Self-Repairing Mechanism of MUF/Epoxy Microcapsules for Epoxy Material

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Abstract: In this paper, a post curing reaction for the microcapsule/epoxy composite material and the conditions of thermal treatment for self-healing process were studied by differential scanning calorimetry (DSC). The condition of thermal treatment for post curing (60°C, 2 hours) was employed to fully cure the epoxy composite. Damage mechanism for the epoxy material was demonstrated via data simulation and three-point bending experiment for the stress distribution reveals that micro-cracks are more likely to be generated on the central region in stress concentration area of two constrained boundaries and the numbers of micro-cracks are reduced from the central area to the two ends of the material. Self-repairing performances of MUF microcapsule/epoxy composite materials were characterized using both destructive bending tests and non-destructive DMA measurements. Self-healing efficiencies of the composites embedded 2% and 5% microcapsule content measured by DMA are 101% and 104% respectively which are close to those results of 104% and 113% correspondingly measured by bending tests. Crack formation and development, core material releasing for MUF microcapsules and physiochemical process of the self-repairing were investigated by using OM, fluorescent technique and infrared microscope. These provide detailed evidences and important information on self-healing mechanism of the microcapsule/epoxy self-repairing material.

1. Introduction

At present, self-repairing properties including fracture toughness or varied mechanical properties of virgin, damaged and healed material are tested by these typical destructive methods at the cost of damaging the sample integrity [1]. In contrast, little work demonstrates a non-destructive test to reasonably investigate the self-healing performance of the materials. Dynamic mechanical performance of the same specimen exhibits a stable response undergoing several DMA non-destructive testings. The changes in dynamic modulus and mechanical loss factor with temperatures and frequency initiated by material molecules or structural modification are simultaneously provided, which is informative to understand the situation of segmental motion for the self-healing material after damaging and self-repairing process on a microscopic level.

Damage mechanism for the epoxy material was demonstrated via data simulation for the stress distribution in three-point bending [2-4]. The post curing reaction for the microcapsule/epoxy composite material, the conditions of thermal treatment to eliminate the interference of the post curing on the evaluation of the self-healing property and self-repairing cross-linking reaction were studied by differential scanning calorimetry (DSC). Self-healing process for MUF microcapsule/epoxy composites
are applied at a high temperature. Damage degrees of these materials were controlled by using constant strain three-point bending fixture. Additionally, self-repairing performance of the MUF microcapsule/epoxy composite material was quantitatively characterized using both bending destructive tests and DMA lossless tests. Core material releasing for MUF microcapsules and physiochemical process of the self-repairing were investigated by using fluorescent technique and infrared microscope. These provide important information on the formation and propagation of micro-cracks in the composite, particularly on self-healing mechanism of the microcapsule/epoxy self-repairing material.

2. Experimental Section

2.1 Preparation of MUF Microcapsules
Refer to previous works, reference 5

2.2 Self-Repairing Efficiency Measurements

2.2.1 Self-Healing Efficiency Characterized by Bending Properties. Self-repairing effects can be evaluated by the variations of bending properties for the microcapsule/epoxy composite subjected to the damages and self-repairing process. The healing efficiency (self-healing), which is the ratio of regained strength for the self-healing material to losing strength for the damaged material.

2.2.2 Self-Healing Efficiency Characterized by Dynamic Mechanical Properties. Self-repairing effects can also be evaluated by the differences in DMA data for the microcapsule/epoxy composites subjected to damages and self-repairing process. The healing efficiency (P’self-healing), which is the ratio of regained storage modulus for the self-healing material to losing storage modulus for the damaged material.

2.3 Data Simulation for The Stress Distribution in Three-Point Bending
ABAQUS finite element software was adopted to compute numerical values of stress distribution in the epoxy composites. The dimension of calculation model was the same as that of tested specimen. A simply-supported edge was selected on a center span of 51mm for long edge and 4 mm vertical displacement was exerted on the center in this model. The data simulation was obtained by C3D8 module with an elastic modulus of 2GPa and a Poisson ratio of 0.38. The stress response at elastic stage was merely considered in order to simplify stress concentration analysis.

2.4 DSC Test
Differential scanning calorimetry (DSC-60, Shimadzu, Japan) calibrated by high purity indium and zinc was employed to study the epoxy post curing reaction and the condition of post curing treatment and self-healing thermal treatment for MUF microcapsule/epoxy composite. The specimens were heated from 25 to 220 °C at a heating rate of 10 °C /min under a nitrogen atmosphere at a flow rate of 50mL/min.

2.5 Testing Sample
At least ten MUF microcapsule/epoxy composite specimens were selected to form a batch of MUF microcapsule/epoxy self-healing sample. Nine specimens were divided into three groups including virgin sample (Type I), damage-controlled sample (Type II) and self-healing sample (Type III). The tenth specimen was used for DMA test. Because these specimens may occur post curing reaction during heat process that interfere with evaluation of the self-healing efficiency, the needed thermal preprocessing conditions were discussed in the relevant DSC measurement.

The Type II and the Type III samples were subjected to a controlled constant strains using three-point bending fixture with deformation of 90% bending fracture strains of the specimens which produce cracks within the materials based on the bending deformation vertical distance of the
specimens in the fixture according to Equation 3. The specimens were clamped at this bending deformation distance for 24 hours. After the cracks observed, the specimens were removed off and flattened under a loading, then tested by universal tensile machine for bending properties and DMA instrument for the dynamic mechanical property.

The Type III samples after subjection to constant strain damaging were heated in vacuum drying oven at 100°C for 1 hour. After the self-healing process, bending properties of these specimens and the dynamic mechanical property were measured through bending tests and DMA test.

2.6 Bending Test
Bending properties for three types of samples were measured by universal tensile machine (model AGX, Shimadzu, Japan). The Type I samples were directly measured without any treatment. In this three-point bending test, bending span was 30 mm and bending speed was 5mm/min. Force-displacement curves were recorded for data analysis. The average value of three specimens data was taken as corresponding type bending strengths (I) and bending fracture strains (ε) [5].

2.7 DMA Test
Storage modulus (E’), loss modulus (E’’) and mechanical loss factor (Tanδ) of the tested specimen were measured via DMA temperature sweep experiment (Q800, TA Instruments, UK). DMA test was performed using a double cantilever beam, heating from 25°C to 140°C at a heating rate of 5°C /min and an oscillator frequency of 1 Hz under nitrogen atmosphere[6].

2.8 Inverted Fluorescence Microscope Analysis
In order to study self-repairing mechanism of the MUFmicrocapsule/epoxy material, a batch of MUF microcapsules was particularly prepared by the addition of sodium fluorescein (F6377, BioReagent, Sigma-Aldrich China) into the epoxy healing agent. These special microcapsules were added into epoxy resin in similar ways to fabricate the composites previously to emphasize self-healing processing and mechanism. Crack propagation of the special specimen after subjected to the damage was observed using inverted fluorescence Microscope (IX70, Olympus, Japan) equipped with a 100W high-pressure mercury lamp as excitation source while a objective lens (40x, N.A. = 0.6, Olympus, Tokyo, Japan) and a blue optical filter were employed. An SIT camera(C2741-08, HAMAMATSU, Shizuoka, Japan) and a digital videocassette recorder (DVCAM, SONY, Tokyo, Japan) were also used for detection of an emission light resulting from the releasing of the healing agent mixed with the fluorescein, recording the rupture and release information of MUF microcapsules in the damaged specimen.

2.9 Infrared Microscope Analysis
Infrared microscope (Nicolet Continuum, ThermoFisher, USA) was employed to analyze the filling materials on the fracture plane of the microcapsule/epoxy composite. Before measurement, cooling container of the microscope was filled with liquid nitrogen. The apparatus was stabilized for 1 hour in order to fully cool MCT detector. The optical gating of the infrared microscope was adjusted to 100μm×100μm. The image observation software attached to the apparatus was used to select the areas of visible fillings within the cracking of the composite specimen and to identify its chemistry. The IR spectrum was obtained in the wave number range 4000-450 cm-1 with a resolution of 4cm-1 from 28 scans.

3. Results and Discussions

3.1 Damage Mechanism for Epoxy Composites by Three-Point Bending
Micro-crack damage within material can be induced via a process of stretching. An effective course is needed for micro-crack propagation, while the pre-stretching damages are hardly controlled by tensile machine for a long time so that crack growth cannot be located using microscope technique. In contrast, constant strain applied in three-point bending fixture was employed to act on the self-healing material.
in a process of pre-bending, which could design various stages of the damage for detailed study.

Figure 1(a) shows the constant strain of three points bending fixture is applied in controlled damage for the epoxy composite specimens. Stress distribution on the composite material was analyzed in the condition of constant strain using three-point bending model. The data analysis in Figure 1(b) predicts the Mises stress distribution of equipotential line for the designed composite material and counter-force on vertical displacement is 0.448N. The Mises stress has a maximum value of 0.684 N/mm² located at the central of the specimen, which is quickly attenuated from the center region along the z direction to the two ends because of small thickness of the specimen, so micro-cracks are more likely to be initiated on the center region of two constrained boundaries in stress concentration area. The OM photograph in Figure 1(c) shows that crack patterns for MUF microcapsule/epoxy self-healing material damaged by constant strain in three-point bending fixture. The region shown in Figure 1(c) is corresponding to the red frame of simulative specimen in Figure 1(b). Cracks are mainly observed in the center of the specimen where the specimen is subjected to the maximum vertical stress. With the gradual reduction of stress along the specimen edge, the numbers and the widths of micro-cracks are decreased from the central area to the two ends of the epoxy composite specimen. Crack initiation of the material subjected to bending by the fixture was located as expected, which could help explain the generation of cracks discussed previously. As shown in Figure 1(d) magnified by using monitoring software, the MUF microcapsule whose diameter is approximate 90μm of the epoxy composite specimen after subjection to constant strain damaging is entirely ruptured by a crack with 20-50μm width that causing adequate epoxy healing agent is capable of releasing into the crack surface. After self-healing process seen in Figure 1(e), the cracking and the MUF microcapsule in the epoxy matrix are entirely rebonded because of the cross-link reaction between the released epoxy healing agent and curing catalyst. However, those 200μm MUF microcapsules incorporated in the epoxy composite after damaging may not be fully ruptured by this width of crack causing less volume of epoxy healing agents flow into the crack plane directly reducing self-healing performance of the material. Therefore, the epoxy composite specimens embedded 90μm MUF microcapsules are merely applied in the following testings.
Figure 1. Data of three point bending testings for the MUF microcapsule/epoxy self-healing materials. (a) constant strain of damaging fixture photo; (b) Mises stress distribution simulation; (c) crack distribution after subjection to constant strain damaging. (d)MUF microcapsule morphology after subjection to constant strain damaging; (e) MUF microcapsule morphology after self-healing process;

3.2 Self-Healing Performance Test

3.2.1 Post Curing Reaction and The Condition of Self-Healing Process. The DSC curves of MUF microcapsule/epoxy composite are shown in Figure 2. The exothermic peak between curve A and baseline (dash line in this figure) over the temperature range above the Tg of epoxy material may demonstrate an existence of post-curing reaction within the epoxy matrix[7]. During the later stage of specimen curing, diffusions of epoxide active centers are difficult. The epoxy composite system transforms to glassy state and the cross-linking reaction is controlled by the chemical diffusion rather than reaction kinetics so that some unreacted active centers still remain within the epoxy matrix[8]. With the increase of testing temperature, chemical diffusions of residual active epoxides within the matrix are improved initiating the post curing reactions within the matrix between the residual epoxy active centers and unreacted curing agent DETA. Therefore, a small exothermic peak of post curing for the epoxy composite specimen cured at room temperature is observed in curve A. Because the post curing reaction may interferes with the self-healing efficiency evaluation for MUF microcapsule/epoxy composites, a post-curing thermal treatment (60°C, 2 hours) for the epoxy composite specimen was applied in order to fully cure the epoxy specimen used in following self-healing performance testings. The curve B exhibits there is no exothermic peak over the temperature range above the Tg of the MUF microcapsule/epoxy composite specimen after the post-curing heat treatment applied, which demonstrates the epoxy composite specimen was fully cured in this thermal treatment condition. The curve C indicates that multiple exothermic peaks of the specimen Type II are observed at approximate 100°C which illustrates significant curing reaction between the epoxy resin and the MC120D catalyst occurs at this temperature. In addition, a broad exothermic peak over the temperature range above the Tg of the epoxy composite demonstrates an effective cross-linking reaction between the healing agent released from ruptured MUF microcapsules and the latent imidazole curing agent embedded within the matrix occurs at the high temperature range, being capable of automatically repairing the cracks within the materials.
3.2.2 The Self-Healing Efficiencies Characterized by Bending Property. Bending properties for three types of the epoxy composites designed at different microcapsule contents are shown in Figure 3. Bending strength of the specimen Type I with 2wt% content microcapsule is reduced to 90.4% value of the epoxy specimen. Microcapsules as stress concentration points directly cause the generation of cracks in the specimen after the constant bending applied so that bending strength of the specimen Type II is decreased. Bending strength of the specimen Type III is recovered and the self-healing efficiency is 104%, while that of the epoxy specimen is not regained. In the condition of 5wt% microcapsule content, bending strength of the specimen is reduced to 64.3% value of the epoxy specimen and the self-healing efficiency of the specimen achieve 113%, which can be attributed to the higher microcapsule content in the self-healing material, the more volumes healing agents release from the ruptured microcapsules into the cracking so that the self-repairing ability of the material is effectively improved.

3.2.3 The Self-Healing Efficiencies Characterized by Dynamic Mechanical Property. Dynamic mechanical properties with the temperatures and the information of chain segment movement for three types of the epoxy composites can be simultaneously monitored. As shown in Figure 4(a), value of initial storage modulus for epoxy specimen is 2.44GPa. That of the epoxy specimen subjected to damage by constant strain dramatically reduces to 1.82GPa and that of the epoxy specimen after self-healing is 1.84GPa. No obvious repairing effect is founded in the epoxy specimen. Figure 4(b) indicates that the value of initial storage modulus for MUF microcapsule/epoxy composite specimen with 2% microcapsule content is 2.27GPa. That of the epoxy composite specimen subjected to damage is decreased to 1.97GPa and that of the epoxy composite specimen after self-healing process is recovered to 2.27GPa. In Figure 4(c), the value of initial storage modulus for MUF microcapsule/epoxy composite specimen with 5% microcapsule content is 2.04GPa. That of specimen type II declined to

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**Figure 2.** DSC curves of MUF microcapsule/epoxy composites. (a) curing at room temperature; (b) curing at 60°C for 2 hours; (c) heat flow changes in self-healing process.

**Figure 3** Bending strength comparisons of self-healing effects for epoxy composites. (a)2wt% microcapsule content; (b)5wt% microcapsule content.
1.65GPa and that of specimen type III is considerably recovered to 2.06GPa, slightly exceeding original value of the material. These changes indicate that cross-linked network within epoxy matrix subjected to damage process is broken and molecular chains are fractured under loading, thus reducing the ability to resist deformation. After self-repairing, because epoxy composite is incorporated with MUF microcapsules contained epoxy healing agent and latent curing agent which are able to initiate cross-linked reaction in the self-healing process at a high temperature, cracks are rebonded by this chemical reactions and the broken cross-linked epoxy network is repaired in some extent, leading to a recovery for the material properties. The 3.12% self-healing efficiency of the epoxy specimen may result from sample error, experimental error and instrumental error, which need to be taken into consideration in evaluation of self-healing efficiency. The significant self-repairing in the MUF microcapsule/epoxy composite specimen with 2% and 5% microcapsule content achieve high self-healing efficiencies of 101% and 105% respectively.

![Figure 4](image_url)

**Figure 4** Storage modulus for three types of epoxy composite specimens. (a) epoxy specimen; (b) MUF microcapsule/epoxy self-healing specimen (2% microcapsule content); (c) MUF microcapsule/epoxy self-healing specimen (5% microcapsule content);

With regard to thermoset epoxy resin, peak value of Tanδ reflects relative degree of cross-linking density. Peak width of Tanδ is related to the relaxation transition for chain segment[10]. In Figure 5(a), the peak value of Tanδ for epoxy specimen is relatively high with a glass-transition temperature of 80.2°C. Relaxation transition of chain segment in epoxy is difficult due to its high cross-linking density which provides little free volume for molecular movement within the cross-linking network. After the epoxy specimen subjected to damages, both the area and the Tg of Tanδ peak are decreased, Tg is 78.7°C. At this stage, The cross-linked network within the matrix is broken partially and molecular chains may be fractured, thus decreasing the cross-linking density. In addition, because the length of chain segment is shortened, flexibility of molecular chains and mobility of motion units are promoted, resulting in this shift of the peak. After self-healing process, the peak width of Tanδ for the epoxy specimen is broadened and Tg is slightly increased to 79.8°C, whereas the peak value considerably decreases. The rearrangement of chain segment occurs within the epoxy material broadening the peak width. However, the damaged cross-linked network is not repaired so the Tg is not obviously increased. As shown in Figure 5(b), the peak value of Tanδ for MUF microcapsule/epoxy composite specimen with 2% microcapsule content is relatively high with a glass-transition temperature of 77.4°C. After damages applied in the epoxy composite specimen, both the shape and the corresponding temperature of Tanδ peak are reduced with the Tg of 75.9°C. The molecular chains in the composite are fractured during damage and the cross-linked network is partially broken, which reduces the cross-linking density. Flexibility of molecular chains and segment mobility are increased, leading to a movement toward a low temperature of the peak. The peak width of Tanδ for the epoxy composite specimen after self-healing process is broadened and the Tg is significantly increased to 85.3°C, while the peak value is considerably reduced. After self-repairing, it is speculated that non-uniformity of chain segments in the self-healing process at high temperature causes an improved mobility of segments so that peak width of the Tanδ is broadened. Compared with epoxy specimen, glass-transition temperature of MUF microcapsule/epoxy composite specimen is considerably increased by approximate 10°C that could result from the damaged material is more densified by the cross-linking reaction between healing agent
and curing catalyst involved. Additionally, the cross-linked network is introduced with long chain segments from special curing agent molecular of latent imidazole curing catalyst MC120D. The length and the stiffness of the chain segment related to the cured epoxy resin in the cracking location and steric hindrance of internal rotation for the chain segments are increased. As a result, the glass-transition temperature of epoxy composite specimen is considerably raised and the dynamic mechanical performance is also enhanced. Additionally, the changes in the Tg and the peak value of Tanδ for MUF microcapsule/epoxy composite specimen with 5% microcapsule content are similar to that of the epoxy composite specimens with 2% microcapsule content shown in Figure 5(c). The Tg for three types of the epoxy composite specimens are 77.5°C, 73.3°C and 84.8°C respectively.

![Figure 5](image)

Figure 5. Mechanical loss factors for three types of epoxy composite specimens. (a) epoxy specimen; (b) MUF microcapsule/epoxy self-healing specimen (2% microcapsule content); (c) MUF microcapsule/epoxy self-healing specimen (5% microcapsule content).

Healing efficiency of MUF microcapsule/epoxy material with 2% and 5% microcapsule content measured by DMA are 101% and 104%, which are close to those of the materials characterized by bending properties. Because bending testing is destructive, bending properties of three type specimen cannot be directly compared. A large numbers of experimental specimen having same composition and closing property must be used in order to have specimen repeatability and testing reliability. In contrast, DMA which is non-destructive, sensitive, repeatable and reliable provides a new method in the research of self-healing material.

### 3.3 Self-Repairing Mechanism

In this part, formation and propagation of the micro-cracks in MUF microcapsule/epoxy composite, reason and process of core healing agent releasing from microcapsule are observed using OM and particularly emphasized by inverted fluorescence microscope. The course of autonomic repairing micro-crack and the chemistry of the crack filling material are studied specifically by infrared microscope discussing the reaction mechanism between epoxy healing agent and latent imidazole curing catalyst.

Figure 6 shows a series OM image of a cracking development for MUF microcapsule/epoxy self-healing composite damaged by a constant strain. The rupture process of MUF microcapsule and the release process of healing agent contained sodium fluorescein can be observed using inverted fluorescence microscope directly. The crack of the specimen is marked so that the rupture details of MUF microcapsules can be observed. The cracking is produced from a bubble as an edge defect at location A. Bubbles are coated in epoxy material due to a high viscosity of epoxy resin, which are difficult to be entirely removed during the process of specimen preparation. When the crack is propagated through MUF microcapsule, the microcapsule is ruptured by cracking stress to release epoxy adhesives that wet crack plane owing to the capillary effect. Epoxy resin is an oligomer and its molecular chains have numerous oxygen and hydrogen active atoms. Viscosity of epoxy E-51 is relatively high at room temperature because of strong intermolecular forces such as hydrogen bond are existed, which restrict molecular motion. This could be a possible reason that those reported self-healing materials have a limited self-repairing efficiency. BGE is used to dilute the epoxy so that the epoxy viscosity is reduced considerably as shown in Figure 7. When BGE content is increased to
10wt%, the viscosity of epoxy resin is considerably reduced. In addition, the micro-crack passes two microcapsules at location B and C in Figure 6 then extending continuously until the crack ruptures a microcapsule at location D. Modified epoxy healing agent in this microcapsule flows into the micro-crack and wets micro-crack surface then contacts curing agent previously incorporated in the matrix triggering a curing chemistry to produce a cured substance that is capable of repairing the cracks more effectively. It is observed that width of the crack is obviously decreased at location D and the crack is finally terminated at location E, which illustrate functions of both that the healing agent released from the ruptured microcapsule at location D bonds the crack surfaces and that the process prevents the crack propagation.

**Figure 6.** Crack propagation of MUF microcapsule/epoxy self-healing material. (40x lens of fluorescence microscope)

Fracture surface for the healed region of the MUF microcapsule/epoxy self-healing composite was analyzed by SEM. Figure 8(a) shows that a cured layer material within matrix visibly repairs the micro-crack. In Figure 8(b), infrared spectrum of this filling material (spectrum a) in comparison with that of epoxy diluted by BGE and cured with MC120D curing agent (spectrum b) show shapes and locations for the characteristic peaks of epoxy resin, mainly include hydroxyl at 3482 cm⁻¹, benzene ring at 1610 and 1506 cm⁻¹ and aryl ether at 1252 cm⁻¹. The characteristic peak of epoxide in 772 cm⁻¹ is almost disappeared in two spectra and that of imidazole ring is seen at 1510 cm⁻¹ in both spectra, which illustrate an expected chemical reaction occurred. These results prove that the cross-link reaction occurred between the epoxy shelled by MUF resin and imidazole chemical inside the matrix cracking which formed an effective bonding between cracked surfaces[11].

**Figure 7.** Viscosity of epoxy resin with BGE diluents.
Figure 8 Characterizations for the filling material within the crack. (a) SEM photo for the filling material; (b) Infrared spectra of the filling material and the reference material.

Molecular structure of the selected latent imidazole hardener contains a relatively long molecular chain positioned at the 3-substituted nitrogen, increasing the molecular steric hindrance so that the reaction between released epoxy adhesive and the hardener cannot be initiated at low temperature, whereas the latency is disappeared under a high temperature of heating discussed previously. BGE as reactive diluent has ether group and epoxide functional group that can join into the reaction of epoxy resin and imidazole hardener, together forming the cross-linking network. Because of charge polarization on epoxide group, it is formed that higher electron cloud density at the oxygen atom and lower electron cloud density at the carbon atom. The oxygen atom with negative charge on epoxide group of the epoxy resin or BGE initiates electrophilic addition for the reactive hydrogen atom of amine from the hardener, which results in ring-open polymerization (a) that the adduct (1:4) is generated by a ring-opened of epoxide group. Besides that, the lone pair electrons positioned at the 1-substituted nitrogen of imidazole ring attack the epoxide group, taking place the 1:1 addition reaction (b) and forming clathrate with coexistence of positive and negative ions. The negative ions of clathrate as active centers catalyze the epoxide ring open, triggering copolycondensation of epoxy resin, BGE and imidazole[12].

4. Conclusion
The conditions of thermal treatment for post curing (60°C, 2 hours) and cross-linking reaction (100°C, 1 hour) are important factors in the measurement of self-healing efficiency for MUF microcapsule/epoxy composite. The data simulation of stress distribution for epoxy composite reveals that micro-cracks are more likely to be initiated on the center region of stress concentration area for two constrained boundaries and the amounts of micro-cracks are decreased from the central area to the two ends of the material. Self-healing efficiencies of the epoxy self-healing composites embedded 2% and 5% microcapsule content measured by DMA are 101% and 104% respectively which are close to those results (104% and 113% correspondingly) measured by bending tests. This demonstrates DMA can be used to characterize self-healing efficiency of composite material. The self-repairing mechanism of the MUF microcapsule/epoxy composite materials is that numbers of micro-cracks within the composite after subjection to constant strain damages are generated. Special prepared microcapsules are ruptured by cracking stress releasing adhesive with fluorescence substance, flowing along crack propagation direction into cracking gaps and wetting its surface. The infrared analysis of the cured filling material inside the cracking captured by SEM prove that the cross-link reaction between the epoxy shelled by MUF resin and imidazole chemical inside the matrix cracking occurred, forming an effective bonding between cracked surfaces.

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