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Analytical Model for the Probability Characteristics of a Crack Penetrating Capsules in Capsule-Based Self-Healing Cementitious Materials

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Autonomous crack healing using pre-embedded capsules containing healing agent is becoming a promising approach to restore the strength of damaged structures. In addition to the material properties, the size and volume fraction of capsules influence crack healing in the matrix. Understanding the crack and capsule interaction is critical in the development and design of structures made of capsule-based self-healing materials. Continuing our previous study, in this contribution a more practical rupturing mode of capsules characterizing the rupturing manner of capsules fractured by cracks in cementitious materials is presented, i.e., penetrating mode. With the underlying assumption that a crack penetrating capsules undoubtedly leads to crack healing, geometrical probability theory is employed to develop the quantitative relationship between crack size and capsule size, capsule concentration in capsule-based self-healing virtual cementitious material. Moreover, an analytical expression of probability of a crack penetrating with randomly dispersed capsules is developed in two-dimensional material matrix setup. The influences of the induced rupturing modes of capsules embedded on the self-healing efficiency are analyzed. Much attention is paid to compare the penetrating probability and the hitting probability, in order to assist the designer to make a choice of the optimal rupturing modes of capsules embedded. The accuracy of results of the theoretical model is also compared with Monte-Carlo numerical analysis of crack interacting with capsules. It shows that the developed probability characteristics of a crack interaction with capsules for different rupturing modes is helpful to provide guidelines for designer working with capsule-based self-healing cementitious materials.
Keywords: self-healing, capsules, crack, penetrating probability, cementitious materials.

1. INTRODUCTION

Structural degradation in materials is generally attributed to the initiation and propagation of cracks at different length scales. The demand for continual improvement of material performance is a common feature of many modern engineering projects and fast repair of the cracks is desirable. However, repair costs are usually large and in some cases repair is impossible due to inaccessibility. A possible solution is self-healing of cracks in materials without external intervention. Autonomic self-healing of cracks in engineering materials is indispensable to maintain the load-bearing capacity, reliability of structure, and further the service life of structure by preventing micro-crack growth into catastrophic macro-cracks. Self-healing materials can be thought of as materials which respond to damage by automatically initiating some form of repair mechanism. A great achievements have been made in the self-healing materials such as polymer materials with encapsulated healing agent [1, 2], cementitious composites healed via embedded capsules containing healing agents [3], etc.

A group from the University of Illinois at Urbana-Champaign first reported autonomous self-healing material by incorporating a microencapsulated healing agent and a catalyst chemical trigger in an epoxy matrix [1]. It is extremely important to determine the effect of capsule concentration, embedded capsule size and crack size on the self-healing effectiveness in the development of the self-healing system. The effect of microcapsule size and crack length/volume on the performance of self-healing polymer was experimentally examined by Rule et al. [4]. The self-healing performance scales with the amount of healing agent delivered to the crack and is proportional to the product of the microcapsule concentration and diameter. They also experimentally verified that when the crack volume exceeds the amount of available healing agent, less successful healing is achieved. Knowing the cracks can occur at various length scales, sub-micron to several inches or meters in length, the determination of capsule geometry characteristics (shape, size) and material quality should be in accordance with the particular application of self-healing materials. For concrete structures, cracks with lengths of the order of few millimetres or inches are of concern and the embedded capsule size in the same order is required [5 – 8]. The fundamental requirement for the self-healing process to work is that cracks need to reach the capsules and cause to be ruptured. For smaller volume fraction of capsules, the chances of cracks intersecting the capsules may be low. For larger volume fraction of capsules, the intersecting probability improves only at the expense of losing the integrity of the matrix. The same explanation holds true for capsule size.

Understanding the crack and capsule interaction in addition to the self-healing chemistry agent and

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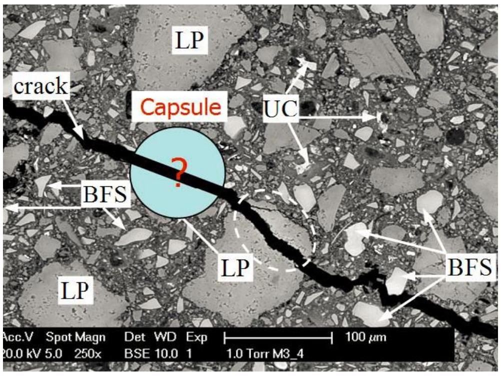
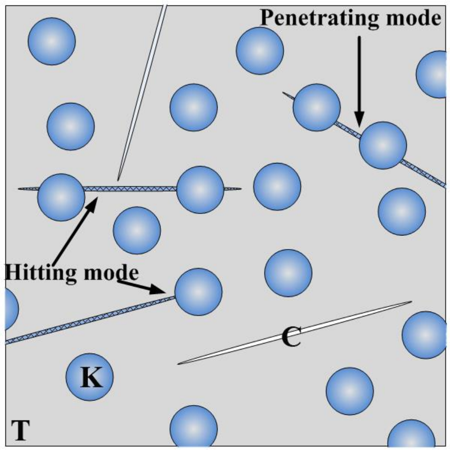
mechanical characterization studies will provide critical insight in the selection of optimal self-healing material system [9]. In essential, the ruptures of capsules are a primary step for self-healing action. Recently Lv et al. [10 – 13] applied geometrical probability theory to determine the exact amount of capsules required to completely or partially repair the cracks in self-healing materials for certain specific scenarios. Similar probabilistic certification was found to be highly desirable for efficient application of self-healing in advanced aerospace structures [14]. Using elementary probability principles, Zemskov et al. [6] recently developed two-dimensional analytical self-healing models and computed the probability of crack intersecting an encapsulated particle in the self-healing materials. These analytical models provided guidelines to estimate critical crack lengths, ideal capsule size, and mean inter-capsule distance in order to improve the efficiency of capsules in a self-healing material.

As Yang [15] reviewed that one of the key issues of self-healing composites by means of microencapsulation lies in rigidity of the shell substance and matrix, and crack trigger in capsules embedded composites depends on matching of deformation characteristics of the related materials. Route of crack propagation is a function of the stiffness ratio of microcapsules and matrix. If the capsules have higher modulus than that of the matrix, the approaching crack tends to pass by the capsules; conversely, the crack could penetrate the microcapsules when the matrix is stiffer [1, 15]. The physical characteristics of compliant interface between capsules shell substance and matrix have been investigated by both theoretical and numerical schemes [16 – 18]. Actually, the bonding between capsule and the bulk matrix is related to interfacial transition zone (ITZ). The ITZ between capsule and bulk matrix varies with materials of capsule shell and mixture of the bulk paste. It can be investigated with similar methods for studying the ITZ between aggregate and bulk paste [17]. When the capsules which consists of saturated SAP particles as the core and sealing shell as the outer surface are incorporated into engineered cementitious composites (ECC), the bond strength at interfacial transition zone (ITZ) between the sealing shell and the cement-based matrix needs to be stronger than the strength of capsule itself, to ensure the artificial cracks propagate through (i.e. penetrate) the capsules instead of passing around the capsules [19]. Thus high bond strength at the interface is one of the important factors contributed to cracks penetrating the capsules. This can be illustrated in Fig. 1 a. For the sake of occurrences of self-healing the penetrating way is expected and more attractive. Intuitively, we name it by penetrating mode that a crack penetrates the capsule(s) where the causes of cracking in matrix are not considered as shown in Fig. 1 b. When capsules are randomly distributed in the matrix and a crack occurs and grows, the chance of a crack penetrating into the capsules in the material is helpful to investigate the self-healing efficiency of capsules. On the other hand, another probability of capsules that a crack hits or meets the embedded capsules has been studied and much progress has been made by modeling methods and numerical simulation [6, 9, 20, 21].



a



b

Fig. 1. The crack pattern interaction with capsule in capsule-based material and the induction of penetrating mode: a – ideal pattern of the crack penetrating saturated SAP particles in capsule-based self-healing engineered cementitious composites (ECC) [19]; b – the penetrating mode is differentiated from and included in the hitting mode in an intuitive way

We here name it by hitting mode that a crack hits or meets the capsule(s) in the matrix, as shown in Fig. 1 b. Zemskov [6] has developed two two-dimensional analytical models for the computation of the probability that a crack hits an encapsulated particle and applied the models into the self-healing cementitious material where the bacteria were used as healing agent. Under the assumptions that the capsules are randomly dispersed in the matrix and the dispersed cracks occur independently in matrix, Lv [9, 11] developed a probabilistic model of capsules hit by the cracks to obtain the theoretical solution on amount of capsules required for self-healing technology via embedded capsules in two- and three-dimensional self-healing system. Based on the probability of a crack hitting capsules with a certain diameter calculated by Monte Carlo simulations, Huang [21, 22] attempted to investigate the effects of the dosage and the size of capsules on self-healing efficiency in capsule-based self-healing cementitious material. Evidently, it was too idealistic that once a crack hits/approaches a capsule in hitting mode and the one ruptures then the self-healing in working in these published articles [6, 9, 11, 12]. It is known that a crack hitting with a capsule does not always break the capsule (i.e. in the hitting mode), while the chance in the penetrating mode will be increased. As shown in Fig. 1 the penetrating mode can be regarded as a subset of the hitting mode, the rupturing chance of capsules in penetrating mode is believed to be higher than that in the hitting mode. It is important to note that the rupturing chance of a capsule is distinguished from the probability of a crack hitting or penetrating the capsules. The rupture of a capsule is determined by many factors, such as loading, adhesion between microencapsulated healing agent and the matrix, capsule shell thickness and the fracture energy emerged from cracks. And yet, the probability of a crack hitting or penetrating the capsules is just investigated from morphology, even a viewpoint of geometry structure.

In this contribution, continuing our previous study, an analytical expression of the probability of a crack penetrating with randomly dispersed capsules (i.e. for the penetrating mode) is developed via geometrically probabilistic theory in two-dimensional capsule-based self-healing virtual cementitious material. Meanwhile, the developed hitting probability model that a segment crack occurring at random in matrix meets at least one disc-shaped capsule of the self-healing system will be in retrospect. Much attention is paid to compare the penetrating probability and the hitting probability and the accuracy of results of the theoretical model is also compared with Monte-Carlo numerical analysis.

2. DEVELOPMENT OF PENETRATING PROBABILITY MODEL

In order for the self-healing process to be effective the ratio of crack size to capsule size needs to be of the same length scale. White [1] used microcapsules to heal cracks at microscale in a self-healing based composite system. If the unhydrated cement nuclei are analogized to be microcapsules in autogenous healing cementitious material, the healing process is in microscale [23 – 25]. To achieve large scale healing in case of self-healing cementitious materials, the internal diameter of the tubes carrying healing agent usually ranges between 0.8 mm to 4 mm and even one capsule hit by a (relatively) small crack may release a sufficient amount of the healing agent to seal off the crack [26 – 28]. In order for the self-healing process to be effective it is welcome that the healing agent released from the ruptured capsule is capable to fully heal each crack that triggers the self-healing process in the model material matrix. In other words, it is unrealistic that a macro-crack can be repaired by several microcapsules because of the limited healing agent released from microcapsules. Also, it is seldom that a micro-crack can be repaired by several macrocapsules since the energy emerged from micro-crack is not enough to rupture the macrocapsules.

Typical volume fraction of capsules embedded in self-healing cementitious material matrix is relatively low. At such low volume fractions, the capsule particle presents in a state of chaos or three-dimensional “randomness” from a viewpoint of spatial distribution. Furthermore, at such low volume fraction it could be supposed to have the independence among capsules instead of capsule clusters in a large scale of matrix. Hence, the capsules embedded can be considered to have a uniform distribution in the matrix. In this model, cracks can randomly generate and propagate in a matrix (although in physical terms they originate from flaws upon action of stress) and not all cracks intersect the capsules, a necessary condition for self-healing. But what is the probability that a crack can hit or penetrate the capsules?

From the perspective of modeling and design of self-healing cementitious materials it is therefore necessary to determine the probability of crack intersecting the capsules (hitting probability) or penetrating the capsules (penetrating probability) as illustrated in Fig. 1. The probability model for hitting mode has been developed and more details can be referred in author’s published paper [9, 11]. The hitting probability should not be confused with the rupturing rates of capsules or the self-healing effect which is commonly characterized by the recovery of macro-scale mechanical/transport performance and other indicators or self-healing capacity related to the healing agent. At the same time, since the capsule wall substances itself has certain rigidity, the behavior of a capsule rupturing immediately is rarely in the hitting probability model once a crack approaches a capsule [9, 11].

In the following sections we develop analytical models to predict a crack penetrating the capsules and show how to determine the penetrating probability for an arbitrary crack in a virtual cementitious material matrix with randomly distributed embedded capsules. It is assumed that the cracks randomly appear and then directionally grow in a two-dimensional matrix setup.

Consider a countable number of congruent capsules K in the square sample T of side h and $N_T(h)$ denotes the number of capsules intersecting the sample T in Fig. 1. The capsule-based self-healing cementitious material is considered to be a homogeneous matrix and the number density of capsules incorporated is σ. Namely,

$$\lim_{h\to\infty}\frac{N_T(h)}{S_T(h)}=\sigma \ , \tag{1}$$

is independent of the square sample with edge length h, where $S_T(h)$ is the area of sample T. Once a segment crack C randomly occurs in T, the probability that it penetrates at least one capsule of the self-healing system is a critical factor for the rupturing rates of capsules and the self-healing efficiency. Assume that a disc-shaped capsule K of radius r is contained inside a square sampling region T of side length h. Now a segment crack C of length l randomly happens in the region, then the probability that the segment crack C penetrates the disc-shaped capsule K could be developed as demonstrated in the Appendix, i.e., Eq. A.10 and Eq. A.11.

For $2r < l << h$. For m randomly dispersed disc-shaped capsules K inside the square sampling region T, the probability that exactly s of them are penetrated by the segment crack C is

$$P_s(m,h)=\binom{m}{s}p^s(1-p)^{m-s} \ , \tag{2}$$

where p comes from Eq. A.11 in the Appendix.

Specifically, from Eq. 1 and Eq. A.11

$$\lim_{m,h\to\infty} mp=\sigma(2rl-\pi r^2) \ . \tag{3}$$

Allowing both m, $h\to\infty$, it obtains the asymptotic limit relation from Eq. 2 and Eq. 3, by applying the interrelationship between binomial distribution and Poisson distribution

$$P_s = \lim_{m.h\to\infty} P_s(m,h) = \frac{\left[\sigma\left(2rl-\pi r^2\right)\right]^s}{s!}\exp\left[-\sigma\left(2rl-\pi r^2\right)\right] \tag{4}$$

Hence, for randomly distributed capsules with number density in the self-healing virtual cementitious material matrix the probability that a random segment crack penetrates disc-shaped capsule(s) is

$$P = 1-\exp\left[-\sigma\left(2rl-\pi r^2\right)\right]. \tag{5}$$

If the probability is expressed by the area fraction A_A of capsules embedded and denote $x = l/r$ ($x \geq 2$) by the ratio of crack size to capsule radius, Eq. 5 becomes

$$P = 1-\exp\left[-A_A\left(\frac{2x}{\pi}-1\right)\right], \tag{6}$$

where $A_A = \pi r^2\sigma$ is the area fraction of capsules embedded in the sampling region.

For $l<2r<h$. A similar process is carried out when p from Eq. A.10 is employed in the Eq. 2. Then, the probability that a random segment crack of length l could penetrates the disc-shaped capsule(s) of radius r in the two-dimensional model self-healing materials is

$$P = 1-\exp\left[-\sigma\left(2rl-\frac{1}{2}l\sqrt{4r^2-l^2}-2r^2\arcsin\frac{l}{2r}\right)\right] \tag{7}$$

and

$$P = 1-\exp\left\{-\frac{A_A}{\pi}\left(2x-\frac{1}{2}x\sqrt{4-x^2}-2\arcsin\frac{x}{2}\right)\right\}, \tag{8}$$

where $x = l/r (x < 2)$ is the ratio of crack size to capsule radius.

3. DISCUSSIONS

In essences, the rupturing modes of capsules are ascribed to the different fracture energy produced by cracks within the material while one of the results of fracture energy is the differences among the cracks size. That is the reason why the ratio of crack size to capsule size should be selected as a key parameter in the development of probability of a crack hitting/penetrating capsules. In author's published paper [11], the probability that a segment crack occurring at random in matrix meets at least one disc-shaped capsule of the self-healing system, i.e. for the hitting mode, has been developed. It should be noticed that the crack is not necessary to penetrate the capsules. If we denote by x the ratio of crack size to capsule radius, the probability that a segment crack hits capsules for hitting mode in capsule-based self-healing virtual cementitious material is expressed by [11]:

$$P = 1-\exp\left[-A_A\left(1+\frac{2x}{\pi}\right)\right]. \tag{9}$$

The plots of hitting probability and penetrating probability P with x from Eq. 6 and Eq. 9 are shown in Fig. 2, respectively. In Fig. 2, *hitting* represents the hitting probability that a random crack meets capsules, while *penetrating* denotes the penetrating probability. According to the Eq. 6 and Eq. 9, it is clear that the changing trends of the probability for these two modes are consistent as shown in Fig. 2. Namely, as the x increases the probability P also does. It is agreed that long cracks have more opportunities to hit or penetrate the capsules compared with short cracks.

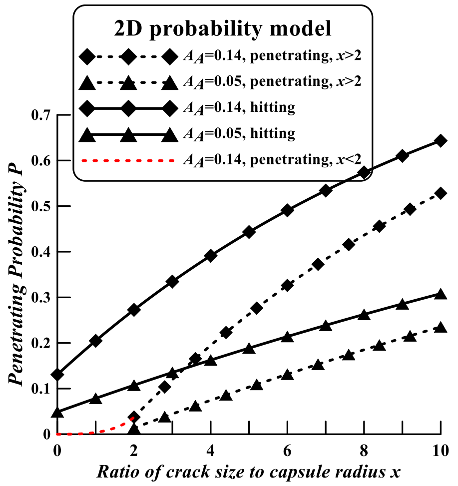


Fig. 2. A comparison between hitting probability and penetrating probability in a two-dimensional capsule-based self-healing virtual cementitious material (*penetrating* presents the model of crack penetrating the capsules, while *hitting* denotes the model of crack hitting the capsules)

When $2 < x < 10$ and $A_A = 0.05$, 0.14, the hitting probability is significantly higher than the penetrating probability as shown in Fig. 2. These consequences are embodied from Eq. 6 and Eq. 9, and reflect that the carriers containing healing agent are easier being hit than being penetrated for a given crack. That is, the probability will be influenced by the different rupture modes of capsules. Ideally, if once a crack penetrates capsules the capsules will rupture and then the self-healing stage will happen and the self-healing efficiency is solely determined by the rupturing rates of capsules, the presented probability can be used to express or even characterize the self-healing efficiency or the releasing efficiency of healing agent encapsulated in capsules. When $x < 2$ and $A_A = 0.14$, the probability of random cracks penetrating the capsules is very low and lower than 0.02 (red dotted line in Fig. 2). This shows that when the length of crack is less than the diameter of capsule embedded, the chance that a crack penetrates the capsules is rare and the penetrating probability can be even ignored in this situation. In practical capsule-based self-healing materials, when the crack is small, the fracture energy emerged from cracking is not often enough to force the rupture of capsules under the conditions that the size of cracks and capsules are in the same order of magnitude. It is also important to note that the chance of rupture of a capsule is different from the probability of a crack hitting/penetrating the capsules.

Generally, it is difficult to repair all the cracks occurred in the matrix and most of the time it is also not necessary to do. Base on this fact a risk-based healing strategy tolerating some defects in the matrix was presented in the design of self-healing materials [14]. Appling the risk-based healing strategy, the amount of capsules required to achieve a certain self-healing level in the matrix can be calculated and analyzed. In other words, if the disc-shaped capsules are uniformly dispersed in the capsule-based self-healing virtual material and the expected penetrating probability P is hypothetically designated, the exact concentration of capsules σ or A_A can be determined from Eq. 5 or Eq. 6 as shown in Fig. 3 when $2r < l$.

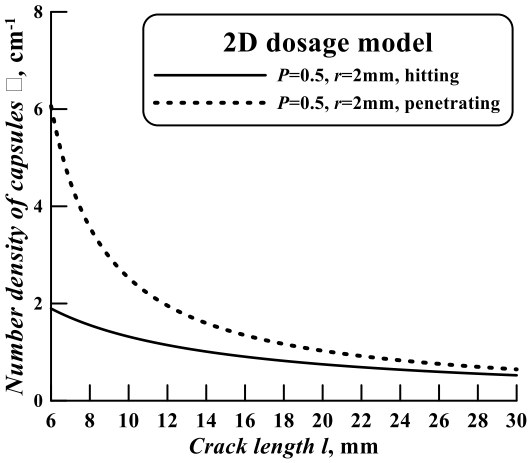


Fig. 3. The amount of capsules required to repair a crack in the hitting probability model and penetrating probability model for a given targeted healing level

The plot of number of capsules incorporated in per cm^3 with the size variant of crack from Eq. 5 is shown in Fig. 3 when the targeted healing level is designated to be 0.5. It shows that as the crack grows the amount of capsules required will decreases because the penetrating probability increases. It should be pointed out here that the healing capacity of a capsule containing healing agent is supposed to sufficiently repair the crack(s) which meet the capsule in the model development. This claim is legal based on that the size of cracks and capsules are in the same order of magnitude or the ratio of crack size to capsule radius follows in a certain range [6, 20, 29, 30].

As an example, Fig. 3 shows that if the targeted healing level P is set to 50 % and the radius r of the capsule is set to 2 mm, the concentration of capsules required for the penetrating model (dotted line) in the per unit area is significantly higher than that for the hitting probability model (solid line). For a given targeted healing level and the crack length satisfying within a specific length (this length is related to the capsules size) many more capsules should be mixed in the matrix when the penetrating mode works comparable to the hitting mode. From Fig. 3 it can be seen that as the crack size increases gradually and reaches a certain value, the dosage of capsules required in the two cracking modes will close. When the capsule size and the dosage of capsules embedded are fixed, both the penetrating probability and the hitting probability are increasing with the increase of crack size as illustrated in Eq. 5 and Eq. 9. At the same time, if the ratio of crack size to capsule size gets so large, the gap between the two probabilities become smaller. Further, imagine that when the segment crack is so long, capsules under the penetrating mode constitute the major components of capsules belonging to hitting mode. So in this sense the penetrating mode can be included in the hitting mode from the given definition of the cracking modes in a two-dimensional capsule-based self-healing virtual material. That is the reason why the amounts of capsules required in the two cracking modes become close to heal a relatively long crack when the targeted healing level and the geometrical characteristics of capsules are given. By the way, a crack whose length overpasses a critical value in practical material matrix is obviously not welcome and expected by designer. This also reveals that the ratio of crack size to capsule radius restricted in a certain range is well-defined and meaningful.

4. VERIFICATION OF THE PENETRATING PROBABILITY MODEL VIA MONTE CARLO ANALYSIS

Simulation and modeling tools as well as various analytical approaches provide a powerful extension of traditional experimental investigations and offer an alternative to at least partly overcome some deficiencies in terms of experimental methods. As illustrated in Fig. 4, an algorithm is developed to numerically model the probability of a segment crack penetrating disc-shaped capsules in a two-dimensional capsule-based self-healing cementitious material matrix.

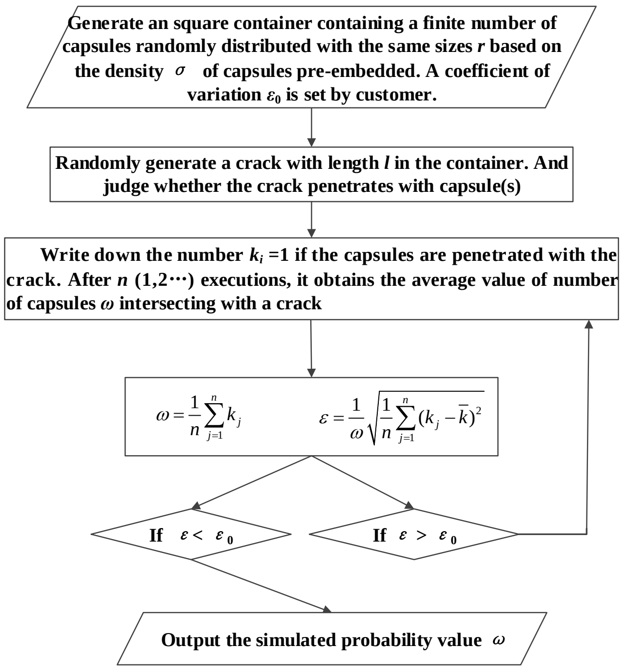


Fig. 4. The scheme of algorithm description of the accuracy of penetrating probability

The details of the numerical algorithm are as follows:

1. Generate a square sampling region with side length of L and provide a variation coefficient ε_0 to control the

accuracy of the simulated value. According to the number density σ or the area fraction A_A a given number N of disc-shaped capsules with radius r are generated and uniformly distributed in the sampling region (r is designated by customer, $r << L$). The location of a capsule K with radius r is determined by its center coordinates (x_i, y_i).

2. Randomly generate a segment crack C in the square sampling region, and the location of the crack C with a given length l (l is designated by customer, $l << L$) is determined by its center coordinates (x, y) as well as the orientation angle θ. For each execution j, i.e., for each segment crack, judge whether the crack C penetrates the capsule (may be more than one capsule). If does, $k_j = 1$. Otherwise, $k_j = 0$. After n executions, the simulated probability of the events of a segment crack penetrating with capsule(s) will be obtained as follows:

$$\omega = \frac{1}{n}\sum_{j=1}^{n} k_j \ , \tag{10}$$

and the variation coefficient

$$\varepsilon = \frac{1}{\omega}\sqrt{\frac{1}{n}\sum_{j=1}^{n}(k_j-\omega)^2} \ , \tag{11}$$

where n=1, 2, ….

3. Continue Step 2 until it reaches $\varepsilon<\varepsilon_0$, then stop Step 2 and write down the current average value ω. At this point, ω is fluctuating with the executions and the variation coefficient ε_0, capsule size r, number density σ of capsules and crack size l designated by user in the simulation procedure. Therefore, the accuracy of the penetrating probability P can be estimated by ω via Monte-Carlo simulation.

As we have demonstrated in reference [9], when capsule size and crack size are on the same length scale, the proposed penetrating probability model is an universal expression in the self-healing model materials and is expected to be valid for various practical self-healing systems such as microcracks and microcapsules based self-healing system in polymer composites [1] and large-scale self-healing cementitious materials [6]. A good agreement could be found between model predictions and computer simulations as demonstrated in Fig. 5.

As shown in Fig. 5 a, for a given crack of length l = 5 mm and disc-shaped capsules of radius r = 0.5 mm in the simulation procedure, simulation results suggest that the change of the probability P of a crack penetrating at least one capsule vs. the area fraction A_A of capsules (A_A =0.05, 0.0657, 0.0814, 0.0971 and 0.1128) is consistent with the trend as demonstrated in Eq. 6. As shown in Fig. 5 b, when crack length l = 2, 3, 5, 7 and 9 mm, capsule size r = 0.5 mm and area fraction A_A = 0.0657, the penetrating probability P with respect to the ratio x of crack length to capsule radius can be verified as the corresponding relation implied in Eq. 6.

Given A_A = 0.097, l = 5 mm, r = 0.5 mm and ε_0 = 0.05, the fluctuating process of the simulation values of penetrating probability P in terms of executions is shown in Fig. 5 c.

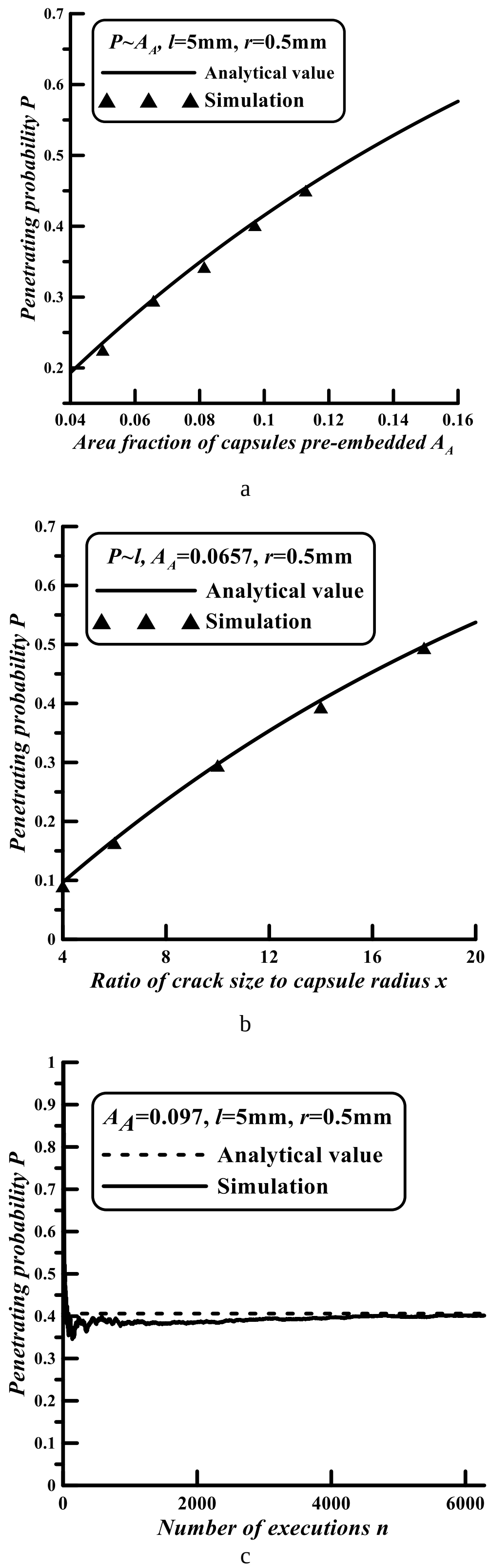


Fig. 5. Computer simulation of the probability of a segment crack penetrating disc-shaped capsules in the two-dimensional capsule-based self-healing virtual cementitious material: a – verification of the penetrating probability when area fraction of capsules pre-embedded is designated; b – verification of the penetrating probability with respect to the ratio of crack size to capsule radius; c – fluctuation of the penetrating probability with the executions for a given variation coefficient

As the executions go on the simulated results reach a stable value which is approximated to the theoretical value from the presented mathematical model. Attenuations should be paid that the executions are dominated by the variation coefficient ε_0.

5. CONCLUSIONS

In mathematics, when the ratio of crack size to capsule size falls in a certain range which is also related with the distribution of capsules embedded, the hitting probability is significantly higher than the penetrating probability as a function of capsules volume. At the same time, if the ratio gets so large, the gap between the two probabilities become smaller and closer. If the crack is long enough, the penetrating mode constitutes the major components of hitting mode in a capsule-based self-healing virtual material. However, a big crack whose length overpasses a critical value in practical material matrix is obviously not welcome and expected by designer. In practical capsule-based self-healing cementitious materials, when the crack is small, the fracture energy emerged from cracking is not often enough to force the rupture of capsules under the conditions that the size of cracks and capsules are in the same order of magnitude. Therefore, it is concluded that the ratio of crack size to capsule radius should be restricted in a certain range in the design of capsule-based self-healing materials.

Furthermore, from a mathematical model standpoint the penetrating mode can be regarded as a subset of the hitting mode, the rupturing chance of capsules in penetrating mode is believed to be higher than that in the hitting mode. So, the penetrating mode is favour of capsule rupture and is more help for the occurrences of self-healing. It should be noted that the rupturing chance of a capsule is distinguished from the probability of a crack hitting or penetrating the capsules. Besides the hitting/penetrating probability of a crack interaction with capsule from the viewpoint of geometry, many factors, such as loading, capsule wall and the fracture energy emerged from cracks should be considered to determine whether a capsule ruptures and improves the self-healing effect.

The next step is to develop the probability of crack penetrating capsules in three-dimensional capsule-based self-healing virtual cementitious materials.

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APPENDIX A

The geometric probability that a random segment penetrates a circle in a two-dimensional convex domain within which lies the circle and the segments can be developed from the theory of integral geometry as follows:

A formula for the kinematic measure of a segment contained in a convex domain was established by Ren [31].

Theorem [31] Let Ω be a bounded convex domain of perimeter L and area F, and let C be a segment of length l in the plane. Denote by $m(l)$ the kinematic measure of C contained in Ω, then

$$m(l) = \pi F - lL + \int_{\substack{G\cap\Omega\neq\varnothing \\ (\upsilon\leq l)}} (l-\upsilon)dG \ , \tag{A.1}$$

where υ is the chord length of Ω intercepted by the line G.

Here, $m(G\cap\Omega = 2) = \int_{\substack{G\cap\Omega\neq\varnothing \\ (\upsilon\leq l)}} (l-\upsilon)dG$ represents the kinematic measure of C exactly intersecting Ω with two points. Hence, the kinematic measure that a segment C penetrates the convex domain Ω is

$$\int_{\substack{G\cap\Omega\neq\varnothing \\ (\upsilon\leq l)}} (l-\upsilon)dG = m(C\cap\Omega = 2) = m(l) - \pi F + lL \tag{A.2}$$

Corollary [31] Let d be the diameter of Ω. If $l \geq d$, then $m(l) = 0$ (line segment N cannot be included in Ω). That is

$$\pi F - lL + \int_{G\cap\Omega\neq\varnothing} (l-\upsilon)dG = 0 \ . \tag{A.3}$$

Hence, the kinematic measure of the segment of length l passing through Ω is

$$\int_{G\cap\Omega\neq\varnothing} (l-\upsilon)dG = lL - \pi F \ . \tag{A.4}$$

Remark [32] The kinematic measure $m(l)$ that a segment of length l is contained in a circle of radius r in the two-dimensional plane has been developed as follows

$$m(l) = \pi^2 r^2 - \frac{1}{2}\pi l\sqrt{4r^2 - l^2} - 2\pi r^2 \arcsin\frac{l}{2r} \tag{A.5}$$

We embody the theorem and the corollary to the hitting probability model of the crack penetrating with capsules in the self-healing materials. Here the convex domain Ω is considered to be a disc-shaped capsule K of radius r and the segment N to be a crack C of length l in the two-dimensional model.

Then, for $l < 2r < h$, combining Eq. A.2 and Eq. A.5, we obtain

$$\begin{aligned} & m(C\cap K = 2) \\ & = 2\pi r l - \frac{1}{2}\pi l\sqrt{4r^2 - l^2} - 2\pi r^2 \arcsin\frac{l}{2r} \end{aligned} \ . \tag{A.6}$$

For $\boldsymbol{l} > 2r$, from Eq. (A.4), it gives

$$\begin{aligned} m(C\cap K = 2) &= \int_{C\cap K\neq\varnothing} (l-\upsilon)dG \\ &= lL - \pi F = 2\pi r l - \pi^2 r^2 \end{aligned} \ . \tag{A.7}$$

Generally, the problem of finding the kinematic measure of the segments of a constant length that are contained in a convex domain has no simple solution and depends largely on the shape of the domain [33]. Fortunately, if the convex domain is a regular region, i.e., a square or a circle, the kinematic measure may be obtained. For example, the kinematic measure that a segment of length l is contained in a square of side length h is based on the classical Laplace extension of the Buffon problem [34]

$$m(C;C\subset T)=\pi h^2-4hl+l^2 . \tag{A.8}$$

For a detailed discussion of Eq. A.8, readers are referred to author's published work [11].

Assume that a disc-shaped capsule of radius r is contained inside a square sampling region T of side length h. Now a segment crack C of length l is randomly placed in the region, then the probability that the segment C penetrates the disc-shaped capsule can be expressed as

$$p=\frac{m(C\cap K=2)}{m(C\subset T)} , \tag{A.9}$$

where $m(C\cap K = 2)$ represents the kinematic measure that a segment crack penetrates a capsule.

Therefore, for $l < 2r < h$, from Eq. A.6, Eq. A.8 and Eq. A.9, it obtains

$$p=\frac{2\pi rl-\frac{1}{2}\pi l\sqrt{4r^2-l^2}-2\pi r^2\arcsin\frac{l}{2r}}{\pi h^2-4hl+l^2} . \tag{A.10}$$

For $l > 2r$, from Eq. A.7, Eq. A.8 and Eq. A.9, it gives

$$p=\frac{2\pi rl-\pi^2r^2}{\pi h^2-4hl+l^2} . \tag{A.11}$$

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