Journal of Solid Mechanics and Materials Engineering
Vol. 4, No. 6, 2010

Durability Estimates of Copper Plated Electrodes for Self-sensing CFRP Composites*

Akira TODOROKI**, Kensuke SUZUKI***, Yoshihiro MIZUTANI****
and Ryosuke MATSUZAKI****
**Department of Mechanical Sciences and Engineering, Tokyo Institute of Technology
2-12-1 Ookayama, Meguro-ku, Tokyo, Japan
E-mail:atodorok@ginza.mes.titech.ac.jp
***Tokyo Institute of Technology
****Department of Mechanical Sciences and Engineering, Tokyo Institute of Technology
2-12-1 Ookayama, Meguro-ku, Tokyo, Japan

Abstract
This study deals with the durability tests of a new copper plated electrode process for self-sensing or self-healing Carbon Fiber Reinforced Plastic (CFRP) composites. The self-sensing or self-healing CFRP composites adopt carbon fibers as sensors or actuators. The electrical contacts at the electrodes have a significant effect on the self-sensing or self-healing ability. This research proposes a new electric copper plated process for fabricating reliable electrodes. Using single-ply specimens, electrical contact resistances after cyclic loading are measured. The cyclic loading test is performed to investigate the long term durability of these new electrodes. The new copper plated electrode was found to have high durability after cyclic loading and is appropriate for the practical self-sensing or self-healing CFRP composites.

Key words: Composites, Cyclic Loading, Durability, CFRP, Electrical Resistance

1. Introduction

Carbon Fiber Reinforced Plastics (CFRP) composites have been applied to many aerospace structures and components of transportation vehicles. Laminated CFRP composites made by stacking unidirectional prepreg sheets are widely used for the structural components. The laminated CFRP, however, has invisible defects such as delamination cracks and matrix cracking. For the detection of these defects, a self-sensing method has attracted attention in recent times. The self-sensing method adopts carbon fibers as sensors. In previous studies, the electrical resistance of the target CFRP components has been measured, and damage was detected as increases in electrical resistance [(1)-(8)]. The self-sensing method does not cause strength reduction and the method can be applied to existing structures with the simple construction of electrodes on the surface of the CFRP components. For self-sensing CFRP composites, it is important to make durable electrodes.

Park et al., proposed self-healing composites using mendable polymer and resistive heating of carbon fiber composites [(9)]. For self-healing composites, carbon fibers are used as sensors to detect the damaged area, and the fibers are used again as actuators to heat the target part of the CFRP composites for activation of the mending process. For the self-healing composites, the fabrication process for making durable electrodes is one of the significant issues.

Silver paste or conductive epoxy is the most commonly adopted technique for making electrical contact between a lead wire and the carbon fibers. The material, however, is weak and depends on the techniques of the workers, although the material does enable us to make

*Received 26 Mar., 2010 (No. 10-0127)
[DOI: 10.1299/jmmp.4.610]

electrical contact without complicated fabrication processes. The authors have adopted a copper plating method for making electrical contacts with carbon fibers (10). The copper plating method enables us to make electrical contacts without the dependence on individual techniques and with small variance. The durability of the copper plating electrodes, however, has not been investigated for the CFRP components.

This study focuses on the electrical copper plated electrodes for self-sensing or self-healing CFRP composites. Even for the copper plated electrodes, lead wires must be attached to measure electrical resistances. When the lead wires are soldered, the heat of soldering causes debonding between the copper and carbon fibers. This study proposes a new integrated wire copper plating method to avoid the debonding caused by soldering. Tensile and cyclic loading tests are performed. The contact resistance of the copper plating method is measured before and after cyclic loading to confirm its durability for practical self-sensing or self-healing systems.

2. New copper plating electrode

Figure 1 shows the cross-section of the previous copper plating method described in reference (10). In the previous study, the process for making copper plating electrodes was as follows:

(1) Polish the target CFRP laminate surface with rough sandpaper #240.
(2) Mask everything but the target area with vinyl chloride tape.
(3) Remove sizing around carbon fibers, using several drops of concentrated sulfuric acid.
(4) Clean the specimen surface.
(5) Polish the surface with fine (#400) sandpaper.
(6) Use silver paste to make electrical contact.
(7) Perform the first electrical copper plating at low electric current density (0.66 A dm^{-2}) in a solution of copper sulfate for 30 minutes.
(8) Perform the second copper plating at high electric current density (2.0 A dm^{-2}) for 30 minutes.
(9) Clean the specimen.
(10) Dry the specimen.
(11) Bond lead wire by soldering.
(12) Cover the electrodes with epoxy adhesive.
(13) Post-cure treatment for the epoxy adhesive.

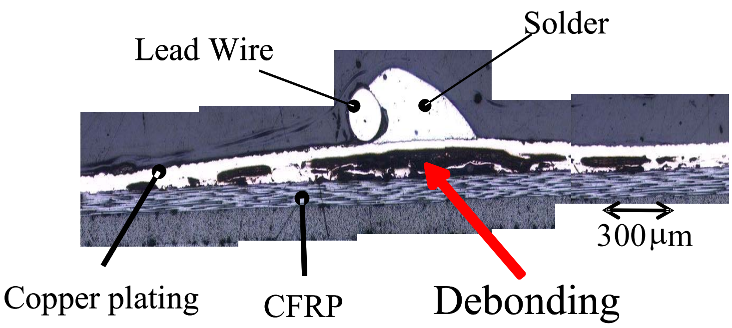


Fig.1 Cross-section of the electrodes made by previous method

As shown in Fig.1, debonding between the copper plating and carbon fibers is observed under the soldered area. This debonding may affect the measured electrical resistance after cyclic loading. Since the debonding occurs because of the thermal stress induced by the soldering, a new method to prevent debonding is proposed here. To prevent the use of soldering, a lead wire is integrated with the copper plating. After the second copper plating, a copper lead wire is placed on the surface of the electrode. A small part of the lead wire is attached to the copper electrode using epoxy adhesive. After curing the adhesive, the set of the lead wire and the specimen is placed again in a solution of copper sulfate. A small electric current density of 0.66 A dm^{-2} is applied first for two hours, followed by a large electric current density of 2.0 A dm^{-2} for 90 minutes. This process creates an integrated lead wire without debonding as shown in Fig.2. After making the integrated electrodes, the new electrodes are coated with epoxy resin to protect them from any handling damage. As shown in Fig.2, no debonding is observed under the lead wire.

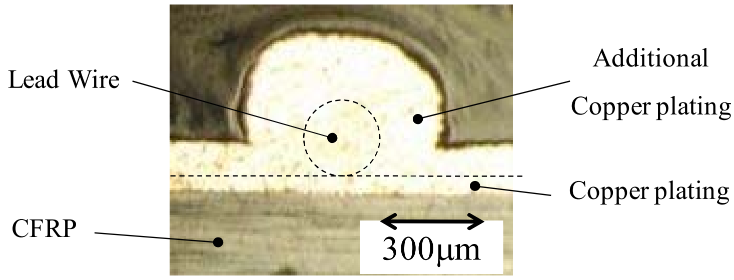


Fig.2 Cross-section view of the new copper plating electrode

3. Specimens and experimental method

The material used is unidirectional CFRP: Toray T300/3601 prepreg # P3060-15. The cure condition of the prepreg is 180°C×0.7MPa×6hr. Two types of specimens are used. The first type is a single ply unidirectional specimen. This type is used to measure the electrical contact resistance of the electrodes. The second type of specimen is a four-ply unidirectional specimen usually used for tensile tests and long cyclic loading tests.

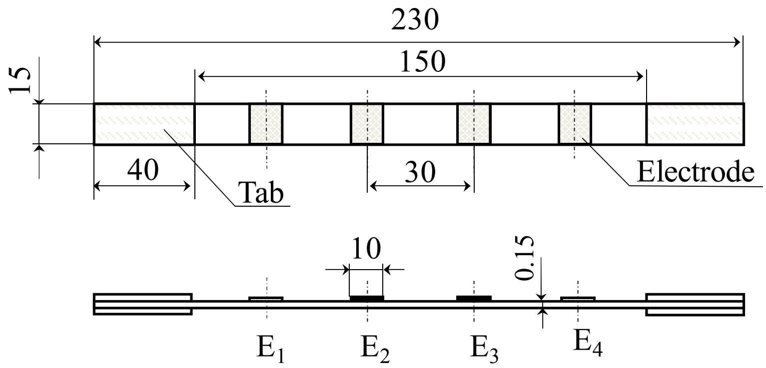


Fig.3 Configuration of a single-ply specimen

The single ply specimen is shown in Fig.3. This specimen is used to measure the contact resistance of the electrodes using the two-probe method. CFRP laminates have a resin rich layer between the plies. The resin rich layer has very low conductivity across the thickness, and a non-uniform electric current may be obtained for the thick laminated CFRP. The single-ply CFRP does not have the resin rich layer. Since the specimen thickness is only 0.15 mm, the electrodes of 10 mm are long enough to make the electric current uniform across the thickness even for the strong orthotropic conductive materials. The measured electric conductivity ratio is $\sigma_{90}/\sigma_0=8.1\times10^{-4}$ from reference [10].

Using the two-probe method, six patterns of electrical resistance can be measured: E_1-E_2, E_2-E_3, E_3-E_4, E_1-E_3, E_2-E_4, E_1-E_4. The first three patterns are the measurements of the individual segments between the electrodes. The next two patterns are the measurements of the sets of two segments. The last measurement is the set of three segments. The measurement of electrical resistance with the two-probe method is illustrated in Fig.4.

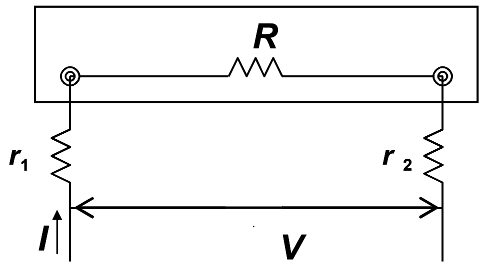


Fig.4 Schematic representation of two-probe method

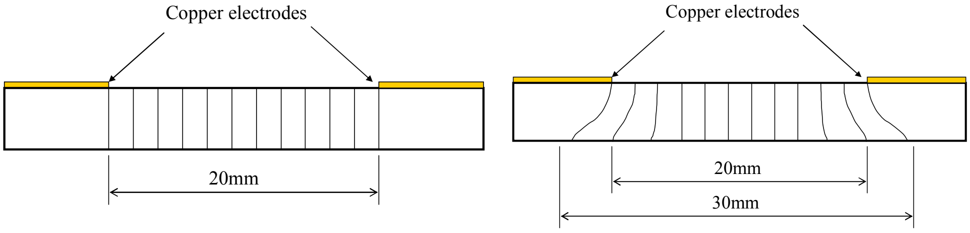
Let R be the electrical resistance of the segment of CFRP, and r_1 and r_2 be the contact resistances of the electrodes. The measurement of the electric current I and voltage change V gives electrical resistance of $V/I=R+r_1+r_2$. The measured electrical resistance includes the two contact resistance of r_1 and r_2. The electrical resistance R of CFRP is expressed as follows:

$$R = \rho \frac{L}{S} \tag{1}$$

Where ρ is resistivity, L is length and S is area of cross section.

When the length L is doubled, the true electrical resistance of the CFRP R doubles. If we assume that the electrical contact resistance at each electrode is almost the same: $r_1+r_2=2r$ (where r is the average contact resistance of each electrode). This enables us to estimate the contact resistance at the electrode. When the measured electrical resistance for contacts of every pattern are plotted against the length L_s (the spacing between the two electrodes), the measured experimental data must exist on a straight line and the initial (constant) value of the straight line at zero spacing is equal to $2r$.

In this study, the spacing L_s is set to 30 mm for E_1-E_2, E_2-E_3 and E_3-E_4. For conventional metallic materials, the spacing between the adjacent electrodes should be 20 mm in this specimen configuration. For the orthotropic CFRP composites, however, a uniform voltage is not obtained at the edge of the electrode. This means the true spacing between the electrodes (uniform electric voltage areas) exists in the area from 20 mm to 30 mm. The illustrated contour plot of electric voltage is shown in Fig.5. This non-uniform electric voltage makes it quite difficult to obtain the exact contact resistance at the electrodes. However, the longer spacing brings a smaller slope to the plots. The smaller slope brings the higher intercept at the perpendicular axis: the higher intercept means the smaller higher resistance. This results in the maximum estimation value of the contact resistance when 30 mm is used as spacing.



(a) Isotropic material (b) Orthotropic material

Fig. 5 Illustrated contour plot of electric voltage between electrodes

After measuring the initial contact resistances for all the cases, tensile stress is applied to the specimen. The maximum stress is 400 MPa, which is approximately 20 % of the tensile strength of the unidirectional CFRP. For the laminated CFRP structures, the compression strength is approximately 70 % of the tensile strength. The most important fracture mode of CFRP structures is a compression after impact (CAI). For the CAI, the fracture strain with visible damage is approximately 30 % of the compression fracture strain of an intact CFRP plate. This means almost 20 % (= 0.7×0.3) of the tensile strength is the practical maximum load of actual CFRP structures. Cyclic loading of 20 % of the tensile strength is applied to the specimen during the measurement of the contact resistance. An LCR meter of Hioki E.E. Corp. Type 3522-50 is used to measure the electric resistances using an alternating current of 30 mA at 450 Hz. Since the phase angle of the alternating current is almost 0°, the measured impedance can be regarded as the electrical resistance. Usually, direct current measurement is affected by floating electric potential, and the measured value is not stable. That is why an alternating current is applied for the measurement of the electric resistance.

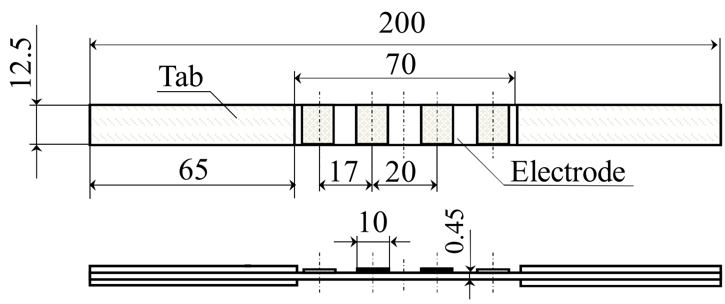


Fig. 6 Specimen configuration of tensile and cyclic loading tests

To confirm the durability of the new electrode, a tensile test and a cyclic loading test of 20 %-tensile strength are performed using a normal laminated unidirectional CFRP specimen. The specimen configuration is shown in Fig.6. A four-probe method is adopted here. The outer two electrodes are used to apply the electric current and the inner two electrodes are used to measure the electric voltage change. The stacking sequence of the specimen is $[0_4]_T$. The curing condition and process for making electrodes are the same as that previously mentioned. Since the tests adopt the four-probe method, partial debonding of the electrodes may not affect the measurement of the electrical resistance.

4. Results and discussion

Figure 7 shows the measured electric resistance before loading. The abscissa is the distance between the two electrodes used for the two-probe method. The ordinate is the measured change in electrical resistance. These plots are approximated by a straight line using the least-square-error method. The linear expression in Fig. 7 defines the relationship obtained, and R^2 is the coefficient of determination. The R^2 is 0.99, which means the linearity of the plots is very good. The y-intercept is 132.59 mΩ. This means the initial contact resistance is 132 mΩ.

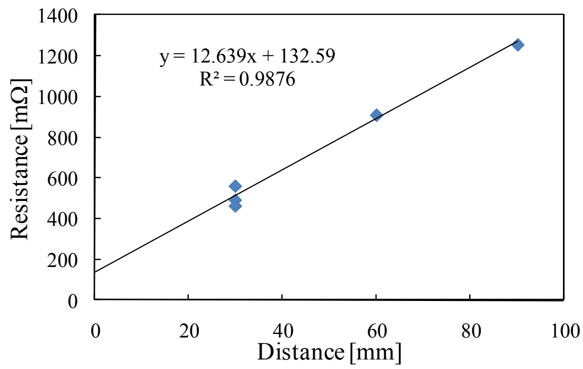


Fig. 7 Resistance values for the various distances for the initial condition

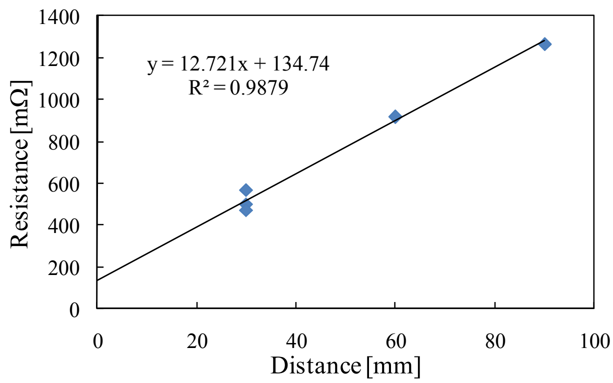


Fig. 8 Resistance values for the various distances after the first cycle loading

Figure 8 shows the measured results of the electrical resistance after the first cycle loading. The R^2 of the line obtained by using the least-square-error method is 0.99. The linearity of the measured data is still very good. The y-intercept is 134 mΩ. Figure 9 shows the results of the electrical resistance after the second cycle loading. The y-intercept is almost the same as that of the initial one.

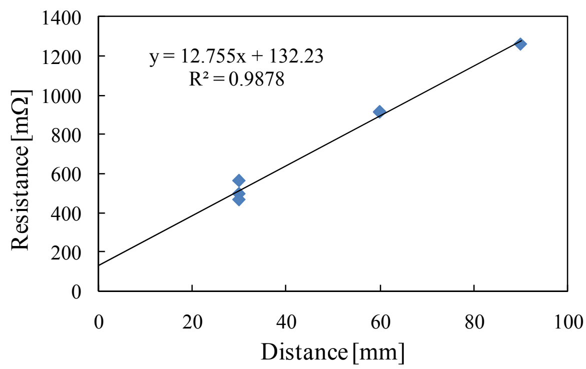


Fig. 9 Resistance values for the various distances after the second cycle loading

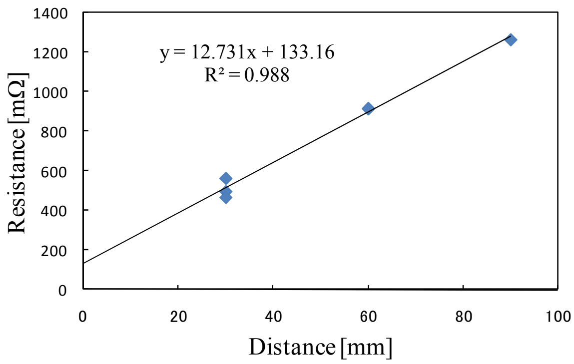


Fig. 10 Resistance values for the various distances after the third cycle loading

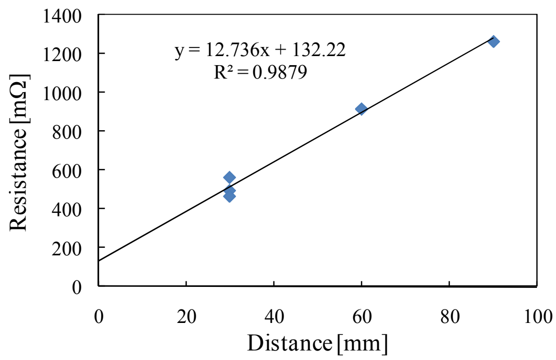


Fig. 11 Resistance values for the various distances after the 100th cycle loading

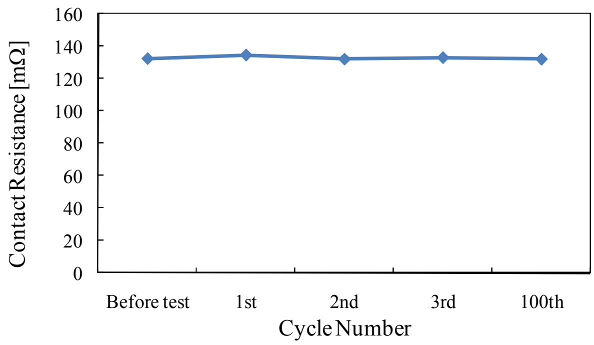


Fig.12 Measured contact resistance after various cyclic loadings

Figures10 and 11 are the measured results obtained after the third and 100th cycle loading. From these results, the contact resistances are calculated from the y-intercept values of the lines obtained with the least-square-error method. Figure 12 shows the results of the calculated contact resistances. The abscissa is the number of cycles and the ordinate is the calculated contact resistances. This figure shows there are no differences between the calculated contact resistances.

Since the contact resistance is calculated from the extrapolations, the calculated values may include experimental errors. To make sure that there is no difference in the results after the cyclic loading a statistical similarity test between the initial linear equation and the linear equation obtained after the cyclic loading is performed using the F-statistic (see reference [11]). The results are listed in Table 1. Since all of the F_0 (similarity test F-values) are smaller than the limit level ($F_{(2,8)}^{0.05}$=4.46) under a significant level of 5 %, all the linear equations are judged to be similar to the initial linear equation. From this statistical test, we can assume that the contact resistance does not change at all.

Table 1 F-test results of contact resistance after cyclic loading (limit level $F_{(1,9)}^{0.05}$=5.12 under significant level of 5 %)

Cycle Number	Contact Resistance [mΩ]	F_0
Before test	132.59	-
1st	134.74	0.087
2nd	132.23	0.066
3rd	133.16	0.060
100th	132.22	0.044

Figure 13 shows the results of the tensile test using the specimen shown in Fig.6. The abscissa is the applied strain. The ordinate of the solid curve is the measured fraction of the electric resistance change, and the ordinate of the broken curve is the applied stress. The stress-strain relationship is almost linear up to the fracture stress (approximately 2400 MPa). As shown in Fig. 13, the fraction of the electrical resistance change increases linearly with the increase of the applied strain up to 4000 μ tensile strain. Over the 4000 μ strain, the fraction of the electrical resistance change increases non-linearly. This increase after 4000 μ of the fraction of electrical resistance change may indicate fiber breakages as shown in the reference [(1)]. Up to the fracture stress, the electrodes do not show a break by visual inspection. Figure 13 shows that the new electrode is quite effective up to the tensile fracture.

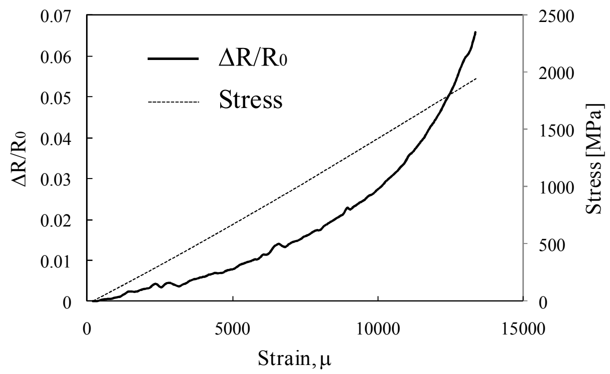


Fig. 13 Fraction of electrical resistance change for the tensile test of $[0_4]_T$.

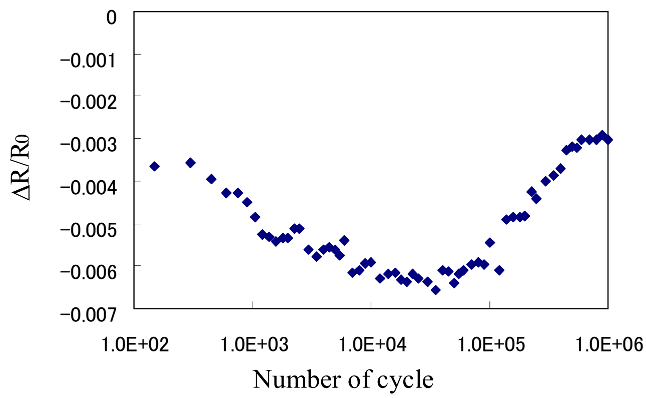
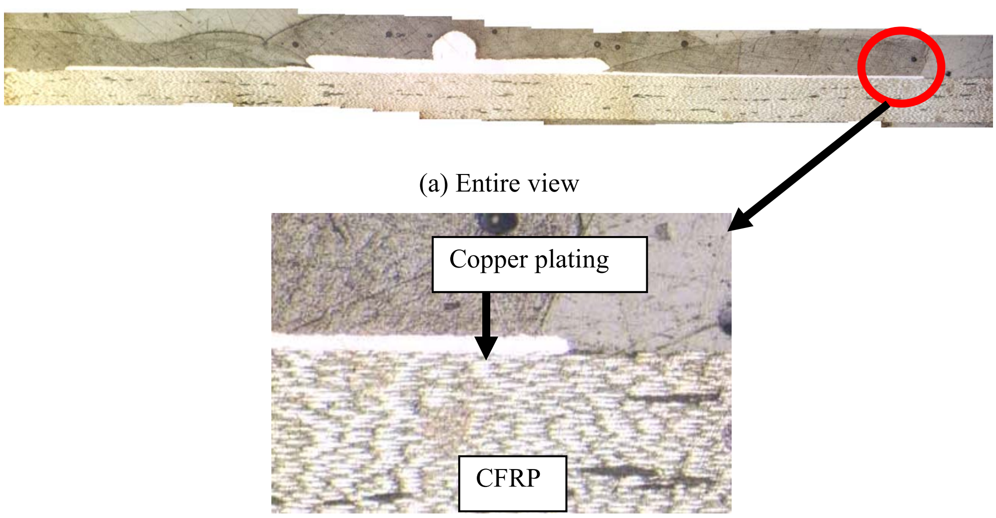


Fig.14 Fraction of electrical resistance change of cyclic loading test of $[0_4]_T$.



(a) Entire view

(b) Cross section at the end of the electrode

Fig. 15 Cross sectional view of electrode after cyclic loading of 10^6 cycles

Figure 14 shows the results of the fraction of electrical resistance change for the cyclic loading test. The abscissa is the number of the cycle, and the ordinate is the fraction of the electrical resistance change. For measurements of the electrical resistance, the cyclic loading test is stopped, and the average tension load is applied to measure the electrical resistance. The measured electrical resistance is normalized by the initial electrical resistance.

Figures 15 (a) and (b) show the cross sectional view of the electrode after the cyclic loading of 10^6 cycles. The entire view of Fig.15 (a) shows there is no debonding after the cyclic loading. The edge of the electrode is shown in Fig. 15 (b). This figure shows there is no debonding even at the edge of the copper plating.

Since the four-probe method is applied to measure the electrical resistance in this test, partial debonding of electrode does not affect the measurements. The decrease of the initial fraction of electrical resistance change may be caused by the increase in the specimen temperature. Reference [12] indicates that the fraction of electrical resistance $\Delta R/R_0$ of CFRP decreases by approximately 0.01 for a 10°C increase in temperature. Since a hydraulic servo testing machine was used here, heat of the hydraulic actuator was conducted into the specimen, and caused a decrease in electrical resistance. The decrease $\Delta R/R_0$ of 0.007 comes from the increase in temperature of 7°C. This slight increase cannot be recognized without using a thermocouple.

Since the applied maximum stress is only 20 % of the tensile strength, the load does not cause breakage of the carbon fibers. Since the measurement is performed by stopping the cyclic loading manually, the temperature may be different at each measurement. There are some voids as shown in Fig.15. The increase in the fraction of the electrical resistance change may indicate the voids affect the increase of electrical resistance change at the high cyclic number. This must be investigated in our future work. We note that, at least, the new electrode does not debond for up to 10^6 cycles of 20 % of the tensile strength. As described before, 20 % of the tensile strength is the maximum load of the actual CFRP structure.

It is useful to know how to calculate the contact resistance for electrodes having different area. Let us assume that all the electrical copper plating has the same thickness L_c and resistivity ρ_c. Equation (1) tells us that the contact resistance of the two electrodes $2r$ can be calculated when we know the value of $2\rho_c L_c=2rA$. For the copper electrode, the value of $2\rho_c L_c=3.96\times10^{-5}$ [Ωm^2]. We can calculate the contact resistance of the copper electrode by dividing the value of $2\rho_c L_c$ by the area A of the electrode.

For compression loading, effect of the shear stress is not different from the obtained results of the present study. In the thickness direction, epoxy coating is done here to prevent debonding. Although experimental researches for the compression loading are required for the practical application to confirm the effectiveness of the electrodes, the method may be effective for the compression loading. This is our future work.

5. Conclusions

This study focuses on the durability tests of a new copper plated electrode. The ability to make a durable electrode is important for self-sensing and self-healing CFRP composites. This research proposes a new electric copper plating method to fabricate the electrodes. Electrical contact resistances after cyclic loading were measured using single-ply specimens. A fatigue test was performed to investigate the long term durability of the new electrodes. The results obtained were as follows:

(1) The new wire-integrated electrode was free from debonding.
(2) The new electrode had very small initial contact resistance.
(3) Even after loading of 100 cycles, the contact resistance of the new electrode kept constant at a low value (the applied maximum stress is 20 % of tensile strength).
(4) Up to the tensile strength, the new electrode was effective in measuring electrical resistance change.
(5) Even after the 10^6 cycles of 20 % of the fracture strength, the new electrode was free from debonding. This indicates the new electrode is suitable for actual self-sensing and self-healing CFRP composites.

References

(1) Schulte K., and Baron Ch., Load and Failure Analyses of CFRP Laminates by Means of Electrical Resistivity Measurements, Comp. Sci Tech, Vol.36, No.1, (1989), pp.63-76.
(2) Muto N., Yanagida H., Nakatsuji T., Sugita M., Ohtsuka, Y., Preventing Fatal Fractures in Carbon-Fibre – Glass-Fibre-Reinforced Plastic Composites by Monitoring Change in Electrical Resistance, J. Amer. Cer. Soc., Vol.76, No.4, (1993), pp.875-879.
(3) Wang X., Chung D.D.L., Continuous carbon fibre epoxy-matrix composite as a sensor of its own strain, Smart Mater. Struct., Vol.5, No.6, (1996), pp.796-800.
(4) Irving P.E., Thiagarajan C., Fatigue Damage Characterization in Carbon Fibre Composite Materials using an Electrical Potential Technique, Smart Mater. Struct.,Vol.7, No.4, (1998), pp.456-466.
(5) Seo D.C., Lee J.J., Damage Detection of CFRP Laminates using Electrical Resistance Measurement and a Neural Network, Comp. Struct.,Vol.47, No.1-4, (1999), pp.525-530.
(6) Abry J.C., Choi Y.K., Chateauminois A, Dalloz B, Giraud G, Salvia M. In-situ Monitoring of Damage in CFRP Laminates by using AC and DC measurements, Comp. Sci. Tech., Vol.61, No.6, (2001),pp.855-864.
(7) Park J.B., Okabe T., Takeda N., Curtin W.A., Electromechanical modeling of unidirectional CFRP composites under tensile loading condition, Composites Part A,Vol.33, No.2, (2002),pp.267-275.
(8) Todoroki A. Tanaka Y., Delamination identification of cross-ply graphite/epoxy composite beams using electric resistance change method, Comp. Sci. & Tech., Vol.62, No.5, (2002) pp.629-639.
(9) Park J.S., Takahashi K., Z. Guo, Wang Y., Bolanos E. Hanann-Schaffner C., Murphy E., Wudl F. and Hahn H.T., Towards development of a self-healing composite using a mendable polymer and resistive heating, J. Comp. Mater., Vol.42, No.26, (2008),

pp.2869-2881.

(10) Todoroki A., Samejima Y., Hirano Y., Matsuzaki R., Mizutani Y., Mechanism of electrical resistance change of a thin CFRP beam after delamination cracking, J. of Solid Mech.& Mater. Eng., JSME, Vol.4, No.1,(2010), pp.1-11

(11) Iwasaki A., Todoroki A., Shimamura Y., Kobayashi H., Unsupervised Structural Damage Diagnostic Method Using Judgment of Change of Response Surface by Statistical Tool (Application for Damage Detection of Composite Structure), JSME International Journal, Series A, 47, 1, (2004) pp.1-7.

(12) Todoroki A., Tanaka Y. and Shimamura Y., Electric resistance Change method for Identification of Embedded Delamination of CFRP Plates
Materials Science Research International, Special Technical Publication-2, JSMS, (2001) pp.139-145.