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A probabilistic micromechanical framework for self-healing polymers containing microcapsules

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Abstract. A probabilistic micromechanical framework is proposed to quantify numerically the self-healing capabilities of polymers containing microcapsules. A two-step self-healing process is designed in this study: A probabilistic micromechanical framework based on the ensemble volume-averaging method is derived for the polymers, and a hitting probability model combined with a crack nucleation model is then utilized for encountering microcapsules and microcracks. Using this framework, a series of parametric investigations are performed to examine the influence of various model parameters (e.g., the volume fraction of microcapsules, microcapsule radius, radius ratio of microcracks to microcapsules, microcrack aspect ratio, and scale parameter) on the self-healing capabilities of the polymers. The proposed framework is also implemented into a finite element code to solve the self-healing behavior of tapered double cantilever beam specimens.

Keywords: microcapsules; micromechanics; polymers; probabilistic approach; self-healing

1. Introduction

Polymers are among the most widely used materials in the fields of composite structures, such as aerospace, automotive, and building construction (Jang *et al*. 2020, 2021, Khalid *et al*. 2021, Kil *et al*. 2021, 2022, Kim *et al*. 2020, Moghadam *et al*. 2015). However, these structures deteriorate because of the external environment (e.g., excessive and repeated loadings or freeze-thaw cycles) (Awaja *et al*. 2016; Grellmann and Langer 2010, Huseien *et al*. 2022, Kil *et al*. 2023, Muñoz-Abella *et al*. 2012). In particular, the microcracks formed in polymers during deterioration can severely degrade their mechanical performance (Chen *et al*. 2020, Gamstedt and Talreja 1999, White *et al*. 2001, Talreja 1989). To recover the load-carrying capacity and serviceability of damaged polymers, they should be manufactured in advance using appropriate self-strengthening measures.

Since the 1980s, the concept of self-healing polymers has been proposed, and studies on self-healing polymers containing microcapsules have been actively performed (Faravelli and Marzi 2010, Fifo *et al*. 2015, Jud and Kausch, 1979, Meure *et al*. 2009, Yang *et al*. 2020, Zhang *et al*. 2021). White *et al*. (2001) first proposed the use of microcapsules to recover the fracture of polymers. They reported that microcapsules embedded in the polymers are ruptured when crack nucleation occurs, and subsequently, the microcracks are healed via polymerization in contact with the catalyst in the polymers (Blaiszik *et al*. 2010, Brown *et al*. 2002, 2003, 2004, 2005, White *et al*. 2001, Verberg *et al*. 2007). In addition, Pang and Bond (2005) used fiber-type microcapsules filled with a liquid healing agent to heal microcracks in the polymers by embedding hollow glass fibers and reported that microcracks caused by extended volume damage effectively are alleviated.

Numerous theoretical studies have been performed to quantify numerically the self-healing capabilities of self-healing materials containing microcapsules (Blaiszik *et al*. 2010, Brown *et al*. 2002, Lee *et al*. 2004, White *et al*. 2001, Yang *et al*. 2019). Barbero *et al*. (2005) proposed a continuum damage-healing model derived from continuum damage mechanics, which considers the healing evolution variable as a counterpart to the damage evolution variable. Davis and Jefferson (2017) predicted a self-healing performance in materials using a two-phase micromechanical model based on the principle of solidification. Although theoretical modeling has been conducted to predict the self-healing performance of polymers containing microcapsules (Brown 2011, Brown *et al*. 2002, Perelmuter 2020, White *et al*. 2001), accurately determining the shape and amount of effective healed inclusions that form when microcapsules and microcracks encounter remains a major challenge.

Probabilistic models have been proposed to consider the intersection of microcapsules and microcracks (Lin *et al*. 2018, Lv and Chen 2013, 2014, Wang *et al*. 2021). Lin *et al*. (2018) developed a two-dimensional probability model to characterize the intersection of microcapsules and

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microcracks by using geometric probability. They considered the radius of the microcapsules and the size of the microcrack as the main parameters in their model and reported that the microcapsule dosage is significantly affected by the spatial distribution of the microcracks and the ratio of the microcapsule length to the microcrack size (Lin *et al*. 2018). Lv and Chen (2014) proposed a probability model to estimate collision probability in three dimensions, which is utilized to determine the microcapsule dosage by calculating the probability of microcapsule and microcrack intersection. However, few studies have presented a probabilistic micromechanical approach for predicting the self-healing capabilities of polymers containing microcapsules, considering the shape and size of the intersection between microcapsules and microcracks.

Against this backdrop, a probabilistic micromechanical framework is proposed in this study to quantify numerically the self-healing capabilities of polymers containing microcapsules. A two-step self-healing process is designed in this study. First, a probabilistic micromechanical framework based on the ensemble volume-averaging method is derived for self-healing polymers containing microcapsules. Next, the hitting probability model combined with a crack nucleation model is utilized for encountering microcapsules and microcracks. A series of parametric investigations based on the proposed framework are conducted to examine the influence of model parameters on the self-healing performance of the polymers. Subsequently, the proposed framework is implemented into the finite element code ABAQUS to solve self-healing behavior of tapered double cantilever beam (TDCB) specimens under tensile loading.

2. Proposed micromechanical framework

Fig. 1 shows a schematic of the two-step self-healing process for self-healing polymers containing microcapsules. In the first step, it is assumed that microcracks are nucleated in the elastic matrix under the uniaxial tension, and some microcracks encounter microcapsules in accordance with the hitting probability model explained in Section 2.2. In the second step, the microcapsules that encounter microcracks are likely to have been ruptured, releasing the healing agent to fill the space between the microcracks and microcapsules via polymerization (Brown *et al*. 2004, White *et al*. 2001). This self-healing process results in the transformation of the microcracks that have encountered microcapsules into effective healed spheroidal inclusions, which can be divided into healed and unhealed spheroidal inclusions based on the healing probability. Following this two-step self-healing process, the five-phase micromechanical framework can be constructed: (1) an elastic matrix (phase 0), (2) unreacted microcapsules (phase 1), (3) microcracks that have not encountered microcapsules (phase 2), (4) healed spheroidal inclusions resulting from

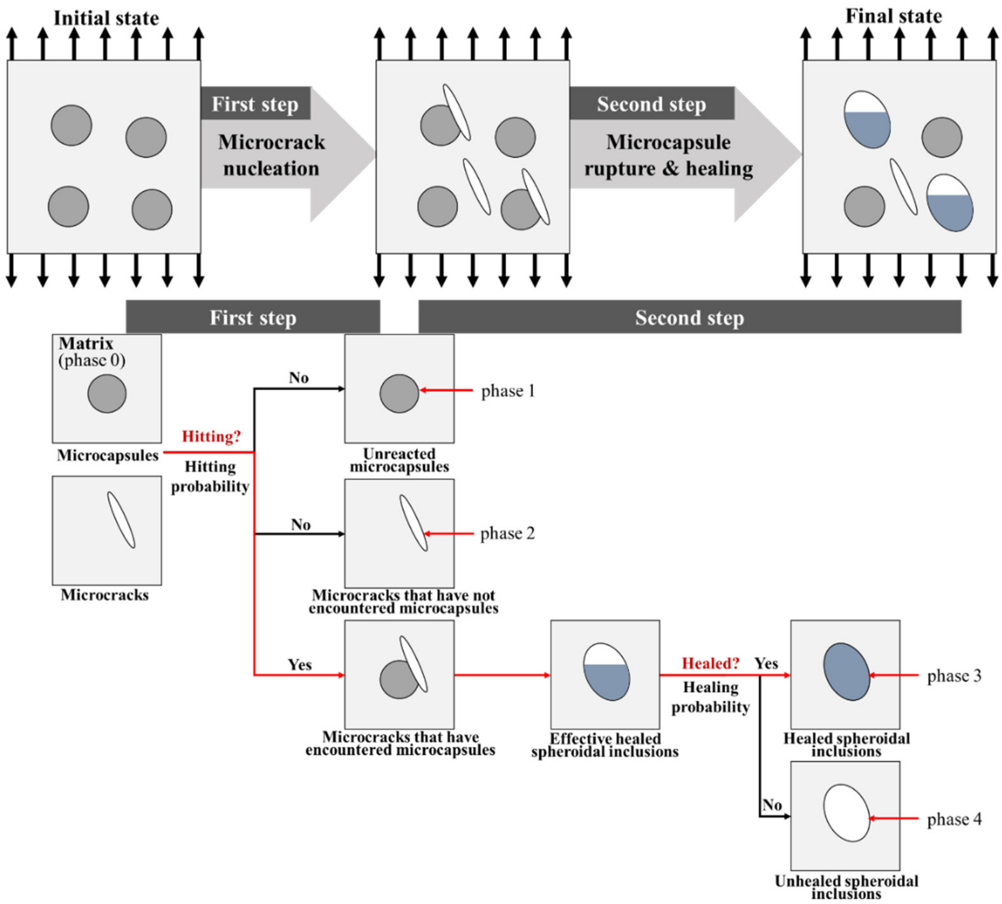


Fig. 1 A schematic of the two-step self-healing process for self-healing polymers containing microcapsules

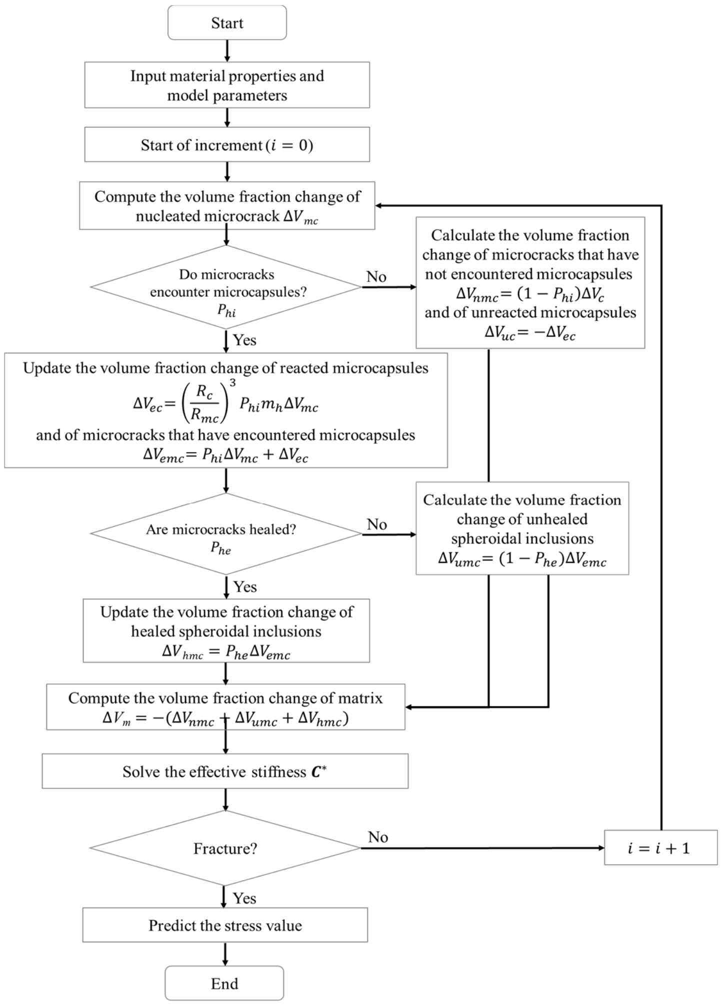


Fig. 2 A flowchart of the probabilistic micromechanical framework for self-healing polymers containing microcapsules

the release of healing agents (phase 3), and (5) unhealed spheroidal inclusions (phase 4). It is assumed in the present model that the catalyst is sufficiently pre-embedded in the polymer matrix referring to the literature (Lv and Chen 2013, Gao *et al*. 2021). Accordingly, it should be noted that the effect of the amount of catalyst for polymerization on the self-healing performance is not considered in this study.

2.1 Effective stiffness of self-healing polymers

Fig. 2 shows a flowchart of the probabilistic micromechanical framework for self-healing polymers containing microcapsules. The ensemble volume averaging method proposed by Ju and Chen (1994) is adopted to predict the self-healing capabilities of polymers with randomly distributed ellipsoidal inhomogeneities. Following Ju and Chen (1994), the effective stiffness $\boldsymbol{C}^*$ for five-phase self-healing polymers containing randomly distributed microcapsules can be expressed as

$$\boldsymbol{C}^* = \boldsymbol{C}_0 \cdot \left[\boldsymbol{I} + \sum_{r=1}^{4}\{\phi_r(\boldsymbol{A}_r + \boldsymbol{S}_r)^{-1} \cdot [\boldsymbol{I} - \phi_r\boldsymbol{S}_r \cdot (\boldsymbol{A}_r + \boldsymbol{S}_r)^{-1}]^{-1}\}\right] \tag{1}$$

where $\boldsymbol{A}_r$ is defined as $\boldsymbol{A}_r = (\boldsymbol{C}_r - \boldsymbol{C}_0)^{-1} \cdot \boldsymbol{C}_0$; $\boldsymbol{I}$ and $\boldsymbol{C}_r$ are the fourth-rank identity tensor and fourth-rank elasticity tensor of the r-phase, respectively; and ϕ_r and $\boldsymbol{S}_r$ are the volume fraction and fourth-rank Eshelby's tensor of the r-phase inclusion, respectively (Ju and Chen 1994). In this study, unreacted microcapsules (phase 1) and microcracks that have not encountered microcapsules (phase 2) are assumed to be spherical and oblate spheroidal-shaped inclusions, respectively. In addition, it is assumed that these inclusions are randomly dispersed and oriented. The Eshelby's tensor $\boldsymbol{S}_r$ for randomly dispersed spherical inclusions can be found in Ju and Chen (1994), while that for randomly dispersed and oriented spheroidal inclusions can be found in Ju and Sun (2001).

Furthermore, it is essential to define the size and shape of the effective healed inclusions. Fig. 3 shows schematics of the assumed effective healed spheroidal inclusions. The size and shape of the assumed effective healed spheroidal inclusions can be determined by the radii of microcracks and microcapsules. When the radius of the microcrack is larger than that of the microcapsule, the effective healed spheroidal inclusions are assumed to be oblate spheroids (as shown in Fig. 3(a)). Conversely, when the radius of the microcapsule is larger than that of the microcrack, the effective healed spheroidal inclusions are assumed to be

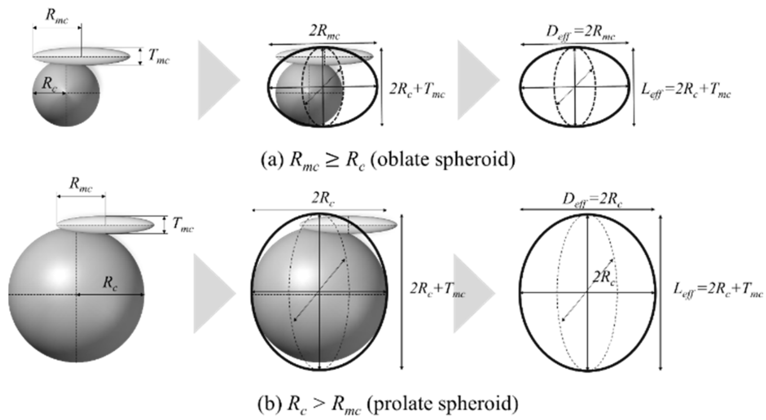


Fig. 3 Schematics of the assumed effective healed spheroidal inclusions (a) in case the radius of microcracks is larger than that of microcapsules; and (b) in case the radius of microcapsules is larger than that of microcracks

prolate spheroids (as shown in Fig. 3(b)). The major axis D_{eff}, minor axis L_{eff}, and aspect ratio α_{eff} of the assumed effective healed spheroidal inclusions can be defined as

$$D_{eff} = \begin{cases} 2R_{mc}, & R_{mc} \geq R_c \\ 2R_c, & R_c > R_{mc} \end{cases} \tag{2}$$

$$L_{eff} = 2R_c + T_{mc} \tag{3}$$

$$\alpha_{eff} = \frac{L_{eff}}{D_{eff}} \tag{4}$$

where R_{mc}, R_c, and T_{mc} are the radius of microcracks, the radius of microcapsules, and the thickness of microcracks, respectively.

2.2 The hitting probability model combined with a crack nucleation model

The crack nucleation model suggested by Karihaloo and Fu (1989) is adopted in this study to model the microcrack nucleation. Note that this model assumes that microcracks having identical shapes and sizes are uniformly dispersed in the matrix. Following Karihaloo and Fu (1989), the volume fraction of the nucleated microcrack, V_{mc}, can be expressed as

$$V_{mc} = \begin{cases} V_{mc}^0, & \varepsilon^a \leq \varepsilon^{th} \\ V_{mc}^0 + c_1\left(1 - \dfrac{\varepsilon^{th}}{\varepsilon^a}\right)^{c_2}, & \varepsilon^a > \varepsilon^{th} \end{cases} \tag{5}$$

where V_{mc}^0, ε^a, and ε^{th} are the initial volume fraction of microcracks, current-accumulated effective macroscopic strain, and threshold effective strain below which no nucleation occurs, respectively; and the model parameters c_1 and c_2 are related to the shape and distribution of microcracks, respectively (Karihaloo and Fu 1989, Lee and Pyo 2009). Therefore, when the effective strain increases by $\Delta\varepsilon^a$, the volume fraction change of microcracks, ΔV_{mc}, can be derived as follows (Karihaloo and Fu 1989).

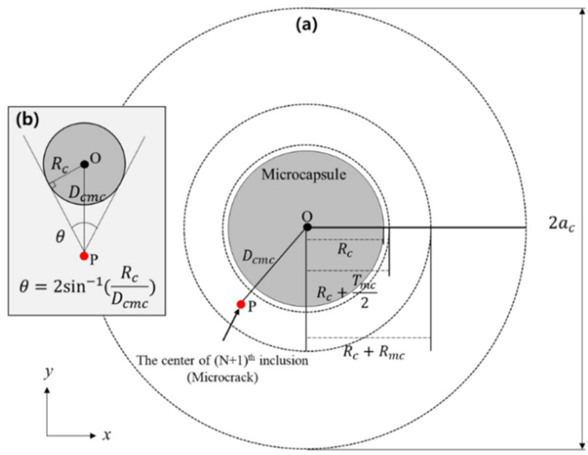


Fig. 4 Schematics of (a) an arbitrary sphere containing a single microcapsule; and (b) the radian angle θ of the tangent line between the center of microcrack and the microcapsule

$$\Delta V_{mc} = \begin{cases} c_1\left(1 - \dfrac{\varepsilon^{th}}{\varepsilon^a + \Delta\varepsilon^a}\right)^{c_2} - c_1\left(1 - \dfrac{\varepsilon^{th}}{\varepsilon^a}\right)^{c_2}, \\ \qquad \varepsilon^a + \Delta\varepsilon^a > \varepsilon^a > \varepsilon^{th} \\ c_1\left(1 - \dfrac{\varepsilon^{th}}{\varepsilon^a + \Delta\varepsilon^a}\right)^{c_2}, \\ \qquad \varepsilon^a + \Delta\varepsilon^a > \varepsilon^{th} \geq \varepsilon^a \\ 0, \\ \qquad \varepsilon^{th} \geq \varepsilon^a + \Delta\varepsilon^a \geq \varepsilon^a \end{cases} \tag{6}$$

The hitting probability model is proposed here to consider the probability of encountering microcapsules and microcracks based on the distance between them. Fig. 4(a) illustrates an arbitrary sphere containing a single microcapsule. When the distance between the microcapsule and microcrack, denoted by D_{cmc}, is less than $R_c + T_{mc}/2$, the hitting probability is considered to be 1. For D_{cmc} between $R_c + T_{mc}/2$ and $R_c + R_{mc}$, the hitting probability is determined by dividing the radian angle of the tangent line, denoted by θ, between the center of microcrack and the microcapsule by π (rad). Note that the value of θ can be estimated geometrically as shown in Fig. 4(b). When D_{cmc} is greater than the sum of the radius of the microcapsule and the radius of the microcrack $R_c + R_{mc}$, the hitting probability is assumed to be 0. Therefore, the hitting probability, denoted by P_{hd}, depending on D_{cmc} can be expressed as

$$P_{hd} = \begin{cases} 1, & R_c + \dfrac{T_{mc}}{2} \geq D_{cmc} \geq 0 \\ \dfrac{\theta}{\pi}, & R_c + R_{mc} \geq D_{cmc} > R_c + \dfrac{T_{mc}}{2} \\ 0, & a_c \geq D_{cmc} > R_c + R_{mc} \end{cases} \tag{7}$$

with

$$\theta = 2\sin^{-1}\left(\frac{R_c}{D_{cmc}}\right), \tag{8}$$

The mean inter-particle distance method (Bagheri and Pearson 2000, Chandrasekhar 1943) is adopted to probabilistically estimate the distance between microcapsules, and the probability, denoted by P_d, of finding a microcapsule at the distance of r can thus be estimated as (cf. Bagheri and Pearson 2000, Chandrasekhar 1943))

$$P_d(r) = \frac{3}{a_c}\left(\frac{r}{a_c}\right)^2 e^{-(r/a_c)^3} \tag{9}$$

with

$$a_c = \frac{R_c}{\sqrt[3]{V_c}} \tag{10}$$

where V_c denotes the volume fraction of microcapsules, and a_c represents the radius of an arbitrary sphere containing a single microcapsule with V_c. Hence, if a microcrack encounter a microcapsule, the value of V_c decreases leading to an increase in a_c.

The hitting probability, denoted by P_{hi}, of encountering spherical microcapsules and spheroidal microcracks can be derived by combining Eqs. (8)-(10), where the microcrack can be considered as the (N+1)th inclusion when it is nucleated in the matrix containing microcapsules of N. Thus, P_{hi} can be expressed as

$$P_{hi} = \frac{2}{\pi}\sin^{-1}\left(\frac{R_c}{r}\right)\int_{R_c+\frac{T_{mc}}{2}}^{R_{mc}} \frac{3}{a_c}\left(\frac{r}{a_c}\right)^2 e^{-\left(\frac{r}{a_c}\right)^3} dr + \int_0^{R_c+\frac{T_{mc}}{2}} \frac{3}{a_c}\left(\frac{r}{a_c}\right)^2 e^{-\left(\frac{r}{a_c}\right)^3} dr \tag{11}$$

Using the aforementioned hitting probability model combined with the crack nucleation model, the volume fraction change of each phase can be derived as the strain increases by $\Delta\varepsilon^a$. The volume fraction change of the reacted microcapsules ΔV_{ec}, the volume fraction change of the unreacted microcapsules ΔV_{uc}, the volume fraction change of the microcracks that have encountered microcapsules ΔV_{emc}, and the volume fraction change of the microcracks that have not encountered microcapsules ΔV_{nmc} can be expressed as

$$\Delta V_{ec} = \left(\frac{R_c}{R_{mc}}\right)^3 P_{hi} m_h \Delta V_{mc}, \quad \sum \Delta V_{ec} \leq V_c \tag{12}$$

$$\Delta V_{uc} = -\Delta V_{ec} \tag{13}$$

$$\Delta V_{emc} = P_{hi}\Delta V_{mc} + \Delta V_{ec} \tag{14}$$

$$\Delta V_{nmc} = (1 - P_{hi})\Delta V_{mc} \tag{15}$$

where m_h represents a scale parameter which is related to the speed of the self-healing reaction.

The volume fraction change of the healed spheroidal inclusions ΔV_{hmc} and the volume fraction change of the unhealed spheroidal inclusions ΔV_{umc} can be expressed as

$$\Delta V_{hmc} = P_{he}\Delta V_{emc} \tag{16}$$

$$\Delta V_{umc} = (1 - P_{he})\Delta V_{emc} \tag{17}$$

where the probability term P_{he} is newly introduced to account for the degree of healing of the assumed effective healed spheroidal inclusions. Using Eqs. (15)-(17), the effective volume fraction change of the matrix ΔV_m can be derived as

$$\Delta V_m = -(\Delta V_{nmc} + \Delta V_{hmc} + \Delta V_{umc}) \tag{18}$$

The symbols used in the present probabilistic micromechanical framework are listed in Table 1.

3. Parametric investigations

3.1 Effect of volume fractions and radii of microcapsules

A series of parametric investigations using the proposed framework are conducted to examine the influence of model parameters (e.g., the volume fraction of microcapsules, microcapsule radius, radius ratio of microcracks to microcapsules, microcrack aspect ratio, and scale parameter) on the self-healing capabilities of the polymers. Specifically, we examined the mechanical properties of the self-healing polymers consisting of an EPON828 epoxy matrix, microcapsules, and healed materials (polydicyclopentadiene) which are formed after encountering the microcapsules and microcracks, with the input values for the present parametric investigations listed in Table 2 (Brown *et al*. 2004, Keller and Sottos 2006, Vallons *et al*. 2015). The following parameters values for the self-healing polymers are assumed for the parametric investigation: $V_c = 5.0$ vol.%, $R_c = 120$ μm, $T_{mc} = 10$ μm, $m_h = 0.1$, and $P_{he} = 0.5$. The threshold effective strain ε^{th}, radius ratio of microcracks to microcapsules R_{mcc}, and microcrack nucleation model parameters c_1 and c_2 are also assumed to be 0.002, 0.12, 0.3, and 1.5, respectively (Lee and Pyo 2009). Previous studies have shown that a volume fraction of microcapsules between 5.0 and 22.0 vol.% is effective for determining the dosage of microcapsules in self-healing materials (Lin *et al*. 2018, Lv and Chen 2013). Therefore, the self-healing polymers containing 1.0 ~ 10.0 vol.% of microcapsules are used for the present parametric investigations.

Table 1 Symbols used in the present probabilistic micromechanical framework

Symbol	Description
D_{eff}	Major axis of assumed effective healed spheroidal inclusions [μm]
L_{eff}	Minor axis of assumed effective healed spheroidal inclusions [μm]
α_{eff}	Aspect ratio of assumed effective healed spheroidal inclusions
R_{mc}	Radius of microcracks [μm]
R_c	Radius of microcapsules [μm]
R_{mcc}	Radius ratio of microcracks to microcapsules
T_{mc}	Thickness of microcracks [μm]
α_{mc}	Aspect ratio of microcracks
a_c	Radius of an arbitrary sphere containing a single microcapsule
P_{hd}	Hitting probability depending on D_{cmc}
$P_d(r)$	Probability of finding a microcapsule at the distance of r
D_{cmc}	Distance between microcapsule and microcrack
P_{hi}	Hitting probability of encountering spherical microcapsules and spheroidal microcracks
P_{he}	Healing probability
c_1, c_2	Microcrack nucleation model parameters
ε^{th}	Threshold effective strain below which no nucleation occurs
ε^{a}	Current-accumulated effective macroscopic strain
V_m	Volume fraction of a matrix
V_c	Volume fraction of microcapsules
V_{ec}	Volume fraction of reacted microcapsules
V_{uc}	Volume fraction of unreacted microcapsules
V_{mc}	Volume fraction of microcracks
V_{emc}	Volume fraction of microcracks that have encountered microcapsules
V_{nmc}	Volume fraction of microcracks that have not encountered microcapsules
V_{hmc}	Volume fraction of healed spheroidal inclusions
V_{umc}	Volume fraction of unhealed spheroidal inclusions
m_h	Scale parameter which is related to the speed of self-healing reaction

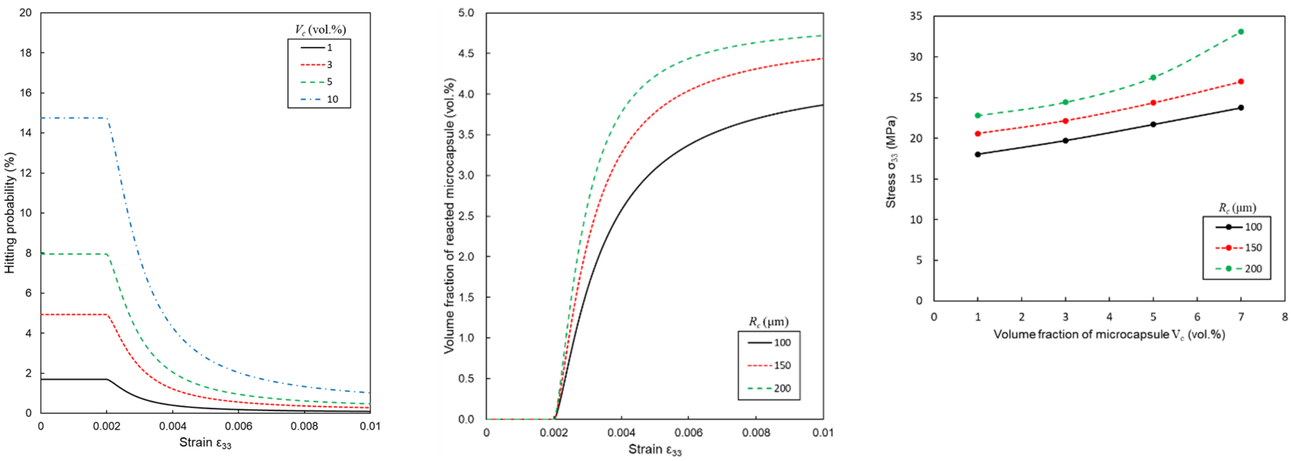


Fig. 5 (a) Hitting probability P_{hi} versus strain for various volume fractions of microcapsules V_c; (b) volume fraction of reacted microcapsules versus strain for various microcapsule radii R_c; and (c) effective stress versus V_c for various R_c at the strain of 0.01

Fig. 5(a) shows the hitting probability P_{hi} versus strain for various volume fractions of microcapsules V_c. The results indicate that P_{hi} in the strain range 0 ~ 0.002 increases to approximately 15% when V_c increases to 10 vol.% after which it decreases significantly as the strain approaches 0.01. Fig. 5(b) presents the volume fraction of the reacted microcapsules versus the strain for various microcapsule radii R_c. The results indicate that the volume fraction of the reacted microcapsules increases significantly in the strain range 0.002 ~ 0.01, and this trend becomes more prominent as R_c increases. For $R_c = 200$ μm, almost all the 5.0 vol.% the microcapsules are reacted at the strain of 0.01. These results suggest that high values of R_c increase the number of reacted microcapsules after the threshold strain of 0.002. The effective stress versus volume fraction of microcapsules V_c for various R_c at the strain of 0.01 is shown in Fig. 5(c), showing that the stress in the polymers increases as V_c and R_c increase. These trends are nearly consistent with the experimental results of Rule *et al.* (2007), who reported that the self-healing performance in the polymers is primarily affected by the volume fraction and the diameter of microcapsules.

3.2 Effect of radius ratios of microcracks to microcapsules and aspect ratios of microcracks

The volume fraction of the reacted microcapsules versus strain for various radius ratios of microcracks to microcapsules R_{mcc} is shown in Fig. 6(a). Note that the microcapsule radius is assumed to be 50 μm; hence, an increase in R_{mcc} indicates an increase in the microcrack radius resulting in a decrease in the number of microcracks. The volume fraction of the reacted microcapsules of the polymers subjected to a strain exceeding 0.002 increases rapidly as R_{mcc} decreases since the number of microcracks decreases. The volume fraction of the reacted microcapsules versus the strain for various aspect ratios of the microcrack α_{mc} is shown in Fig. 6(b). As α_{mc} increases, the slope decreases at a strain exceeding 0.002, and the volume fraction of the reacted microcapsules decreases at the strain of 0.01. Increase in α_{mc} value indicates that the

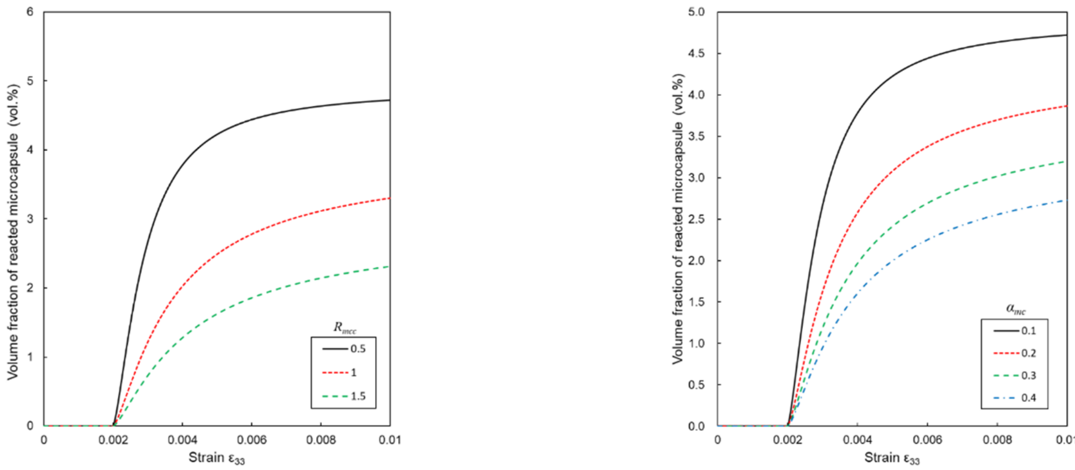


Fig. 6 Volume fraction of reacted microcapsules versus strain for (a) various radius ratios of microcracks to microcapsules R_{mcc}; and (b) various aspect ratios of microcracks α_{mc}

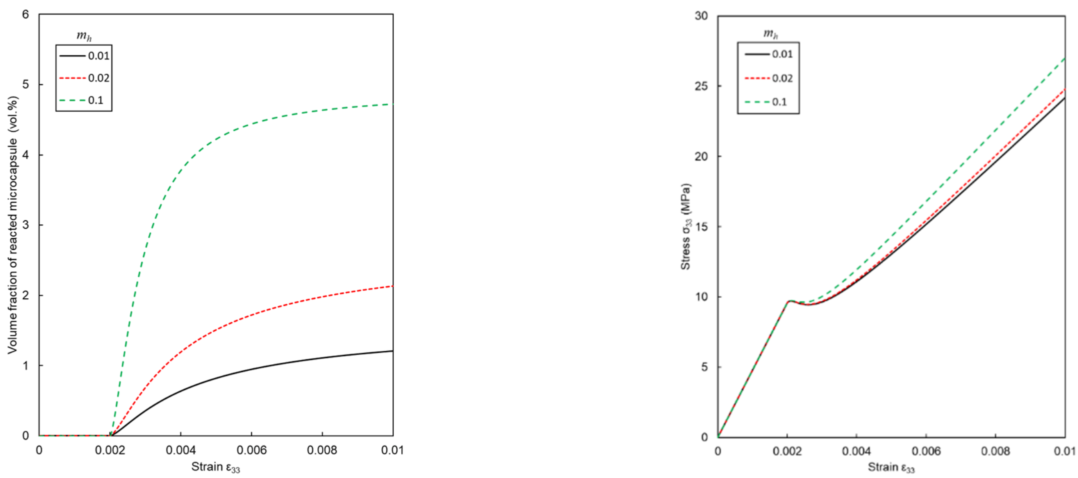


Fig. 7 (a) Volume fraction of reacted microcapsules versus strain for various scale parameters m_h; (b) predicted stress–strain response of polymers containing microcapsules under uniaxial tension for various m_h corresponding to Fig. 7(a) microcracks are transformed into a spheroidal shape, leading to an increase in the number of microcracks since the volume fraction of the spherical microcracks is lower than that of the spheroidal microcracks. The volume fraction of reacted microcapsules decreases as α_{mc} increases, and it can thus be said that polymers having microcracks with higher R_{mcc} and α_{mc} values are likely to consume microcapsules faster than polymers having microcracks with lower R_{mcc} and α_{mc} values.

3.3 Effect of the scale parameter

An additional parametric investigation is carried out to analyze the self-healing performance of polymers containing microcapsules, in terms of the scale parameter which is related to the speed of the self-healing reaction m_h. Fig. 7(a) shows the volume fraction of the reacted microcapsules versus strain for various m_h. R_{mcc} and α_{mc} are assumed to be 0.5 and 0.1, respectively, as used in Section 3.2. The results indicate that the volume fraction of reacted microcapsules increases significantly at a strain exceeding 0.002 with an increase in m_h. Fig. 7(b) shows the predicted strain–stress response of the self-healing polymers containing microcapsules under uniaxial tension for various m_h corresponding to Fig. 7(a). The stress value noticeably increases at the strain of 0.01 as the value of m_h increases.

4. Comparisons between the present predictions and experiments

The proposed probabilistic micromechanical framework is implemented into the finite element code ABAQUS via UMAT (Bian *et al.* 2018, Gao *et al.* 2021) to solve the self-healing behavior of a TDCB specimen under tensile loading (Brown *et al.* 2002, 2004, Garoz Gómez *et al.* 2015, Tsang 2020). The TDCB specimen commonly used for observing fractures and evaluating self-healing capabilities in polymers containing microcapsules (Brown *et al.* 2002) is being considered here. The values of the parameters utilized in the present prediction are identical to those used in the parametric investigations which are listed in Table 2 (Brown *et al.* 2004, Keller and Sottos 2006, Vallons *et al.* 2015).

The TDCB specimen made of EPON828 polymers containing microcapsules with a pre-crack length of 50 mm, subjected to a pin loading at the rate of 5.0 μm/s (Brown *et al.* 2002, Garoz Gómez *et al.* 2015), is modeled and its self-healing behavior is examined in this section. The meshes employed in the models of the TDCB specimen with parts and constraints (cf. Brown *et al.* 2004, Garoz Gómez *et al.* 2015, Tsang 2020)) are shown in Fig. 8. The details including dimensions of the TDCB specimen can be found in Brown *et al.* (2004). The specimen consists of upper, lower, and central parts with the pre-crack located in the central part. The central part is connected to the upper and lower parts via tie constraints, and perfect bonding is assumed between them, provided that the interfacial area is compatible with the parts until the peak strength of the specimen is reached (Garoz Gómez *et al.* 2015). The central part is modeled with 3D eight-node linear brick solid elements (C3D8), while the upper and lower parts use 3D twenty-node linear brick solid elements (Garoz Gómez *et al.* 2015). The central part with the crack comprises 8,470 elements, while each of the upper and lower components contains 16,038 elements.

The present predicted uniaxial stress-strain response of the self-healing polymers containing microcapsules is shown in Fig. 9(a). In addition, the predicted volume fractions of unreacted microcapsules V_{uc}, microcracks that have not encountered microcapsules V_{nmc}, healed spheroidal inclusions V_{hmc}, and unhealed spheroidal inclusions V_{umc} corresponding to Fig. 9(a) are shown in Fig. 9(b). The input values listed in Table 2 (Brown *et al.* 2004, Keller and Sottos 2006, Vallons *et al.* 2015) and the following parameters values obtained through the parametric investigations are adopted in the predictions: $V_c = 5.0$ vol.%, $R_c = 120$ μm, $T_{mc} = 10$ μm, $m_h = 0.1$, $c_1 = 0.3$, $c_2 = 1.5$, and $P_{he} = 0.9$. Comparisons of the load-displacement behaviors of the TDCB specimen between the present prediction and experimental results by Garoz Gómez *et al.* (2015) are shown in Fig. 10. The

Table 2 Input values for the present parametric investigations (Brown *et al.* 2004, Keller and Sottos 2006, Vallons *et al.* 2015)

Material properties		Values
Epoxy matrix (EPON828) [a]	Young's modulus [MPa]	3400
	Poisson's ratio	0.38
Microcapsules [b]	Young's modulus [MPa]	3200
	Poisson's ratio	0.33
Healed materials (polydicyclopentadiene) [c]	Young's modulus [MPa]	1900
	Poisson's ratio	0.39

[a]Data adopted from Brown *et al.* (2004)
[b]Data adopted from Vallons *et al.* (2015)
[c]Data adopted from Keller and Sottos (2006)

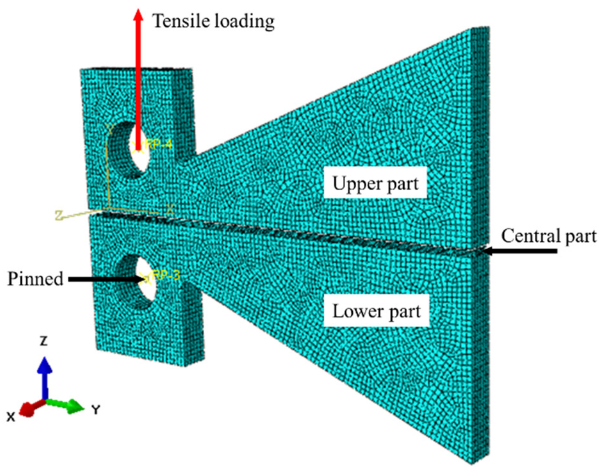
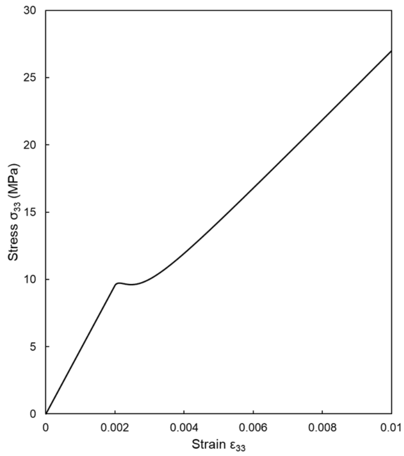


Fig. 8 The meshes employed in the models of the TDCB specimen with parts and constraints (Brown *et al.* 2004, Garoz Gómez *et al.* 2015, Tsang 2020)



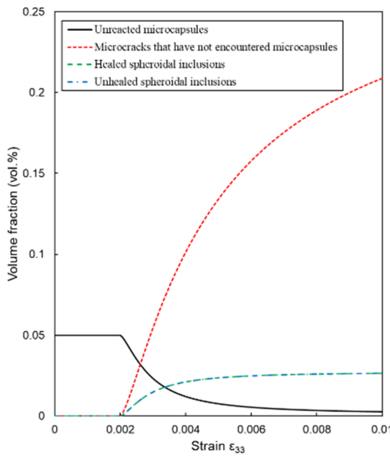


Fig. 9 (a) The present predicted uniaxial stress-strain response of self-healing polymers containing the microcapsules; (b) predicted volume fractions of unreacted microcapsules V_{uc}, microcracks that have not encountered microcapsules V_{nmc}, healed spheroidal inclusions V_{hmc}, and unhealed spheroidal inclusions V_{umc} corresponding to Fig. 9(a)

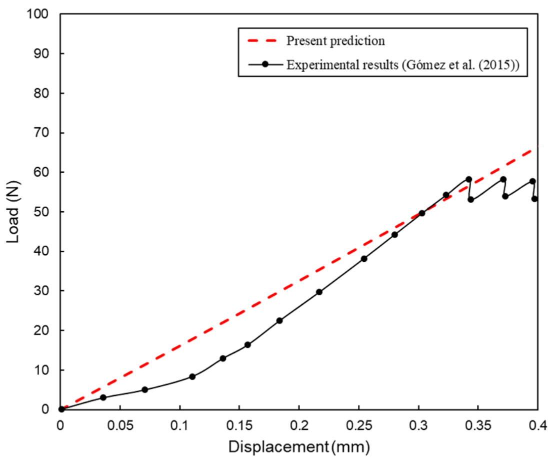


Fig. 10 Comparisons of the load–displacement behaviors of TDCB specimen between the present prediction and experimental results by Garoz Gómez *et al*. (2015)

predicted load is shown to be slightly higher than the experimental results. One of the causes that affects this discrepancy may be due to the fact that the present model does not consider large cracks which can be observed on the surface of the TDCB specimen.

5. Conclusions

A probabilistic micromechanical framework for self-healing polymers containing microcapsules has been presented herein. A five-phase micromechanical model based on the two-step self-healing process is proposed, and the hitting probability model combined with a crack nucleation model is utilized for encountering microcapsules and microcracks. A series of parametric investigations are conducted to examine the influence of various model parameters on the self-healing capabilities of the polymers. The proposed probabilistic micromechanical framework is implemented into the finite element code ABAQUS to solve the self-healing behavior of the TDCB specimen, and the prediction results are compared with the experimental results by Garoz Gómez *et al*. (2015). The observations and findings of this numerical study can be summarized as follows:

(1) In parametric investigations, the effective stress values of the self-healing polymers at the strain of 0.01 are increased due to an increase in both the volume fraction of microcapsules and the radius of the microcapsules. It can be said that the volume fraction (e.g., 1.0 ~ 10.0 vol.%) and the radius of microcapsules (e.g., 100 ~ 200 μm).
(2) The volume fraction of reacted microcapsules is significantly influenced by the radius ratio of microcracks to microcapsules and the aspect ratios of microcracks. Additionally, a higher scale parameter leads to a substantial increase in the stiffness of the polymers.
(3) The proposed micromechanical framework is implemented into a finite element code to solve the self-healing behavior of TDCB specimens. The predicted load-displacement behavior of the TDCB specimen is shown to be slightly higher than the experimental results (Garoz Gómez *et al*. 2015).

The applicability of a probabilistic micromechanical framework for predicting the self-healing characteristics of the self-healing polymers containing microcapsules has been investigated. Some model parameters (e.g., the radius of microcracks, the aspect ratio of microcracks, and the scale parameter) utilized in the series of the present predictions are numerically obtained. Therefore, it is worth mentioning that the model parameters used in this work need to be further calibrated through a series of experimentation on target self-healing polymers for a more realistic prediction. While the present study does not consider the effect of the amount of catalyst on the self-healing performance, this could be considered in future extensions of the proposed modeling framework.

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