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Correlation Between Mechanical Behavior and Electrical Resistance Change in Carbon Particle Dispersed Plastic Composite

D-Y. Song [1], N. Takeda [1] and J-K Kim [2]

[1] Graduate School of Frontier Science, The University of Tokyo c/o Komaba Open Laboratory (KOL), The University of Tokyo, 4-6-1 Komaba, Meguro-ku, Tokyo 153-8904, Japan
[2] Dept. of Mechanical Engineering, Hanyang University, 17 Haengdang-dong, Sungdong-ku, Seoul 133-791, Korea

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Abstract

Mechanical behavior and electrical resistance change of CPDP (carbon particle dispersed plastic) composite consisting of epoxy resin and conductive carbon particle were investigated under monotonic loading and repeated loading-unloading. The electrical resistance almost linearly increased with increasing strain during loading and the residual electrical resistance was observed even after removing load. The value of the residual electrical resistance was dependent on the maximum strain under the applied stress. This result suggests that the estimation of maximum strain (i.e., damage) is possible by the measuring electrical resistance of composite. The behavior of electrical resistance change during and after loading was discussed on the basis of the results of microscopic deformation and fracture observation. Moreover, the relationship between the volume fraction of carbon particle and the electrical resistivity of CPDP was investigated in relation to the percolation theory. Simulation model of percolation structure was established by Monte Carlo method and the simulation result was compared to the experimental results. The electrical resistance change under applied loading was analyzed quantitatively using the percolation equation and a simple model for the critical volume fraction of carbon particle as a function of the mechanical stress. It was revealed that the prediction was in good agreement with the experimental result except in the region near the failure of material.

1. Introduction

The electrical resistance method is expected to be effective for foreseeing damage and preventing fatal fracture of composite material structure. Recently, the relationship between the mechanical parameters (i.e., stress and strain) and the changes in electrical resistance of CFRP composite has been experimentally demonstrated [1-2]. Moreover, the damage detection utilizing the percolation structure of conductive particle instead of carbon fiber has been made [3], but this study was limited to the qualitative evaluation, and the systematic evaluation with regard to the cor relation between mechanical behavior and electrical resistance change is not yet made. In the present study, the relationship between mechanical behavior and the change of electrical resistance for CPDP (carbon particle dispersed plastic) composite was evaluated using both experimental and theoretical methods. To this end, the monotonic loading and repeated loading-unloading tensile tests were carried out using CPDP consisted of epoxy resin and conductive carbon particle, and its microscopic deformation and fracture were observed. Moreover, the relationship between the volume fraction of carbon particle and the electrical resistivity of

CPDP was investigated in relation to the percolation theory. Simulation model of percolation structure was established by Monte Carlo method and this result was compared to the experimental results. Finally, the electrical resistance cha nge under applied load was analyzed quantitatively using the percolation equation and a simple model for the critical volume fraction of carbon particle as a function of the mechanical stress (strain).

2. Experimental Procedure

The materials used were the conducting carbon particle with the flake shape of average size 5μm and an insulting epoxy matrix (Epikote 828 Yuka Shell Co.). Specimens were prepared as follows: firstly, carbon particle and epoxy resin were mixed in a given proportion and the pore in this mixture was removed under vacuum. Then, the mixture was injected into the teflon-made mold of dumbbell shape and cured in the electrical furnace under the condition of precure at 50°C, 80min, and aftercure at 100°C, 60min (hereafter, this composite was referred to CPDP). The electrodes for measuring electrical resistance were attached on the opposite surfaces, which were first polished matrix resin layer to achieve electrical contacts, within the gage length of specimen using silver paste. The dimensions of specimens were 105mm in length, 10mm in width and 1mm in thickness. Tensile tests were conducted at the crosshead speed of 0.5mm/min. The electrical resistance was measured using two-probe DC method. A constant current of 0.1μA was applied on the specimen, and the changes in electrical resistance under loading and unloading were measured simultaneously with stress and strain. Microscopic deformation on specimen surfaces was observed using polarization microscope under loading and fracture surfaces were also observed using scanning electron microscope (SEM) after tensile test.

3. Experimental Results and Discussion

Fig. 1 shows the electrical resistance change/strain and stress/strain curves for CPDP with 10vol.% carbon particle. The change in electrical resistance exhibited nearly linear behavior up to the final failure of specimen with increasing strain. In particular, CPDP indicated the change in electrical resistance from a considerably small strain level compared to the case of

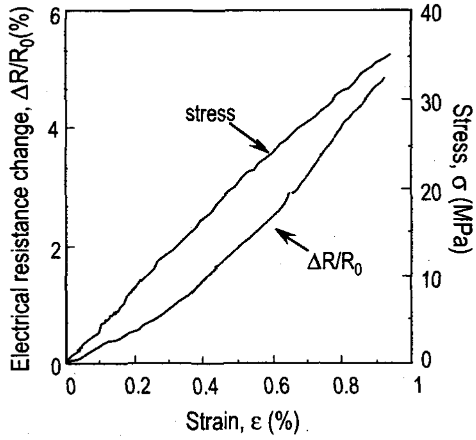


Fig. 1 Relations of $\Delta R/R_0$ vs. ε and σ vs. ε for CPDP with 10 vol. % carbon particle.

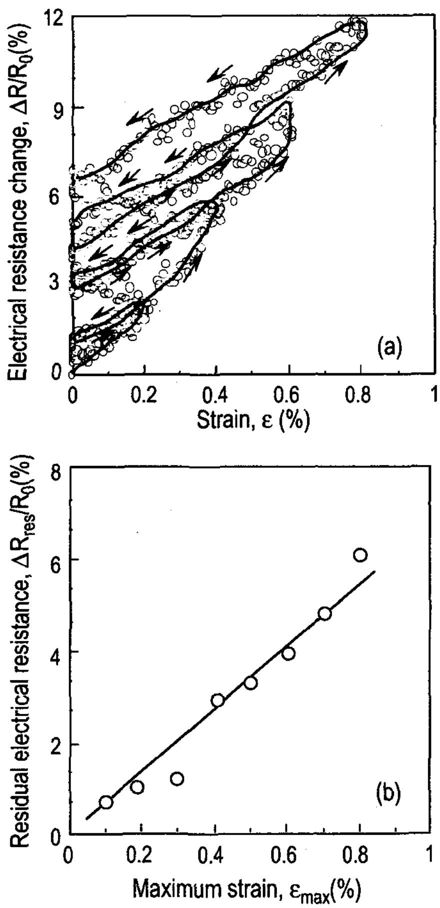


Fig. 2 Relations of (a) $\Delta R/R_0$ vs. ε and (b) $\Delta R_{res}/R_0$ vs. ε_{max} for CPDP with 10 vol. % carbon particle.

CFRP showing electrical resistance change due to the fracture of carbon fiber mainly. This was the reason why the percolation structure formed with carbon particle showed the sensitive response (i.e., the change of conduction path) against applied loading. Fig. 2(a) shows theelectrical resistance change/strain curve for CPDP with 15vol.% carbon particle obtained from repeated loading and unloading test. It was seen that the change in electrical resistance was left after unloading at a relatively small 0.2% strain and increases with increasing strain. Fig. 2(b) shows the relation between maximum strain applied in the past and residual electrical resistance. The value of residual resistance was dependent on the maximum strain under the applied load. This means that the CPDP has the ability to memorize the maximum strain applied in the past as a residual electrical resistance. This change in electrical resistance under applied load is considered to be caused by the rearrangement of carbon particles with percolation structure due to the microdeformation and cracking of matrix. Fig. 3 shows a SEM micrograph of a fracture surface of the CPDP specimen with 15vol.% carbon particle. It was seen that matrix cracking and plastic deformation were significantly observed on the fracture surface, but carbon particles were not observed on the fracture surface. This can be considered that most of carbon particles dropped out when the final fracture of specimen occurred because of the weak adhesion between particle and matrix. Fig. 4 shows a polarization micrograph for the surface of CPDP specimen with 0.1vol.% carbon particle. The plastic deformation of matrix surrounding carbon particles wasclearly observed as indicated by arrows in this figure. Therefore, the existence of residual electrical resistance as described above is considered to be attributed to this plastic deformation of matrix, leading to the change of geometrical arrangement of carbon particle from its initial state (i.e., the change of conduction path).

Fig. 3 SEM micrograph of fracture surface of specimen with 15 vol. % carbon particle.

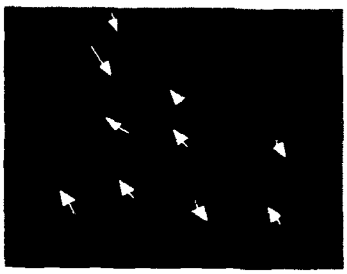


Fig. 4 Polarization micrograph of specimen with 0.1 vol. % carbon particle.

4.Theoretical Analysis of Electromechanical Property of CPDP

4.1. Percolation theory

The property of composite system conposed of conductive carbon particle dispersed in insulting polymer matrix are explained as percolation phenomena i.e., when the amount of carbon particle is sufficiently high, the composite transforms from an insulator to a conductor as the result of continuous contacts between

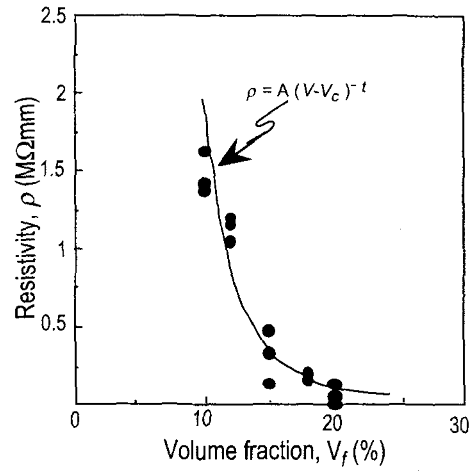


Fig. 5 Relations of ρ vs. V_f for 5 μm carbon particle.

carbon particles (i.e., percolation) within the polymermatrix. Generally, close to the percolation threshold, the concentration dependence of resistivity in a conductor-insulator composite has been well expressed with percolation theory [4-6].

$$\rho(V) = A(V - V_c)^{-t} \tag{1}$$

where, V_c is the critical carbon volume fraction required to form a continuous conduction path, $\rho(V)$ is the resistivity at carbon volume fraction,V, A and t are the constants. According to literature [7-9], t was found to exist between 1.65 and 2.0 for three dimensional system. Fig.5 shows the variation of resistivity with volume fraction of 5 μm carbon particle. It is seen that the resistivity decrease abruptly when the carbon volume fraction exceeds a critical value. This suggests that the percolation theory can hold in the present experimental results. The constants in Eq. (1) were obtained using a least square fit to the experimental data. The obtained values are V_c= 0.065, t = 1.9, A = 0.003 MΩmm The curve calculated with these values is also plotted in Fig. 5, showing a good agreement with experiments.

4.2. Simulation model of percolation structure

A simulation model of percolation structure was established by Monte Carlo method. Here, two dimensional random array problem was simulated as follows: At first, the sites of n =1000, which correspond to carbon particles, were randomly arranged in a square of side unity. In order to satisfy the normal distribution with the standard deviation of 0.2R, the circles of the radius, $R=1/(\pi n)^{0.5}$ were randomly assigned to each site. Next, for the constructed sites, we examined whether the conduction path between any two sites was formed. Here, we firstly introduced the distance between sites as follows:

$$d_{ij} = [(x_i - x_j)^2 + (y_i - y_j)^2]^{0.5} \tag{2}$$

where, x_i and y_i, x_j and y_j are the coordinates at sites i and j. Then, setting $R_{ij} = R_i + R_j$, the conduction path between sites could judge by the following equation:

$$H(x) = R_{ij} - d_{ij} \tag{3}$$

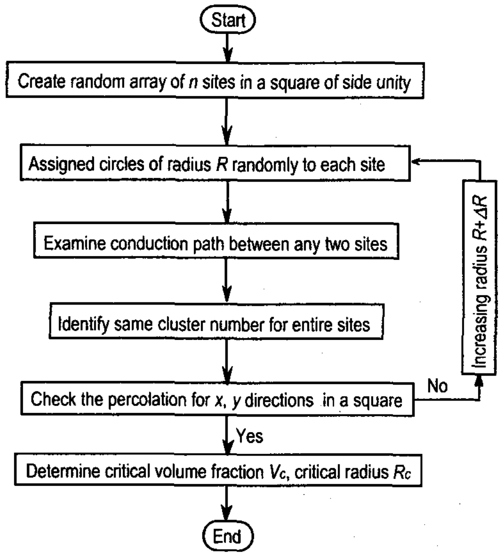


Fig. 6 Flow chart of simulation.

where, $H(x)$ is Heaviside function ($H(x)$ =0: $x < 0$, $H(x)$ =1: $x \geqq 0$). Thus, $H(x)$ = 1 is the formation condition of conduction path between sites. The condition that two sites exists in the same cluster connected by conduction path was given by

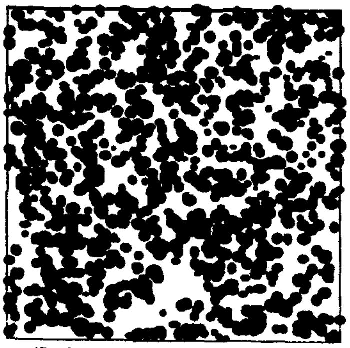
$$B_{ij} = \prod_{a=1}^{A} H_a(x) = 1 \tag{4}$$

where, A is the number of Heaviside functions between sites i and j.

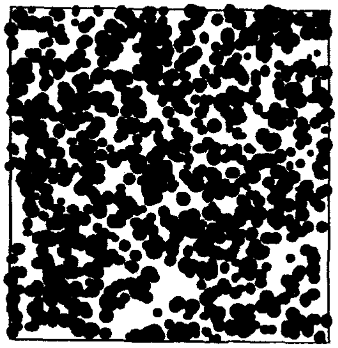
Thus, as the check of the overall percolation in the square, when at least one site having the same cluster number exists at the edges of both directions of x and y in a square (i.e., spanning state), we regard this state as the starting point of percolation. However, during the entire conduction path was not identified, the same way as described above was repeated until the percolation reaches as the radius R increases at the rate of 0.05R. The radius of site at percolation state was referred to the critical radius R_c. This simulation procedure is summarized in Fig.6.

4.3. Simulation result

The process of percolation formation obtained by two dimensional simulation is shown in Fig. 7. The critical volume fraction, V_c obtained from the simulation



(Before percolation, $V_f \cong 40\%$)



(At percolation, $V_f \cong 48\%$)

Fig. 7 Process of percolation formation.

was about 48vol.%. This value is veryhigh compared to the value of about 7vol.% obtained from the experimental results. This difference can be explained by the tunneling effect of particle. For conductive particle-polymer composites, it has been recognized that although the particles were not completely interconnected, the conductive phase was activated and the electrical network was formed (that is, tunneling effect) [10-12]. Considering such a tunneling effect, it can be seen that the site area of this simulation is not the area of particle but corresponds to the region affected by the tunneling effect. Thus, it was simply assumed that the tunneling effect existed to a certain distance on the particle size. That is, for the particle of radius R, if the tunneling effect was effective to the range of kR, the value of k, which might be a constant representing the extent of tunneling effect, was found to be about 2.71 using the average critical volume fraction of 48vol.%. Thus, it can be said that the tunneling effect existed to the distance of about 8.3μm from the surfaces of particles for the particle of average diameter 5μm.

4.4. Approximate analysis of electrical resistance change under applied load

The change of electrical resistance of CPDP with tensile loading will be developed using Eq. (1). Here, the critical volume fraction, V_c, as a first approximation, was assumed to change linearly with applied stress. Moreover, in the case of conductive particle-polymer composite the average number of contacts per particle changes linearly with stress, and was known to be inversely proportional to the critical volume fraction [13, 14]. Thus, the critical volume fraction with stress can be expressed as a simple form as follows:

$$V_c(\sigma) = \frac{V_c(0)}{1 - \alpha\,\sigma} \tag{5}$$

where, $V_c(0)$ was 0.065, the critical volume fraction when the applied stress was absent. This value was obtained from the relationship between the resistivity and the volume fraction described in Fig. 6. α is a constant. Then, substituting Eq. (5) into the percolation Eq. (1), the resistivity and its change of CPDP under applied stress were finally determined from the following Eqs. (6) and (7).

$$\rho = A\left[V - \frac{V_c(0)}{1 - \alpha\,\sigma}\right]^{-t} \tag{6}$$

$$\frac{\Delta\rho}{\rho_0} \approx \frac{\Delta R}{R_0} = \left[\frac{V - \dfrac{V_c(0)}{1-\alpha\,\sigma}}{V - V_c(0)}\right]^{-t} - 1 = \left[\frac{V - \dfrac{V_c(0)}{1-\alpha\,E\,\varepsilon}}{V - V_c(0)}\right]^{-t} - 1 \tag{7}$$

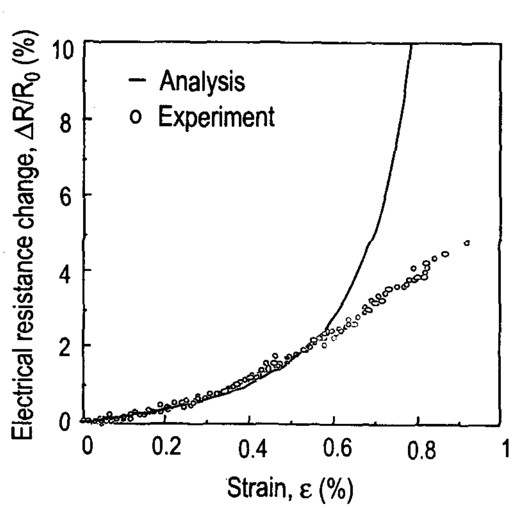


Fig. 8 Comparison between analysis and experimental results.

Fig. 8 shows a comparison between the analysis result by Eq. (7) and experimental result for the CPDP of 5μm carbon particle (in this study the resistivity of CPDP is equivalent to the its resistance, i.e., $\rho \fallingdotseq R$). Here, the constant α was obtained by fitting Eq. (7) to the experimental result and its value was 0.008. The predicted result agreed well with the experimental result below 0.6% strain near the failure of material, where damage and electrical resistance greatly changed. It can be therefore said that the electrical resistance change under applied loading is evaluated approximately using the percolation equation and an appropriate model for the critical volume fraction of carbon particle as a function of the stress (or strain).

5. Conclusions

Mechanical behavior and electrical resistance change for CPDP (carbon particle dispersed plastic) composite were characterized under tensile loading and repeated loading-unloading. The electrical resistance almost linearly increased with increasing strain during loading and the residual electrical resistance was observed even after removing load. The value of the residual electrical resistance was dependent on the maximum strain under the applied stress. These results suggest that estimation of maximum strain (damage state) is possible by measuring electrical resistance of composite. The electrical resistance change during loading was attributed to geometrical rearrangement (i.e., the separation of conduction path) of carbon particles with percolation structure due to the plastic deformation and/or the cracking of matrix. The residual resistance was found to be resulted from the plastic deformation of matrix remaining after removing loading. The relation between the volume fraction of carbon particle and the electrical resistivity of CPDP was well expressed by the percolation equation. Percolation process was simulated by Monte Carlo method. The electrical resistance change of CPDP under applied loading could be approximately analyzed using the percolation equation and a simple model for the critical volume fraction as a function of the stress. This electrical resistance method using the conductive carbon particle is considered to be effective for damage estimation of composite material and structure.

References

1. O. Ceysson et al.,1996, Scripta Mater. 34-8, pp.1273-1280.
2. K. Schulte et al, 1995, Proc. ICCM, pp. 349-395.
3. M. Takada et al., 1999, J. Japan Soc. Comp. Mater., 25, pp. 225-230.
4. D. S. McLachlan et al., 1990, J. Am. Ceram. Soc., 73-8, pp. 2187-2203.
5. S. Kirkpatrik, 1973, Rev. Mod. Phys., 45, pp. 574 - 578.
6. G. E. Pike et al., 1974, Phys. Rev., B10, pp. 1421-1434.
7. B. Abeles et al., 1975, Phys. Rev. Lett., 35, pp. 247-250.
8. F. Carmona et al., 1987, J. Appl. Phys., 61-7, pp. 2550-2557.
9. R. Zallen, 1983, The Physics of Amorphous Solids; Ch.4., Willey New York.
10 I. Balberg, 1987, Phys. Rev. Lett., 59-12, pp. 1305-1308.
11. E. K. Sichel et al., 1978, Phys. Rev. B, 18-10, pp. 5712-5716.
12. G. R. Ruschau et al., 1992, J. Appl. Phys., 72-3, pp. 953-959.
13. J. Gurland, 1966,Trans. Metall. Soc. AIME, 236, pp. 642-646.
14. S. M. Aharoni, 1972, J. Appl. Phys., 43, pp. 2463-2465.