UNIVERSITI PUTRA MALAYSIA

STRENGTH RECOVERY AND HEALING EFFICIENCY OF CEMENT-BASED MATERIALS CONTAINING AUTOGENOUS HEALING BINDERS AND AUTONOMOUS ENCAPSULATED HEALING AGENTS

ABDULMOHAIMEN IMAD MOHAMMED

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By

ABDULMOHAIMEN IMAD MOHAMMED

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Doctor of Philosophy

June 2021

DEDICATION

This thesis is dedicated to my beloved parents, my sister and my brother, I wouldn't be able to make it happen without your affection, love and prayers; which were like a light of hope in a valley of darkness. I LOVE YOU!

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Doctor of Philosophy

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Chairman : Noor Azline Binti Mohd. Nasir, PhD
Faculty : Engineering

Self-healing is a new technology that is about promoting the recovery of mechanical properties of concrete without any external aid, to reduce maintenance costs. However, previous studies have reported some issues related to the encapsulation of healing agent, as capsules are unable to survive mixing process, or may slip without breaking due to their low bonding strength. Moreover, it was reported that autogenous healing has very limited ability in term of healable crack widths, while autonomous healing had an issue where capsules may not break and release the healing agent. Even more, it was reported that MgO expansive agent may cause strength reduction to concrete, which is disadvantageous. Therefore, this study aims to propose new surface modifications to polystyrene capsules; smooth, rough and weakness lines cylindrical sphere polystyrene capsules, that helps to enhance their ability to survive mixing process without being damaged, and with high bonding strength. In addition to that, this study is suggesting an enhancement to the autogenous and autonomous healing systems by including silica fume. Also, an investigation and comparison to the healing and sealing performance of autogenous, autonomous and a combination of both systems is conducted, together with an assessment to the healing compounds developed on the microstructure level for each healing system.

In this study, there are four mix combinations consists of OPC, silica fume and MgO expansive agent, with three variables which are smooth, rough and weakness lines capsules to carry sodium silicate (Na_2SiO_3) healing agent. The scope of work focuses on two main issues; (i) study of mineral admixtures in autogenous self-healing, and (ii) study the potential improvement to the encapsulation method used for autonomous self-healing. For the first issue, the evaluation was based on; (i) the healing ability of cement compounds by expansive admixture only, and (ii) the healing ability of cement compounds by a combination of mineral and expansive admixtures. Whereas the second issue focuses on; (i) capsule modification, (ii) evaluation of healing ability of cement compounds by encapsulated sodium silicate only, and (iii) evaluation of healing ability

of cement compounds by a combination between encapsulated method and mineral admixtures.

The evaluation of capsules was conducted based on their compressive strength, bonding strength, mixing survivability and the ability to survive elevated temperatures. Whereas for self-healing assessment, specimens were divided into two sets; the first set was left un-cracked, while the second set of specimens were pre-cracked after 7 days of water immersing curing using two different pre-cracking methods; the first method used for cubes by applying 80% of ultimate compressive strength, while the second method used for prisms by bending using 3-points flexural. Thereafter, specimens of both sets were re-immersed in water for further curing durations of 28, 56, 90 and 120 days. The healing and sealing performance of cement paste, mortar and concrete were assessed based on; the compressive and flexural strength regain, and crack sealing based on crack depth and area. Thereafter, microstructure assessment was conducted to determine the healing compounds developed on the crack planes using SEM and FTIR tests. Moreover, statistical analysis using ANOVA and multiple linear regression (MLR) were conducted to evaluate the effect of using silica fume, the difference in performance of each healing mechanism and to formulate prediction equations for strength recovery.

In conclusion, this research confirmed the efficiency of the new encapsulation surface modifications in term of mixing survivability and bonding strength, which was reflected on the self-healing performance. In addition to that, silica fume was proved to be able to successfully enhance the autogenous healing, and the combined self-healing system was proved to be superior in comparison to both autogenous and autonomous self-healing systems in term of strength recovery and crack sealing performance.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

PEMULIHAN KEKUATAN DAN PRESTASI PENYEMBUHAN BAHAN BERASASKAN SIMEN YANG MENGANDUNGI PENGIKAT PENYEMBUHAN AUTOGENOUS DAN EJEN PENYEMBUHAN BERKAPSUL AUTONOMOUS

Oleh

ABDULMOHAIMEN IMAD MOHAMMED

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Pemulihan diri ialah teknologi baharu yang direka untuk menggalak pemulihan ketermampatan konkrit tanpa sebarang bantuan luar, untuk mengurangkan kos penyelenggaraan. Walau bagaimanapun, kajian terdahulu telah melaporkan beberapa isu yang berkaitan dengan enkapsulasi agen pemulihan, kerana kapsul tidak dapat bertahan lama dalam proses pencampuran, atau mungkin tergelincir tanpa pecah kerana kekuatan ikatannya yang rendah. Selain itu, juga dilaporkan bahawa keupayaan permulihan automatik sangat terhad dari segi lebar keretakan yang boleh dipulih, manakala pemulihan autonomous mempunyai isu di mana kapsul mungkin tidak dapat pecah dan melepaskan agen pemulihann. Lebih-lebih lagi, dilaporkan bahawa agen ekspansif MgO boleh menyebabkan kemerosotan pada kekuatan konkrit, yang merupakan satu kelemahan. Atas sebab-sebab ini, kajian ini bertujuan untuk mencadangkan modifikasi permukaan yang baharu; sejenis kapsul bulat silinder yang licin, kasar dengan garis kelemahan yang diperbuat dari polistirena, yang mampu bertahan dalam proses pencampuran tanpa sebarang kerosakan, dengan kekuatan pecahan yang tinggi. Di samping itu, kajian ini juga ingin buat peningkatan baharu kepada proses pemulihan autogenous, dengan menggunakan kapsul agen pemulihan. Justeru, kajian terhadap prestasi pemulihan dan pengedap pemulihan autogenous, autonomous dan autogenous yang baru dicadang untuk dijalankan, bersama-sama dengan penilaian kepada sebatian pemulihan yang dibina pada tahap struktur mikro untuk setiap sistem pemulihan.

Dalam kajian ini, terdapat empat kombinasi campuran yang terdiri daripada OPC, asap silika dan agen ekspansif MgO, bersama dengan tiga pembolehubah iaitu kapsul licin, kasar dan dengan garis kelemahan untuk bungkus agen pemulihan natrium silikat (Na_2SiO_3). Skop kajian fokus pada dua isu utama; (i) kajian campuran mineral dalam proses pemulihan diri autogenous, dan (ii) kajian potensi peningkatan kepada kaedah enkapsulasi yang digunakan untuk pemulihan diri autonomous. Bagi isu pertama, penilaian adalah berdasarkan; (i) keupayaan pemulihan campuran simen melalui

campuran ekspansif sahaja, dan (ii) keupayaan pemulihan campuran simen dengan gabungan mineral dan campuran ekspansif. Manakala isu kedua fokus pada; (i) pengubahsuaian pada kapsul, (ii) penilaian keupayaan pemulihan campuran simen oleh natrium silikat terkapsul sahaja, dan (iii) penilaian keupayaan pemulihan campuran simen dengan gabungan antara kaedah terkapsul dan campuran mineral.

Penilaian kapsul telah dijalankan, kemampatan, kekuatan ikatan, ketahanan campuran dan ketahanan suhu tinggi. Manakala untuk penilaian pemulihan diri, spesimen dibahagikan kepada dua set; set pertama tanpa diretak, manakala set kedua di pra-retak, spesimen telah di pra-retak selepas 7 hari proses pengawetan dibawah air menggunakan dua kaedah pra-retak yang berbeza; kaedah pertama digunakan pada kubus dengan menggunakan 80% kekuatan kemampatan yang tertinggi, manakala kaedah kedua digunakan pada prisma dengan membongkok menggunakan lenturan 3-titik. Selepas itu, spesimen kedua-dua set direndam semula dalam air untuk teruskan proses pengawetan selama 28, 56, 90 dan 120 hari. Prestasi pemulihan dan pengedap pes simen, mortar dan spesimen konkrit dinilai berdasarkan kekuatan mampatan dan kelenturan, dan kesan pengedap retak berdasarkan kedalaman dan keluasan retakan. Selepas itu, penilaian struktur mikro telah dijalankan untuk menentukan sebatian pemulihan yang ada pada bahagian keretakan menggunakan ujian SEM dan FTIR. Selain itu, analisis statistik menggunakan ANOVA dan regresi linear berganda (MLR) telah dijalankan untuk menilai kesan penggunaan asap silika, perbezaan prestasi setiap mekanisme pemulihan dan untuk dapat formulasi persamaan ramalan untuk kekuatan pemulihan.

Kesimpulannya, penyelidikan ini mengesahkan prestasi modifikasi permukaan yang baharu dari segi ketahanan campuran dan kekuatan ikatan, yang telah dicerminkan pada prestasi pemulihan diri. Di samping itu, asap silika telah terbukti dapat berjaya meningkatkan proses pemulihan autogenous, dan gabungan proses pemulihan telah terbukti lebih baik berbanding dengan sistem pemulihan diri autogenous dan autonomous dari segi pemulihan kekuatan dan prestasi pengedap keretakan.

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Finally, no amount of 'thanks' on a page can describe my gratefulness for my parents, my sister and my brother. This thesis is dedicated to them, for their unconditional love and support, and for influencing who I am today.

This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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Date: 10 February 2022

Declaration by Members of Supervisory Committee

This is to confirm that:

- the research conducted and the writing of this thesis was under our supervision;
- supervision responsibilities as stated in the Universiti Putra Malaysia (Graduate Studies) Rules 2003 (Revision 2012-2013) are adhered to.

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TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

LIST OF APPENDICES

LIST OF ABBREVIATIONS

$(Fe_5Al_3)(SiAl)O_{10}(OH)_5$	Polymorphic Iron-aluminum-silicate
°C	Degree Celsius
ACI	American Concrete Institute
AG	Age of Specimen
ANOVA	Analysis of Variance
ASTM	American Society for Testing and Materials
ATR	Attenuated Total Reflection Mode
BRE	Building Research Establishment
BS EN	British Standard European Norm
C10	Concrete Cubes with Capsules Under 10 Mm Cover
C30	Concrete Cubes with Capsules Under 30 Mm Cover
C50	Concrete Cubes with Capsules Under 50 Mm Cover
CA(%)	Crack Sealing
CA	Cyanoacrylate
$Ca(NO_3)_2$	Calcium Nitrate
$Ca(OH)_2$	Calcium Hydroxide (Portlandite)
CA_0	Cross-Sectional Area Directly After Cracking
$Ca_4(AlO_2)_6SO_4$	Calcium Sulfoaluminate
$CaCO_3$	Calcium Carbonate
CaH	Dalcium Hydroxide
CaO	Calcium Oxide
$CaSO_4$	Anhydrite
CA_t	Crack Cross-Sectional Area After T Duration
Cd	Crack Depth

cm	Centimetre
CMOD	Crack Mouth Opening Displacement
CO_2	Carbon Dioxide
CS	Colloidal Silica
C-S-H	Calcium Silicate Hydrate Gel
d	Diameter of The Capsule
DCPD	Dicyclopentadiene
df	Degree of Freedom
DRIFTS	Diffuse Reflectance Spectroscopy
ECC	Engineered Cementitious Composites
EDX	Energy Dispersive X-Ray
F	Maximum Force at Failure
FA	Fly Ash
FTIR	Fourier Transform Infrared Spectroscopy
g	Gram
GGBS	Ground Granulated Blast-furnace Slag
GU	General Use
H	Healing Degree
H_2O	Water
IW	Initial Crack Width
kg	Kilogram
kHz	Kilohertz
kN	Kilonewton
L	The Length of The Capsule
LR	Load Regain
LVDT	Linear Variable Differential Transformer

MEA	Magnesium Oxide Expansive Agent
$Mg(OH)_2$	Magnesium Hydroxide (Brucite)
$MgCO_3$	Magnesium Carbonate
MgO	Magnesium Oxide
min	Minutes
mL	Millilitre
mm	Millimetre
MMA	Methyl Methacrylate
MPa	Megapascal
MS	Mean Squares
M-S-H	Magnesium Silicate Hydrate Gel
N	Newton
Na_2CO_3	Sodium Carbonate
Na_2O	Sodium Oxide
Na_2SiO_3	Sodium Silicate
$NaHCO_3$	Sodium Bicarbonate
n_i	Number of Intact Capsules
n_T	The Total Number of Capsules
OPC	Ordinary Portland Cement
p-value	Probability Value
P_1	Peak Load Before Healing
P_2	Peak Load After Healing
PA	Polyacrylate
PC	Portland Cement
PF	Phenol Formaldehyde
PFA	Pulverized Fly Ash

pH	Potential Hydrogen
PLA	Polylactic Acid
$P_{max,h}$	The Maximum Load Attained By The Healed Sample
$P_{max,i}$	The Load at Which Initial Cracking Occurred
PMMA	Poly Methyl Methacrylate
POFA	Palm Oil Fuel Ash
PP	Polypropylene
PS	Polystyrene
PSR	Predicted Strength Recovery
PU	Polyurethane
P_{un}	The Point of The Residual Load After Failure
P_{un}	Residual Load After Failure
PVA	Polyvinyl Alcohol
R	Load Regain
S	Compressive Strength at Loading
s	Seconds
SAP	Superabsorbent Polymer
SDDW	Short Distance Detour Wave
SEM	Scanning Electron Microscope
SF	Silica Fume
S_h	Compressive Strength After Healing
SiO_2	Silica Oxide
SMF	Sodium Monofluorophosphate
SR	Strength Recovery
SS (ANOVA)	Sum of Squares
SS	Sodium Silicate

SUR	Survival Ratio
t_1	Time of wave travel to position 1
t_2	Time of wave travel to position 2
TEOS	Tetraethyl Orthosilicate
UF	Urea-Formaldehyde
UFF	Urea Formaldehyde Formalin
UPV	Ultrasonic Pulse Velocity
USB	Universal Serial Bus
w/c	Water to Cement Ratio
XRD	X-Ray Diffraction
β	Bonding Strength
μm	Micrometre
$\sigma_{p,7}$	Stress of Pre-Cracked Samples at The Age of 7 Days
$\sigma_{p,n}$	Stress of Pre-Cracked Samples at The Age of n Days
$\sigma_{u,n}$	Stress of Un-Cracked Samples at The Age of n Days

CHAPTER 1

INTRODUCTION

1.1 Introduction

Concrete is a material that is susceptible to cracking due to different causes such as; compression, tension, shrinkage, fire, weathering and others. Durability of concrete is affected by cracks and becomes weaker due to the leaking of water and acids that comes with the rain, causing damage to the core and reinforcements of concrete, which as a result, reduces the age of concrete and causing various problems to the structure. In the current time, maintenance works are conducted to seal these cracks and stop them from getting wider to prevent water and chemicals from leaking to the concrete core. However, maintenance works are costly and need much effort, which turn the eyes of researchers towards other methods to contain this problem. In the recent years, studies about designing new concretes containing other materials in order to enhance its properties and increase the lifetime of structures. However, due to the unavailability of such materials in all regions, normal concrete is the most widely used material up to now, and the need to enhance it is essential. Recently, a new method was presented to overcome the durability problem, this method called as self-healing, which is defined as the restoration of mechanical properties without any human intervention. This method is still considered very new and started to get researchers' attention during the last 2 decades, as the "self-healing" term started to become more common as shown in Figure 1.1.

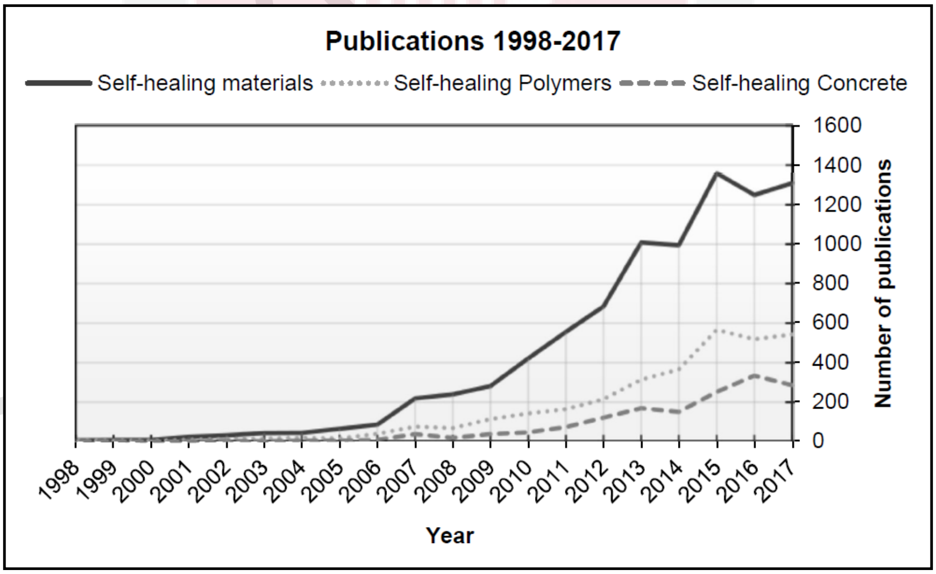

Figure 1.1: Yearly Publications Containing "self-healing" Words Extracted from Google Scholar (Roig-Flores, 2018)

Self-healing currently is still being developed to overcome any obstacles and enhance its mechanism. Different types of self-healing were studied over the previous years, such as autogenous, bacterial, chemical and mineral admixtures self-healing mechanisms, each type has its own pros and cons which will be discussed in this study. However, this study presents a new modification to the self-healing by enhancing the autogenous healing using encapsulated sodium silicate (Na_2SiO_3) and magnesium oxide (MgO) expansive agent. Moreover, new encapsulation designs and modifications were also proposed to overcome the encapsulation problems faced by other researchers in term of mixing resistance, bonding and strength of the capsules used.

1.2 Problem Statement

The durability is the most important challenge facing normal concrete, no matter what, cracks normally happen in concrete used in structural components such as beams, slabs, foundations, columns and other elements, which may lead to structural failures afterwards. For this reason, continuous maintenance works are required to be conducted to prevent such cracks from worsening and cause permanent damage. However, these maintenance works are costly and require skilled labour to deal with, and in addition to that, some cracks are not visible or accessible. Therefore, researchers have been paying attention to develop new materials that have the ability to heal the damage and restore the original condition of concrete depending on a number of chemical reactions, these materials are referred to as self-healing agents.

Normal concrete has an autogenous ability to heal microcracks under special conditions, however; its autogenous healing ability is limited to cracks of very small sizes. Van Tittelboom et al., (2012), Sisomphon et al., (2012) and Qureshi & Al-Tabbaa (2016) have studied different types of expansive minerals and crystalline admixtures to enhance the autogenous healing ability of concrete. MgO expansive agent showed an extraordinary performance in term of sealing and healing, due to its ability to react with water and produce $Mg(OH)_2$ expansion product, that has larger size than the original MgO particles. However, it was reported that the healable crack width is very limited, due to the amount of un-hydrated MgO particles available on the crack planes when the cracking occurs. In addition to that, it was reported that the addition of MgO had caused a reduction in the strength (Zheng et al., 1991; Cao et al., 2018). For this reason, autogenous healing needs further enhancements to be able to deal with cracks, with minimal strength reduction to the cementitious composite.

On the other hand, other researchers such as; Joseph et al., (2010), Thao (2011) and Vijay et al., (2017) have investigated the application of chemical agents and bacterial species as healing agents by delivering them to the damaged position using extended tubes or capsules as in autonomic healing. Delivery of healing agent using capsules as carrier is the most promising method, as these capsules are added during mixing or casting process. However, a successful encapsulation method depends on several factors such as the size of the capsule, compatibility with the cementitious composite, ability to survive during mixing process and the ability to break and release of healing agent when needed. For instance, microcapsules are a group of capsules with a very small size that ranging from few hundred microns and up to few millimetres in diameter. These capsules were reported to be disadvantageous due to their small size, and the ability to carry very small amounts of healing agent (Nishiwaki, 1997), which limits their ability in term of strength recovery. Moreover, Beglarigale et al., (2021) have reported that the addition of microcapsules may case strength reduction to the sample, which is mostly related to the reduction in the density caused by these capsules (Salman et al., 2021).

Macro capsules, on the other hand, that have larger sizes and are able to carry larger volumes of healing agents, were also used in the recent years by several researchers such as; Escobar et al., (2013) Van Tittelboom & De Belie (2013), Kanellopoulos et al., (2015) and Qureshi et al., (2016), who used glass and ceramic capsules as healing agent carriers. However, a common problem was faced by these capsules which is related to their inability to survive mixing process (Kanellopoulos et al., 2015), due to the brittleness of glass and ceramic. Several researchers have tried to overcome this problem by introducing polymeric capsules (Hilloulin et al., 2015; Araújo et al., 2018), as these capsules have lower brittleness factor than that of glass and ceramic. However, another issue was faced by these capsules related to their bonding strength, as some capsules may slip without breaking (Araújo et al., 2018), and thus, no healing agent is released to the crack, and no healing process takes place. Therefore, an enhancement to the bonding strength of polymeric capsules is needed.

Apart from size and material, the shape of the capsule is also a concern, as it was reported that spherical capsules have better chances in surviving mixing process (Sisomphon & Copuroglu, 2011), but are harder to break when crack occurs due to their spherical shape that can distribute the stresses without breaking. Whereas, cylindrical shape capsules, in general, have lower probability to survive during mixing process due to their shape which make them unable to bear the mechanical stresses (Van Tittelboom & De Belie, 2013), although; they have better chances in breaking when crack occurs. However, cylindrical capsules may also not break when needed, either due to their orientation inside the sample, as it is hard to break if they were oriented parallel to the crack, or due to the wall thickness of the capsule (Thao et al., 2009), or due to the crack size, as very thin cracks are mostly not wide enough to lead to the breakage of the capsule even if the capsule was oriented favourably. This issue raises another problem with autonomic healing where the capsule may not crack, and as result, no healing agent is released. Thus, there is an actual need to enhance the healing system without depending only on the encapsulated healing agent.

Based on the literature, this study is evaluating the enhancement of autogenous healing ability of cement composites with a combination of mineral admixtures (i.e. silica fume and MgO), by adding them directly during mixing process, to overcome the issue related to the strength reduction caused by microcapsules or pellets addition.

This study is also examining the compatibility of the encapsulation method used for autonomous healing in term of bonding strength and ability to survive mixing process, by evaluating the surface modifications.

Furthermore, this study is evaluating the addition of MgO and silica fume alongside encapsulated sodium silicate, to enhance the healing process by not depending on the breakage of the capsules and the release of liquid sodium silicate, instead; MgO and silica fume will have the ability to heal and seal the cracks that are smaller in size and do not cause the crack of the capsule. In addition to that, MgO and silica fume can also contribute to the self-healing process even if the capsules are cracked and released the healing agent, and by this way; additional healing may happen on the microstructure level due to this enhancement in the autogenous healing.

1.3 Objectives of Study

The main goal of this research is to understand the healing mechanism of autogenous and autonomous self-healing mechanisms of cement compounds. The work aims to give new sight of enhancement approaches for both self-healing mechanisms. In order to achieve this goal, the following objectives have been identified:

1. To propose surface modifications on polystyrene capsules to expand its potential, and assess the performance of these modifications in term of characteristics and compatibility within cement-based composites.
2. To propose the inclusion of silica fume alongside MgO to enhance the autogenous self-healing of cement compounds and categorize the healing product of the blended cement compounds.
3. To evaluate the healing efficiency of autonomous self-healing performance using encapsulated sodium silicate with and without incorporation of mineral admixtures.
4. To develop an appropriate combination of self-healing mechanisms and determining the potential self-healing enhancements for cement compounds.

1.4 Scope and Limitations of Study

Basically, the research is broadly divided into two main phases; (i) study of mineral admixtures in autogenous self-healing, and (ii) study the potential improvement to the encapsulation method used for autonomous self-healing. Both phases were examined for potential enhancement in self-healing of cementitious compounds. In the first stage, the autogenous self-healing is divided into two parts, which are; (i) evaluation of healing ability of cement compounds by expansive admixture only, and (ii) evaluation of healing ability of cement compounds by a combination of mineral and expansive admixtures. While in the second stage, the autonomous self-healing is divided into three parts, which are; (a) capsule modification, (b) evaluation of healing ability of cement compounds by encapsulated sodium silicate only, and (c) evaluation of healing ability of cement compounds by a combination between encapsulated method and mineral admixtures. For both stages, the healing and sealing investigations were conducted on three phases, which are; paste, mortar and concrete. Prior to that, the capsule modification for autonomous self-healing mechanism is conducted by modifying the capsule surface texture to enhance the bonding and mixing survivability of the capsule. The assessment of mixing survivability was conducted using three mixing speeds (i.e. low, medium and high) and based on the changes in pH level, Fourier transform infrared spectroscopy

(FTIR), and also the physical observation for any damages. In addition to that, the compressive strength, bonding strength and the ability of capsules to survive elevated temperatures were also investigated. For all 3 phases of testing medium (i.e. cement paste, mortar and concrete), the samples were divided into two groups; the first group was left un-cracked, while the second group of samples was pre-cracked after 7 days of curing by water immersing method. Crack generating was conducted using two different methods; compression and 3-points bending, in order to obtain different crack sizes, depths and positions. After pre-cracking, all samples were re-immersed in water to continue their curing for the durations of 28, 56, 90 and 120 days. Thereafter, samples were re-tested to evaluate their self-healing ability by measuring their performance in term of mechanical properties recovery and sealing after the prementioned curing durations. The evaluation includes testing for cement paste, mortar and concrete for compressive strength, 3-points bending strength, crack depth and area reduction using UPV and digital microscope monitoring respectively. The entire research program is illustrated in Figure 1.2.

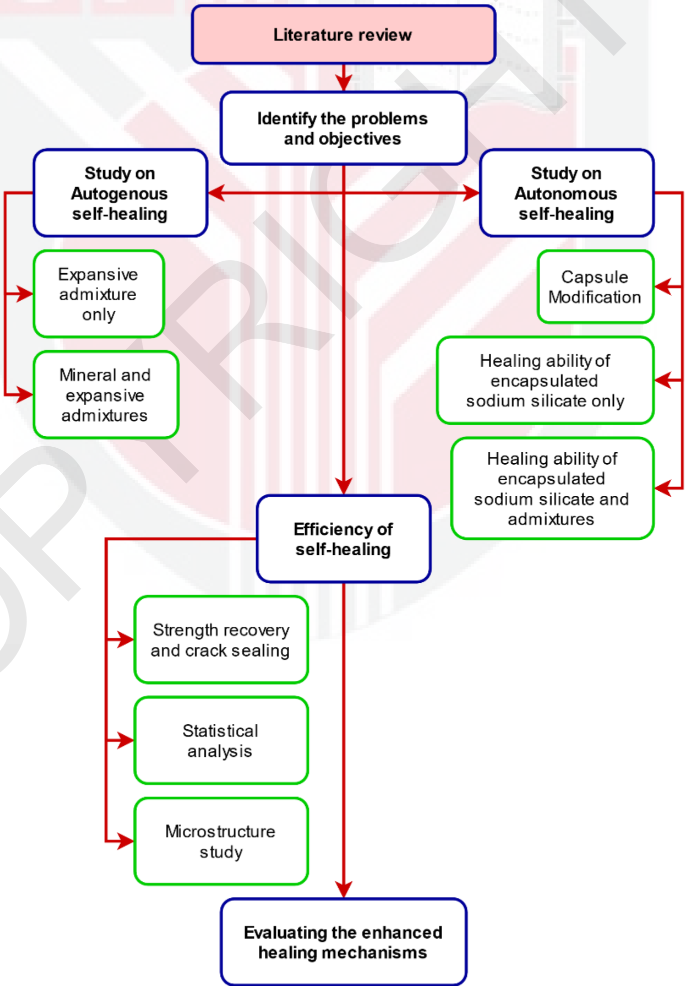

Figure 1.2: Scope of Study

Moreover, microstructure investigation was conducted to define and analyse the chemical compounds developed on the crack planes after healing process, these were assessed using scanning electron microscope (SEM) and Fourier transform infrared spectroscopy (FTIR) tests. The healing was quantified by measuring the regained strength related to the strength of un-cracked specimens, while sealing performance was assessed depending on the changes in depth and area of the crack. The results are then analysed and the performance of each healing combination was compared to control, and to other combinations, to define the best healing system. It is noteworthy that, in this study, there are four mix combinations; OPC only, OPC with silica fume, OPC with MgO, and OPC with both silica fume and MgO. Also, there are two variables which includes two different capsule modifications (rough and weakness lines) carrying sodium silicate solution, alongside the original smooth capsule that is evaluated in the capsule compatibility stage.

Limitations of the study, on the other hand, are as follows:

1. This study does not cover the self-healing performance in term of water absorption, penetration and porosity.
2. The healing performance in structural elements such as columns, slabs or beams need to be studied in future works.
3. Further investigations needed to define the effect of capsule presence on steel reinforcements, and while these reinforcements are damaged after healing process.

1.5 Significance of Study

Based on the existing studies, the use of MgO as expensive agent was found to be promising in autogenous self-healing. Nevertheless, it was also reported that the presence of MgO decreased the strength of the cement-based composites. Thus, this study proposed a combination between mineral admixture (i.e. silica fume) and expansive agent MgO, as the mineral admixture is expected to offset the strength reduction caused by the expansive agent. On the other hand, the literature has also found that there are some critical problems associated with encapsulation method, the main problems are; bonding strength between the capsule and the cement matrix. In relation to this, the study suggests surface modifications for polystyrene capsules to extend the potential usage of polystyrene capsule as a carrier for encapsulation method. Furthermore, this study also claims that the improvements in the cementitious combination of blended cements may help in enhancing the bonding characteristic of capsule. Even more, it was reported that polystyrene capsules have the ability to survive during mixing process, however; some designs are more susceptible to break during mixing than others, thus; this study is presenting round ends capsules to increase their resistance. It was also noticed that a combined approach between encapsulation and blended cements methods, which also known as combination between autonomous and autogenous self-healing mechanisms, is not a common approach in self-healing concept. Thus, this study is basically proposing an enhanced self-healing system depending on the current autogenous and autonomous self-healing systems, which is expected to overcome the limitations of the current self-healing mechanisms. In investigating the efficiency of the healing mechanisms involved with the proposed enhancement methods, this study is assessing the healing performance of cement paste, mortar and concrete composites using strength recovery, crack sealing and microstructural tests. Furthermore, the analysis using statistical approach was also carried out to predict the behaviour of self-healing ability of cement composites.

1.6 Thesis Layout

This thesis consists of 5 chapters, starting by introduction chapter (Chapter 1), which consist of a brief introduction, problem statement, objectives, scope and limitations of work and the significance of the work.

Thereafter, Chapter 2 presents a comprehensive summary to the literature related to the history of self-healing, its concepts, types, mechanisms, healing agents and delivery methods, together with the main findings obtained by previous studies. In addition to that, experimental works conducted by other researchers to assess the healing performance are also included in this chapter.

The methodology used in this study is explained in Chapter 3, including the materials preparations, mix designs, casting, curing, capsule modifications and crack inducing methods. Moreover, this chapter also presents the details of experimental works conducted to evaluate the compatibility of the surface modified capsules and the healing and sealing performances, together with the methods used to collect and prepare the specimens for microstructure assessment. In addition to that, data analysis section is included in this chapter.

After that, analysis and discussion of results are presented in Chapter 4, which is mainly consists of 4 parts. The first part focuses on the results related to the compatibility of the capsules, which includes the strength of the capsules, survivability during mixing, bonding strength and the behaviour under elevated temperatures. The following three parts of this chapter present the analysis and discussion of results for autogenous healing system, autonomous healing system, and a comparison between these two healing systems, respectively. Each part of these three presents the strength recovery and crack sealing results for cement paste, mortar and concrete specimens, together with the findings related to the microstructure. The relationship between strength recovery and crack sealing for each healing system is also discussed in this chapter. At the end, a summary is given for the results obtained at the age of 28 and 120 days for all the three healing systems of cement paste, mortar and concrete specimens.

Finally, in Chapter 5, the conclusion drawn from the study is presented, together with recommendations for future works.

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