Consolidation of Carbon Fiber Laminae During Polymer-Matrix Composite Fabrication, Studied by Electrical Resistance Measurement

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The consolidation of carbon fiber epoxy-matrix laminae during composite fabrication by lamination (involving consolidation, curing and cooling) was found to be hastened and to occur to a greater extent by increasing the pressure. The consequence of better consolidation remained after curing and subsequent cooling. A higher pressure resulted in a lower through-thickness resistivity in a composite after curing and cooling. The extent of consolidation was quantitatively described by N/N_o (the ratio of the number of fiber-fiber contacts between adjacent laminae at a given time, divided by that at the start of consolidation), which was calculated from the measured through-thickness electrical resistivity. From the variation of N/N_o during heating in the process of consolidation, three stages of consolidation were observed. In the first stage, N/N_o increased very gradually; in the second stage, N/N_o increased abruptly; in the third stage, N/N_o increased at a moderate rate.

INTRODUCTION

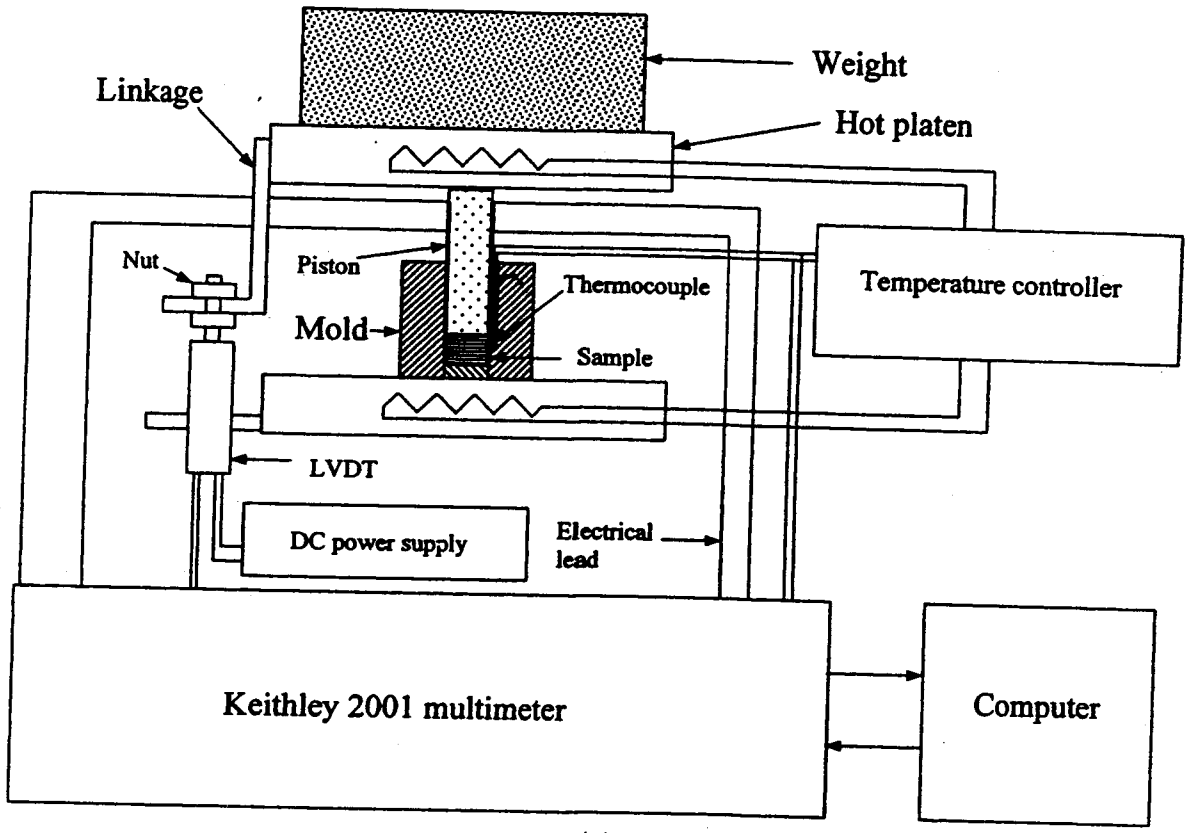
Continuous fiber polymer-matrix composites with thermosetting matrices are important structural materials because of their high strength, high modulus and low density. These composites are commonly made by stacking up layers of fiber prepreg and subsequent consolidation and curing under heat and pressure. Consolidation involves the use of pressure to bring the fiber layers closer to one another and the use of heat to melt the resin in the prepreg, so that the resin flow will allow the layers to come even closer together. A fraction of the resin may be squeezed out during consolidation. Curing occurs subsequent to consolidation and involves the resin's completing its polymerization reaction so that it sets. Curing requires sufficient time and temperature in addition to the recommended pressure for curing. There has been much work on the curing process (1–6), but relatively little work on the consolidation process. As consolidation is an important step in the composite fabrication process, understanding of the consolidation process and characterization of the effectiveness of consolidation are valuable.

During consolidation, the thickness of the prepreg stack decreases. However, thickness change does not provide information on the extent of interaction between the prepreg layers. When the fibers are carbon fibers, which are electrically conducting, the interaction between the prepreg layers leads to contact between fibers of adjacent layers, thereby causing the volume electrical resistivity in the through-thickness direction (direction perpendicular to the plane of the layers) to decrease. Hence, the resistivity provides information on the extent of the fiber-fiber contact. In this paper, this resistivity was measured during consolidation for the purpose of studying the consolidation process in detail. Measurement of the resistivity requires measurement of the resistance as well as the thickness.

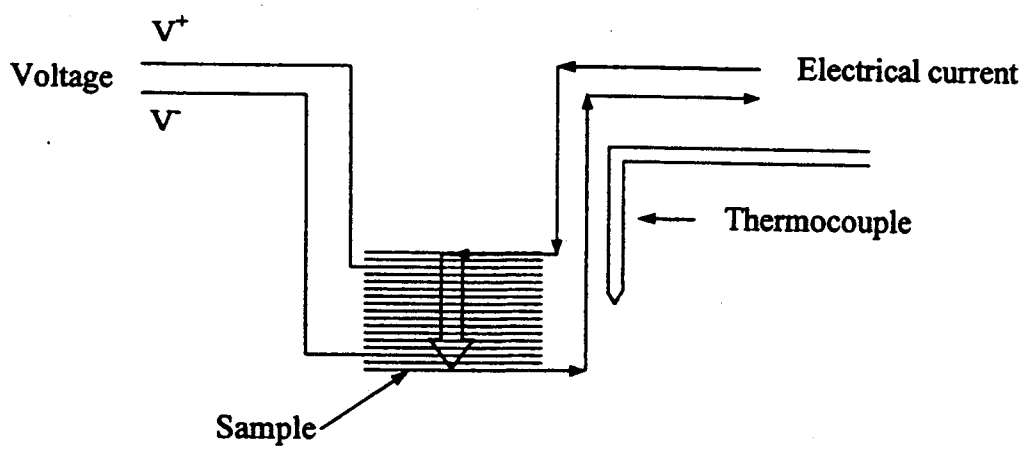
There is prior work on using the through-thickness resistance to indicate delamination in a carbon fiber polymer-matrix composite (7), but there is no prior work on using the resistance or resistivity to indicate the extent of consolidation.

EXPERIMENTAL METHODS

The raw material used to fabricate the samples was a unidirectional carbon-fiber epoxy prepreg, provided by Cape Composites Incorporated (San Diego, CA). The matrix was C2002 series epoxy thermosetting resin in the amount of 37% by weight. The continuous carbon fibers were Type 555 of Fortafil Fibers Inc. (Knoxville, TN); the fibers exhibited tensile strength 3795 MPa, tensile modulus 231 GPa, ultimate elongation 1.7%, density 1.8 g/cm^3, diameter 6.2 μm and no twist.



(a)



(b)

Fig. 1. (a) The experimental setup. (b) The configuration of the four-probe resistance measurement.

Figure 1 is a schematic of the experimental setup. The prepreg was cut into strips that were 10 mm in width (fiber direction) and 100 mm in length (transverse direction). The strips were stacked unidirectionally in a steel mold. Each laminate contained 16 layers (laminae). Four of the layers were 200 mm rather than 10 mm in width (fiber direction) so that they protruded out of the stack to serve as electrical leads (probes). The four protruding layers consisted of the two outermost layers, which served as current probes, and the third layer from the top and the third layer from the bottom, which served as voltage probes (*Fig. 1b*). The epoxy at the tips of the four protruding layers was burned out to expose the carbon fibers for the purpose of making better electrical contacts. These exposed fibers were wrapped by copper foil, with silver paint between the copper and the fibers. A copper wire was soldered on the copper foil at each of the four leads.

The laminate was put into a steel mold lined with a release film to facilitate demolding. The mold was put between the two platens of a Carver hot press, where the sample was cured. A constant pressure (either 0.56 or 1.10 MPa) was provided by the weight of the upper platen plus extra weights put on the top of the upper platen; the pressure was not provided by the hydraulic device of the hot press. The temperature and the heating rate were controlled by a Watlow model 981C-10CA-ARRR temperature controller. Five curing temperatures, namely 100, 110, 120, 130 and 140°C, were used for different samples. All samples were given the same heating rate of 2°C/min, the same constant temperature period of 4 h, and the same cooling rate of 0.37°C/min.

The through-thickness electrical resistance between the two voltage probes and the temperature of the sample were measured respectively by a Keithley 2001 multimeter and a T-type thermocouple, which was placed close to the sample. The thickness change of the sample was measured by an LVDT (Omega, LD600-25), which had been carefully calibrated. The LVDT was fixed on the two platens of the hot press. Through the voltage output of the LVDT, the distance between the two platens was measured. This distance varied with the thickness of the sample. A blank test was performed under the condition of no sample in the mold. The distance with sample present minus the distance with sample absent (blank test) gave the thickness of the sample.

ANALYSIS METHODOLOGY

The through-thickness resistance (R) together with the sample thickness (d) gave the through-thickness resistivity (ρ) according to the equation

$$\rho = R\frac{A}{d}, \tag{1}$$

where A is the area of the sample in the plane of the laminate. This area was assumed to be constant during consolidation and curing.

Since the through-thickness resistance is dominated by the contact resistance between laminae, the resistivity change during consolidation and curing is assumed to be due to the change in the number of contacts between fibers of adjacent laminae. Let the average number of contacts between fibers of two adjacent laminae be m. For a laminate with two laminae, the number of through-thickness conduction paths is m. For a laminate with three laminae, there are two interlaminar interfaces, each having m fiber-fiber contacts. Then the number of through-thickness conduction paths is m^2. In general, if the number of laminae is n, and the number of fiber-fiber contacts between adjacent laminae is m, the number of through-thickness conduction paths is given by

$$N = m^{n-1} \tag{2}$$

The number of conduction paths at the start of consolidation is

$$N_o = m_o^{n-1} \tag{3}$$

The total number of interlaminar contacts in the laminate is $(n-1)m$.

From *Eq 2* and *3*, the ratio of the number of contacts at a certain time during consolidation to that at the start of consolidation is given by

$$\frac{(n-1)m}{(n-1)m_o} = \frac{m}{m_o} = \left(\frac{N}{N_o}\right)^{\frac{1}{(n-1)}} \tag{4}$$

Let the total through-thickness resistance at a certain time during consolidation be R and that at the start of consolidation be R_o. Let the average resistance of each through-thickness conduction path be R_i. Since the various through-thickness conduction paths are electrically equivalent to resistors in parallel, the total resistance R is given by

$$R = \frac{R_i}{N} . \tag{5}$$

At the start of consolidation, the resistance is

$$R = \frac{R_i}{N_o} . \tag{6}$$

The change in resistance during consolidation is given by

$$\Delta R = R - R_o = \frac{R_i}{N} - \frac{R_i}{N_o} . \tag{7}$$

Thus,

$$\frac{\Delta R}{R_o} = \frac{N_o}{N} - 1 \tag{8}$$

or

$$\frac{N}{N_o} = \frac{R_o}{\Delta R + R_o} \tag{9}$$

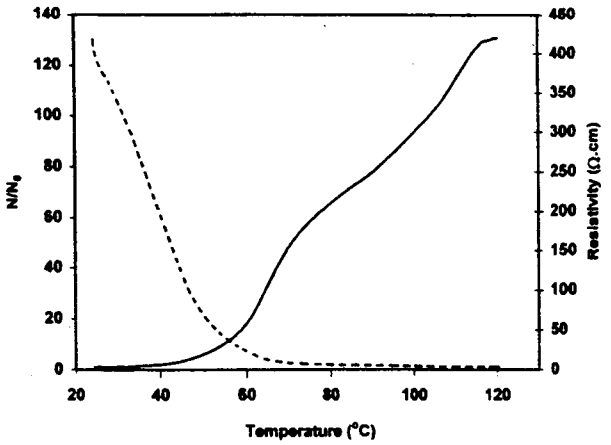


Fig. 2. Variations of N/N_o (solid line) and through-thickness electrical resistivity (dashed line) with temperature during consolidation heating to 120°C at a pressure of 0.56 MPa.

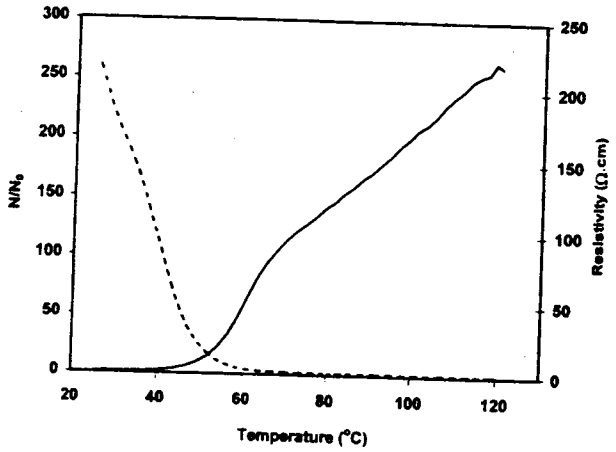


Fig. 3. Variations of N/N_o (solid line) and through-thickness electrical resistivity (dashed line) with temperature during consolidation heating to 120°C at a pressure of 1.10 MPa.

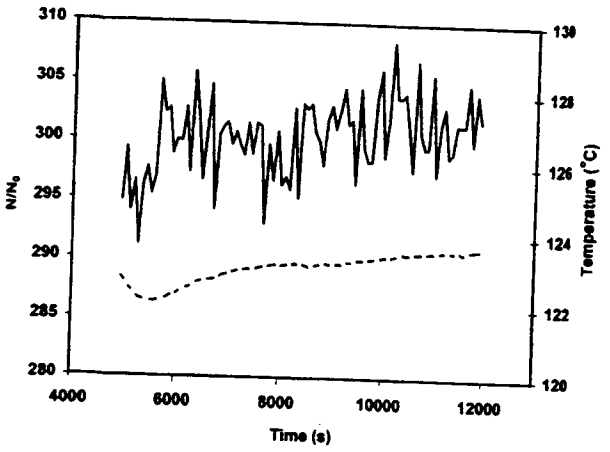


Fig. 4. Variations of N/N_o (solid line) and temperature (dashed line) with time during curing at a pressure of 1.10 MPa.

Hence, N/N_o can be calculated from R_o and ΔR, which was measured during consolidation, using *Eq 9*.

RESULTS AND DISCUSSION

Figure 2 and *3* show the variation of the through-thickness electrical resistivity and N/N_o during consolidation at pressures of 0.56 and 1.10 MPa respectively. During consolidation, the temperature was raised linearly and reached 120°C, which was the curing temperature. In both *Figs. 2* and *3*, the resistivity decreased and N/N_o increased during consolidation, such that the N/N_o curve revealed three stages of consolidation. The first stage was characterized by a very gradual increase in N/N_o (due to the solid form of the resin); the second stage was characterized by an abrupt increase in N/N_o (due to the molten form of the resin); the third stage was characterized by a moderately gradual increase in N/N_o (due to the thickening of the resin as the temperature increased). Similar changes in curvature of the N/N_o plot were observed for consolidation conducted at ramped temperatures that reached 100, 110, 120, 130 and 140°C, although only the results for a maximum temperature of 120°C are shown in *Figs. 2* and *3*. The curve of N/N_o vs. temperature was quantitatively quite independent of the maximum temperature of consolidation, but was quantitatively different for the two pressures, as shown by comparing *Figs. 2* and *3*, which show that, at the higher pressure, (i) N/N_o reached much higher values, (ii) the second stage began and ended at higher temperatures, and (iii) the slope of the N/N_o vs. temperature curve in the second stage was higher (7.18/°C for *Fig. 3* and 3.31/°C for *Fig. 2*). This means that pressure hastened consolidation and promoted the extent of consolidation, as expressed by the quantity N/N_o. In contrast, increasing the maximum temperature did not promote the extent of consolidation.

Figure 4 shows the variation of N/N_o with time in the period of nearly constant temperature (nominally 120°C). The period was for curing, and took place after the consolidation period (*Fig. 3*). The pressure was 1.10 MPa, as in *Fig. 3*. The ratio N/N_o changed very little in the curing period (*Fig. 4*), compared to the large change in the consolidation period (*Fig. 3*). This means that the curing of the resin has little effect on the number of fiber-fiber contacts.

In the cooling period (*Fig. 5*) subsequent to the curing period (*Fig. 4*), at a pressure of 1.10 MPa, N/N_o decreased and the resistivity increased as the temperature decreased (partly owing to thermal stress buildup resulting from the thermal expansion mismatch between carbon fibers and the epoxy matrix), but the variations of both N/N_o and resistivity were small. Similar behavior was observed during cooling at a pressure of 0.56 MPa (*Fig. 6*). This means that the thermal stress has little effect on the number of fiber-fiber contacts.

Two pressures were used in this work for the consolidation-curing-cooling process. The lower pressure (0.56 MPa) was close to the curing pressure recommended by the manufacturer. The higher pressure, though less standard, gave better consolidation, the consequence of which remained after curing and cooling, as shown by both resistivity and N/N_o after cooling. Thus, the extent of consolidation affects the through-thickness electrical property of the resulting composite, and likely affects the mechanical properties (e.g., interlaminar shear strength) as well.

It should be mentioned that the resistivity of the carbon fibers also varies with temperature. However, the resistivity of the carbon fiber is much smaller (by orders of magnitude) than the through-thickness resistivity measured, so the influence of the fibers' dependence on temperature to the results reported here is negligible.

The electrical method described in this paper for monitoring consolidation is limited to composites with electrically conducting fibers at a high volume fraction. Since a high volume fraction of continuous fibers

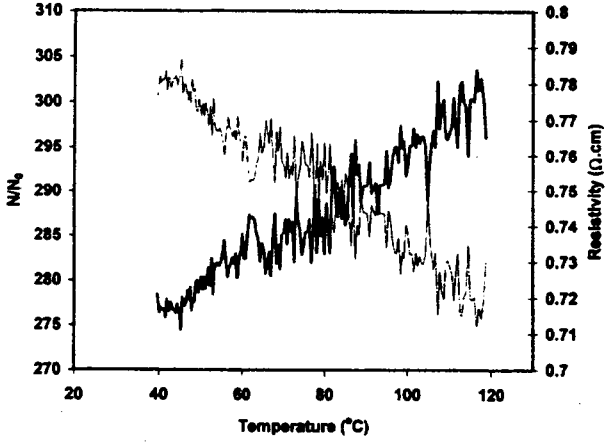


Fig. 5. Variations of N/N_0 (thick line) and through-thickness electrical resistivity (thin line) with temperature during cooling after curing at 120°C and 1.10 MPa.

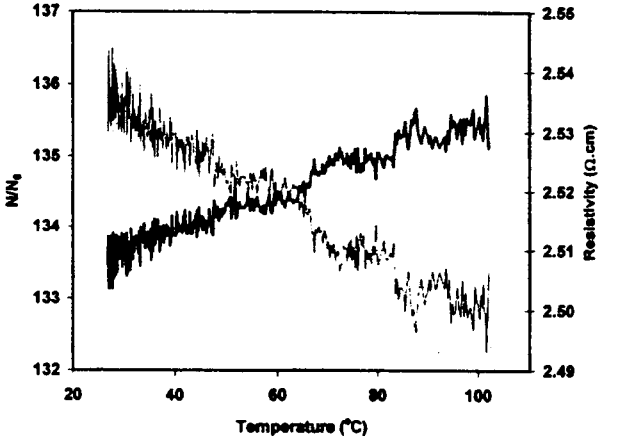


Fig. 6. Variations of N/N_0 (thick line) and through-thickness electrical resistivity (thin line) with temperature during cooling after curing at 100°C and 0.56 MPa.

is typically used in structural composites and carbon fibers are widely used in advanced composites, the method of this paper is of wide applicability.

CONCLUSION

The consolidation of carbon fiber epoxy-matrix laminae during composite fabrication by lamination (involving consolidation, curing and cooling) was found to be hastened and to occur to a greater extent by increasing the pressure. The consequence of better consolidation remained after curing and subsequent cooling. A higher pressure resulted in a lower through-thickness resistivity in a composite after curing and cooling.

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