A method for self-healing of CFRP using optical fibers

BAO Limin *, UCHIO Chika, RUAN Fangtao, KEMMOCHI Kiyoshi

Faculty of Textile Science and Technology, Shinshu University,
3-15-1 Tokida, Ueda, Nagano 386-8567, Japan

Received 16 August 2014, accepted for publication 1 June 2015

Abstract

CFRP has been applied in various fields where it is mostly used as laminates. CFRP laminates are likely to fracture between the interlaminar surfaces. To resolve this problem, we proposed a self-healing system for CFRP using optical fibers and UV-curable resin. In this study, self-healing CFRP samples were prepared and the effectiveness of repairing the inter-laminar fractures was confirmed by using optical fibers and UV-curable resin. In addition, we have also clarified the factors that affect the repair rate by determining the bonding strength of epoxy resin and UV-curable resin. From the experiment results, we were able to confirm the feasibility of the proposed self-healing system.

Key Words: CFRP, Optical fibers, Self-healing, Bonding strength

1. Introduction

Carbon Fiber Reinforced Plastic (CFRP), a composite material, is used in a wide range of fields because of its excellent specific strength and specific rigidity. Much CFRP is molded into laminated plates. CFRP is strong in each layer, particularly in the fiber direction; however, it is slightly weaker between layers and, therefore, it will experience degradation over time or develop delamination and/or cracks when it receives any external shock. When delamination and/or cracks develop, CFRP becomes unable to share the load as a whole and the portion with delamination and/or cracks will be destroyed by a lower load than undamaged material would be. Because any delamination and/or cracks that occur while CFRP components are in use are very dangerous, it is necessary to clarify the influence of such defects on the material strength and to improve the reliability and safety of that material.

We note that organisms have self-healing mechanisms against their wounds. An example of such a mechanism is the healing of bones. When a fracture occurs, the bone cells around the fractured part die. Blood is then carried from the heart via the blood vessels and capillaries, the hematoma formed around the fractured part due to hemorrhage from the damaged tissue fill the bone fracture gaps, fibroblasts from the inner layer of the outer periosteum and the endosteum infiltrate into the hematoma and become a cartilaginous callus by forming granulation tissue, and the cartilaginous tissue in the callus becomes bony tissue. The bones in the fractured part will thus exhibit bone healing. If the material can heal any damage by itself as in organisms, such damage will not develop into fatal destruction. In other words, if structural materials can be given a self-healing function and its effect is demonstrated, it will contribute to improving the reliability of structures. Attention is thus paid to earlier studies of self-healing CFRP that enables automatic repair of delamination and/or cracks.

Achieving self-healing of materials requires a sensor section to detect any damage to the material, a controller section to perform response and control based on information from the sensor, and an actuator section to implement the repair. Materials that have these three elements are defined as self-healing materials [1].

One self-healing method for CFRP uses a microcapsule as proposed by White et al [2, 3] and the authors [4]. Another method uses hollow glass fibers as proposed by Bond et al [5]. These methods propose that the damage be repaired with a repair agent that flows out of a microcapsule or hollow glass fiber embedded in the CFRP if any damage occurs. However, there is a problem in that a sufficient volume of repair agent cannot be loaded in the microcapsule or hollow glass fiber embedded in the CFRP if any damage occurs. We note that organisms have self-healing mechanisms against their wounds. An example of such a mechanism is the healing of bones. When a fracture occurs, the bone cells around the fractured part die. Blood is then carried from the heart via the blood vessels and capillaries, the hematoma formed around the fractured part due to hemorrhage from the damaged tissue fill the bone fracture gaps, fibroblasts from the inner layer of the outer periosteum and the endosteum infiltrate into the hematoma and become a cartilaginous callus by forming granulation tissue, and the cartilaginous tissue in the callus becomes bony tissue. The bones in the fractured part will thus exhibit bone healing. If the material can heal any damage by itself as in organisms, such damage will not develop into fatal destruction. In other words, if structural materials can be given a self-healing function and its effect is demonstrated, it will contribute to improving the reliability of structures. Attention is thus paid to earlier studies of self-healing CFRP that enables automatic repair of delamination and/or cracks.

Achieving self-healing of materials requires a sensor section to detect any damage to the material, a controller section to perform response and control based on information from the sensor, and an actuator section to implement the repair. Materials that have these three elements are defined as self-healing materials [1].

One self-healing method for CFRP uses a microcapsule as proposed by White et al [2, 3] and the authors [4]. Another method uses hollow glass fibers as proposed by Bond et al [5]. These methods propose that the damage be repaired with a repair agent that flows out of a microcapsule or hollow glass fiber embedded in the CFRP if any damage occurs. However, there is a problem in that a sufficient volume of repair agent cannot be loaded in the microcapsule or hollow glass fiber (50% or less of the internal capacity of the microcapsule), and the strength of the CFRP decreases considerably if a large volume of repair agent is loaded in the microcapsule. The authors [6] devised a capillary that does not decrease the CFRP strength very much, manufactured a prototype, and tried a method of injecting repair agent into the damaged part from outside of the CFRP. In this method, we considered that the shortage of repair agent in the microcapsule method can be overcome by injecting the repairing agent continuously into the...
damaged part. However, two-liquid mixed epoxy resin is used as the repair agent, and there are some problems, such as that it requires a long time for after-curing before hardening and caution is required for its storage.

In this study, therefore, we will propose a self-healing system for CFRP that can continuously inject a repair agent from the outside using optical fibers with a light-transmission function as well as photocoagulation resin as the repair agent. An outline of the proposed self-healing system is presented in Fig. 1. The flow of repair is as follows:

1. Embed an optical fiber in the CFRP and incorporate the mechanism for injecting repair agent. Pass infrared light through the optical fiber to detect any damage.
2. When damage occurs in the CFRP, the optical fiber ruptures, and this will be detected by the infrared-light-receiving sensor. A pump is actuated based on information from the sensor to continuously inject repair agent into the damaged part. At the same time, the infrared light is converted into light corresponding to the hardening of resin that will be radiated from the tip of the ruptured optical fiber.
3. The repair agent injected into the damaged part will be cured by light, repairing the damage.

In the proposed self-healing system, repair begins immediately after the damage occurs through sensing of the damage with optical fibers and radiation of light in response to it. Furthermore, photo-curable resin hardens quickly and does not require after-curing, so the repair is also completed within a short time. By continuously injecting repair agent, a sufficient volume of repair agent can be injected to repair the damage.

To confirm the feasibility of the proposed self-healing system, we will mold CFRP that has a self-healing structure and try self-healing using optical fibers and ultraviolet-curable resin (UV-curable resin), a kind of photocurable resin, as the repair agent. We will also clarify the factors that influence the repair rate by investigating the mechanical characteristics of the resin of the base material and of the repair agent.

2. Propagation of UV via optical fibers

UV-curable resin is hardened by the energy of light for polymerization when it is irradiated with UV rays. This resin is used in a wide range of fields because it hardens quickly and does not require much energy for hardening. An optical fiber is a thin fiber made of a transparent insulator such as glass or plastic that is composed of a central core that passes light and the clad surrounding the core. Optical fiber is used mainly in the field of digital communications. Takeda et al. [7] developed structure-health monitoring technology that uses optical fibers to improve the reliability of CFRP structures for use in aerospace applications. Damage such as cracks and delamination caused by an external load near the sensor section has been detected using an optical fiber sensor embedded in the CFRP structure. The authors [8] proposed measuring and evaluating the erosion state of FRP by embedding an optical fiber in the FRP and using it as a health-monitoring sensor. We prepared a prototype and conducted tests to confirm its usefulness as an erosion- and wear-detecting sensor for FRP.

We conducted a confirmation test for the methods as depicted in Fig. 2 to determine if UV light that passes through the optical
One end of an optical fiber is inserted into UV-curable resin (80MFA, Kyoeisha Chemical Co., Ltd.) poured into a non-transparent plastic cup to L=2mm above the bottom of the cup, and the other end of the optical fiber is connected to the tip of a UV LED (NSPU510CS, Nichia Chemical Industries Co., Ltd., Wavelength: 375nm). The UV LED radiates UV rays into the resin to harden it, with 13mA of electric current supplied to the LED.

Common, commercially available quartz optical fibers are made for communications and produce a large loss at UV wavelengths. Experiments indicated that hardening takes a long time, so such fibers are not suitable for UV curing. Instead, the three types of plastic optical fibers listed in Table 1 were used. The core diameter differs from one type to another.

After the UV LED is turned on, the resin at the end of the optical fiber hardens and the diameter of the hardening portion of resin increases. Fig. 3 depicts the relationship between the UV irradiation time and the diameter of the hardened resin as measured using optical fiber BK-CK20. The diameter increases rapidly with time initially but the increase slows after 60min. This indicates that hardening of resin is possible by transmitting UV light via optical fibers.

As the diameter of the optical fiber core increases, the diameter of the hardened resin increases, and thus the hardening time decreases. In this study, it is necessary to insert optical fibers into the fiber bunch of carbon textile, and gaps in the fiber form easily when the diameter of the optical fiber increases, so the strength of the CFRP might decrease. Therefore, we decided to use optical fiber BK-CK20 in this study.

### 3. Characteristics of UV-curable resin and two-liquid epoxy resin

When repairing cracks in CFRP, the mechanical characteristics of the resin of the base material and of the repair agent influence the characteristics after the repair. Mechanical characteristics of the resin used for the base material and of the repair agent will be evaluated with the Double Cantilever Beam (DCB) test. The DCB test will be conducted in compliance with JIS7086. A schematic diagram of the DCB test is presented in Fig. 4. The test speed will be 0.5mm/min. The test will be continued on acrylic specimens until the crack opening displacement (COD) reaches 10mm and on the CFRP until the optical fibers rupture.

#### 3.1 Acrylic specimens

Properly speaking, the mechanical characteristics of the resin shall be evaluated using CFRP. However, CFRP is not permeable to UV light, so it is impossible to harden UV-curable resin. In this experiment, therefore, we will investigate the mechanical characteristics of the resin using transparent acrylic boards that are permeable to UV light.

Acrylic boards permeable to UV light (SUMIPEX: 2-mm thick, made by Sumitomo Chemical Co., Ltd.) will be used for the acrylic specimens. A mixture of UV-curable resin (80MFA, made by Kyoei Chemical Co., Ltd.) and 5% of photoinitiator (DAROCUR117, made by BASF Japan Co., Ltd.) will be used as the repair agent. The same epoxy resin as the base material of CFRP (DENATITE XNR 6815, made by Nagase Chemtex Corporation) will be used for comparison. A set of two acrylic boards with surfaces treated with corona discharge will be prepared, and each resin will be
applied to the gap between the acrylic boards. In this case, a 40µm thick film will be inserted as an initial crack. The acrylic boards applied with UV-curable resin will be exposed to UV light (wavelength: 375nm) for 1h. UV light permeates the acrylic boards, so the UV-curable resin will harden. The acrylic boards coated with epoxy resin will be subjected to after-curing under the same conditions as for CFRP molding (i.e., 55°C, 6h). The specimen is 140mm long, 25mm wide, and 4mm thick, as seen in Fig. 5, and is bonded to a block for pin load. Five specimens of each resin will be prepared.

3.2 DCB tests using acrylic boards

We conducted DCB tests using acrylic boards (hereinafter called the acrylic DCB test). An example of Load-COD (Crack Opening Displacement) curves for a specimen coated with epoxy resin and a specimen coated with UV-curable resin is depicted in Fig. 6. Here, we confirmed that the resin was destroyed but there was no peeling at the interface between the acrylic board and the resin. Let us assume the maximum loads of epoxy resin (P_{Ep}[N]) and UV-curable resin, (P_{UV}[N]) as the bond strength of each resin. The average bond strength and standard deviation of each resin are depicted in Fig. 7. The UV-curable resin used in this experiment has a lower bond strength than that of epoxy resin, accounting for 35.46% of epoxy resin.

4. Self-healing CFRP specimens

CFRP is molded by the vacuum resin-transfer molding (VaRTM) method using carbon-fiber cross material (Torayca cross CO6343, made by Torey Corporation) and epoxy resin as the base material (DENATITE XNR 6815). A schematic diagram of molding CFRP is presented in Fig. 8. The cross material will be made of 12 layers and a 7.5µm-thick polyimide film will be inserted between the 6th and 7th layers as an initial crack. Resin-injecting holes will be opened while molding the CFRP without rupturing reinforcing fibers by utilizing the method in reference [4]. After being impregnated with resin, each specimen will be subjected to after-curing at 55°C for 6h. The specimen will be 140mm long, 25mm wide, and 3mm long and will be bonded with a block for pin load. A schematic diagram.
5. Test results and consideration of self-healing

5.1 CFRP self-healing test

An example of Load-COD curves from the DCB test on CFRP with self-healing structure is presented in Fig. 10. In an ordinary DCB test, the load decreases in line with the development of cracks as indicated by the broken-line arrow. Using these results, the interlamellar fracture toughness in mode I will be obtained. Optical fibers are embedded in the load-bearing direction. As the crack approaches an optical fiber, the load increases as indicated by the solid-line arrow. Therefore, this study assumes that the load during development of the crack before the influence of the optical fiber becomes noticeable ($P_{\text{Normal}}$) is assumed as the load before the repair. The measured $P_{\text{Normal}}$ is listed in Table 2.

Cracks found in the specimen after the DCB test were repaired. A schematic diagram of a specimen being repaired as viewed from the side is presented in Fig. 11. The repair procedure is as follows.

(1) Fasten the film and tube on the resin-injecting hole with sealant tape. Connect the UV LED (Nichia Chemical Industries Co., Ltd., wavelength: 375mm) to the tip of the optical fiber.

(2) Inject repair agent into the crack via the tube using an injector.

(3) Turn on the UV LED and radiate UV light from the tip of the ruptured optical fiber for 1h.

The DCB test was conducted again on repaired specimens. Fig. 10 plots an example of a Load-COD curve from the DCB test after the repair (dotted line). For $P_{\text{Repair}}$, the limit load at the repair site during the DCB test was used. The load after the repair, $P_{\text{Repair}}$, is listed in Table 2. The repair rate $\eta$ [%] was calculated from $P_{\text{Normal}}$ and $P_{\text{Repair}}$ using Eq. (1) to evaluate the repair effectiveness. The calculated repair rate is listed in Table 2 together with the above.

$$\eta = \frac{P_{\text{Repair}}}{P_{\text{Normal}}} \quad (1)$$

The average repair rate was 9.94%, demonstrating that cracks can be repaired with optical fibers and UV-curable resin.

5.2 Elements that influence self-healing

The repaired part of the specimen after undergoing the DCB test following the repair was observed, and the repaired area was considered. A digital microscope was used to observe the repaired part. Fig. 12 presents an example of these observations. The round
portion is the optical fiber, and the portion surrounded by a line is a trace of the repaired portion of the resin that was observed. Here, as indicated in the diagram in Fig. 13, repaired area $B'$ [\%] will be defined as in Eq. (2).

$$B' = \frac{B_1 + B_2 + B_3}{B}$$  \hspace{1cm} (2)

$B'$ : Repaired area [\%]
$B$ : Width of the specimen [mm]
$B_{1,3}$ : Width of the repaired part [mm]

The greatest lateral length of the repaired part was measured as the width. The measured repaired area is listed in Table 3. The repair rate depends on the bonding strength between the base material and the repair agent, as well as on the repaired area. The equivalent of the repair rate $\eta_{C.V}$ [\%] was obtained from the ratio of bonding strengths ($P_{UV}/P_{Ep}$) obtained in Section 3 and the repaired area ($B'$) using Eq. (3).

$$\eta_{C.V} = \frac{P_{UV}}{P_{Ep}} \times B'$$  \hspace{1cm} (3)

The equivalent value was close to the measured value in Table 3. In other words, both the bonding strength between the base material and the repair agent and the repaired area significantly influenced the repair rate.

### 5.3 Increase in self-healing ability

We tried increasing the repaired area to confirm the factors that significantly influence the repair rate. We conducted repair tests with three to ten optical fibers inserted in the lateral direction of the DCB test specimen and then measured and observed the results. Two specimens were used.

Fig. 14 presents the test results. The horizontal axis represents the repaired area and the vertical axis represents the measured repair rate. A dot (●) represents the measurement results with three optical fibers, and a square (■) represents the measurement results.

![Table 3 Repaired area rate and corresponding value.](image)

| No. | $\eta$ [\%] | $P_{UV}/P_{Ep}$ | $B'$ [\%] | $\eta_{C.V}$ [\%] |
|-----|-------------|-----------------|----------|-----------------|
| 1   | 13.98       | 35.46           | 40.99    | 14.54           |
| 2   | 11.43       | 35.46           | 33.12    | 11.74           |
| 3   | 7.64        | 35.46           | 33.44    | 11.86           |
| 4   | 9.30        | 35.46           | 28.97    | 10.27           |
| 5   | 7.95        | 35.46           | 30.22    | 10.72           |
| 6   | 10.20       | 35.46           | 33.02    | 11.71           |
| 7   | 9.05        | 35.46           | 25.75    | 9.13            |
| Ave | 9.94        | 35.46           | 32.22    | 11.42           |
with ten optical fibers. The slope of the solid line represents the ratio of bonding strengths of UV-curable resin and epoxy resin. The measured values plot an almost straight line, where the repair rate improves in proportion to the increase in the repaired area as seen in the figure. Thus, we identified a factor that influences the repair rate. The dotted line indicates that complete repair is possible if UV-curable resin having a bonding strength close to that of epoxy resin is used. With the cooperation Kyoei Chemical Co., Ltd., we pursued greater rigidity of the resin by adding plasticizer to UV-curable resin 80MFA. The measured ratio of bonding strengths \((P_{UV[N]} / P_{EP[N]})\) improved to 0.834, and we used this resin as the repair agent. The repair rate with this resin improved considerably along the two-dot broken line (Fig. 14).

6. Concluding remarks

To confirm the feasibility of the proposed self-healing system for CFRP, we prepared CFRP with a self-healing structure and tried self-healing against cracks. The major findings from this study are as follows.

- We developed a mechanism to continuously inject repair agent and successfully repaired cracks without encountering any shortage of the repair agent.
- Cracks can be repaired using optical fibers and UV-curable resin.
- The bonding strengths of the resin of the base material and the repair agent, along with the repaired area, affect the repair rate.

This system measures the delamination between CFRP layers and radiates UV light onto the damaged part, so it is desirable to arrange the optical fibers perpendicular to the gap between CFRP layers.

Based on the above results, we were able to demonstrate the feasibility of the proposed self-healing system for CFRP.

Acknowledgments

This study was conducted using subsidies for scientific research granted by the Ministry of Education, Culture, Sports, Science and Technology (24656084). While conducting this study, we received UV-curable resin as well as useful advice and cooperation from Mr. Kazuya Akanuma and others of Kyoeishi Chemical Co., Ltd. We express our hearty thanks to these persons.

References

[1] Self-Healing Materials Research Society (2003) “The self-repair materials which came to here”, Chap 7-8, Japan Kogyo Chosakai Publishing Co Ltd
[2] White SR, Sottos NR, Moore J, Geubelle P, Kessler M, Brown E, Suresh S, Viswanathan S (2001) Nature, 409, 794-797
[3] Kessler MR, White SR (2001) Composites Part A, 32, 683-699
[4] Bao L, Kemmochi K (2007) Mechanical materials and material processing technology proceedings, 15, 351-352
[5] Williams GJ, Trask RS, Bond IP (2007) Composites Part A, 38, 1525-1532
[6] Arao Y, BAO L, Kemmochi K (2011) Japan Society of Mechanical Engineers academic conference processing, 701-702
[7] Takeda N (2009) Japan Society of Mechanical Engineers, 112, 544-547
[8] Bao L, Akahane K, Kemmochi K (2012) Adv Compos Mater, 21,133-145
[9] Akamatsu K (2002) “Photopolymer”, pp450-510, CMC Publishing Co Ltd
