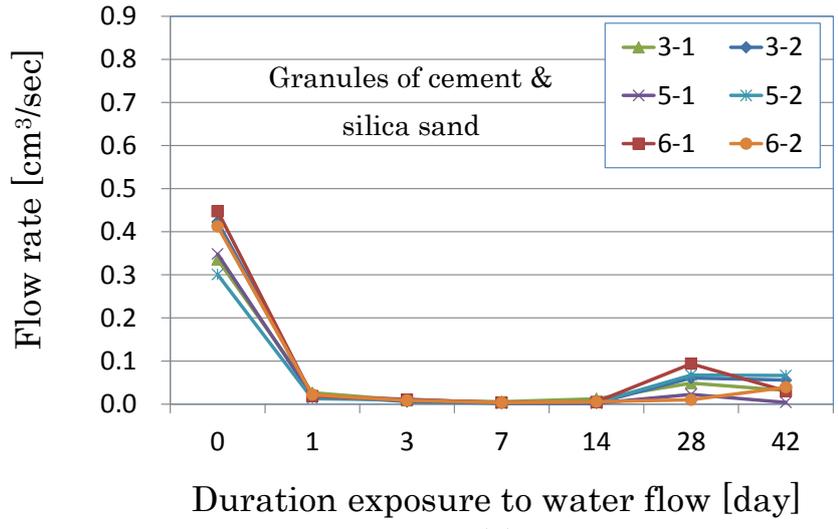
**(c) Result of water flow rate**

After sealed curing in air for two weeks, all the specimens were induced a penetrated crack by applying a splitting load. Then, it took one week to prepare the specimens readily subjected to continuous water supply. At this time, the crack width at the bottom of specimen was controlled (0.32-0.39)mm and each trial had two specimens for water pass test. Water was supply continuously to concrete specimens until 42 days testing.

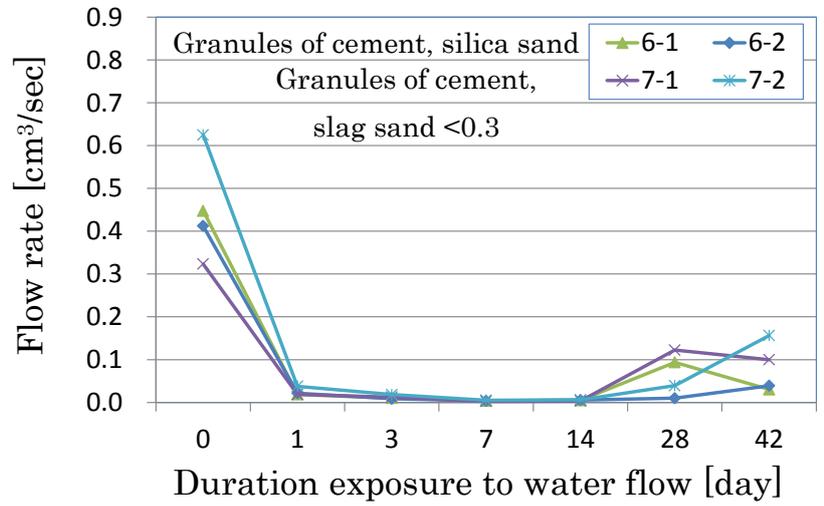
The results of time-dependent water flow rate for each specimen can be seen in *Figure 5.13*.



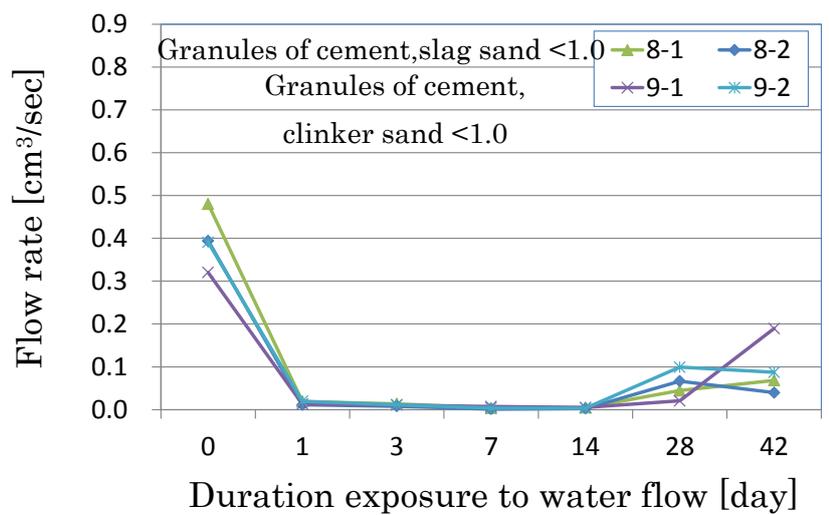
TRIALS TO ENHANCE SEMI-CAPSULATION TECHNIQUE FOR CEMENT/POZZOLAN



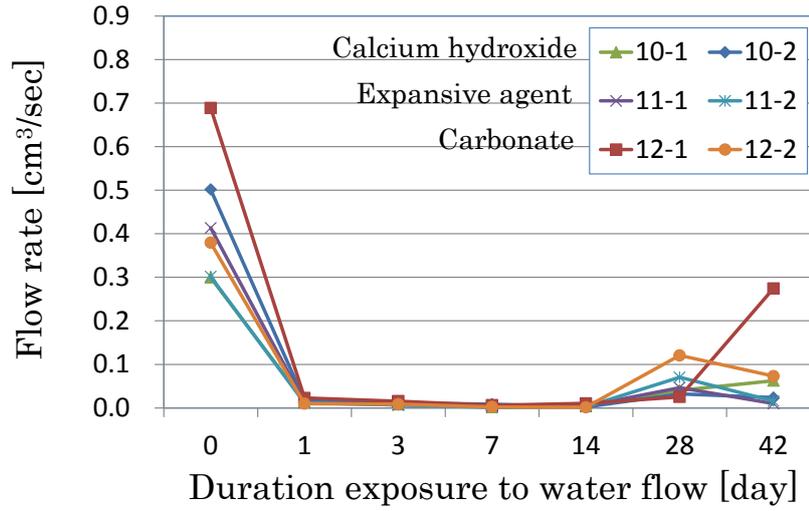
(b)



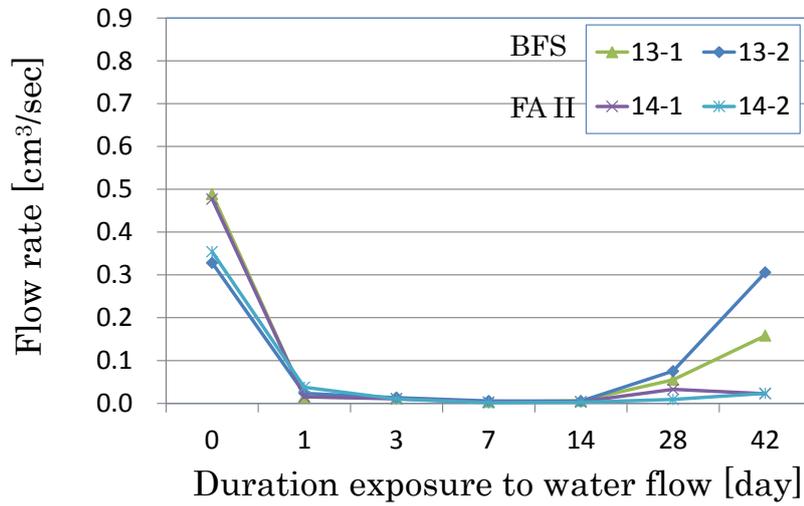
(c)



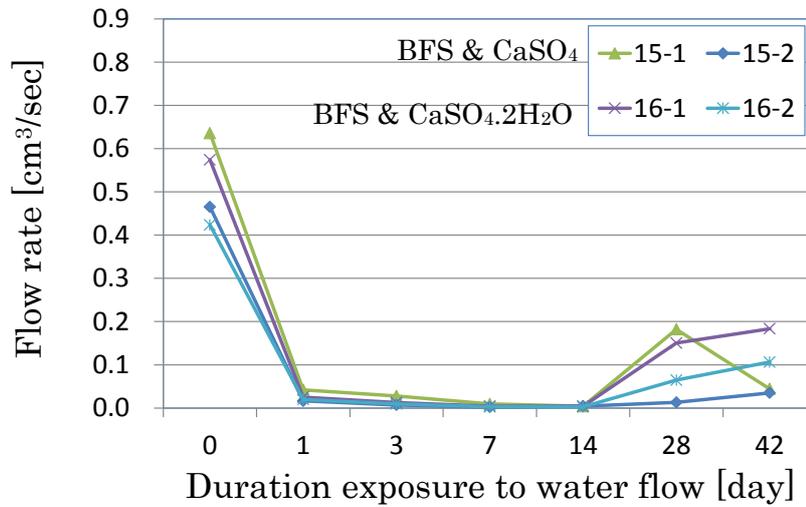
(d)



(e)



(f)



(g)

Figure 5.13 Time-dependent water flow rate through a crack

As can be seen in *Figure 5.13*, the same tendency of water flow through a crack could be observed with regardless of the types of materials used. In the first stage, the flow rate was decreased significantly due to the saturation and densification of the concrete matrix. Then the water flow became stable until two or four weeks exposed to continuous water supply. After that it was found that there was a increase in the flow rate in all trials. However, depending on the types of granules used and the initial values of water flow rate and crack width, the water flow would be changed with different rate. The possible reason was due to the relative large crack width of the crack in concrete, some components or healing products would be washed away or lost with the water flow.

Based on the results, it can be seen that granules using silica sand and slag sand <1.0mm showed better results in case of using fine sand as nuclei for granulation. Moreover, the use of functional ingredients such as Calcium hydroxide (easily dissolve), expansive agent, fly ash type II and compound of blast furnace slag and anhydrous calcium sulfate also brought promising effect in reduction of flow rate. However, further investigation should be performed to verify the effect of each ingredient.

In general, even though it was observed several healing product deposit at the crack surface (at the bottom and along the height of concrete specimens), the self-healing performance of concretes incorporating those granules was still not as good as expectation.

Moreover, there was no significant differences in self-healing performance between normal concrete and self-healing concretes when a crack was induced to concrete specimen at a young age.

## 5.6 SUMMARY

(1) Based on the proposed requirements and basic design concepts of granules having semi-capsulation effect, several trials have been conducted in the laboratory with an attempt to implement this effect in this stage. There

were a varieties of granules fabricated by different functional powders and granulation techniques. However, it still has not obtained the satisfied result yet.

According to the obtained results of fresh properties and compressive strengths of concretes in this stage, it can be found that there was an improvement in slump loss and strength variance between controlled and self-healing concretes using granules by introducing a waterproofing property to coating layer of granules. Especially in those trials cast in April 2013, an effective coating layer was induced to granule by using early hardening strength cement and superplasticizer (a kind of water-reducing agent). Moreover, a better distribution of granules in concrete matrix was expected because the size of granules was controlled as smaller than that of in the past. It indicates that the performance of granules will depend on the quality of inner material.

Up to this stage, it could be identified that there was a high quantity of cement or pozzolan used as the main ingredient contributing to the self-healing effect of granules. Due to chemical reactions between cement/pozzolan with water, there was a high possibility that the inner material would be hardened inside granules when exposed to water. That meant inner material could not fully satisfy its requirement: self-healing material was difficult to release into the crack surface. As a result, the healing capability of concrete incorporating granules was reduced significantly.

It can be concluded that both the requirements of the coating layer and inner material of granule should be satisfied so that the healing performance of concrete embedding granules having semi-capsulation effect will be substantially improved.

## **(2) Self-healing ingredients**

### **(a) Selection of inner material**

At the beginning of this stage, the strategies to improve self-healing performance via selecting functional ingredients were proposed as seen in *Figure 5.4*. At that moment, it was thought that the self-healing effect of granules were mainly contributed by the hydration of cement and/or pozzolanic reactions of supplementary cementitious materials. However, based on the experimental results and also due to immature technique of granulation, the quantity of cementitious or supplementary cementitious material should be reduced and acts as an additional component of inner material. In other words, when the technique is still under development, the amount of those materials should be small enough to provide enough strength for the granule to withstand the impact of coarse aggregates while mixing and casting concrete.

On the other hand, self-healing additives (proposed by AHN) in terms of swelling, expansion and precipitation seem to be preferable to use as inner material. These agents may be weak enough and also easy to diffuse into crack surface. Therefore, it is expected that the combination of using self-healing additives (proposed by AHN) and semi-capsulation technique (developed in this study) may bring to better performance of self-healing concretes. And the inner material should be designed as following proposal (*Figure 5.14*).

The basic idea for designing inner material is inner material should not be hardened even if exposed to water. In the future, further improvements in techniques should be developed so that it is possible to make the inner material of other materials, such as Portland cement or pozzolan, etc. ,weak enough to be easily released.

At this moment, the spreading effect of self-healing material has not verified yet and there was no evident about it in healing process. However, this function is preferable to introduce to inner material so that it can fulfill the requirements of semi-capsulation effect. Further investigation should be performed.

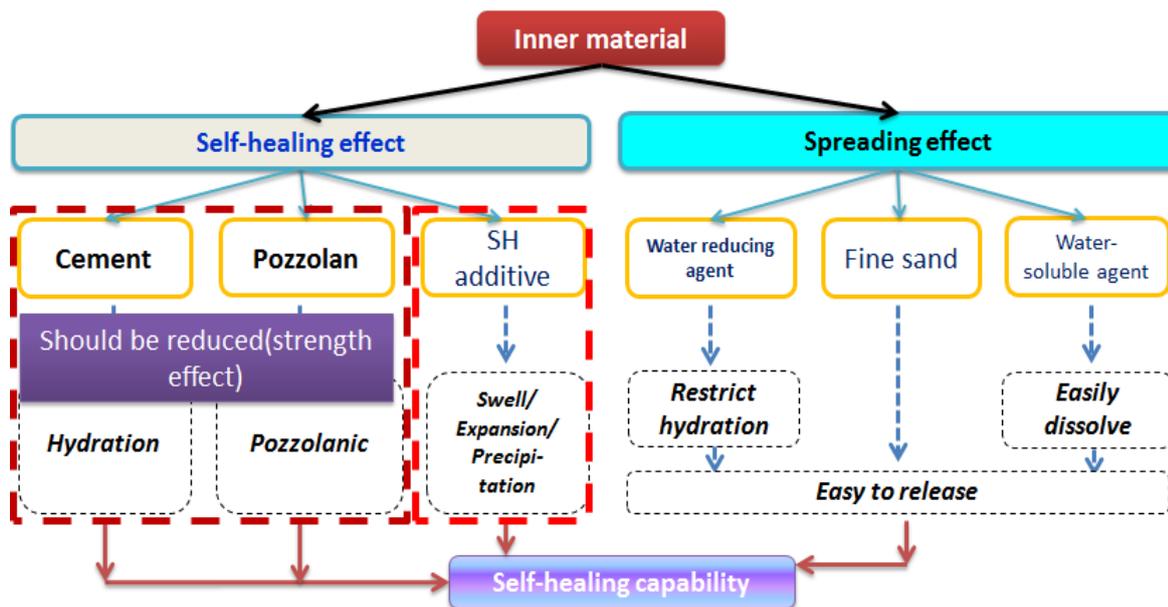


Figure 5.14 Changes in selecting the ingredients of inner material

**(b) Selection of coating material**

Throughout the trials in the 2<sup>nd</sup> stage, it was identified that using early high strength cement (or rapid hardening material) and water-reducing admixture could create an effective outer layer of granule. The resultant layer has sufficient thickness and strength (good bonding with concrete matrix) to protect the granule from unexpected events (or to reduce the possibility of broken granules) during mixing and casting concrete. In addition, by using these materials a dense outer shell was formed that also brought to a waterproofing effect.

Another point is, it is possible to introduce a waterproof property to the coating layer by using waterproofing agents. However, due to its high cost and an insignificant improvement in healing performance, it seemed that waterproofing agents were not considered as a good selection.

**(3) Granulation technique**

Besides improvement in selecting effective self-healing ingredient (aspect of chemical effect), the granulation technique, kind of physical treatment, for powder material should be considered. It was identified that only using good

self-healing ingredient or only applying good physical granulation technique, the excellent performance of granules will not be obtained. They are both important and should be simultaneously satisfied to achieve a successful self-healing granules having semi-capsulation effect. In this stage, there was an effort to enhance the semi-capsulation technique to store self-healing ingredients based on a clearer concept and requirements for coating layer and inner material.

In order to improve the distribution in concrete matrix and also increase the self-healing efficiency, finer granules are preferable to use. The approach of using steel balls to control the size of granules could fabricate smaller granules. However, due to its tough work and high possibility to break the granules during preparing process, it was considered as not a good option.

Another method showing a promising effect was using fine sand as nuclei for granulation process. Once mixing inner material in form of powder with fine sand and sufficient liquid (by using water-reducing agent), it was found that it was easier to form the shape of granules in small size. Even though the effect of using fine sand has still not been verified yet in this study, it looks potential due to its own advantages: physical (fine, spherical shape, loose) & chemical properties (pozzolanic or cementitious property).

**(4) Based on the proposed concepts and techniques, it can be explained the reasons why the concrete trial using granules SH-Mix1-1 in 1<sup>st</sup> stage showed the best healing performance.**

As seen in the 1<sup>st</sup> stage, trial using granules of Portland cement compound; self-healing additives in terms of swelling, expansion, precipitation; and a special material possessing both water-reducing and water-soluble effect, showed good results in both reduction of water leakage and crack closing process. Moreover, it also has a potential to reverse the healing capability for long-term (at least one year). However, at that time there was no clear concept to explain the result. Based on current knowledge and further study in the 2<sup>nd</sup>

stage, it can be explained well now.

Firstly, the inner material of granule SH-Mix1-1 were composed of such materials that provide both self-healing effect and spreading effect to granule as shown in **Table 5.4**. For example, Portland cement compound of low-heat (LC) and high-early strength cement (HC) and self-healing additives can contribute to self-healing capacity. Moreover, a special material having both water-reducing and water-soluble effect was added to inner material as a component to reduce the strength of core and make it easily to release into the crack surface.

**Table 5.4 Ingredients of granule SH-Mix1-1**

Ingredients	Percentage [%]	
	Inner	Coating
Portland cements (HC&LC)	37.63	16.13
SHA & Water-soluble agent (water reducing agent)	37.63	-
Liquid	5.38	3.23

In addition, it is believed that another reason, related to the granulation technique, also makes the inner material weak enough to be easily ruptured and dissipated out of the granules. In that time, inner material were manufactured by gradually adding the self-healing powder and spraying the liquid in the roller mixer (drum mixer) as seen in **Figure 5.15 (upper-left)**. Due to this procedure, it was assumed that flocs of self-healing powder would be loose or weak (just pointed or small area of surface contact with each other) so that it is easier to be broken and diffuse out of the granule (**Figure 5.15-lower**).

Secondly, an effective coating layer was introduced to granules by using steamer as seen in **Figure 5.15 (upper-right)**. Under the effect of steaming: high temperature & moisture, it was expected that a dense & hardened coating layer would be formed by the hydration products of cement compound. This layer will protect the weak inner material and also create a barrier that makes it difficult for water to pass through.

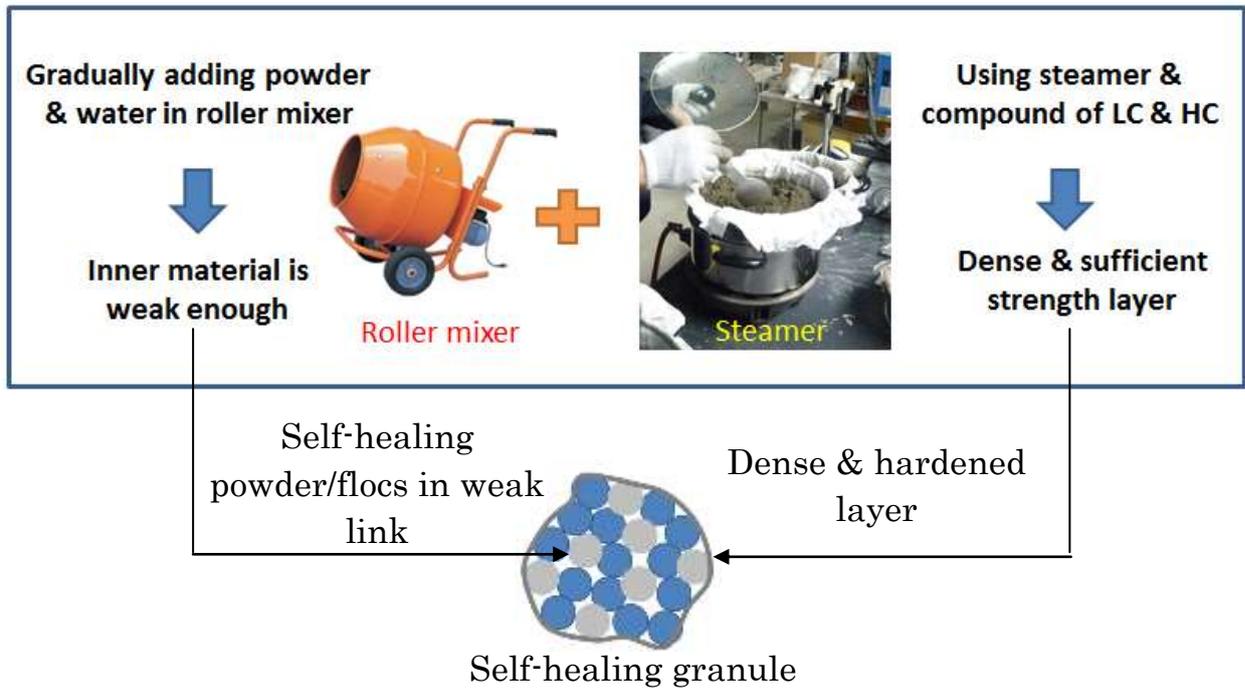


Figure 5.15 Granulation techniques to manufacture granule SH-Mix1-1

Thirdly, due to the small size of granules (as can be seen in *Figure 5.16-right*: about 80% of granules could pass the sieve of 2.36mm), it was believed the distribution of granules in concrete matrix became better. The higher possibility that a crack ruptures the granules, the higher healing capacity of concrete it is.

Sample of granule SH-Mix1-1



Particle size distribution of SH granules

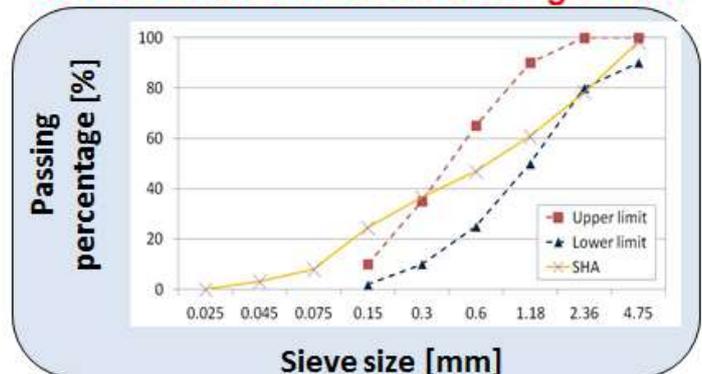


Figure 5.16 Sample (left) & particle size distribution of granule SH-Mix1-1 (right)

6-FIELD TRIAL-

**CHAPTER 6**

APPLICATIONS IN SLAB

MOCK-UP

## 6.1 PURPOSE

The purpose of this chapter was to investigate the applicability and the real healing performance of self-healing concrete incorporating pre-fabricated granules, which were designed based on the basic design concepts proposed in this study (*Chapter 5*), in real structures- a portion of slab mock-up in Chiba experimental station, the University of Tokyo.

## 6.2 PROCEDURES

### 6.2.1 Step 1-Manufacture & cure self-healing granules in the laboratory

*Table 6.1* showed the self-healing ingredients used to manufacture the granules.

**Table 6.1 Self-healing ingredients in percentage**

Ingredient	Inner material						Coating material	
	W	Water-soluble (water-reducing)	LC	EA	CH	Silica sand	HC	W
Percentage [%]	7.5	7.5	16.7	16.7	16.7	16.7	16.7	1.7
Notification		Easily dissolved & weak material	Hydra-tion	Expansion	Dissolved agent	Nuclei	Rapid hardening	

(*Note:* W-tap water; LC-Low-heat Portland cement; EA-Expansive agent-CH-Calcium hydroxide; HC-Early high strength Portland cement)

Self-healing granules were manufactured in advance in laboratory by using a normal concrete mixer, as following: firstly, mix all inner ingredients and then add early high strength cement and water to concrete mixer in order to introduce a coating layer to granule (*Figure 6.1-left*).

After granulation process, pre-fabricated granules were stored in a plastic bag for curing to ensure the effectiveness of coating layer (*Figure 6.1-right*) and then transported to the construction site.



Figure 6.1 Pre-fabrication of granules and curing condition

6.2.2 Step 2- Prepare a ready-mixed concrete at a plant and then transport it to construction site by ready-mixed concrete truck (Figure 6.2-left)

6.2.3 Step 3- Add pre-fabricated granules to the drum of ready-mixed concrete truck and mix, then discharge and cast the portion of slab mock-up

Just before casting, pre-fabricated self-healing granules were added to the ready-mixed concrete truck and mixed together with ready-mixed concrete in the drum agitator (Figure 6.2-right). It was expected that the granules would be well distributed in concrete matrix and restrained the possibility of broken granules during mixing concrete.



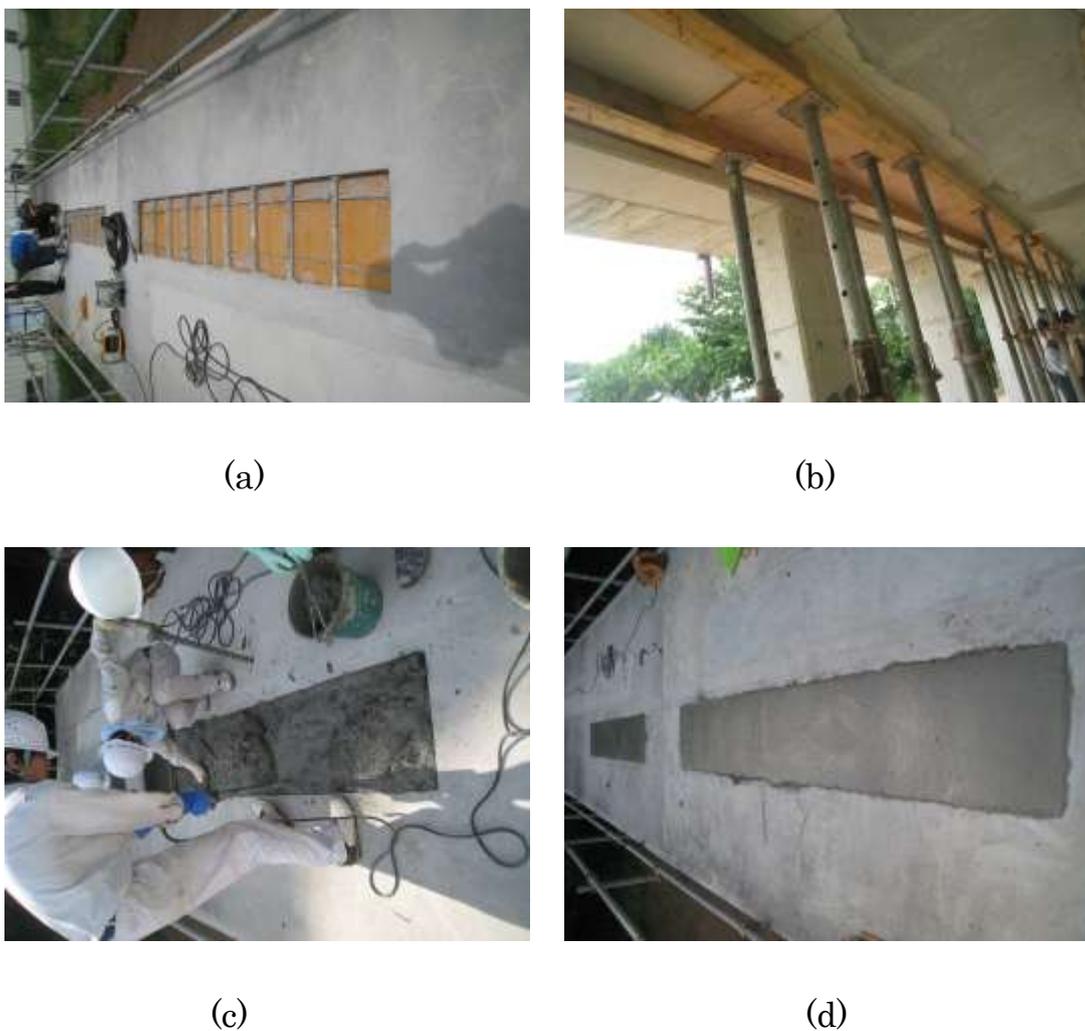
Figure 6.2 Ready-mixed concrete truck & addition of granules into drum agitator

After discharge from a drum agitator, the affect of inclusion of granules into

concrete mixture were examined by doing the slump test and air content measurement of fresh concrete (*Figure 6.3*). And then, cylindrical concrete specimens and a portion of slab were cast (*Figure 6.4*).



**Figure 6.3** Investigations on the fresh properties of concrete using granules



**Figure 6.4** Concrete casting for a portion of slab mock-up

### 6.3 FUTURE WORKS

After finish casting, the whole structure was exposed to surrounding environment for a period of time: under wet/dry conditions, changes of temperature, etc. Later on, several cracks will be induced to the portion of slab by using a jack and then long-term investigation of crack healing capability of concrete will be done.

It is expected that this self-healing granule approach will provide a wide range of applications in practice depending on the required self-healing performance, manufacturing cost, exposure conditions and types of structures, by improvement of the granulation technique, selection of self-healing ingredients and dosage.

One of advantages of this approach is that concrete incorporating granules can preserve the healing capability as long as the granules would be ruptured by a crack penetration and exposed to water. Moreover with a smaller crack width and small flow rate of water through a crack, higher possibility for a crack healed itself. On the other hands, under such conditions: larger crack width, high water pressure, high water flow rate and relatively flat crack plane, the healing capacity of concrete will restrained.

It is important to point out that the healing performance of granules in concrete is affected by the quality of granulation process (techniques and ingredients), its compatibility of granules with concrete matrix, and the possibility of broken granules due to crack appearance (healing process is only activated when a crack ruptures granules and water is available at crack).

Furthermore, other issues should be considered and investigated before apply this technique to construction industry are the thermodynamic/chemical conditions of granules and cracking patterns or crack initiation of concrete/reinforced concrete in different structures, under various exposure conditions, etc.

# CHAPTER 7

7-CONCLUSIONS,  
RECOMMENDATIONS  
AND  
FURTHER RESEARCHES

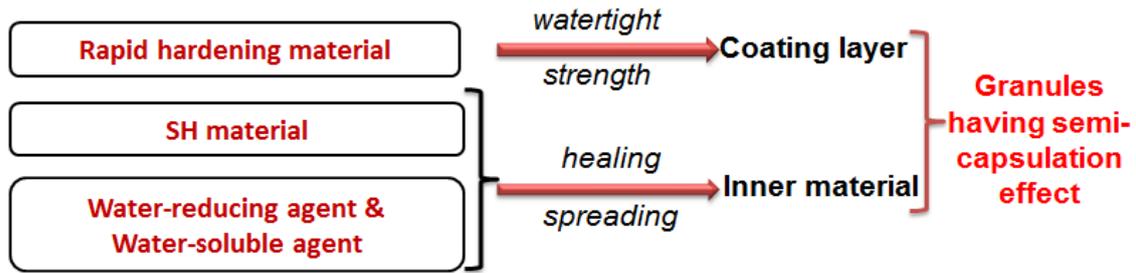
## 7.1 CONCLUSIONS

In this research, the normally used water-cement ratio concretes were designed with the self-healing capability in terms of reduction of water flow rate through a penetrated crack and crack-closing process by incorporating the granules of cement replacement materials and/or Portland cement and other additives, considered as a kind of engineered self-healing approach.

Even though the optimum ratio of effective ingredients of granules still have not been confirmed yet and it is still difficult to verify the self-healing performance in concrete, the requirements and basic design concepts for granules having semi-capsulation effect were established. The basic ideas are embedded granules should be broken by a penetration of crack and inner self-healing material should be released into crack surface. In order to obtain the aboved targets, both requirements of coating layer and inner material should be satisfied simultaneously: coating layer is strong and dense enough; inner material containing reactive agent is weak enough and easy to spread away out of the granule. Based on these proposed concepts, it is expected that self-healing performance of concrete using granules having semi-capsulation effect will be significantly improved

It was found that concrete incorporating granules of the functional ingredients has no adverse effects on the properties of concrete such as the workability of fresh concrete and the compressive strength of hardened concrete; has a possibility of recovering the water tightness property of cracked concrete, especially in young concrete; and can preserve the healing ability of concrete for a long period of time.

In order to achieve the granules having semi-capsulation effect, it was proposed that following materials should be used as self-healing ingredients (*Figure 7.1*).



**Figure 7.1 Functional ingredients in granule**

Granules can be manufactured by a conventional simple granulation method (a common approach to make capsules for polymer materials or used in food/medicine industry), however in order to possess a semi-capsulation function, it is necessary to develop semi-capsulation technique for powder materials. Throughout this study, it can be concluded that it is feasible to fabricate finer granules to improve the distribution of granules in concrete matrix and healing capability of concrete with the considerations of simple & cheap fabrication of granules. 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 by using a typical mortar/concrete mixer for granulation process and using fine sand as nuclei for granulation.

Finally, it is important to bear in mind that both self-healing ingredients of granule (term of chemical effect) and granulation technique (term of physical effect) are paramount importances to ensure the performance of concrete incorporating granules having semi-capsulation effect.

## 7.2 RECOMMENDATIONS

Several trials have been done to investigate the self-healing performance of concretes using various types of granules through the water pass test. In this study, a penetrated crack was introduced to concrete specimen and then after binding again, concrete specimen was exposed to continuous water supply with the ranges of crack width of 0.2-0.4mm. It was considered as severe

conditions.

Some recommendations to improve the experiment of water pass test were proposed, as follows:

First, due to mentioned-above severe conditions, water flow rate through a crack should be restrained as small as possible to stimulate the healing process. It is a fact that water flow rate through a concrete crack is very complicated phenomenon. There are various factors influencing the characteristics of flow rate, such as crack width, roughness of crack surface, saturation degree of crack surface, pressure of water head, temperature, materials used, etc. Especially, the water flow through a cracked self-healing concrete was thought more sophisticated. Even though with successful granules, the healing effect of concrete is unsecured if concrete specimen is under continuous water supply, a relatively large crack width and less roughness of crack plane. In that case, when crack ruptures embedded granules, self-healing additives will be released into crack surface and chemical reactions producing new product will be activated once contact with water flow. However, newly form products will be washed away with the flowing water.

In another word, there was a relationship between water flow rate and crack healing capability. Due to this fact, the characteristics of water flow though a crack in conventional concretes with/without incorporating granules and in low water-cement ratios or high strength concrete with/without granules will be different (as can be seen in Appendix 4). Moreover, under different water supply (static/ continuous water flow/wet&dry conditions), the performance of healing capability of concrete are also affected (as can be seen in Appendix 4).

Second, it should be careful to control the loading rate when inducing a crack to conventional concrete specimen with/without using granules. Because it will affect the crack pattern and initial conditions of crack surface,

the appearance of broken granules on crack surface, considered as important factors influencing the results of water pass test.

### 7.3 FURTHER RESEARCHES

In this study, even though there was a improvement in the performance of self-healing granules, the final target has still not obtained yet. Further deep investigations should be done:

+ To identify the functional effective ingredients for granules.

In this research, the combination of self-healing material proposed by Ahn,2008 (as inner material) and enhanced semi-capsulation technique (to induce an effective coating layer) showed the promising performance. However, based on new proposed concepts and further improvements in granulation technique , there is a high potential to use other materials, such as commercially available Portland cement or supplementary cementitious materials that possessing self-healing properties, instead of materials proposed by Ahn. The key point is to make the reactive materials weak enough and easy to release.

There are several promising techniques needed to investigate, such as:

- Using roller mixer (drum mixer): to make a weak link (bonding) among flocs of powder material.
- Using admixtures such as water-reducing or water-soluble agents
- Using microbubble techniques: to make the inner material more porosity, etc.

+ To confirm the self-healing performance of concrete incorporating granules

under various exposure conditions.

In this study, the self-healing performance of concrete was investigated through observation of water flow rate through a static penetrated crack and crack closing under continuous water supply. However to apply this material in real structures, further test with different exposure conditions and types of structures should be performed, such as wet/dry conditions, high pressure water, water submersion, chloride environment, dynamic crack, high temperature environment, period of crack appearance and exposed to water, size or dimensions of specimen, crack pattern or crack initiation, etc.

+ To develop techniques to verify self-healing performance of concrete using granules.

The influences of adding granules to concrete mixture were examined based on the performances of the workability of fresh concrete, compressive strength of hardened concrete and crack-healing capability through water pass test. However, it is necessary to do deeper investigations on the time-dependent properties of slump loss, strength variance and heat generation and healing capability.

One of the new contributions to develop self-healing granules having semi-capsulation effect in this research was to provide spreading effect to inner material. Unfortunately, there was no clear evident about this effect or also other other effects of granule. There should be a standard procedures to examine or verify the effectiveness of granules in each step of application: from granule fabrication process, casting concrete and in hardened concrete.

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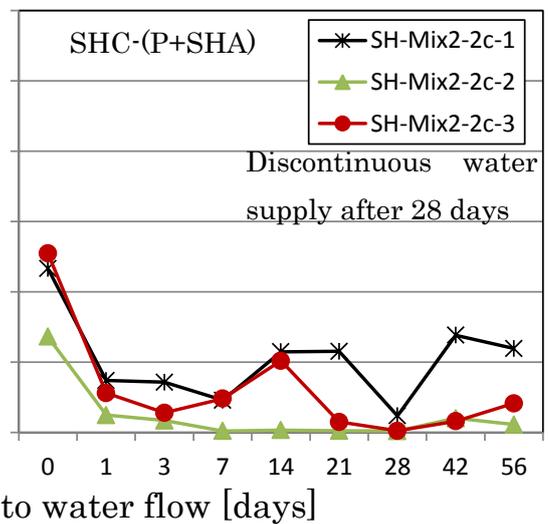
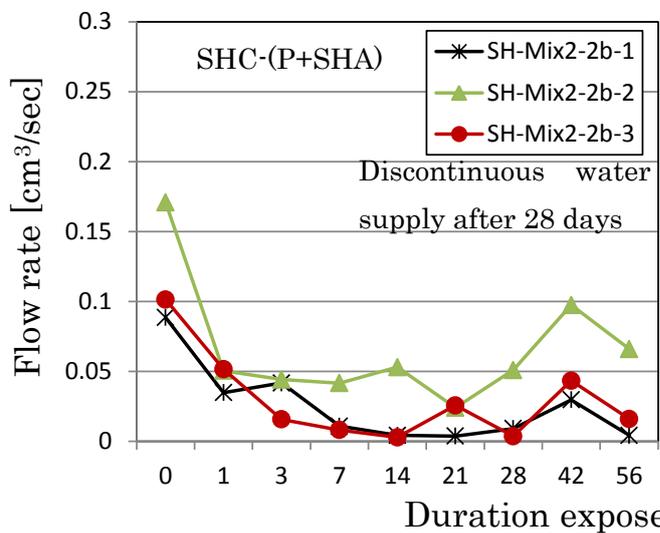
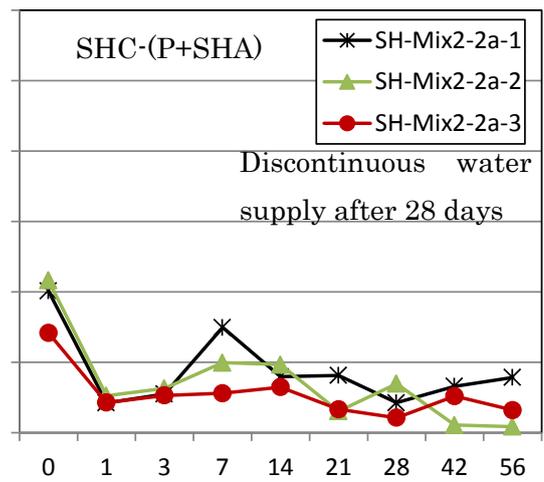
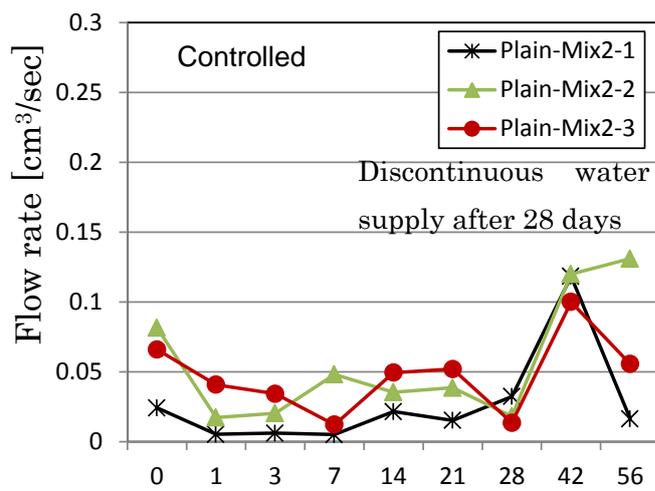
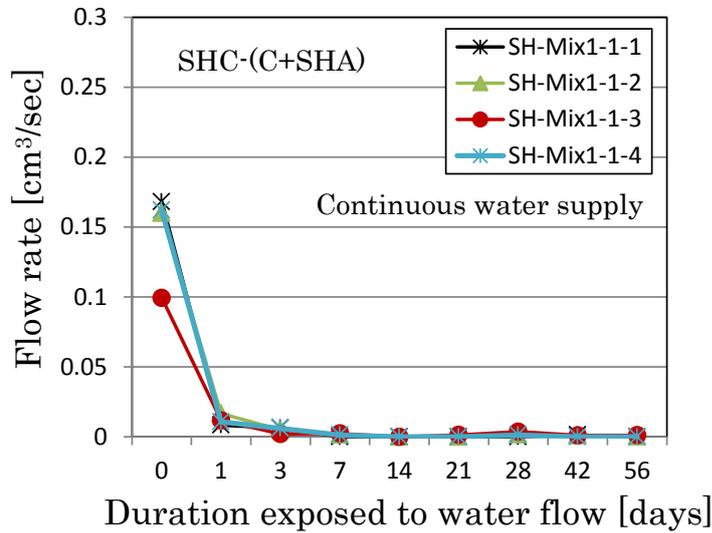
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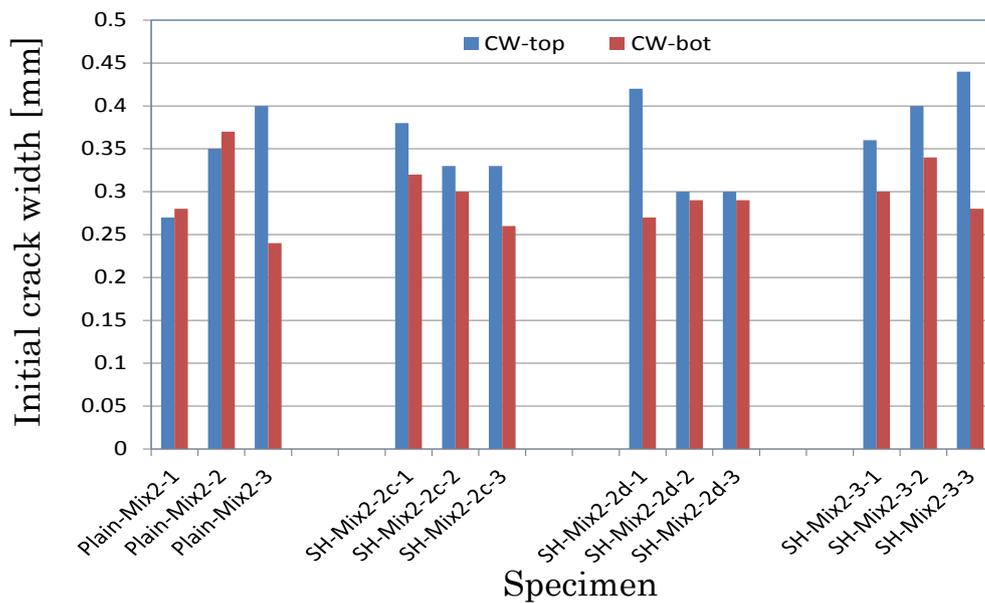
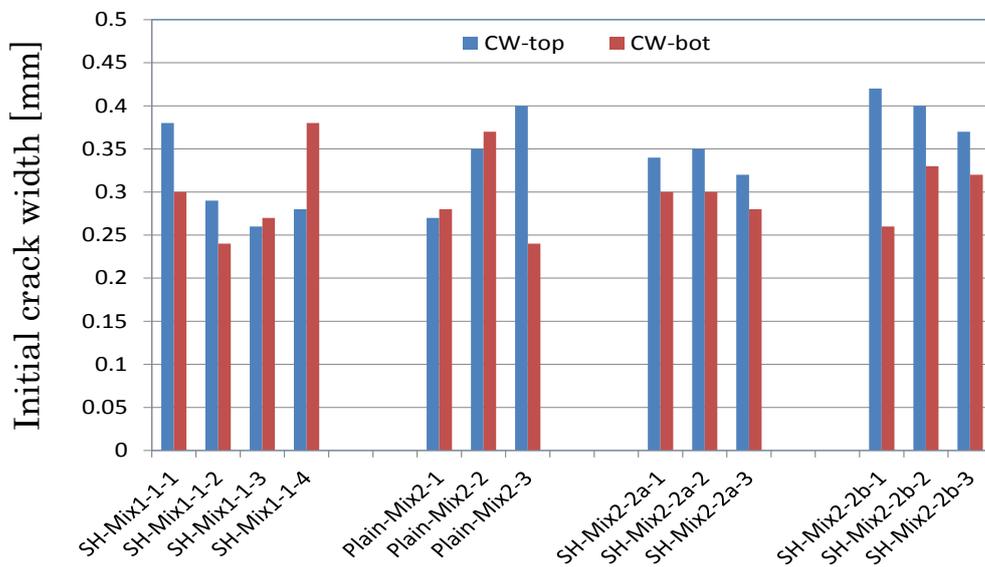
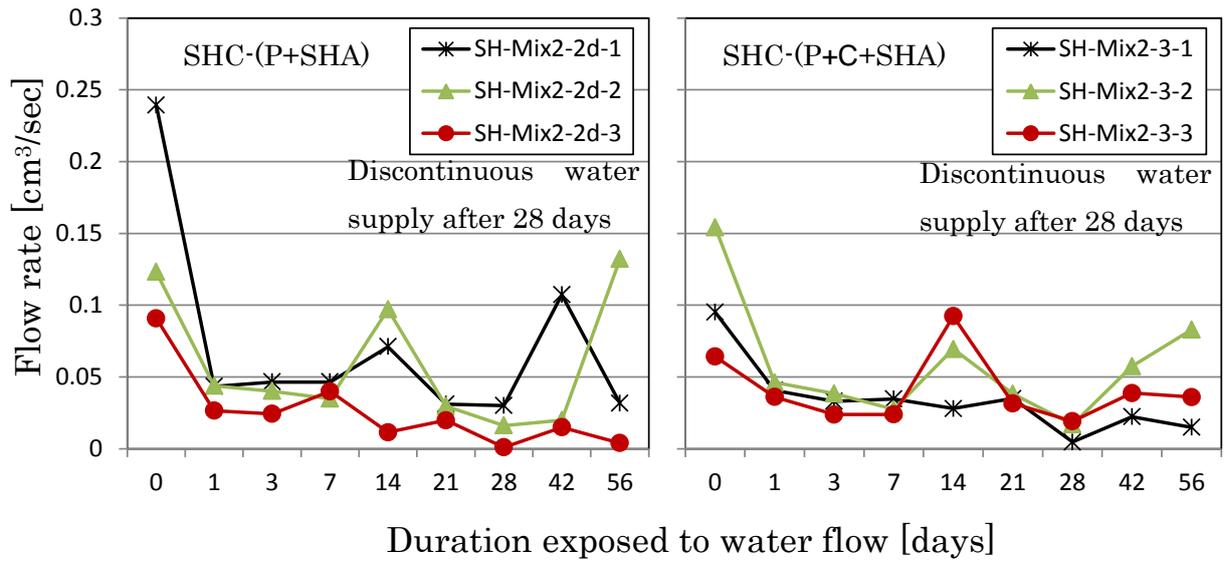
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## APPENDIXES

1. WATER FLOW RATE & INITIAL CRACK WIDTH AT THE TOP & BOTTOM IN EACH SPECIMEN FOR EACH TRIAL (Chosen trials in the 1<sup>st</sup> stage)





## 2. PERFORMANCE OF SELF-HEALING TRIALS CAST IN AUGUST 2012

### 2.1 Concrete mix proportions

Concrete	W/C [%]	s/a [%]	Air [%]	kg/m <sup>3</sup>					SP [Bx%]	AE [Bx%]
				W	C	SH	S	G		
Plain	49	42.3	4.5	171	349	-	802	953	0.4	0.02
SH	49		4.5	171	349	70	732	953		

### 2.2 Materials of granules for each trial & Fresh properties of concrete

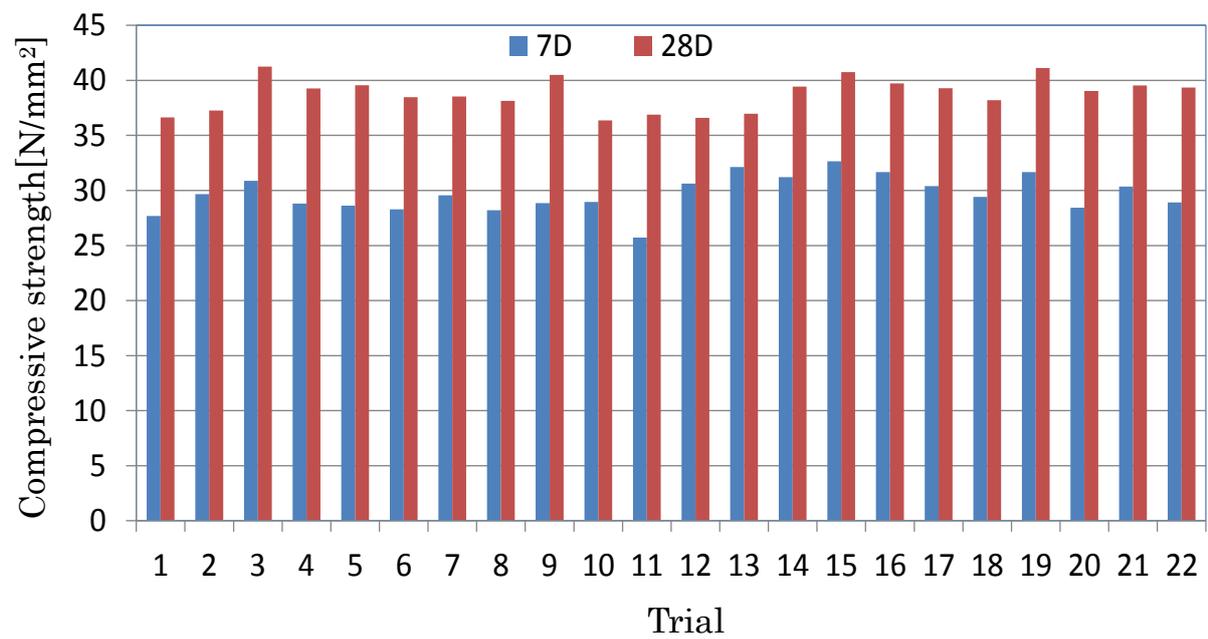
Trial	SH ingredients [%]	SP [Bx%]	AE [Bx%]	Slump [cm]	Air [%]	Temp [°C]
1	Plain	0.4	0.02	14	4.7	21
2	W15.3,HC21.2,LC63.5	0.6	0.02	17	5.5	21.1
3	Liquid15.3,HC21.2,LC63.5	0.5	0.01	13	5	21.7
4	Granules in Trial 3+steamer			13.5	5.1	21.7
5	Liquid15.2,W7.6,HC19.3,LC57.9			13.5	5.4	21.1
6	Granules in Trial 5+steamer			14.5	4.5	21.3
7	Liquid17.4,HC17.2,LC51.5,AD13.9			11.5	3.5	21.4
8	Granules in Trial 7+steamer			13.5	3.8	21.8

9	Liquid16.7,W8.3HC15,LC45,AD15			12	3.7	21.6
10	Granules in Trial 9+steamer			13	3.8	21.8
11	Similar ingredient as SH-Mix1-1			13.5	3.7	21.4
12	Granules in Trial 11+steamer			11	3.6	21.7
13	Liquid10.9,W3.6HC31,LC54.5			13.5	4.5	21.8
14	Granules in Trial 13+steamer			15	4.2	22.2
15	Liquid,W,HC,LC,BFS,SF,AD			12.5	5.2	21.9
16	Granules in Trial 15+steamer			14.5	4.5	21.4
17	Liquid,W,HC,LC,BFS,SF,AD			13	4.3	21.4
18	Granules in Trial 17+steamer			14	5	21.7
19	Liquid,W,HC,LC,BFS,SF,AD			12.5	4.6	21.6
20	Granules in Trial 19+steamer			13.5	4.5	21.4
21	Liquid,W,HC,LC,BFS,SF,AD			12.5	4.5	22.1
22	Granules in Trial 21+steamer			14	5	21.9

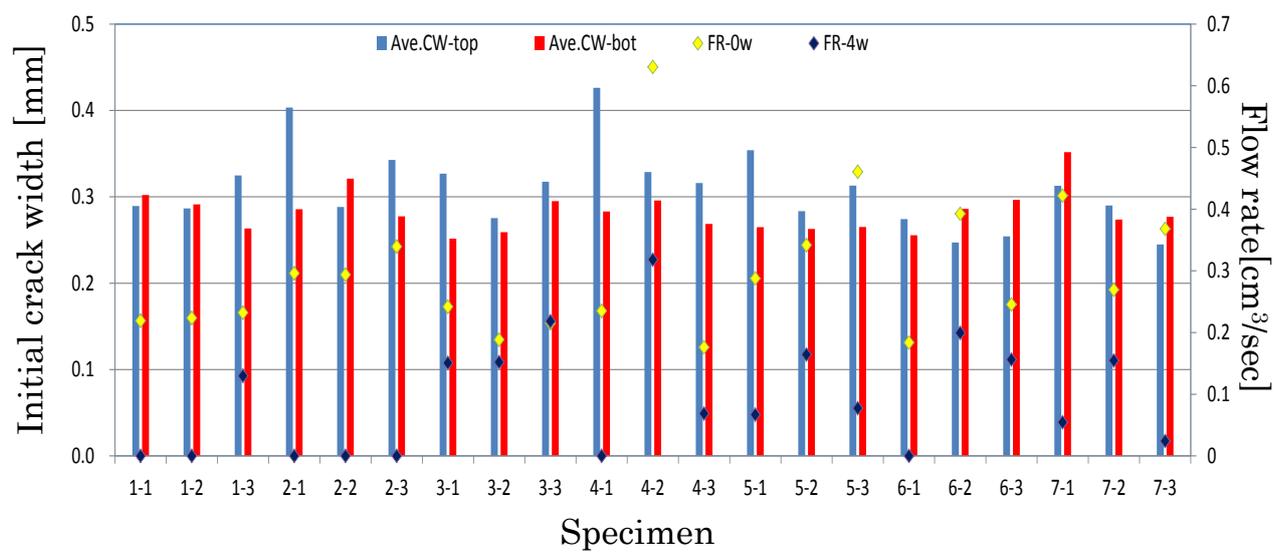
Trial 15-22: two-step granulation technique

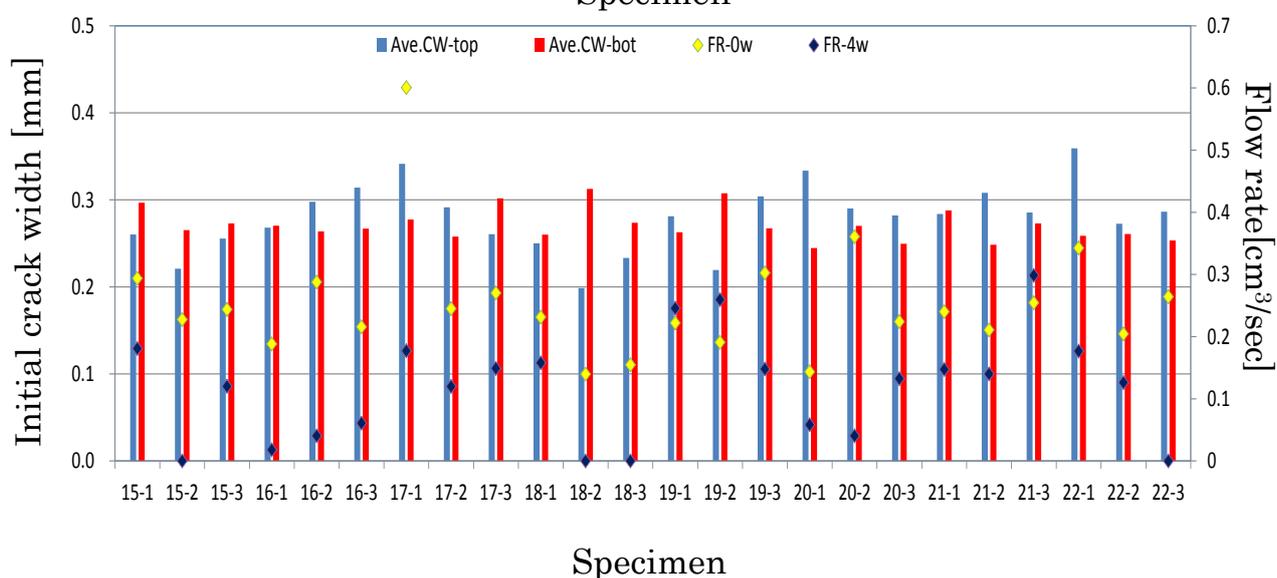
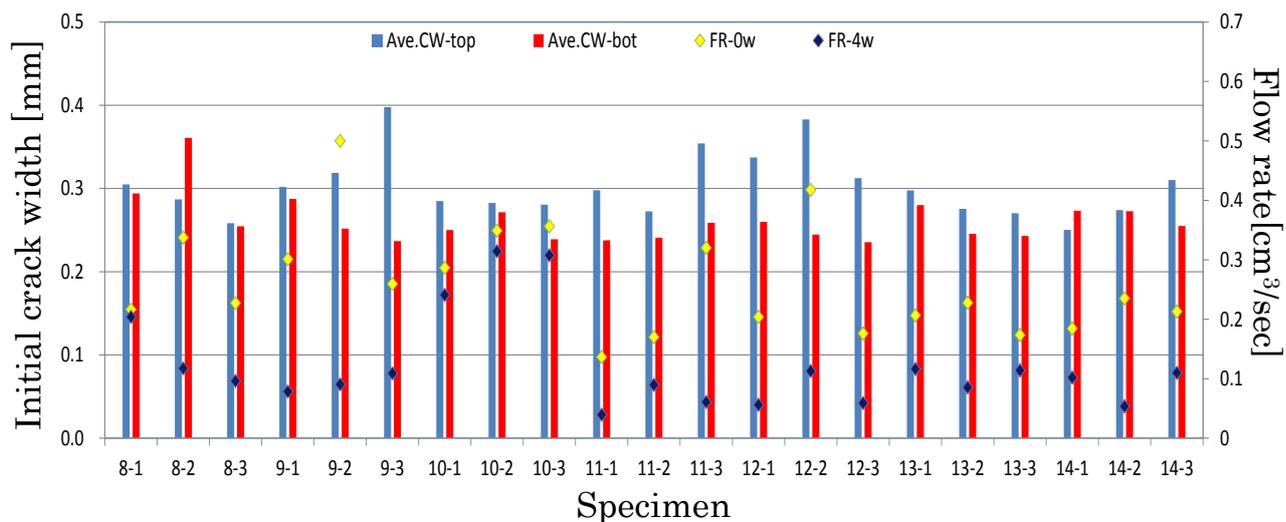
Note: HC-Early high strength cement; LC-Low-heat cement; W-tap water; BFS-Blast furnace slag;SF-Silica fume;AD-specific additives

### 2.3 Compressive strength



### 2.4 Results of water pass test





### 3. PERFORMANCE OF SELF-HEALING TRIALS CAST IN NOVEMBER 2012

#### 3.1 Concrete mix proportion

Concrete	W/C [%]	s/a [%]	Air [%]	kg/m <sup>3</sup>					SP [Bx%]	AE [Bx%]
				W	C	SH	S	G		
Plain	55	46	4.5	168	306	-	848	949	0.9	0.01
SH	55		4.5	168	306	70	778	949		

### 3.2 Materials & fresh properties of concrete

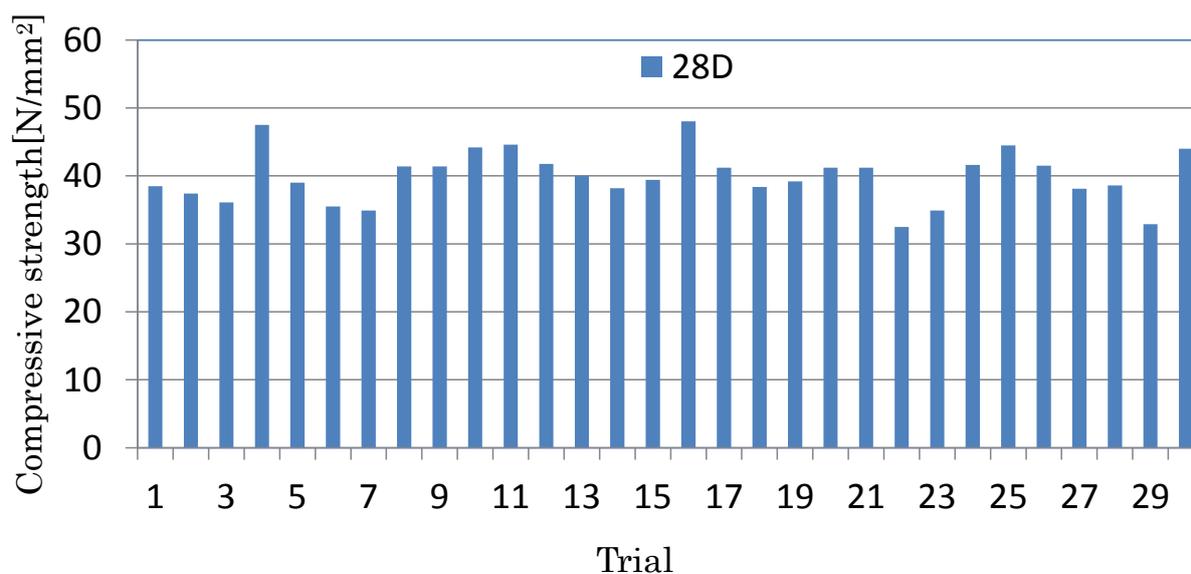
Trial	SH ingredients [%]	SP [Bx%]	AE [Bx%]	Slump [cm]	Air [%]	Temp [°C]
1	Plain	0.9	0.01	18	3	19.3
2	W/C15,HC0,LC100	1	0.015	12	5.8	19.5
3	W/C15,HC12.5,LC87.5	1	0.015	11	5.8	19.6
4	Powder HC25,LC75, Calcium Stearate 3%	1.2	0.01	15	2.5	19.6
5	W/C15, HC25,LC75, Calcium Stearate 3% (mixed)	1.1	0.015	15.5	2.6	19.5
6	W/C15,HC25,LC75	1.1	0.02	15	5.9	19.7
7	W/C15,HC25,LC75, stearic acid+ethanol 10%	1.1	0.02	15	5.4	19.8
8	W/C15, HC25,LC75, Calcium Stearate 3% (coating)	1.1	0.02	13	4	19.7
9	W/C15, HC25,LC75, Liquid paraffin 10%	1.1	0.02	12	2.8	19.6
10	W/C15,HC37.5,LC62.5	1.1	0.02	9.5	2.7	19.8
11	W/C15,HC50,LC50	1.1	0.02	8.5	2.9	19.8

12	W/C15,HC75,LC25	1.1	0.02	14	4.1	19.6
13	W/C15,HC100,LC0	1.1	0.02	14.5	5	19.7
14	W/C17,HC25,LC75	1.1	0.02	16	4.9	19.6
15	W/C15,HC23.25,LC69.75,EA7	1.1	0.02	14	6	19.6
16	Powder HC22.5,LC67.5,EA10 Calcium Stearate 3%	1.2	0.02	15	2.5	19.9
17	W/C15,HC22.5,LC67.5,EA10 Calcium Stearate 3%	1.1	0.02	14	3	19.7
18	W/C15,HC22.5,LC67.5,EA10	1.1	0.02	16	5.1	20
19	W/C15,HC22.5,LC67.5,EA10, stearic acid+ethanol 10%	1.1	0.02	17	3.1	20
20	W/C15,HC22.5,LC67.5,EA10, Ca Stearate 3% (coating)	1.1	0.02	14	3.4	19.7
21	W/C15,HC22.5,LC67.5,EA10, Liquid paraffin 10%	1.1	0.02	13	2	19.8
22	W/C15,HC22.5,LC67.5,EA10, Aqua seal 10%	1.1	0.02	9.5	2.1	19.8
23	W/C15,HC22.5,LC67.5,EA10, Protect seal 10%	1.1	0.02	13.5	2.9	19.8
24	W/C15,HC22.5,LC67.5,EA10,	1.1	0.02	15	2.7	19.6

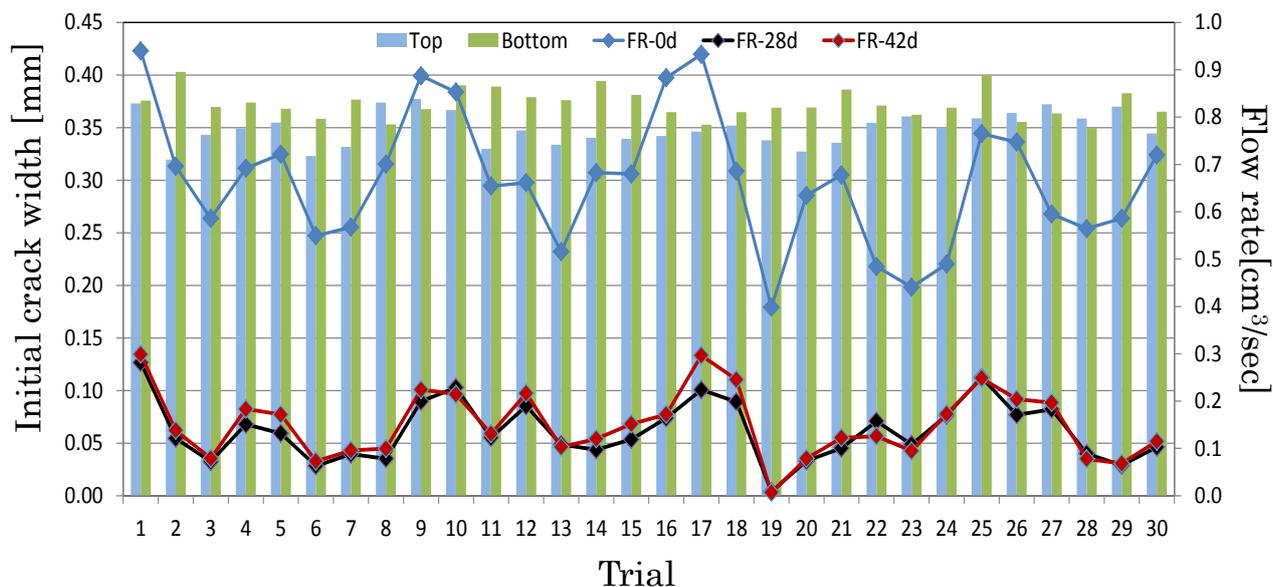
	Environ seal 10%					
25	W/C15,HC23.75,LC71.25,CaO5	1.1	0.02	7.5	2.5	19.8
26	W/C15,HC22.5,LC67.5,CH10	1.1	0.02	10	3.1	19.7
27	W/C15,HC20,LC60,CH20	1.1	0.02	12	5.1	19.7
28	W/C15,HC,LC,EA,AD	2	0.02	19	5.1	19.1
29	W/C15,HC,LC,EA,AD, acid+ethanol 10%	2	0.02	20	5.5	19.6
30	W/C15,HC,LC,EA,AD, paraffin 10%	2	0.02	20.5	2	19.5

Note: HC-Early high strength cement; LC-Low-heat cement; W-tap water; EA-Expansion agent; AD-specific additives; CH-Calcium hydroxide

### 3.3 Compressive strength



### 3.4 Result of water pass test



## 4. PERFORMANCE OF OTHER SELF-HEALING TRIALS CAST IN APRIL 2013

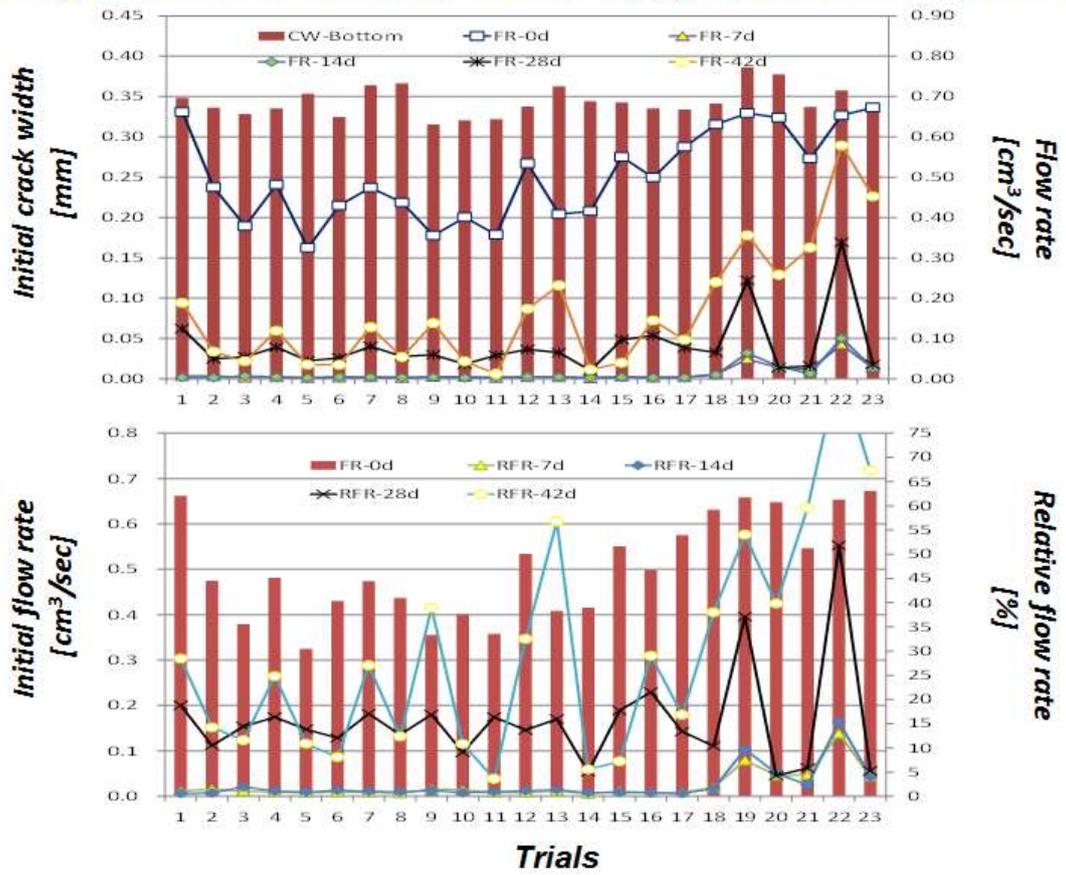
### 4.1 Mix proportion of concrete

Concrete	Trial	Mix proportion						
		W	NC	LS	SH	S	G	AD
SDC	17	175	389	-	-	842	868	6.6
	18	175	389	-	70	772	868	6.6
	19	175	389	-	-	842	868	6.6
	20	175	389	-	70	772	868	6.6
SCC	21	165	350	200	-	786	789	8.8
	22	165	350	200	39	747	789	8.8
	23	165	350	200	70	716	789	8.8

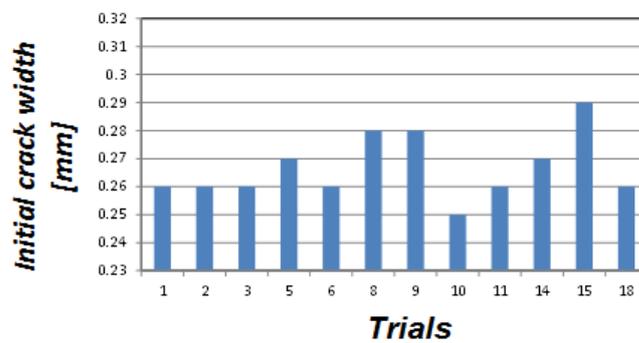
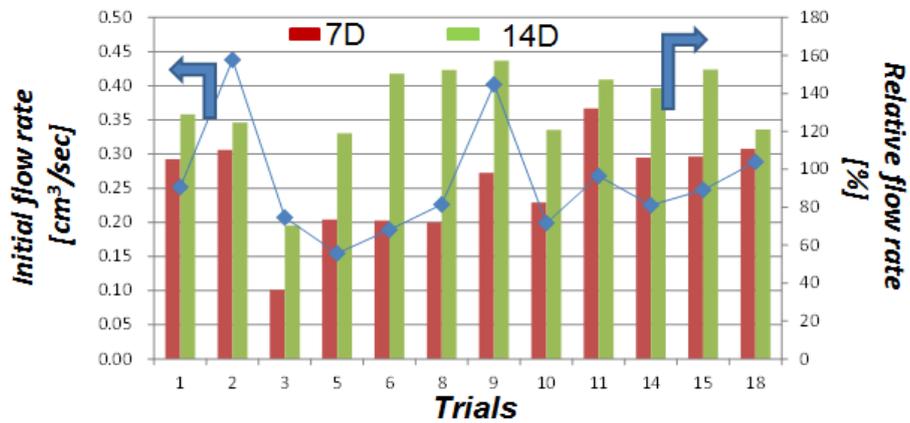
Note: LS-Limestone powder for strength restraint; SDC-Smart Dynamic Concrete which has semi-compactability with normal concrete's mix proportion; SCC-Self-Compacting Concrete

### 4.2 Results of water flow rate under different exposure

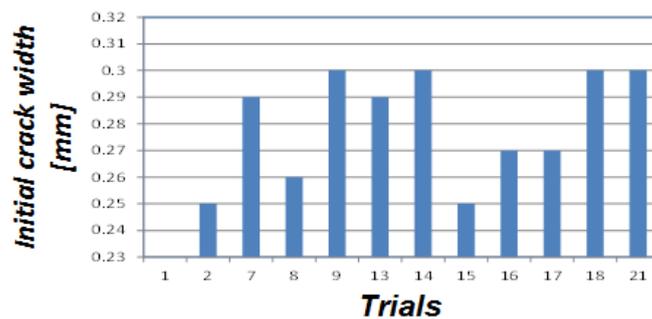
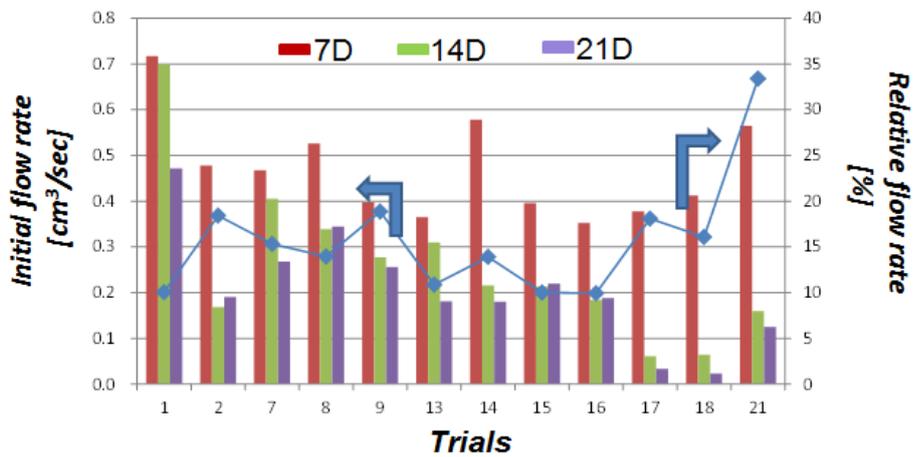
2 week age cured in air – Continuous water supply - Crack width = (0.32-0.39)mm



**3 week age cured in hot water – Continuous water supply**  
**Crack width = (0.25-0.29)mm**

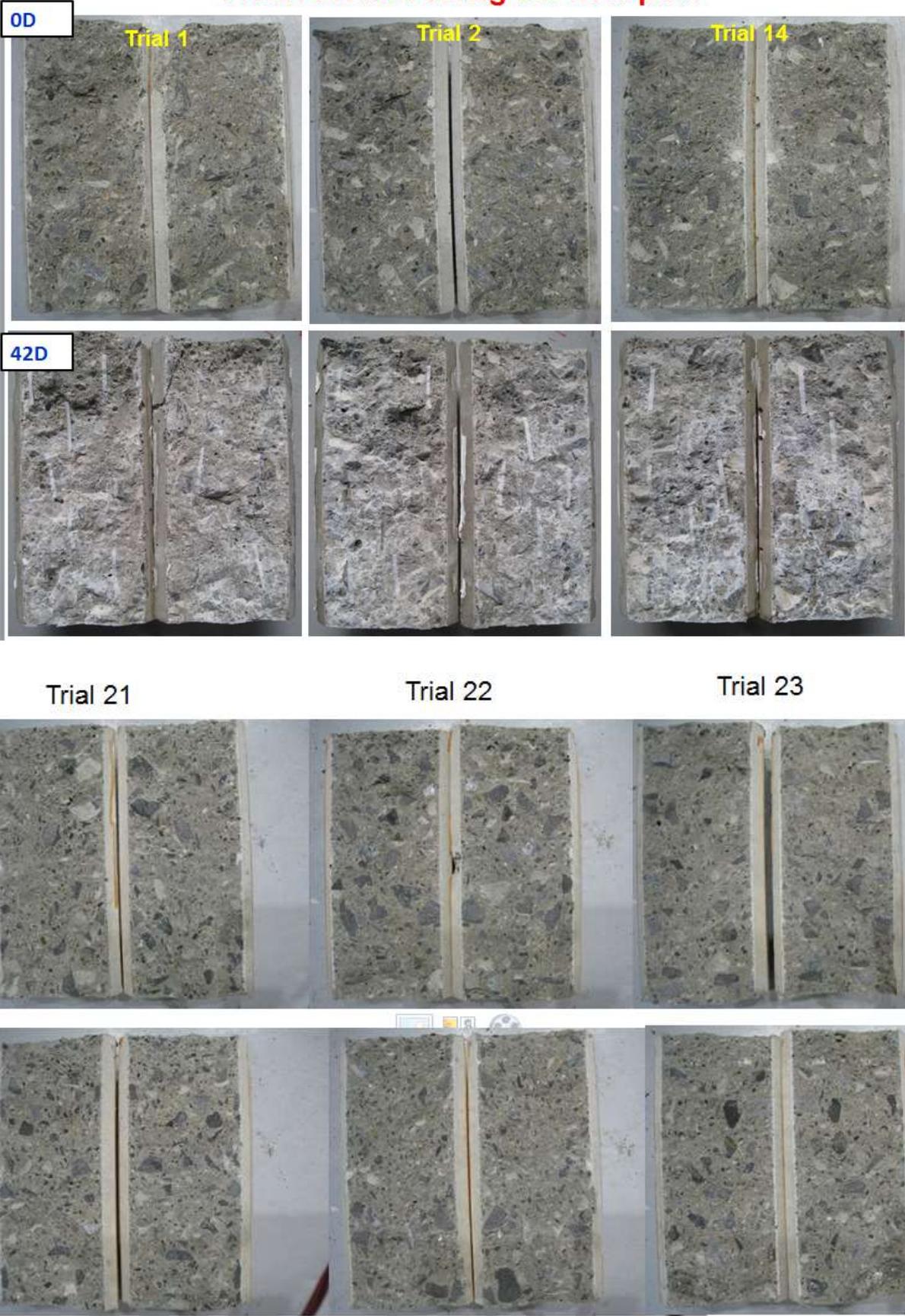


**3 week age cured in water – Static water curing**  
**Crack width = (0.25-0.30)mm**





**Crack surface along the flow path**



## 5. PERFORMANCE OF CONCRETES INCORPORATING SAME DOSAGE OF SH INGREDIENTS BUT IN DIFFERENT FORMS (GRANULES IN W/C55 CONCRETE (Apr.2013)& POWDER IN W/C30 CONCRETE)

### 5.1 Mix proportion of concrete

Concrete	W/C [%]	Air [%]	kg/m <sup>3</sup>					SP [Bx%]	AE [Bx%]
			W	C	SH	S	G		
SCC	30	5	193	640	-	704	748	0.7	0.2
SCC-SH-P	30	5	193	640	70	634	748	0.7	0.2

### 5.2 Materials & concrete properties

Trial	SH ingredients [%]	Slump [cm]	Air [%]	Compressive strength at 7 <sup>th</sup> day [N/mm <sup>2</sup> ]	Note
1	SCC-Plain	61x62	6.1	58	
2	HC18,LC18,Silica sand40, FA(II)24	57x58	5.6x	62.5	Trial 14 (Apr2012)
3	HC20,LC20,Silica sand40, EA20	57x58	5.0	62	Trial 11 (Apr2012)
4	BFS9.2,Fine FA23.1,CaSO <sub>4</sub> 6.2,Ca(OH) <sub>2</sub> 30.8,Slag sand30.8	44x43	4.5	61.1	

5	Clinker sand100	59x59	5.0	61.4	
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### 5.3 Result of water pass test (induce a crack at 7<sup>th</sup> day after casting)

