Piezoresistivity in Continuous Carbon Fiber Polymer-Matrix Composite

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Piezoresistivity involving the volume resistivity of a continuous unidirectional carbon fiber epoxy-matrix composite in the fiber direction decreasing reversibly upon tension in the fiber direction was observed by the four-probe method, due to an increase in the degree of fiber alignment. Use of the two-probe method resulted in measurement of the contact resistance rather than the volume resistance. The contact resistance increased reversibly upon tension.

INTRODUCTION

Piezoresistivity refers to the phenomenon in which the volume electrical resistivity of a material changes with strain in the material. It is unrelated to piezoelectricity. It is to be distinguished from the change in electrical resistance (not resistivity) because of dimensional changes during strain. This phenomenon allows the use of the material as a strain sensor, as the measured resistance relates to the strain. Piezoresistivity in a structural material, such as a continuous fiber polymer-matrix composite, is particularly attractive, since the structural material becomes an intrinsically smart material that senses its own strain without the need for embedded or attached strain sensors. Not needing embedded or attached sensors means lower cost, greater durability, larger sensing volume (with the whole structure being able to sense) and absence of mechanical property degradation (which occurs in the case of embedded sensors).

Piezoresistivity has been previously reported in continuous carbon fiber epoxy-matrix composites (1–6), which are important for lightweight structures. Tensile strain in the fiber direction of the composite results in reversible increase in the resistivity in the through-thickness direction (perpendicular to the fiber layers in the composite) (3, 4), as measured by the four-probe method. This is due to the increase in the degree of fiber alignment and the consequent decreased chance of fibers of adjacent layers touching one another. Tensile strain in the fiber direction also results in reversible decrease in the resistance in the fiber direction, as measured by using the four-probe method in which two current (outer) and two voltage (inner) contacts are around the entire perimeter of the composite at four planes that are perpendicular to the fiber direction (1, 2, 4). This was attributed to the increase in the degree of fiber alignment (1, 2, 4), just as the phenomenon observed in the through-thickness direction. However, by using the two-probe method in which the common current/voltage contacts are at the ends of the fibers in the composite, the resistance in the fiber direction was observed to increase reversibly upon tension in the fiber direction (5). Irving and Thiagarajan (5) attributed this phenomenon to the dimensional changes during tension.

The opposite trends described above in the change in resistance in the fiber direction upon tensile strain in the fiber direction (1, 2, 4, 5) are due to the difference in electrical contact configurations, so a study of the effect of electrical contact configuration is needed. The four-probe method (1, 2, 4) is in general better than the two-probe method (5), owing to the elimination of the contact resistance from the measured resistance. Moreover, practical implementation of strain sensing (particularly strain distribution sensing) is more convenient when the contacts do not have to be at the ends of the fibers. However, having the current contacts at the ends of the fibers (5) ensures that current goes through all the fibers. Therefore, this paper extends previous work (1, 2, 4, 5) to provide a systematic comparison of the results obtained on the same composite with four contact configurations, namely (i) four-probe method with all four contacts around the entire perimeter at four planes that are perpendicular to the fiber direction, (ii) four-probe method with two voltage contacts around the entire perimeter at two planes that are perpendicular to the fiber direction and two current contacts at the fiber ends, (iii) two-probe method with both contacts around the entire perimeter at two planes that are perpendicular to the fiber direction, and (iv) two-probe method with both contacts at the fiber ends. The objectives of this work are to clarify the discrepancy and to understand the origins of the observed phenomena.

EXPERIMENTAL METHODS

The composite materials used are the same as those used in Ref. 2. They were constructed from individual layers cut from a 305 mm (12 inch) wide unidirectional carbon fiber prepreg tape manufactured by ICI Fiberite. The product used was Hy-E 1076E, which consisted of a 976 epoxy matrix and 10E carbon fibers. The fiber and matrix properties, which are taken from the manufacturer data sheet, are shown in *Table 1*. The matrix was electrically insulating, whereas the fibers were electrically conducting, with a resistivity of 2.2×10^{-5} Ω.m.

The composite laminates were laid up in a 102 mm × 178 mm (4 inch × 7 inch) platen compression mold with laminate configuration $[0]_8$ (i.e., eight unidirectional fiber layers in the laminate). The individual 102 mm × 178 mm fiber layers were cut from the prepreg tape. The layers were stacked in the mold with a mold release film on the top and bottom of the layup. No liquid mold release was necessary. The density and thickness of the laminate were 1.52 ± 0.01 g cm^{-3} and 1.1 mm, respectively. The volume fraction of carbon fibers in the composite was 58%. The volume resistivity of the laminate in the fiber direction was 4.1×10^{-5} Ω.m, as measured by the four-probe method and silver paint electrical contacts around the perimeter of the sample at four planes perpendicular to the fiber direction. The resistivity calculated by using the Rule of Mixtures was 3.8×10^{-5} Ω.m. That the measured resistivity was higher than the calculated value is attributed to the limited degree of fiber alignment. The laminates were cured using a cycle based on the ICI Fiberite C-5 cure cycle. Curing occurred at 179 ± 6°C (355 ± 10°F) and 0.61 MPa (89 psi) for 120 min. Afterward, they were cut to pieces of size $176 \times 8.9 \times 1.1$ mm. Glass fiber reinforced epoxy end tabs for gripping the sample during subsequent tension were applied to both ends on both sides of each piece, such that the inner edges of the end tabs on the same side were 100 mm apart.

Table 1. Carbon Fiber and Epoxy Matrix Properties (According to ICI Fiberite).

10E—Torayca T-300 (6K) untwisted, UC-309 sized	
Diameter	7 μm
Density	1.76 g cm^{-3}
Electrical resistivity	2.2×10^{-3} Ω.cm
Tensile modulus	221 GPa
Tensile strength	3.1 GPa
976 epoxy	
Process temperature	350°F (177°C)
Maximum service temperature	350°F (177°C) dry 250°F (121°C) wet
Flexural modulus	3.7 GPa
Flexural strength	138 MPa
T_g	232°C
Density	1.28 g cm^{-3}

The electrical resistance R was measured in the fiber (longitudinal) direction while cyclic tension was applied in the same direction. Silver paint was used for electrical contacts.

In the four-probe method, the four probes consisted of two outer current probes and two inner voltage probes. The measured resistance is the sample resistance between the inner probes. In one four-probe contact configuration (2), the four electrical contacts were around the whole perimeter of the sample in four parallel planes that were perpendicular to the fiber direction, such that the inner probes were 45 mm apart. In another four-probe configuration, the current probes were at the end faces containing the fiber ends, such that these faces were flush with the outer edges of the end tabs and the current contacts were not gripped during subsequent tensile testing, while the voltage probes were the same as those in the other four-probe configuration.

In one two-probe configuration, the two contacts were at the end faces containing the fiber ends, such that these faces were flush with the outer edges of the end tabs and the current contacts were not gripped during subsequent tensile testing. In the other two-probe configuration, the two contacts were around the whole perimeter of the sample in four parallel planes that were perpendicular to the fiber direction, such that the contacts were 45 mm apart. In either two-probe configuration, each of these two contacts served as both current and voltage contacts, though separate wires were used for passing current and for voltage measurement at each contact in order to eliminate the resistance of the wires from the measured resistance.

The current-voltage characteristic curves were obtained by the method described in Ref. 7. During the tensile testing, the impedance and phase angle were measured by using a QuadTech 7600 precision RLC meter. A strain gauge of type EA-06-120LZ-120, produced by Measurements Group, Inc., with strain limit 3% and gauge factor 2060 ± 0.5% (at 24°C), was attached to the center of one of the largest opposite faces. A Keithley 2001 multimeter was used for DC resistance measurement. The displacement rate was 0.5 mm min^{-1}. A hydraulic mechanical testing system (MTS 810) was used for cyclic tensile loading in the fiber direction.

RESULTS AND DISCUSSION

The gauge factors and the initial resistances (before loading) for different contact configurations are summarized in *Table 2*. *Figure 1a* shows the change in resistance during cyclic tension for the four-probe configuration in which all contacts were around the whole perimeter of the sample in four parallel planes that were perpendicular to the fiber direction. The resistance decreased reversibly upon tension, as in Ref. 2. *Figure 1b* is the corresponding graph of resistance vs. strain in the first cycle. It shows the hysteretic nature of the relationship. The gauge factor (fractional reversible change in resistance per unit strain) was −23.

Table 2. The Gauge Factor and the Initial Resistances Before Loading for Different Contact Configurations. The Standard Deviations of the Gauge Factors are Shown in Parentheses.

Configuration of Contacts	Gauge Factor*	Initial Resistance (Ω)	
		Measured	Calculated
4-probe, perimeter contacts	−23 (1.9)	0.243	0.19
4-probe, 2 perimeter contacts (voltage) + 2 fiber end contacts (current)	−4.6 (0.55)	0.206	0.19
2-probe, perimeter contacts	+3.0 (0.69)	0.774	0.19
4-probe, fiber end contacts	+8.6 (0.35)	6.79	0.74

*Obtained from regression analysis of the curve of fractional change in resistance vs. strain in the first loading cycle.

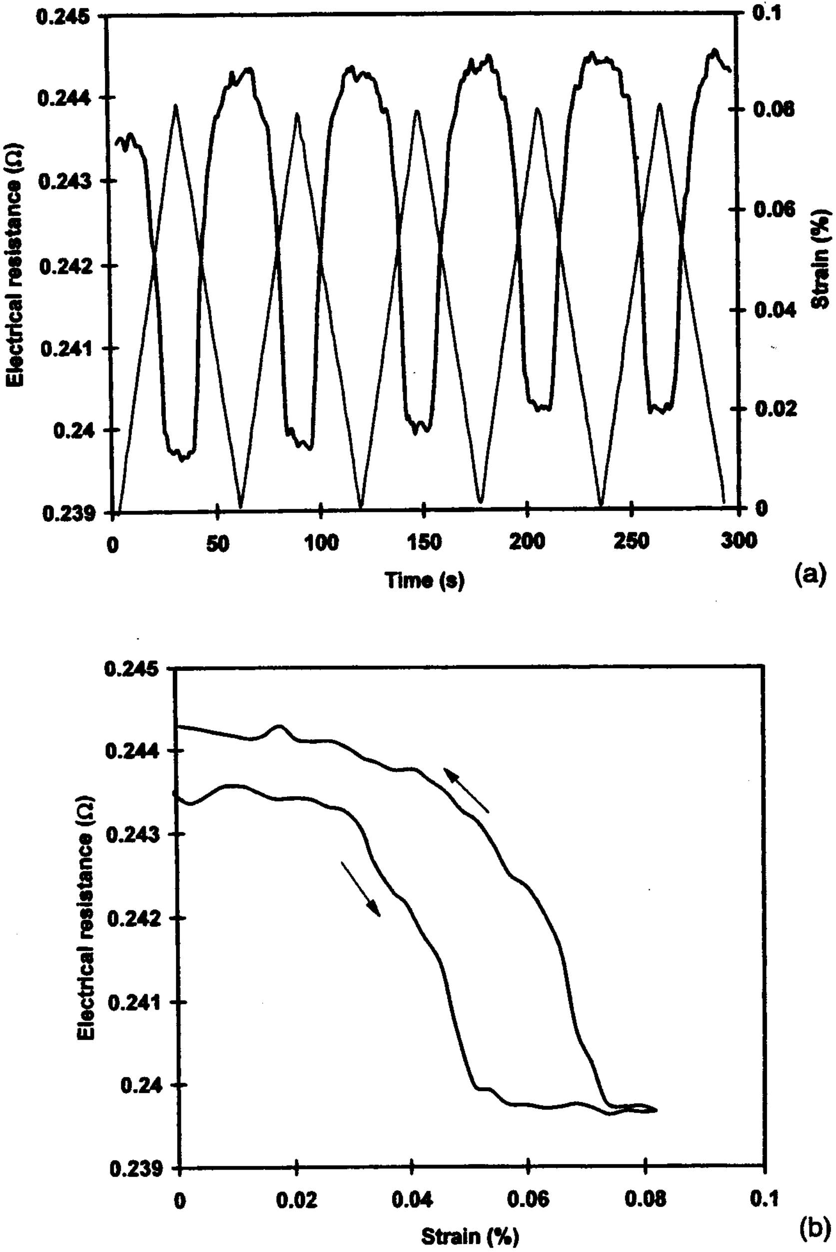

Fig. 1. (a) Plots of resistance (four-probe method with all four contacts perimetric) vs. time and of strain vs. time during cyclic tension. Resistance: thick line. Strain: thin line. (b) Plot of resistance (four-probe method with all four contacts perimetric) vs. strain in the first loading cycle.

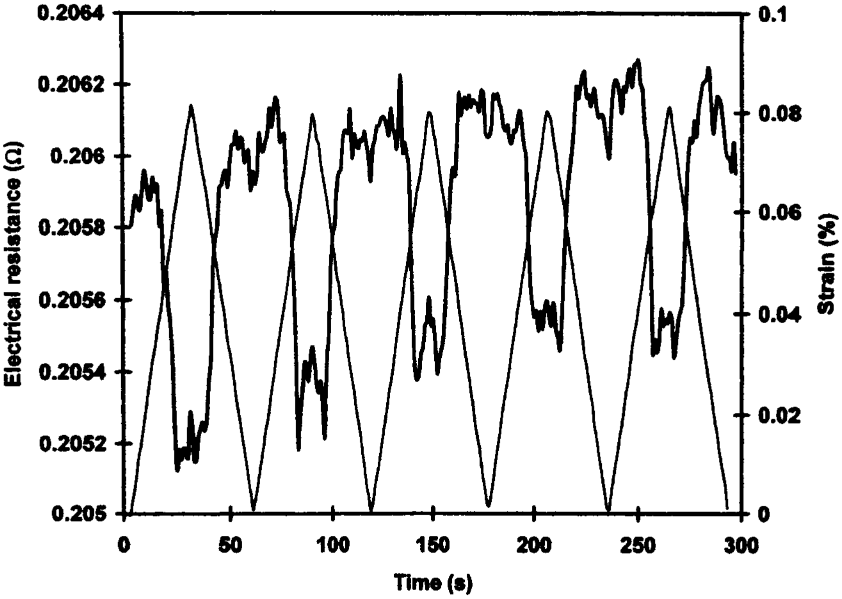

Fig. 2. Plots of resistance (four-probe method with two voltage contacts perimetric and two current contacts at the fiber ends) vs. time and of strain vs. time during cyclic tension. Resistance: thick line. Strain: thin line.

Figure 2 shows the corresponding result for the other four-probe configuration, in which the current contacts were at the fiber ends and the voltage contacts were around the whole perimeter of the sample in two parallel planes that were perpendicular to the fiber direction. The resistance decreased vertically upon tension, as in *Fig. 1*, but the gauge factor was only −4.6, the resistance was smaller (resistance before loading = 0.206 Ω in *Fig. 2*, but was 0.243 Ω in *Fig. 1*) and the resistance change upon loading was more noisy. The higher resistance in *Fig. 1* is attributed to the perimetric current contacts of *Fig. 1* being not able to get the current to penetrate the entire cross-section of the sample evenly, whereas the current contacts at the fiber ends (*Fig. 2*) ensured current penetration throughout the sample cross section.

For both two-probe configurations (*Figs. 3* and *4*), the resistance increased upon tension, in contrast to the opposite trend for both four-probe configurations (*Figs. 1* and *2*). The resistance before loading was 0.774 Ω for *Fig. 3* (perimetric contacts) and 6.79 Ω for *Fig. 4* (contacts at fiber ends). This means that the resistance of the contacts at the fiber ends was much higher than that of the perimetric contacts. The noisiness and the low magnitude of the gauge factor in *Fig. 2* compared to *Fig. 1* are attributed to the high resistance of the contacts at the fiber ends compared to that of the perimetric contacts.

Based on the resistivity of the composite in the fiber direction (4.1×10^{-5} Ω.m), the volume resistance of the whole sample of *Fig. 4* in the fiber direction was calculated to be 0.74 Ω. This means that the high

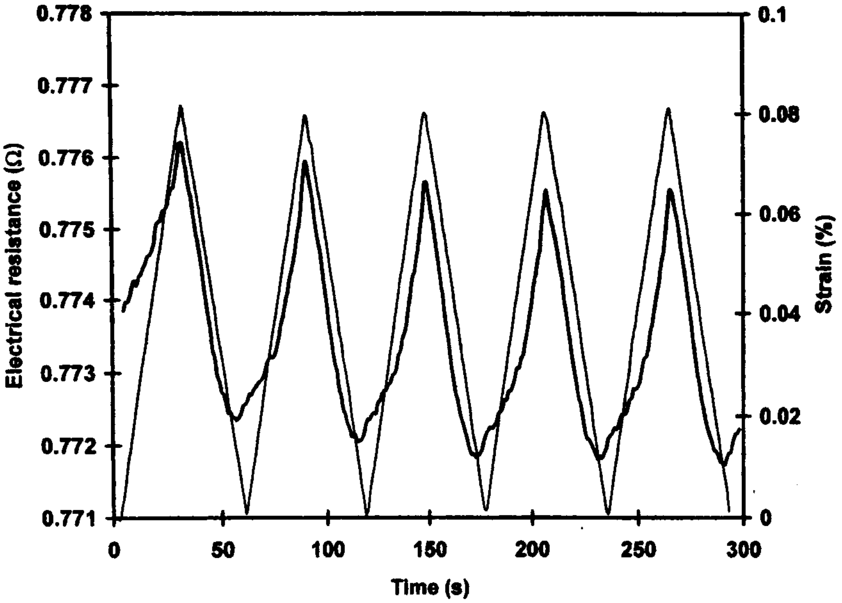

Fig. 3. Plots of resistance (two-probe method with contacts perimetric) vs. time and of strain vs. time during cyclic tension. Resistance: thick line. Strain: thin line.

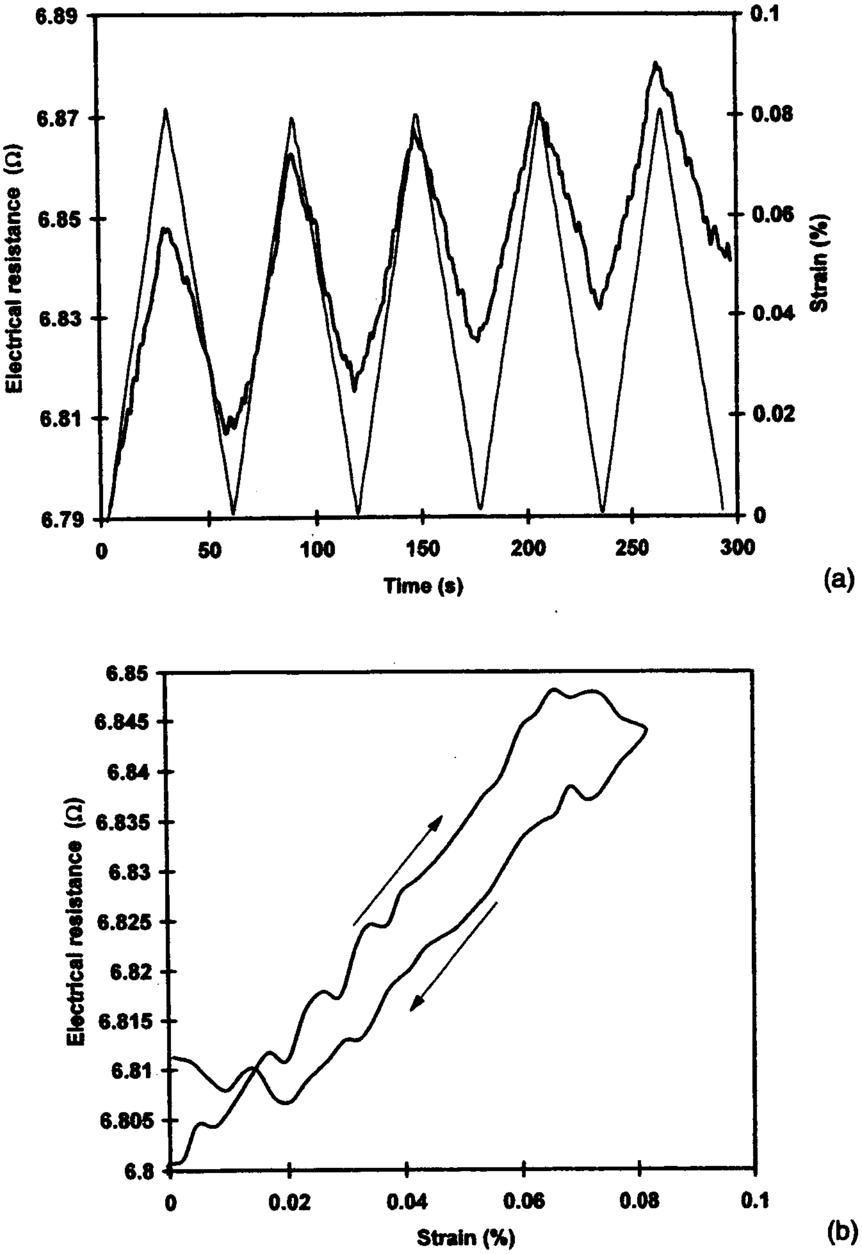

Fig. 4. (a) Plots of resistance (four-probe method with contacts at the fiber ends) and of strain vs. time during cyclic tension. Resistance: thick line. Strain: thin line. (b) Plot of resistance (four-probe method with contacts at the fiber ends) vs. strain in the first loading cycle.

resistance in *Fig. 4* is almost all due to the contact resistance. Based on the resistivity of the composite, the volume resistance of the sample of *Figs. 1–3* between the voltage probes (45 mm apart) was calculated to be 0.19 Ω. This means that the resistance in *Figs. 1* and *2* is essentially the volume resistance of the sample, and the resistance in *Fig. 3* is dominated by the contact resistance. Hence, the four-probe method (*Figs. 1* and *2*) yields the volume resistance, whereas the two-probe method (*Figs. 3* and *4*) yields mainly the contact resistance.

That the resistance before loading was much lower in *Fig. 1* than *Fig. 3* means that the resistance in *Fig. 3* was dominated by the contact resistance. That the resistance before loading was very much lower in *Fig. 2* than *Fig. 4* means that the resistance in *Fig. 4* was much dominated by the contact resistance. That the resistance increased upon tension in *Figs. 3* and *4* is attributed to the contact resistance increasing (i.e., contacts degrading) upon tension. The gauge factor was +3.0 and +8.6 for *Figs. 3* and *4*, respectively. The higher gauge factor in *Fig. 4* compared to *Fig. 3* is

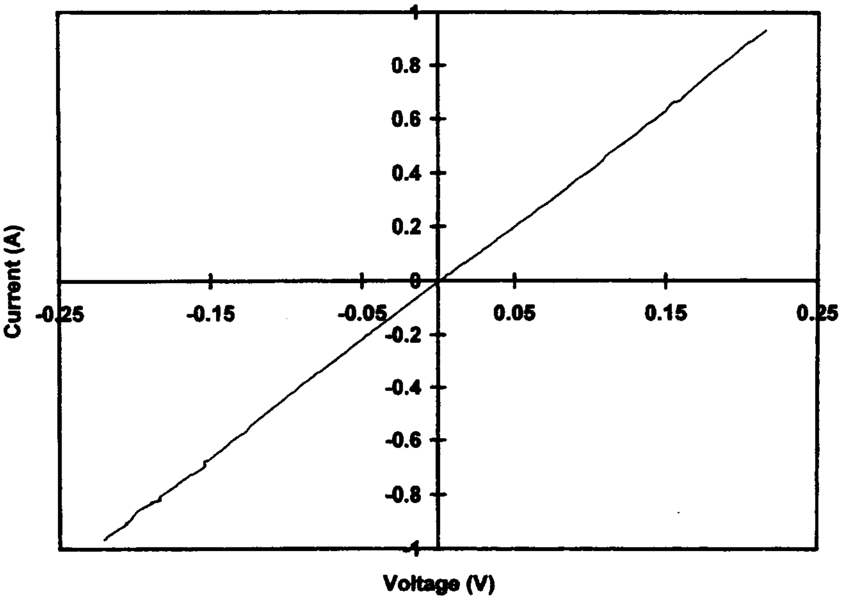

Fig. 5. Plot of current (four-probe method with all four contacts perimetric) vs. voltage under no load.

attributed to the greater dominance of the contact resistance in *Fig. 4* than in *Fig. 3*. The resistance change upon loading was more noisy in *Fig. 4* than *Fig. 3*, due to the noise associated with changes in the quality of the contacts at the fiber ends. The reason is similar to that for the greater noisiness in *Fig. 2* than in *Fig. 1*.

The stress (not shown in *Figs. 1–4*) is linear with strain, such that the strain is totally reversible and is 0.08% at a stress of 140 MPa.

The resistance increase upon tension in *Fig. 4* cannot be just due to dimensional changes (in contrast to the claim in Ref. 5), since the gauge factor would have been only +2 if that were the case. The observed large gauge factor of +8.6 for *Fig. 4* is attributed to the degradation of the electrical contacts during tension and the consequent increase in contact resistance. Although the contacts at the fiber ends were not gripped during tension, slight pull-out of the fibers away from the contact and into the composite probably occurred during tensile loading of the composite, thereby resulting in electrical contact degradation (i.e., contact resistivity increase). This degradation is nearly reversible, as shown by the reversibility of the resistance increase (*Fig. 4*), because of the reversibility of the contact degradation mechanism. Because the resistance increase in *Figs. 3* and *4* is not due to a change in volume resistivity, but a change in contact resistivity, the phenomenon of *Figs. 3* and *4* is not true piezoresistivity.

Although the phenomenon of *Figs. 3* and *4* is not true piezoresistivity, it is still an electromechanical effect. However, this effect is not suitable for use in strain sensing, because the quality of an electrical contact is hard to control in practice (especially in a real structure) and the gauge factor associated with this phenomenon depends on the contact quality, which is reflected by the contact resistance before loading.

The resistance decrease upon tension in *Figs. 1* and *2* is attributed to the increase in the degree of fiber alignment (1, 2, 4) (as supported by the concurrent increase in through-thickness resistance (3)). This interpretation is consistent with the fact that the measured resistivity of the composite in the fiber direction is higher than that calculated by using the Rule of Mixtures. Note that the resistance decrease upon tension cannot be due to an increase in the depth of current penetration, since the through-thickness resistance increases upon tension. An increase in the degree of fiber alignment is expected to decrease the resistivity, because the misaligned fibers may not be at the same potential as the aligned fibers at the same cross-sectional plane perpendicular to the current direction, so that the misaligned fibers may contribute less to conduction than the aligned fibers.

The two-probe method is simpler to implement than the four-probe method. However, the two-probe method and the four-probe method measure different quantities. Therefore, for use of the composite as a strain sensor, the four-probe method is necessary.

The current-voltage characteristic curves for all the four-probe configurations are quite linear over a current range of 0-1 A, as shown in *Fig. 5*. This means that the electrical contacts are basically ohmic.

The A.C. resistance is nearly equal to the magnitude of the impedance and the reactance and phase angle are negligible at frequencies below 1 kHz.

CONCLUSION

Piezoresistivity in continuous unidirectional carbon fiber epoxy-matrix composites was observed upon tension in the fiber direction. The phenomenon involved the volume resistivity of the composite in the fiber direction decreasing reversibly upon tension, due to an increase in the degree of fiber alignment, as observed by using the four-probe method. Use of the two-probe method resulted in measurement of the contact resistance rather than the volume resistance. The contact resistivity increased reversibly upon tension, but the phenomenon is not true piezoresistivity and is not suitable for practical use for strain sensing because of the need to have the electrical contacts at the fiber ends.

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