

NUMERICAL SIMULATION OF
CAPILLARY FLOW AND CURING BEHAVIOUR OF HEALING AGENT IN
ENCAPSULATED-BASED SELF-HEALING CONCRETE

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DEDICATION

This thesis is dedicated to my father, who taught me that the best kind of knowledge to have is that which is learned for its own sake. It is also dedicated to my mother, who taught me that even the largest task can be accomplished if it is done one step at a time.

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ABSTRACT

A self-healing concrete has emerged as a potential solution for tackling cracking issues in concrete. Self-healing works through the infiltration of healing agents into the cracks, followed by the curing process that prevents further crack penetration in the concrete matrix. As far as the literature is concerned, the effect of the curing reaction on its rheological properties has not been addressed adequately. In addition, a fluid flow model considering the capillary effect and curing reaction has not been established for flow behaviours within discrete cracks in the encapsulation-based self-healing concrete. Therefore, in this study, a coupled fluid flow and curing reaction model was proposed to simulate the concrete encapsulation system's mechanics better. The healing agent flow was modelled using the Volume-of-Fluid (VOF) method. This study proposed using the viscosity function to describe the curing effect using the Castro-Macosko model. The dynamic mechanical analysis experiment cured and changed the cyanoacrylate's rheological properties. The fluid flow and curing reaction models were coupled in ANSYS Fluent in the form of self-developed user-defined functions. Parametric studies were carried out to determine the influence of healing agent rheological properties (surface tension, contact angle and viscosity) and crack geometries (planar, inclined and tapered) on the healing efficiency. The coupled model was validated against available experiment results and the model's capability to predict the healing agent's flow accurately and the curing process was shown. For flows in small cracks driven by capillary action, the simulated VOF outcomes with constant contact angles were in poor agreement with the experiment. The simulation results showed a better prediction of the capillary flow with the use of dynamic contact angles (DCA). For example, when validated against the modified Lucas-Washburn equation (LWE), the VOF predictions considering the velocity-dependent DCA have mean absolute percentage errors of between 3.1 – 5.3%, much lower than that of classical LWE with errors between 17.0 – 42.9%. The results indicated that a DCA influences the initial speed of the capillary flow and plays a vital role in the healing efficiency of fast-curing healing agents. Due to the curing reaction, the increasing viscosity arrests the capillary flow of the healing agent in a small discrete crack. DCA and viscosity control the infiltration speed of capillary flow via frictional dissipation and flow resistance, respectively. However, they do not affect the final equilibrium height in capillary rise. A higher frictional coefficient in the DCA model decreases the infiltration speed at the initial state of the capillary rise. In said capillary flow, the infiltration length of the healing agent depends on the capillary pressure, which is strongly influenced by the surface tension, equilibrium contact angle and crack widths. Based on the Young-Laplace equation, the capillary pressure is directly proportional to the surface tension force and inversely proportional to the crack width. A lower contact angle indicates good wettability and provides faster liquid spreading on a surface. Overall, this study has provided a new coupled self-healing model for predicting the transport and curing processes in encapsulation-based self-healing systems in concrete. The model can provide a better understanding of flow mechanisms and serves as a sound basis for future researchers to design a more efficient concrete self-healing system.

ABSTRAK

Konkrit penyembuhan diri muncul sebagai penyelesaian yang berpotensi untuk menangani masalah retakan dalam konkrit. Penyembuhan diri bertindak melalui penyusupan agen penyembuhan ke dalam retakan, diikuti dengan proses pengawetan yang menghalang penembusan retakan selanjutnya dalam matriks konkrit. Kajian literatur menunjukkan kesan tindak balas pengawetan agen penyembuhan terhadap sifat rheologi masih belum difahami dengan baik. Di samping itu, model aliran bendalir yang mengambil kira kesan daya kapilari dan tindak balas pengawetan belum ditetapkan untuk aliran dalam retakan diskret dalam konkrit penyembuhan diri berasaskan pengkapsulan. Oleh itu, mekanisme yang terlibat dalam sistem pengkapsulan konkrit dapat disimulasikan dengan baik menggunakan model gandingan antara aliran bendalir dan tindak balas pengawetan. Aliran agen penyembuhan dimodelkan dengan menggunakan kaedah Volume-of-Fluid (VOF). Kajian ini mencadangkan penggunaan fungsi kelikatan untuk menerangkan kesan pengawetan dengan menggunakan model Castro-Macosko. Eksperimen menggunakan analisis mekanikal dinamik menunjukkan bukti korelasi antara pengawetan dan perubahan dalam sifat rheologi agen penyembuhan. Model aliran bendalir dan model tindak balas pengawetan telah digandingkan dalam ANSYS Fluent dengan menggunakan fungsi takrifan pengguna. Kajian parametrik telah dijalankan untuk mengenalpasti kesan pengaruh sifat rheologi agen penyembuhan (tegangan permukaan, sudut sentuhan dan kelikatan) dan geometri retakan (planar, cenderung dan tirus) terhadap kecekapan penyembuhan. Keputusan eksperimen yang sedia mengesahkan keupayaan model gandingan tersebut dalam meramalkan aliran agen penyembuhan dan proses pengawetannya. Untuk aliran kapilari dalam retakan kecil, keputusan simulasi VOF dengan sudut sentuhan malar tidak selari dengan keputusan eksperimen. Keputusan simulasi menunjukkan ramalan aliran kapilari yang lebih baik dengan penggunaan sudut sentuhan dinamik. Keputusan tersebut menunjukkan bahawa sudut sentuhan dinamik mempengaruhi kelajuan aliran kapilari di peringkat permulaan dan memainkan peranan penting dalam kecekapan dan tindak balas penyembuhan. Ini disebabkan oleh tindak balas pengawetan dan peningkatan kelikatan yang merencatkan aliran kapilari agen penyembuhan dalam retakan diskret yang kecil. Sudut sentuhan dinamik dan kelikatan mengawal kelajuan infiltrasi aliran kapilari melalui pelepasan geseran dan rintangan aliran. Namun, kedua-dua parameter tersebut tidak memberi kesan terhadap ketinggian keseimbangan dalam peningkatan kapilari. Panjang infiltrasi agen penyembuh bergantung kepada tekanan kapilari yang merangkumi tegangan permukaan dan sudut sentuhan keseimbangan. Dalam aliran kapilari, panjang infiltrasi agen penyembuh sangat dipengaruhi oleh ketegangan permukaan, sudut sentuhan keseimbangan dan lebar retakan. Kesimpulannya, kajian ini telah menyediakan model penyembuhan diri gandingan yang baru untuk meramalkan aliran dan proses pengawetan dalam konteks sistem penyembuhan diri berasaskan pengkapsulan. Model tersebut boleh digunakan untuk memberikan pemahaman yang lebih baik tentang mekanisme aliran dan menyediakan asas kukuh kepada penyelidik di masa hadapan dalam usaha untuk mereka bentuk sistem penyembuhan diri konkrit yang lebih cekap.

TABLE OF CONTENTS

	TITLE	PAGE
	DECLARATION	iii
	DEDICATION	iv
	ACKNOWLEDGEMENT	v
	ABSTRACT	vi
	ABSTRAK	vii
	TABLE OF CONTENTS	viii
	LIST OF TABLES	xii
	LIST OF FIGURES	xiii
	LIST OF ABBREVIATIONS	xix
	LIST OF SYMBOLS	xx
	LIST OF APPENDICES	xxii
CHAPTER 1	INTRODUCTION	1
1.1	Background of the study	1
1.2	Problem Statement	4
1.3	Objectives	5
1.4	Scope of the Study	6
1.5	Significance of the Study	7
1.6	Outlines	9
CHAPTER 2	LITERATURE REVIEW	11
2.1	Roads to Self-Healing Concrete	11
2.2	Autogenous Healing	18
2.3	Autonomic Healing	21
2.3.1	Vascular Network	24
2.3.2	Encapsulation Techniques	25
2.3.2.1	Macro-Encapsulation	27
2.3.2.2	Microencapsulation	28

2.3.3	Healing Agents	29
2.3.4	Capsule Material	30
2.3.5	Evaluation of Self-Healing Efficiency	31
2.3.6	Field Applications of Encapsulation-Based Self-Healing Concrete	38
2.4	Healing Agent Flow in Self-Healing Concrete	40
2.4.1	Experimental and Numerical Investigation of Healing Agents Flow in Self-Healing Concrete	40
2.4.2	Capillary Flow in Discrete Crack	46
2.4.3	Numerical Simulation of Capillary Flow	49
2.5	Curing Process of Healing Agent	51
2.5.1	Curing Mechanisms of Healing Agent	51
2.5.2	Curing Process of Moisture Curing Polymer in Self-Healing Concrete	52
2.5.3	Influence of Curing Reaction on Flow Properties	58
2.5.3.1	Chemo-Rheological Model	58
2.5.3.2	Influence on Viscosity	60
2.6	Summary	63
CHAPTER 3	METHODOLOGY	65
3.1	Introduction	65
3.2	Governing Equations	68
3.2.1	Navier-Stokes Equations	68
3.2.2	Multiphase Flow	69
3.2.3	Surface Tension Force	70
3.2.4	Wall Adhesion (Contact Angle)	73
3.2.4.1	Dynamic Contact Angle Model	74
3.2.4.2	Time-Dependent DCA Model	75
3.2.4.3	Velocity-Dependent DCA Model	77
3.2.5	Degree of Cure and Dynamic Viscosity	80
3.3	Numerical Simulation	81
3.3.1	ANSYS Fluent	81
3.3.2	Adaptive Mesh	87

	3.3.3 Adaptive Time Step	88
	3.3.4 Boundary Conditions	89
3.4	Experimental Characterisation of Material Properties	90
	3.4.1 Material and Specimens	90
	3.4.2 Dynamic Viscosity Measurements	91
3.5	Validation of Castro-Macosko Model	93
3.6	Summary	98
CHAPTER 4	NUMERICAL SIMULATION OF CAPILLARY FLOW IN DISCRETE CRACK	99
4.1	Introduction	99
4.2	Grid Independence Study	99
4.3	Model Validation	101
	4.3.1 Capillary Rise in Cylindrical Tube	101
	4.3.2 Capillary Rise between Parallel Plates	102
4.4	Capillary Rise in Planar Crack in Concrete	105
4.5	Dynamic Contact Angle Model	108
	4.5.1 Time-Dependent DCA Model	109
	4.5.2 Velocity-Dependent DCA Model	113
4.6	Dynamic Viscosity	116
	4.6.1 Increasing Viscosity due to Curing Reaction	116
	4.6.2 Validation of Castro-Macosko Model	120
4.7	Summary	124
CHAPTER 5	PARAMETRIC STUDY	125
5.1	Introduction	125
5.2	Viscosity and Curing Effect	126
	5.2.1 Constant Viscosity	126
	5.2.2 Degree of Cure and Viscosity Change	127
	5.2.3 Curing Effect on Viscosity	132
	5.2.4 Curing Effect on Capillary Rise in Discrete Crack	133
5.3	Surface Tension and Contact Angle	136
	5.3.1 Surface Tension	136

5.3.2	Contact Angle	139
5.3.2.1	Equilibrium Contact Angle	139
5.3.2.2	Frictional Coefficient in Dynamic Contact Angle	142
5.4	Crack Configuration	144
5.4.1	Planar Crack	144
5.4.2	Crack Inclination Angle	147
5.4.3	Tapered Crack	150
5.5	Branched Crack	152
5.6	3D Crack Filling Process in Encapsulation-Based Self-Healing System	158
5.7	Summary	164
CHAPTER 6	CONCLUSION AND RECOMMENDATIONS	169
6.1	Conclusions	169
6.2	Recommendations for Future Work	173
	REFERENCES	175
	LIST OF PUBLICATIONS	209

LIST OF TABLES

TABLE NO.	TITLE	PAGE
Table 2.1	Healing agents for cementitious materials	33
Table 2.2	Capsules used in autonomous self-healing concrete	35
Table 2.3	Evaluation methods of self-healing efficiency for cementitious materials	36
Table 3.1	Named expressions included in the simulation	79
Table 3.2	Parameters used in the simulations.	90
Table 3.3	Material properties of EMC material.	97
Table 5.1	Material properties of cyanoacrylate PC20 (Selvarajoo, 2020)	128

LIST OF FIGURES

FIGURE NO.	TITLE	PAGE
Figure 1.1	Taxonomy in self-healing concrete.	2
Figure 1.2	The schematic concept of encapsulation-based autonomic healing for cementitious materials (Xue <i>et al.</i> , 2019).	3
Figure 2.1	(a) & (b) The comparison between the performance and costs for normal (Line A) and high-quality buildings (Line B). (c) & (d) The performance and cost for a building constructed with self-healing concrete (Schlangen & Joseph, 2009).	12
Figure 2.2	Construction output price indices in United Kingdom, 2014 to 2017 (Office for National Statistics, 2018)	13
Figure 2.3	Schematic chart of self-healing terms (Joseph <i>et al.</i> , 2011).	14
Figure 2.4	Schematic diagram of active mode. Melted methylmethacrylate healing agent is released into the cracks after first heating. The crack is healed when methylmethacrylate cured during second heating. (Dry, 1994)	15
Figure 2.5	Schematic diagram of passive mode. The crack ruptures the healing agent containing capsules embedded in the concrete, then the healing agent, driven by the capillary action, infiltrates through the crack. The crack is healed when the healing agent cures. (Dry, 1994).	15
Figure 2.6	Hierarchy chart of self-healing concrete	16
Figure 2.7	Schematic diagram for encapsulation-based self-healing concrete (Riccardo Maddalena, 2019).	17
Figure 2.8	Schematic diagram for vascular networks system in self-healing concrete (Riccardo Maddalena, 2019).	17
Figure 2.9	Schematic diagram for bacteria-based self-healing concrete (Riccardo Maddalena, 2019).	18
Figure 2.10	Schematic diagram for the methods utilising SMP in self-healing concrete (Riccardo Maddalena, 2019).	18
Figure 2.11	Three main causes in autogenic self-healing of cementitious materials (Rooij, Van Tittelboom, <i>et al.</i> , 2013)	19
Figure 2.12	The schematic diagram of autonomous healing in cementitious materials (Joseph <i>et al.</i> , 2010).	22

Figure 2.13	Self-healing approach. (a) microcapsule and (b) hollow glass fibres (Hall <i>et al.</i> , 2015).	23
Figure 2.14	Self-healing approach. (c) vascular (250-micron-bore, optical micrographic image) and (d) porous hollow vascular (400-micron-bore, SEM image) (Hall <i>et al.</i> , 2015).	23
Figure 2.15	Schematic concept of external healing agent supply system (Joseph <i>et al.</i> , 2007; Mihashi <i>et al.</i> , 2000)	25
Figure 2.16	The schematic concept of encapsulation-based autonomic healing for cementitious materials (Xue <i>et al.</i> , 2019).	26
Figure 2.17	Existing works on encapsulation-based self-healing concrete.	26
Figure 2.18	The main forces involved in the internal encapsulated healing agent in the brittle vessel (Joseph <i>et al.</i> , 2010).	27
Figure 2.19	The microcapsules to be mixed in the concrete (Al-Tabbaa <i>et al.</i> , 2019).	39
Figure 2.20	The self-healing concrete panels in field application (Al-Tabbaa <i>et al.</i> , 2019).	39
Figure 2.21	Experimental setup and sample preparation: (a) sample dimensions, (b) drilling of the hole for vascular capsule installation, (c) crack opening distance, and (d) shearing of the vascular capsule to allow healing agent leakage. (Gilabert <i>et al.</i> , 2017)	41
Figure 2.22	Simulation results for 100 μm crack opening distance at different time steps. (Gilabert <i>et al.</i> , 2017)	42
Figure 2.23	Schematic diagram of the capillary rise experiment setup (Gardner <i>et al.</i> , 2012, 2014; Selvarajoo, 2020; Selvarajoo <i>et al.</i> , 2020)	43
Figure 2.24	Simulation results for healing agent GGBS(S) and cyanoacrylate (PC20) using experimentally determined flow parameters. (Gardner <i>et al.</i> , 2017)	44
Figure 2.25	Comparison of numerical simulation results with experimental data (Freeman & Jefferson, 2020)	45
Figure 2.26	Degree of cure of different thicknesses of cyanoacrylate films on a microscope glass slide (Tomlinson <i>et al.</i> , 2006)	54
Figure 2.27	Experimental setup for monitoring curing front propagation of cyanoacrylate on the concrete substrate (Selvarajoo <i>et al.</i> , 2020)	55
Figure 2.28	Propagation of curing front over time (Selvarajoo <i>et al.</i> , 2020)	56

Figure 2.29	Schematic diagram of the cured cyanoacrylate (white-colour precipitation) on the concrete surface (grey region).	57
Figure 2.30	Custom viscometer used by Gardner <i>et al.</i> (Gardner <i>et al.</i> , 2017)	61
Figure 2.31	Viscosity change of cyanoacrylate over time (Gardner <i>et al.</i> , 2017)	62
Figure 3.1	Schematic diagram of encapsulation-based self-healing concrete.	66
Figure 3.2	Schematic diagram of the capillary-driven flow of healing agent in a discrete crack.	66
Figure 3.3	Overview flow chart in the present study.	67
Figure 3.4	Diagram of the cohesive forces on molecules of a liquid (Ebnesajjad, 2014)	71
Figure 3.5	Arbitrary smooth surface with curvature radius, $R1$ and $R2$.	71
Figure 3.6	Capillary flow between parallel plates.	72
Figure 3.7	Schematic diagram of contact angle on a solid surface.	73
Figure 3.8	The contact angle changes over time during the capillary rise (H. Kim <i>et al.</i> , 2020)	74
Figure 3.9	Schematic diagram of the geometry of the computational domain.	81
Figure 3.10	Integration of numerical models.	82
Figure 3.11	The multiphase model dialog box for the VOF model in ANSYS Fluent and the settings to enable the surface tension and contact angle functions.	83
Figure 3.12	The dialog box shows the expression describing the dynamic contact angle (DCA) of cyanoacrylate.	84
Figure 3.13	The wall dialog box and the settings to enable the dynamic contact angle.	85
Figure 3.14	The materials dialog box and the settings to include user-defined function that describing the dynamic viscosity.	85
Figure 3.15	The overall interface of the ANSYS Fluent software and the geometry domain of the capillary flow in the discrete crack.	86
Figure 3.16	The automatic mesh adaption dialog box in ANSYS Fluent	88

Figure 3.17	Meshed model of the planar crack with adaptive mesh refinement at the free surface.	88
Figure 3.18	Cyanoacrylate Loctite 460 sample	91
Figure 3.19	Rheometer (Anton Paar MCR 302)	92
Figure 3.20	Geometry domain of S-CSP package model (Abdullah <i>et al.</i> , 2007).	95
Figure 3.21	Half geometry domain used in the simulation.	95
Figure 3.22	Plan view of the half S-CSP package model.	96
Figure 3.23	Closer view of the stacking chips.	96
Figure 4.1	Height of capillary rise versus flow time for various mesh sizes.	100
Figure 4.2	Model validation against experimental data (LeGrand & Rense, 1945) for the capillary rise in a cylindrical glass tube with a diameter of 0.57 mm.	102
Figure 4.3	Simulation results for the capillary rise between 0.261 mm parallel plates (dashed line). The solid line represents LWE prediction.	103
Figure 4.4	Simulation results for the capillary rise of water in parallel plates with different widths.	104
Figure 4.5	The capillary rise of water in planar crack with (a) 0.261 mm (b) 0.380 mm (c) 0.650 mm and (d) 1 mm	106
Figure 4.6	Capillary rise curves of water predicted by modified Lucas-Washburn equation (Gardner <i>et al.</i> , 2012).	109
Figure 4.7	Calculated contact angle from macroscopic capillary rise results based on modified Lucas-Washburn equation (Gardner <i>et al.</i> , 2012).	110
Figure 4.8	The capillary rise of water in planar crack with (a) 0.261 mm (b) 0.380 mm (c) 0.650 mm and (d) 1 mm	111
Figure 4.9	The capillary rise of water in planar crack with (a) 0.261 mm (b) 0.380 mm (c) 0.650 mm and (d) 1 mm	114
Figure 4.10	Amplitude sweep results for cyanoacrylate.	117
Figure 4.11	Storage and loss modulus of cyanoacrylate over time.	118
Figure 4.12	Viscosity change of cyanoacrylate over time.	119
Figure 4.13	Comparison between the experimental results (Abdullah <i>et al.</i> , 2007) and the simulation results with the Cross and the Castro-Macosko models.	121

Figure 4.14	Comparison between the experimental results (Abdullah <i>et al.</i> , 2007) and the simulation results with the Cross and the Castro-Macosko models	123
Figure 5.1	Capillary rises of the healing agent in 0.2 mm width discrete crack with various constant viscosities.	127
Figure 5.2	(a)-(d) Schematic diagrams show that the thickness of cured cyanoacrylate increases over time on the concrete surface.	128
Figure 5.3	Propagation of curing front of cyanoacrylate over time.	129
Figure 5.4	Schematic diagram of the distance x_c measuring from the concrete surface.	129
Figure 5.5	The degree of cure at each point between the discrete crack, x_c .	130
Figure 5.6	Total degree of cure across whole discrete crack with different gap sizes.	131
Figure 5.7	Viscosity changes in different crack widths.	133
Figure 5.8	Capillary rise of cyanoacrylate in 0.346 mm crack.	134
Figure 5.9	Capillary rise of cyanoacrylate in 0.1 mm and 0.2 mm discrete cracks.	135
Figure 5.10	Capillary rises of water in the discrete crack with various surface tension forces.	138
Figure 5.11	Capillary rises of cyanoacrylate in the discrete crack with various surface tension forces.	139
Figure 5.12	Capillary rises of water in the discrete crack with various equilibrium contact angles.	141
Figure 5.13	Capillary rises of cyanoacrylate in the discrete crack with various equilibrium contact angles.	142
Figure 5.14	Capillary rises of the healing agent in the discrete crack with various frictional coefficients in dynamic contact angle.	143
Figure 5.15	Capillary rise of water in the discrete crack with different crack widths.	145
Figure 5.16	Capillary rise of cyanoacrylate in the discrete crack with different crack widths.	146
Figure 5.17	Schematic diagram of inclined crack configuration (Gardner <i>et al.</i> , 2014).	147

Figure 5.18	Capillary rise of cyanoacrylate in the inclined crack compared with modified Lucas-Washburn equation (Gardner <i>et al.</i> , 2014).	148
Figure 5.19	Schematic diagram of crack with different inclination angles.	149
Figure 5.20	Capillary rise of cyanoacrylate in the discrete crack with different inclination angles.	149
Figure 5.21	Capillary rises of cyanoacrylate in the discrete crack with the inclination angle of 30° and 0°.	150
Figure 5.22	Crack geometry for tapered crack (Gardner <i>et al.</i> , 2014).	151
Figure 5.23	Capillary rise of cyanoacrylate in the tapered crack with different crack openings.	152
Figure 5.24	Y-shape branched crack geometry.	154
Figure 5.25	The evolution of capillary rise of fluorinated oil in the branched crack.	154
Figure 5.26	(a)-(f): The evolution of capillary rise in the branched crack at different time steps.	155
Figure 5.27	(a)-(i) The propagation of the meniscus in the branched cracks (Sadjadi <i>et al.</i> , 2015).	156
Figure 5.28	The evolution of capillary rise of cyanoacrylate in branched crack	158
Figure 5.29	Computational domain of the crack volume for the simulation of the crack filling process of healing agent (Gilabert <i>et al.</i> , 2017).	159
Figure 5.30	Crack filling of cyanoacrylate in 0.1 mm vertical crack plane at different time steps.	161
Figure 5.31	The pattern of the healing agent released from the ruptured capsule (Gilabert <i>et al.</i> , 2017).	162
Figure 5.32	Spreading distance of cyanoacrylate from the capsule in upward and downward directions (0.1 mm crack plane).	162
Figure 5.33	Crack filling of cyanoacrylate in 0.3 mm vertical crack plane at different time steps.	163
Figure 5.34	Spreading distance of cyanoacrylate from the capsule in upward and downward directions (0.3 mm crack plane).	164

LIST OF ABBREVIATIONS

2D	-	Two-Dimensional
3D	-	Three-Dimensional
ADE	-	Advection-Diffusion Equation
CFD	-	Computational Fluid Dynamic
CIDB	-	Construction Industry Development Board
CSF	-	Continuum Surface Force
CSH	-	Calcium Silicate Hydrates
DCA	-	Dynamic Contact Angle
DSC	-	Differential Scanning Calorimetry
ECCs	-	Engineered Cementitious Composites
EDTA	-	Ethylene Diamine Tetra-acetic Acid
EMC	-	Epoxy Moulding Compound
EPSRC	-	Engineering and Physical Science Research Council
ESM	-	Electron Scanning Microscopy
GGBS	-	Ground-Granulated Blast-Furnace Slag
GNF	-	Generalised Newtonian fluid
LVE	-	Linear Viscoelastic
LWE	-	Lucas-Washburn equation
MMA	-	Methyl Methacrylate
NDT	-	Non-Destructive Tests
RH	-	Relative Humidity
RM4L	-	Resilient Materials 4 Life
S-CSP	-	Stacked-Chip Scale Package
SEM	-	Scanning Electron Microscopy
SIMPLE	-	Semi-Implicit Pressure Linked Equations
SMP	-	Shape Memory Polymer
TGA	-	Thermogravimetric Analysis
UDF	-	User-Defined Function
VOF	-	Volume of Fluid

LIST OF SYMBOLS

F_{st}	-	Surface tension force term in momentum equation
K_p	-	Permeability of cured adhesive layer
P_v	-	Vapour pressure
T_b	-	Activation-energy dependent parameter
c_p	-	Specific heat capacity
\hat{n}	-	Unit normal vector
z_c	-	Wall factor
z_{c0}	-	Critical curing depth
α_g	-	Degree of cure at gel point
$\dot{\gamma}$	-	Shear rate
η_0	-	Viscosity when the shear rate approaches zero
θ_D	-	Dynamic contact angle
θ_E	-	Equilibrium contact angle
τ^*	-	Shear stress at the transition between Newtonian and non-Newtonian flow
Δt	-	Time step
Δx	-	Mesh size
h	-	Capillary rise height
A	-	Area
B	-	Exponential-fitted constant
C	-	Fitting constant
Ca	-	Dimensionless Capillary number
Co	-	Dimensionless Courant number
D	-	Diffusion coefficient
F	-	Scalar for volume fraction in a cell
I	-	Identity matrix
L	-	Length
R	-	Radius
T	-	Temperature

V	-	Equilibrium adhesive volume that involved in reaction
W	-	Lambert W function
b	-	Crack width
g	-	Gravitational acceleration
k	-	Heat conductivity
n	-	Reaction order
p	-	Pressure
t	-	Time
u	-	Velocity at x-direction
v	-	Velocity at y-direction
z	-	Curing front depth
α	-	Degree of cure
β	-	Frictional coefficient in dynamic contact angle model
η	-	Dynamic viscosity
θ	-	Contact angle
κ	-	Mean curvature
μ	-	Constant viscosity
π	-	Mathematical constant, Pi
ρ	-	Density
σ	-	Surface tension
τ	-	Curing rate parameter
ϕ	-	Crack inclination angle

LIST OF APPENDICES

APPENDIX	TITLE	PAGE
Appendix A	User-defined functions	203

CHAPTER 1

INTRODUCTION

1.1 Background of the study

Self-healing concrete is emerging as an innovative construction material to tackle the environmental issues caused by carbon dioxide emissions from concrete manufacturing industries. By mimicking the natural healing ability in the human body, self-healing concrete is designed to heal itself without external human intervention. Self-healing concrete can also play an essential role in addressing concrete structures' durability and serviceability issues, as the inevitable formation of cracks in the concrete matrix allows the penetration of harmful substances and thus reduces the durability of the concrete over time.

Over the past decade, self-healing concrete has been garnering interest from many researchers. With the support from the government and policymakers, the development of self-healing concrete is getting traction in recent years. For instance, in 2017, an ongoing five-year research project, Resilient Materials 4 Life (RM4L), was funded and firmly supported by Engineering and Physical Science Research Council (EPSRC) (Al-Tabbaa *et al.*, 2018; Davies *et al.*, 2018; Paine *et al.*, 2019). To show the extent of support from various parties, the project is in fact joined by Cardiff, Cambridge, Bath and Bradford universities and a whopping amount of 22 industrial companies as well. The project aims to develop a sustainable construction material with self-healing, self-sensing and self-diagnosing abilities, as well as being immune against physical and chemical damages.

Figure 1.1 shows the taxonomy in self-healing concrete. The self-healing process in concrete is classified into two major groups: autogenous and autonomous healing (De Belie *et al.*, 2018; Sidiq *et al.*, 2019; Van Tittelboom & De Belie, 2013; Xue *et al.*, 2019). Autogenous healing occurs via the hydration process of un-hydrated

cement particles in the concrete matrix without any external intervention, while autonomous healing involves the addition of external engineered materials such as polymers, microorganisms, chemical compounds and admixtures for healing purposes. Autogenous healing occurs naturally in concrete, but is limited to the healing of crack widths of less than 300 μm (De Belie *et al.*, 2018). Autonomous healing provides better healing for crack widths of more than 300 μm and can heal cracks up to 1 mm.

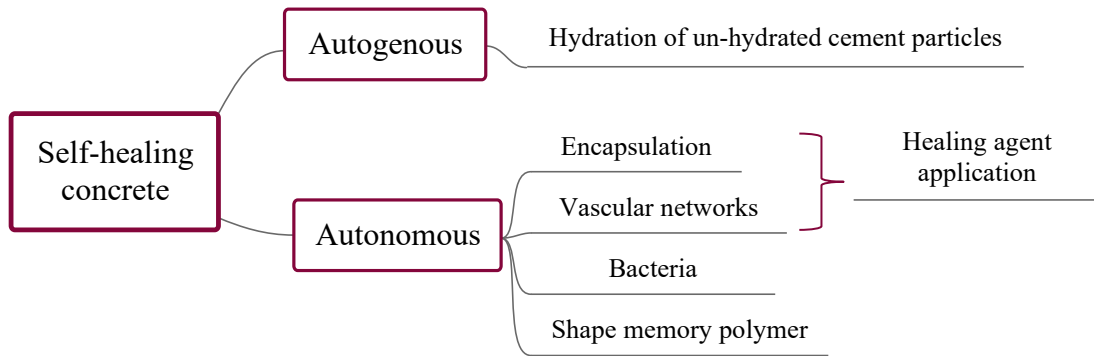


Figure 1.1 Taxonomy in self-healing concrete.

A significant number of studies on self-healing concrete have used encapsulation techniques, in which the healing agent are released and delivered to the damaged areas when cracking occurs (Gupta & Kua, 2016; Xue *et al.*, 2019). Different techniques have been introduced to encapsulate and deliver the healing agent in the self-healing concrete, such as encapsulation and vascular networks methods. Different healing agents have been studied as well, such as polymers, adhesives, mineral admixtures and chemical compounds. Various experimental characterisation techniques have been introduced to evaluate the performance of self-healing systems in concrete too (Ferrara *et al.*, 2018; Muhammad *et al.*, 2016; Sidiq *et al.*, 2019). At the same time, significant research works have been done in the numerical modelling of self-healing systems in concrete as well (T. Jefferson *et al.*, 2018; Mauludin & Oucif, 2019).

As presented in Figure 1.1, self-healing techniques such as encapsulation and vascular networks methods are rely on healing agents to achieve mechanical and durability recovery after healing action. While abundant progress has been done on many aspects in encapsulation-based self-healing concrete, the transportation of

healing agent in the concrete itself has rarely been researched (Z. Dong *et al.*, 2015). In addition, the researchers have acknowledged the complexity of self-healing processes in concrete. They have highlighted that the self-healing process in concrete is a set of multi-physics problems as it involves three interacting physical processes: fracture mechanism, fluid flow, and chemical reactions as presented in Figure 1.2. For the encapsulation techniques in self-healing concrete, the capsules containing a liquid healing agent are mixed in the concrete. If the crack forms, it will rupture the capsules, the glue-like healing agent that will glue together the cracks after the healing agent cures.

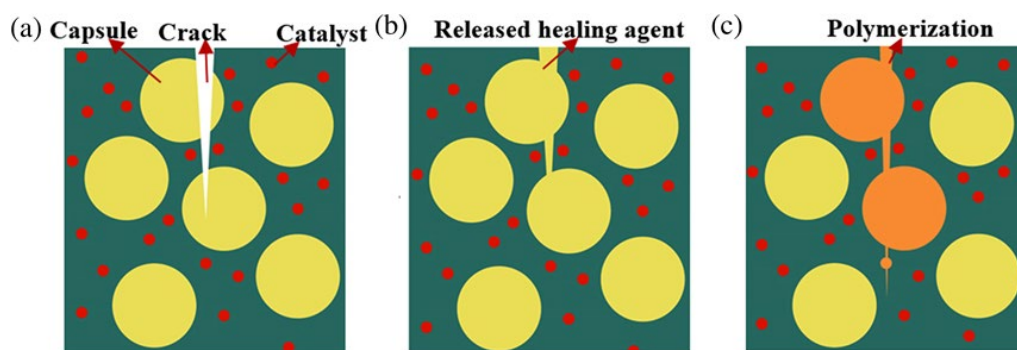


Figure 1.2 The schematic concept of encapsulation-based autonomic healing for cementitious materials (Xue *et al.*, 2019).

Fast-curing healing agent, such as, cyanoacrylate cures rapidly when contacts with the moisture on the concrete surface. During the rapid curing process, the viscosity of the healing agent increases and might retards the healing agent flow in the discrete crack. In order to achieve satisfying healing efficiency, it is required to ensure that a sufficient amount of healing agent is delivered to the discrete crack before the healing agent is fully cured. Therefore, the in-depth investigation on the combination effects of the fluid flow and the curing processes is getting interested and requires further discussion within the field of encapsulation-based self-healing system in concrete.

1.2 Problem Statement

In the autonomic self-healing system, the transportation of the healing agent plays a primary role in determining the concrete healing efficiency. Self-healing techniques like encapsulation and vascular networks store, deliver and release the embedded healing agent to the damaged site when cracking occurs. In order to understand the self-healing mechanisms, it is required to study the transport processes of healing agents in concrete. Several numerical works that address transport processes in self-healing concrete have focused on the transportation of moisture and ions associated with carbonation and autogenous healing (Aliko-Benítez *et al.*, 2015; Freeman, 2017; Huang *et al.*, 2010; Huang & Ye, 2016; Ranaivomanana & Benkemoun, 2017). Although they are related to this study, the present work places more focus on the transportation of healing agents in autonomous self-healing concrete instead.

In the actual healing agent flow in discrete cracks within the concrete matrix, the visualisation of the healing agent flow phenomenon is inherently challenging and is constrained by the limitation of visualisation equipment, small crack size and concrete opaqueness. In terms of flow modelling, the classical Lucas-Washburn equation (LWE) has a limitation in predicting the capillary flow in non-uniform channels and complex porous media since the LWE is developed with the assumption on the fluid flow as one-dimensional laminar flow in a uniform channel with a constant contact angle. The LWE equation is required to be modified to describe the capillary flow in different porous systems. Therefore, a numerical simulation might be an alternative to better characterise the capillary flow process and provide a better depiction of the multiscale physical process of capillary flow. Numerical simulation can also aid with the visualisation and examination of the capillary flow from a microscopic perspective. However, to date, a limited number of numerical models have taken into account the coupled effects of flow and curing processes in self-healing systems.

Apart from the works by previous researchers (Freeman & Jefferson, 2020; Gardner *et al.*, 2012, 2014, 2017; Gilabert *et al.*, 2017; Selvarajoo *et al.*, 2020), there

is no other work on the investigation of healing agent flow in autonomic self-healing concrete. It has been proven that the viscosity of the healing agent increases during the curing reaction process and undergoes a liquid/solid transition (Freeman & Jefferson, 2020; Gardner *et al.*, 2017). To date, the influence of the curing process on its rheological properties has not been adequately addressed during the healing agent infiltration. In addition, a numerical simulation model considering the combination effects between capillary action and curing processes in encapsulation-based self-healing concrete has not been established yet. To consider the curing effect, the inclusion of Castro-Macosko viscosity model in the capillary flow model needs further investigation and validity checking. Thus, in the present study, experimental and numerical investigations are performed to study the influence of the curing reaction on the rheological properties of healing agents during the capillary flow event in the encapsulation-based self-healing system in concrete.

1.3 Objectives

The present study looks into the transportation of healing agents during the curing process in encapsulation-based self-healing concrete. Understanding the transport process of the healing agent is essential to designing a better self-healing system in concrete. Some of the specific objectives in this study include:

- To determine the correlation between curing effect and changes in rheological properties of the healing agent by using dynamic mechanical analysis.
- To simulate the capillary flow of the healing agent in discrete cracks using the Volume-of-Fluid method together with the implementation of the dynamic contact angle model.
- To describe the viscosity change of the healing agent using the Castro-Macosko viscosity model and couple the model with the aforementioned fluid flow model to better simulate the curing effect of healing agents under varying capillary flows.

- To examine the various parametric effects on the capillary flow and corresponding self-healing efficiency by using the coupled model.

1.4 Scope of the Study

In the present study, both experimental and numerical studies are carried out to provide a better insight into the physics of the problems and to allow for the enhancement of the self-healing performance by comprehensively simulating and designing a better self-healing system. This work is not trying to replicate the embedded capsules in any particular vascular or encapsulated self-healing system in concrete. The key target for this study is to investigate the effects of curing reaction on the capillary flow during the infiltration of the healing agent in discrete cracks in concrete. This work focuses on encapsulation-based self-healing systems that use cyanoacrylate as a healing agent but could readily be extended to a wide range of other healing agents as well.

In terms of experimental measurement and analysis, the measurement of rheological properties of the cyanoacrylate-based healing agent are performed by using dynamic mechanical analysis in a rotational rheometer. The rheological measurement results are utilized specifically to show the correlation between the viscosity properties and the curing reaction of the cyanoacrylate adhesive. In terms of numerical modelling, a viscosity function is used to describe the curing effect on the rheological properties. The Castro-Macosko model is selected and specified as the viscosity function for this study. Material constitutive models, such as dynamic contact angle model, Castro-Macosko viscosity model, and degree of cure functions, is written in the form of user-defined functions and coupled via ANSYS Fluent with ANSYS 2021 R1 Student Version. ANSYS Fluent, a Finite Volume-based computational fluid dynamic (CFD) simulation software is selected as the software of choice for this study as it can be used to solve the fluid flow modelling by simulating the transport processes of the healing agent in a discrete crack. A Volume-of-Fluid (VOF) technique is applied to track the moving meniscus in the crack.

In the simulation, healing agent flow is assumed to be laminar and incompressible. When cracking occurs, the embedded capsule ruptures and releases the healing agent into discrete concrete cracks. Therefore, the healing agent flow is assumed to be driven by capillary action (Z. Dong *et al.*, 2015). Surface tension and wall adhesion models are used to determine the capillary pressure in the capillary flow. As the scope of this study encompasses both the healing agent flow and the viscosity change (corresponding to the curing degree of the healing agent in the crack) during the curing reaction (polymerisation), the capillary flow model is coupled with the Castro-Macosko model to comprehensively predict the healing agent flow and its reaction in discrete concrete cracks. The coupled model is validated with available capillary rise data, and hopefully the simulation results will provide a better visualisation for the transport process of the healing agent in discrete cracks. To supplement the numerical modelling and experimental validation, parametric studies are conducted to investigate the influence of the healing agent's rheological properties and crack geometry on the capillary flow and corresponding self-healing efficiency in discrete concrete cracks.

1.5 Significance of the Study

This study contributes to the knowledge development in self-healing concrete with practical and theoretical significances as follows:

- a) Theoretical significance
 - a. This study introduces a VOF multiphase model combined with surface tension and dynamic contact angle models to simulate the capillary rise of liquids in discrete cracks. The velocity-dependent dynamic contact angle model can be determined from capillary rise measurement data.
 - b. This study develops a novel, coupled model consisting of the VOF, surface tension, dynamic contact angle, and viscosity models. The coupled model can comprehensively simulate a reacting healing agent flow in the discrete cracks within self-healing concrete and allows for

a better understanding of the intricate mechanism of reaction-based healing in encapsulation-based self-healing concrete. This, in turn, can serve as a sound basis for future researches in the similar vein.

- c. This study proposes a new experimental measurement method in order to determine the material and rheological properties of cyanoacrylate-based healing agent used in encapsulation-based self-healing concrete. With the novel implementation of said measurement method in this field, it allows for a more comprehensive measurement and analysis of the material and rheological properties of self-healing concrete samples, thus improving related research analysis efforts as a whole.

b) Practical significance

- a. The numerical simulation technique provides a clear visualisation and a reliable prediction of the transport processes of the healing agent in discrete cracks. The result provides a better understanding of the healing agent's flow behaviours and shows how the curing reaction affects the capillary flow with increasing viscosity. Thus, the result can be used for selecting a suitable healing agent with an optimised healing rate and volume. The numerical model is ready to be extended to simulate the healing agent flow in a more complex crack geometry. In addition, the model can be used for a wide range of healing agents, thus expanding the model's inherent practicality.
- b. The parametric studies provide a better understanding of healing agent flow mechanisms and help related industries to design a better self-healing system. The results give a deeper insight into the impacts of the parameters (surface tension, contact angle, viscosity, crack configurations) on the capillary flow of the healing agent in discrete cracks and can help other researchers in their investigations by modifying the healing agent's material properties to improve the healing efficiency of the self-healing system, thus benefiting the concrete manufacturing industry and tackling the ensuing sustainability issues in the long run.

1.6 Outlines

The thesis presents the development of a numerical simulation model for the healing agent's capillary flow in the concrete's discrete crack. This thesis includes six chapters which are organised as follows:

Chapter 2 presents a literature review on self-healing concrete and specifies the knowledge gaps that have to be bridged by this research. The review starts with the background and the types of healing mechanisms in self-healing concrete, such as autogenous and autonomic healings. The review also discusses different techniques used in the autonomic healing system and the evaluation methods for assessing self-healing efficiency. Next, the review discusses the experimental and numerical investigation of healing agent flow in capillary cracks and its curing process.

Chapter 3 presents the detailed methodology of the study. The numerical model and its governing equations for simulating the healing agent's capillary flow in the discrete crack are discussed. The testing methods in order to determine the rheological properties of a cyanoacrylate-based healing agent is presented.

Chapter 4 presents the validation of the VOF capillary flow model and the simulation of the capillary flow with the help of Computational Fluid Dynamic (CFD) packages in ANSYS Fluent simulation software. The inclusion of the dynamic contact angle model in improving the capillary flow prediction is discussed. Later, the validation of the VOF capillary flow model coupling the Castro-Macosko viscosity model is presented and discussed.

Chapter 5 presents the simulation of the healing agent flow in discrete cracks, considering the curing effect after validating the coupled model (coupling of fluid flow model and Castro-Macosko viscosity model) as presented in Chapter 4. The capillary flow of the healing agent in the discrete crack and its flow characteristics are investigated with various flow parameters, which in turn affect the infiltration rate and the final location of the healing agent in the discrete crack.

Chapter 6 summarises and concludes this research with some recommendations for future works.

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