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1d Modelling Of Healing Agent In Self Healing Concrete Using Finite Element Method

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Abstract: The stability and safety of structures are always in the heart of civil engineering disciplines. Thus, the formation and healing of concrete cracks are crucial in terms of monitoring or predicting the subsequent effects. Computational or numerical modelling in the field is still lacking and in the development stage despite the fact of its ability to obtain faster and cost-saving data or results as compared to the experimental approach. This study represents a numerical model to imitate the diffusion of urea into the water-filled crack and then of precipitation of calcite, generated from the hydrolysis process of urea. The precipitation of calcite is a solid that will close the crack. The process depends on the existence of urea in the form of capsules and with the existence of bacteria, water, nutrients and calcium in the crack domain, calcite will be induced. This study aims to provide mathematical modelling using MATLAB to have an accurate simulation of bacteria-based healing and the time required to heal a crack. The model can be developed by discretising Partial Differential Equations using Galerkin Finite Element. The model is developed in 1D with linear elements having 2 nodes each. Then, the assembly of global matrices takes place by overlapping of corner nodes from local matrices. Moreover, initial and boundary conditions are provided to the model. The results are validated with past literature. Hence, it is shown that with initial urea concentration, c_0 of 333 mole/m^3 that is diffusing through the crack with a length, L of 20 mm, a healing ratio, S of 60% can be achieved within 500 days.

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Keywords: Self-Healing Concrete; Finite Element Method; Urea; MATLAB; Galerkin Finite Element.

1. Introduction

The formation process of the micro-cracks in concrete may form linked and connected flow routes as a result of external loads, that may provide a path for undesired chemicals or substances to finding its way into the concrete. Subsequently, this may result in possible corrosion of the rebars, which will vehemently deteriorate the durability of the structure. Cracks are participating in the drop of the strength, stiffness, durability and the lifespan of concrete structures [1]. Over the last few years, a state-of-the-art method to solve this problem, called self-healing, has gained much attention. Simply, we can express and simulate the healing process or the filling of the micro-cracks as to the healing of wounds in the human body. Self-healing concrete has 3 main stages as shown in Figure 1 which are: crack formation (crack propagation), filling the crack with the healing agent, and precipitation of calcite to seal the crack pores. In this study, stage 2 and 3 would be highlighted. The flow of urea in the crack domain is visualized by the diffusion of urea over time. The sealing of crack pores is expressed by precipitation of calcite, p while the efficiency of healing which is represented by the healing ratio, S.

The various benefits and characteristics of concrete made it a booming material all over the world. About 7% of the total CO_2 emission in the anthropogenic atmospheric is often attributed to cement production and construction mechanisms which may enhance the material quality and sustainability. Consequently, this can contribute to a longer service life of concrete structures would make the material not only more durable but also more sustainable. As a consequence, the application of sustainable concrete is a crucial step on the path of mitigation the environmental impacts of massive using of concrete. [2].

Generally, the bacteria are being utilized through the oxidation of the organic acids. The role of the indigenous bacteria to release a urease enzyme would create microbial calcium carbonate. Then, that enzyme will stimulate the urea decomposition to carbonate and ammonium ions. According to Irwan and colleagues [3], Sulphate Reduction Bacteria (SRB) was utilized to generate calcium carbonate to fill the pores in the concrete specimen. Material properties tests were conducted such as compressive strength and water permeability to show the difference in performance before and after introducing the bacteria. A liquid culture containing SRB as replacement of water content was used at different ratios of 1%, 3% and 5%. Concrete specimens were cured at 7, 14 and 28 days under air conditions. Remarkable surge of compressive strength was reported to be 13.0%, while a decrease in water infiltration was 8.5% occurred with the addition of 5% SRB. Such enhancement in the compressive strength and water infiltration performance was attributed to the calcium precipitation within concrete pores.

Another approach introduced by Ersana et al, [4] which is nitrate-reducing bacteria that can suppress corrosion of rebars and promote crack healing, by producing NO_2 and stimulating $CaCO_3$ precipitation concurrently. That study investigated the implementation of one non-axenic and two axenic NO_3 as a way to reduce cultures for the development of corrosion-resistant self-healing concrete. A crack of width 0.46mm was healed with the utilization of nitrate-reducing bacteria in 8 weeks. Both axenic cultures managed to survive in mortar after incorporation in protective carriers. They became active 3 days after the pH dropped below 10. The non-axenic culture named “activated compact denitrifying core” (ACDC) showed comparable revivification efficiency without any additional protection. Oxygen is an agent that can induce corrosion, as bacteria feeds on oxygen, the tendency for the corrosion of reinforcement can be reduced. The utilizing of bacteria in self-healing is considered to have significant economic benefit due to the need to develop a concrete material which can repair itself. According to Bhaskar et al, [5], mineral producing bacteria can be added to cementitious mortars to enhance the self-healing ability. The study proposed the zeolite to be used as carrier substance to protect the bacteria in a high PH environment that usually appears in concrete.

A study by Balazs [6] highlighted three computational models on self-healing materials. The first system is imitated from biological cells or leukocytes that locate the damaged area and promote its repair. Secondly, the study discussed the behaviour of polymeric nanocomposites where the particles migrate to the area of the cracks in the brittle confined materials and enhance the restoration of the properties of multilayer composites. Finally, a model of a polymeric coating was described. Another remarkable 2D mathematical model was developed by Zemskov [7] with utilizing of bacteria-based healing agent.

That study used Galerkin FE analysis on the structured triangular mesh, so it is considered pertain closely with the present study. Various types of healing agents in SHC through the past studies at the laboratory and experimental scale are introduced. There are only a few numerical modelling involving the healing process such as the development of numerical models such as designing of ‘artificial leukocytes’, healing in polymer nanocomposites, and using bacteria as a healing agent. The common objective between the experimental and modelling is the formation of calcium carbonate (CaCo3) to seal the cracks. On the other hand, researchers have used different ways to generate calcium carbonate.

Generally, the main aim we have sought of this study is to manifest a 1D validated numerical model using MATLAB for a crack of self-healing concrete. This model can describe the kinetics of calcite precipitation by microbial calcium carbonate. In addition, to estimate both concentration of urea and time required to fully seal a crack as the healing ratio, *S* is 1.0. a.

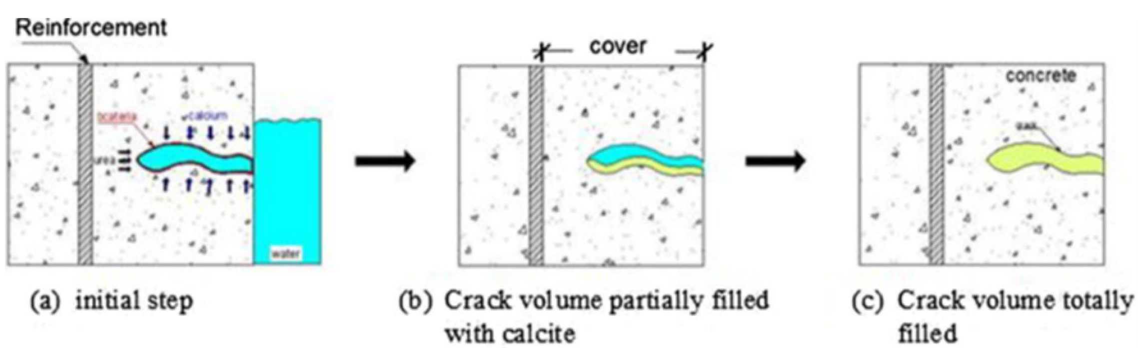


Figure 1. The development of a crack repairing over various stages

2. Material and Methods

Since the experimental work is neither a time-saving technique nor cost-saving, the need for computational modelling is now of importance. That is because modelling has the traits of being:

a) Flexible; to modify the inputs and make changes to the data and monitor or compare between the results.

b) Fast in means of getting the results. Apart from that, computational modelling can give information that is difficult to be measured using experimental such as the movement of urea in the crack itself and the precipitation process over time.

2.1 Finite Element Method (FEM)

This research will produce a 1D numerical model of crack of self-healing concrete. The scopes of study and limitations for this project are as follow: 1. The crack self-healing concrete model will be determined using Finite Element Method (FEM). This model only considers the chemical reaction mechanism. Other mechanisms such as fracture mechanism and fluid mechanism are not considered as it requires more complicated Governing Equations. 2. Crack already exists, and the nutrients, bacteria, urea, and calcium were presumed to be in the crack area for the chemical reaction to occur as shown in Figure 2.

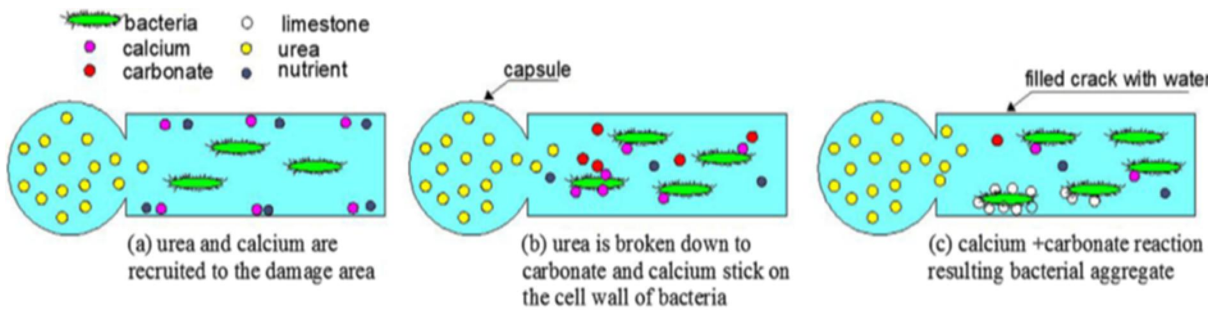


Figure 2. Schematic diagram of evolving microbial calcium carbonate (CaCo3), Algaifi (2018)

FEM is considered as one of the numerical techniques used to find approximate solutions for differential equations with applying adjustable boundary conditions. It divides a structure into many small elements, called 'finite elements', then connects those elements by 'nodes' at their ends considering the compatibility conditions for each element. Hence, two adjacent elements are supposed to share the same Degree of Freedom DOF at their connecting nodes. Through applying this method, a more complicated structure would be analyzed over more elements and nodes. Finally, properties of elements such as displacements, stresses, and nodal forces for structures and velocity and concentration for fluids can be obtained by solving a set of algebraic equations. Despite having various methods in FEM, they share the concept of converting the continuous nature of PDE or ODE to a similar and equivalent simultaneous algebraic equation which can be solved. This research consists of three key activities; description of the proposed model, model discretization, and model development and validation as shown in Figure 2.

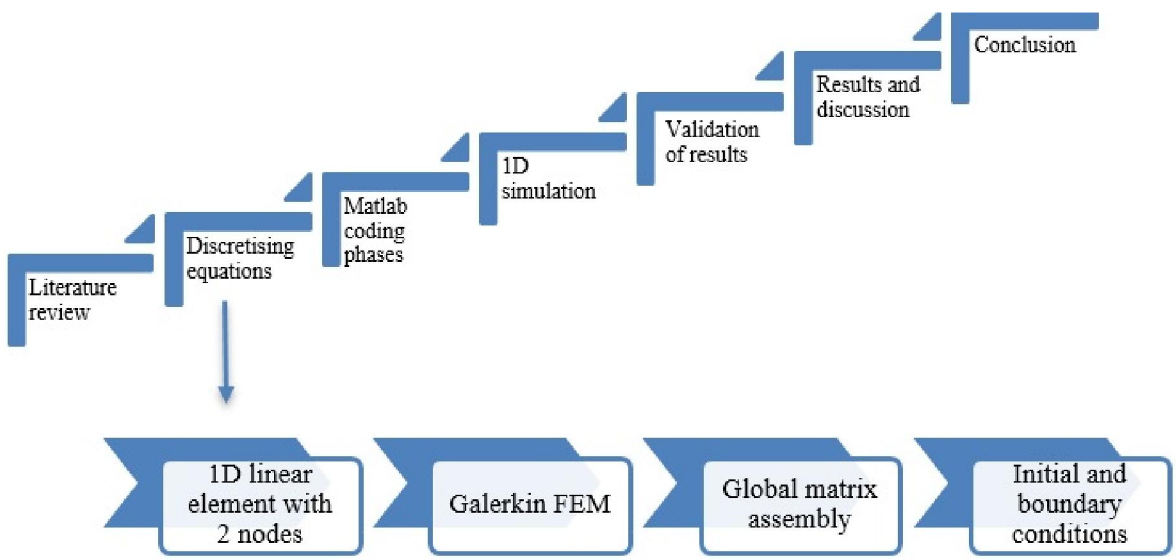


Figure 3. Flow chart of methodology adopted in this study

2.2 Description of the proposed model

A 1D mathematical model would be developed in the present study. The model is based on the following sequence:

- It is assumed to have the urea in the capsules, while bacteria, nutrients, calcium, and water would be available in the crack domain.
- Since the bacteria cells are negatively charged, ions of calcium are attracted and collected by the cell wall of bacteria.
- The hydrolysis process of urea would start only if the capsule is broken after passing through a crack.
- The cells of bacteria would release urease enzyme as a stimulator to the degradation process of urea ($CO\,(NH_2)_2$) to carbonate and ammonium, as expressed in Eq. (1).
- The carbonate would react with calcium which is already available on the cell wall of bacteria and hence, microbial calcium carbonate ($CaCo_3$) is produced, as shown in Eq. (2).
- It is important to mention that the deeper the crack, the less urea hydrolysis to occur. That is due to lack of oxygen which would haul the growth and reproduction of bacteria cells. Hence, that will decline the releasing of urease enzyme and then reduce the degradation of the urea to carbonate and ammonium. List of the parameters used in this study are shown in Table 1.

$$CO\,(NH_2)_2 + 2H_2O \xrightarrow{urease\ enzyme} 2NH_4 + CO_3^{-2} \quad (1)$$

$$Ca^{+2} + CO_3^{-2} \rightarrow CaCo_3 \quad (2)$$

Table 1. Parameters used in the study

Parameter	Symbol	Unit	Value
Length of the crack	L	m	0.02
The affected cracked area	A	m^2	0.004
Initial urea concentration	c_o	mole/m^3	333 and 750
Diffusion coefficient	D	m^2/day	$0.962*10^{-6}$
Ureolysis rate constant	k_1	1/day	1.0
Calcium carbonate precipitation rate	k_2	1/day	1.0
Bacteria cells concentration	α	cell/m^3	0.1
Calcium carbonate precipitation	p	mole	varies
Molar mass of calcite	M	g/mole	100.0869
Density of calcite	ρ	g/m^3	$2.711*10^6$

2.3 Model discretization

The general diffusion equation for fluids was introduced by Joseph Fourier in 1822 which is shown in Eq. (3). The equation is PDE which is defined as differential equation involving more than one independent variable. The independent variables here are time (t) and distance (x).

$$\frac{\partial c}{\partial t} = D\,\frac{\partial^2 c}{\partial x^2} \quad (3)$$

where, *c*, urea concentration (dependent)
t, time (independent)
D, diffusion coefficient (constant)
x, coordinate in space (independent)

In this study, the diffusion flux and the concentration of urea vary with time. Moreover, the hydrolysis rate can be expressed as the depletion rate of urea species which are required to produce calcite through the continuous degradation of urea to ammonium and carbonate. Hence, the depletion rate in fact is a reduction in urea concentration. As a result, the depletion rate will be accompanied by a negative sign to indicate the flow of urea species from high concentration regions to low concentration regions. Hence, Eq. (3) can be written as

$$\frac{\partial c}{\partial t} = D\,\frac{\partial^2 c}{\partial x^2} - k_1\,\alpha\,c \quad (4)$$

where k_1, Ureolysis rate constant and
α, Bacteria cells concentration

Galerkin FEM is applied to solve Eq. (4) by dividing the crack domain into finite length involving 1D finite elements. Galerkin FEM is applied to linear element with two nodes, the first is at x=0 while the second is at x=L, where L is the length of the element and not the length of the whole domain. The concentration, c is then approximate using the two nodes of a linear element multiplied with an interpolation function. In FEM, the interpolation function act as a shape function *N*. Eq. (5) is the approximation in FEM. Figure 4 shows an illustration of a linear 1D element with 2 nodes that has been used in this study.

$$c\,(x) = N_1\,c_1 + N_2\,c_2 \quad (5)$$

$$N_1 = \frac{1-x}{L}\,,\; N_2 = \frac{x}{L} \quad (6)$$

Where

N_1, is the shape function at the first node

N_2 is the shape function at the first node

c_1 is the concentration of urea at the first node

c_2 is the concentration of urea at the second node.

Figure 4. Linear bar element

Inserting the approximation solution of Eq. (5) into Eq. (4), and analysed throughout the crack volume, $V = A\,dx$. The following equation is then obtained.

$$\int_0^L N_i \left(\frac{\partial c}{\partial t} - D\frac{\partial^2 c}{\partial x^2} + k_1\,\alpha\, c\right) A\,dx = 0 \quad (7)$$

where i, is the number of nodes (i=2).

The next step is to perform integration for Eq. (7). Integration is done for the first and third term, while Integration by Parts (IBP) is utilized to derive the second term.

The first term initial derivation steps are:

$$\int_0^L A\,N_i \left(\frac{\partial c}{\partial t}\right) dx \qquad i=1,2 \qquad (8)$$

$$A\int_0^L \left(1-\frac{x}{L}\right)\left(\frac{\partial c}{\partial t}\right) dx = A\int_0^L \left(1-\frac{x}{L}\right)\left(\frac{\partial c_1 N_1 + c_2 N_2}{\partial t}\right) \qquad (9)$$

$$A\int_0^L \left(1-\frac{x}{L}\right)\left(\frac{\partial c_1\left(1-\frac{x}{L}\right) + c_2\left(\frac{x}{L}\right)}{\partial t}\right) dx \qquad (10)$$

Until it becomes $\frac{LA}{6}\begin{bmatrix}2 & 1\\ 1 & 2\end{bmatrix}\begin{Bmatrix}\dot{c}_1\\ \dot{c}_2\end{Bmatrix}$ (11)

The second term;

$$\int_0^L -A\,D\left(\frac{\partial^2 c}{\partial x^2}\right) dx \qquad i=1,2 \qquad (12)$$

By applying integration by parts,

$$\int u\,dv = u.v - \int v\,du \qquad (13)$$

Let u=N_i, Hence, $\frac{du}{dx} = \frac{dN_i}{dx}$ (14)

du= $\frac{dN_i}{dx}\,dx = dN_i$ (15)

Let, $dv = \left(\frac{\partial^2 c}{\partial x^2}\right) dx$, Hence,

v = $\int\left(\frac{\partial^2 c}{\partial x^2}\right) dx = \frac{dc}{dx}$ (16)

$$\int u\,dv = u.v - \int v\,du = N_i\frac{dc}{dx} - \int\left(\frac{dc}{dx}\right) dN_i \qquad (17)$$

Until it becomes $\frac{AD}{L}\begin{bmatrix}1 & -1\\ -1 & 1\end{bmatrix}\begin{Bmatrix}c_1\\ c_2\end{Bmatrix}$ (18)

The third term.

$$\int_0^L \alpha\,A\,k_1\,(N_i\,c)\,dx \quad i=1,2 \qquad (19)$$

$$= \alpha\,A\,k_1\int_0^L (N_1\,c)\,dx \qquad (20)$$

$$= \alpha\,A\,k_1\int_0^L \left(1-\frac{x}{L}\right)\left[c_1\left(1-\frac{x}{L}\right) + c_2\left(\frac{x}{L}\right)\right] d \qquad (21)$$

Until it becomes $\frac{A\alpha k_1 L}{6}\begin{bmatrix}2 & 1\\ 1 & 2\end{bmatrix}\begin{bmatrix}c_1\\ c_2\end{bmatrix}$ (22)

After integration, Eq. (7) becomes.

$$\frac{LA}{6}\begin{bmatrix}2 & 1\\ 1 & 2\end{bmatrix}\begin{Bmatrix}\dot{c}_1\\ \dot{c}_2\end{Bmatrix} + \frac{AD}{L}\begin{bmatrix}1 & -1\\ -1 & 1\end{bmatrix}\begin{Bmatrix}c_1\\ c_2\end{Bmatrix} + \frac{A\alpha k_1 L}{6}\begin{bmatrix}2 & 1\\ 1 & 2\end{bmatrix}\begin{bmatrix}c_1\\ c_2\end{bmatrix} = \begin{Bmatrix}F_1\\ F_2\end{Bmatrix} \qquad (23)$$

For simplicity, Eq. (23) can be expressed as:

$$[m]\{\dot{c}\} + [k]\{c\} + [k_g]\{c\} = \{f\} \qquad (24)$$

Where

1- $\frac{LA}{6}\begin{bmatrix}2 & 1\\ 1 & 2\end{bmatrix} = [m]$

2- $\frac{AD}{L}\begin{bmatrix}1 & -1\\ -1 & 1\end{bmatrix} = [k]$

3- $\frac{A\alpha k_1 L}{6}\begin{bmatrix}2 & 1\\ 1 & 2\end{bmatrix} = [k_g]$

In order to obtain the concentration for the subsequent time, forward finite difference approach was utilized. The approximate derivation of time for the nodal concentration matrix can be shown as:

$$\dot{c} = \frac{\{c_{n+1}\} - \{c_n\}}{\Delta t} \qquad (25)$$

where n is the time counter.

By substituting Eq. (25) into Eq. (23) and getting the concentration along the crack length. The following can be obtained

$$c(x,t) = c + (\Delta t * M^{-1} * (F-(K*c) -(Kg*c)) \quad (26)$$

Initial condition and would be $c(x,0) = 0$,

while boundary conditions; $c(0,t) = c_o$, and $c(L,t) = 0$

Precipitation of calcite, p can be calculated along the crack from; $p = p + (\Delta t * k_2 * K_g * c)$; (27)

Where:

p, precipitation of calcite along the crack

Δt is the time step

k_2, is the Calcium carbonate precipitation rate

$K_g = \frac{A\alpha k_1 L}{6}\begin{bmatrix} 2 & 1 \\ 1 & 2 \end{bmatrix}$ from Eq. (24)

c_o, urea concentration

The efficiency of healing the crack is measured by the healing ratio, S which is shown as follows; $S = V_h/V$.

Where

V_h, volume of the healed portion of the crack

V, volume of the original crack

$V_h = (M*p)/(2.711*10^6)$

M, molar mass of calcite =100.0869 g/mole

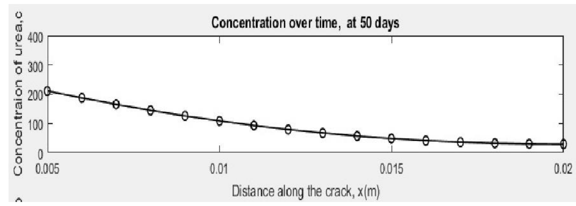
ρ, density of calcite = $2.711*10^6$ g/m^3

3. Results and discussion

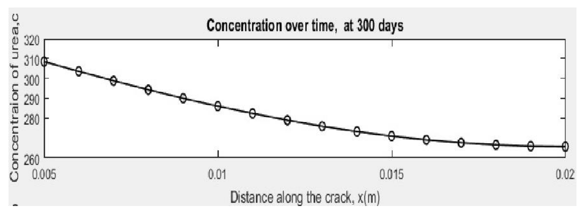
The results from the MATLAB programming are given and explained in detail. The precipitation process is shown through highlighting the concentration of urea which would decrease, precipitation of calcite and healing ratio which would increase.

3.1 Validation of Concentration vs distance

Cases from Algaifi (2018) are compared to the present model to ensure its validation. The results of concentration over the length in the absence of bacteria ($k_1=k_2=0$) are shown here (equivalent to Eq. (3) for diffusion of urea only). The initial urea concentration, c_0 is set to be 333 mole/m^3 at the crack opening, $x=0$ and fixed over time. Physically, this is always presenting the urea at one end of the crack with 333 mole/m3 value. The comparison between Figure 5 and Figure 6 shows a good agreement.



(a)



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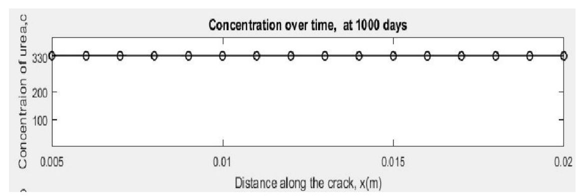
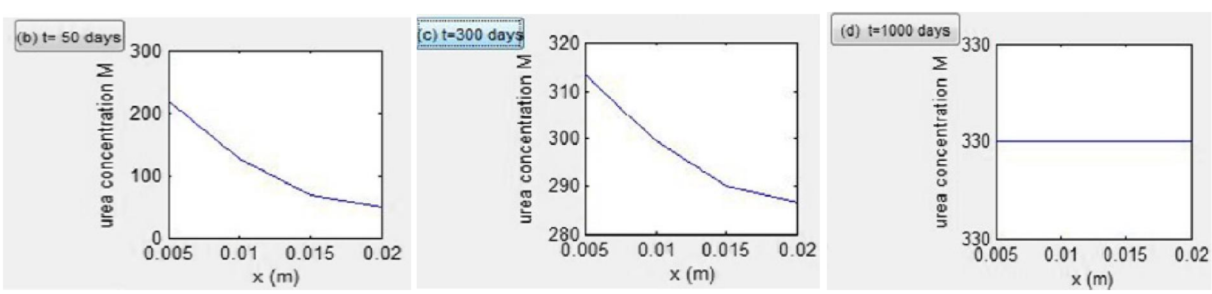


(c)

Figure 5: Concentration of urea diffusion at (a) 50 days, (b) 300 days, (c) 1000 days.



(a) (b) (c)

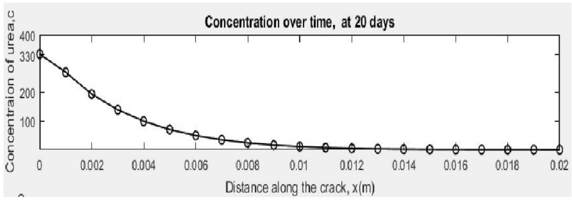
Figure 6: Concentration of urea at (a) 50 days, (b) 300 days, (c) 1000 days, Algaifi (2018)

3.2 Simulation of results

The results here are in case of existence of bacteria ($k_1=k_2=1.0$) and they will be shown at 20, 200, and 500 days (equivalent to Eq. (4)). This means that now the urea diffuses and also deplete due to the ureolysis process.

3.2.1 Concentration vs distance

Here is the first result between concentration of urea on the y axis and the distance along the crack length on the x axis. While the time lapses, the concentration of urea decreased as compared to Figure 2. That reduction happens as the urea is being utilized in the reaction with water to form carbonate, as shown in Eq. (1). This means that as the urea diffuse into the crack, it also reduces.



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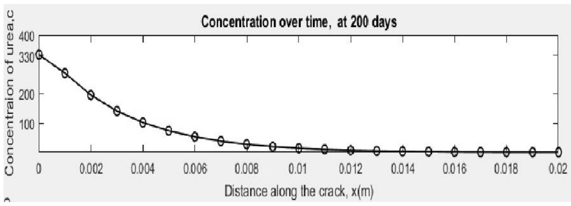

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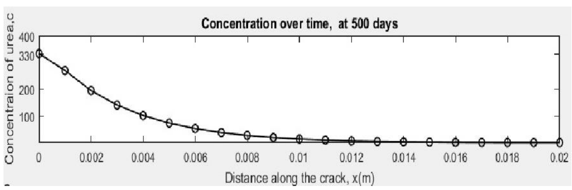
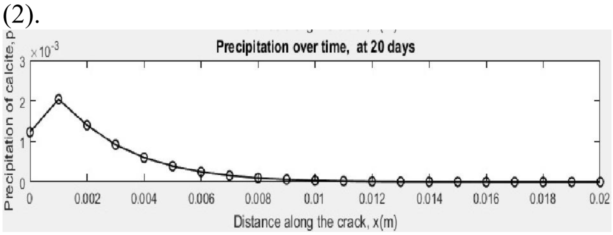


(c)

Figure 7: Concentration of urea at (a) 20 days, (b) 200 days, (c) 500 days with existence of bacteria

3.2.2 Precipitation vs distance

This section shows the relationship between precipitation of calcite on y axis and the distance along the crack length on x axis. As the time lapse, the precipitation of calcite increased. That surge happens as calcite is being formed from the continuous reaction between calcium and carbonate. Refer to Eq. (2).



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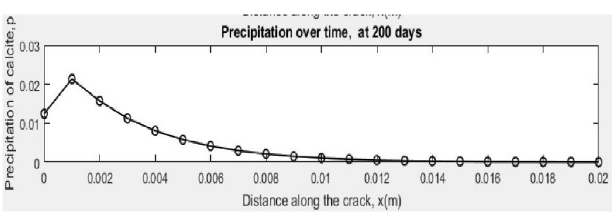

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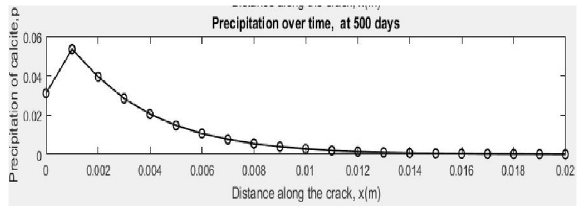
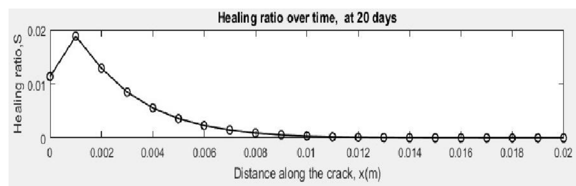


(c)

Figure 8: Precipitation of calcite (a) 20 days, (b) 200 days, (c) 500 days

3.2.3 *Healing ratio vs distance*

Here is the third result which is relationship between healing ratio of the crack, *S* on *y* axis and the distance along the crack length on *x* axis. This process is going on over time. Thus, while the time lapses, the healing ratio of the crack would be increased. That surge happens as the volume of the healed portion of the crack domain, V_h is increasing due to the continuous formation of calcite.



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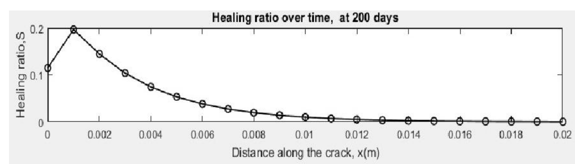

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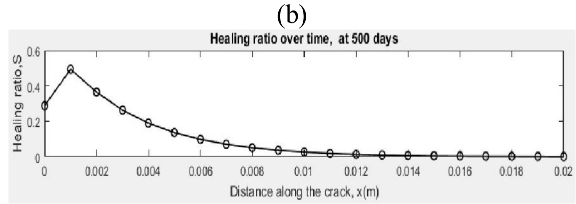

(c)

Figure 9: Healing ratio at (a) 20 days = 17%, (b) 200 days = 20%, (c) 500 days = 50%

The healing ratio has a maximum value of 1.0 as that indicates full healing of the crack domain and then the reaction process should be stopped. In that case, the volume of the healed portion of the crack, V_h would be equal to the original crack volume, V ($S=V_h/V = 1.0$). That could be achieved in this model if we increase the initial concentration of urea, c_0 from 333 mole/m^3 to 750 mole/m^3 within time of 450 days. Figure 10 shows initial concentration of urea, $c_0 = 750$ mole/m^3 at time 450 days, while Figure 11 shows the healing ratio, S=100% at the fully healing case.

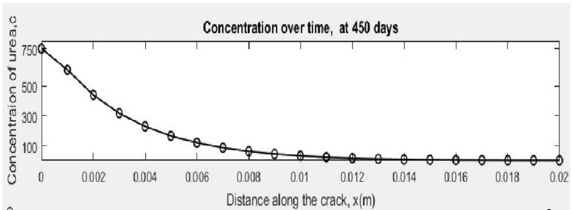

Figure 10: Concentration of urea, c_0=750 mole/m^3

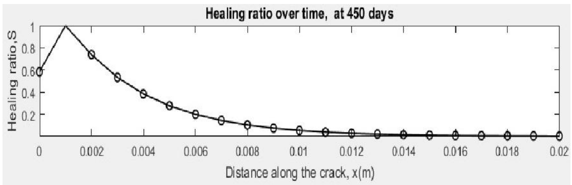

Figure 11: Healing ratio, S=100% at c_0=750 mole/m^3 and time 450 days

4. Conclusion

A numerical model for 1D diffusion and precipitation of the healing agent has been developed and validated. Finite element with 2 nodes linear element is applied in this study to discretize and simulate the problem. Movement of the healing agent and solidification processes are dependent on a few parameters such as c_o, urea concentration, diffusion coefficient, D, M, molar mass of calcite, and ρ, density of calcite. It shows the concentration of urea at different points along the crack which is dependent on the ureolysis of urea. For example, results showed that precipitation of calcite, p at 500 days was 0.57 cell/m^3, while the healing ratio, S at the same time was 50%. In addition, the study highlights the amount of precipitated calcite which depends on the total degraded urea. This model helps to determine the time required to fully heal a crack volume and seal it with calcite. That can be referred from the healing ratio, S. For the current dimensions of crack, there will be an S of 1.0 to indicate a full healing of the crack when c_0=750 mole/m^3 over 450 days.

As a result, this model provides feasible flow which saves engineers and researchers time. It also may ease the process of planning during the early stages of construction or any other related engineering problem. Hence, the study helps to cater the problem of time or facilities required to perform the experimental work at lab. The model can be upgraded to 2D level which caters x and y dimensions of the crack volume or 3D level which caters x, y, and z dimensions of the crack volume. This study can contribute to determine the mechanism of the healing process of cracks in Self-Healing Concrete. In future, the results can be compared with experimental results for better validation. Also, other parameters could be considered such as effect of chemical attack, earthquakes, temperature, and intersecting cracks behaviour.

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