Composites Science and Technology 61 (2001) 885–888

COMPOSITES SCIENCE AND TECHNOLOGY

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Carbon-fiber/polymer-matrix composites as capacitors

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Received 8 February 1999; received in revised form 18 October 1999; accepted 18 July 2000

Abstract

A continuous carbon-fiber/epoxy-matrix composite with a paper interlayer (0.04 mm thick after composite fabrication) was found to exhibit a capacitance of 1.2 μF/m^2 at 2 MHz, in contrast to a value of 0.21 μF/m^2 for epoxy-impregnated paper (0.10 mm thick). The high capacitance is partly a consequence of the large area of the surface of a fiber layer sandwiching the paper interlayer. This area is twice the flat area. Without a paper interlayer, the composite failed to serve as a capacitor, because of the conductivity in the through-thickness direction. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: A. Carbon fibers; A. Polymer-matrix composites (PMCs); A. Structural materials; B. Electrical properties; Capacitor

1. Introduction

Capacitors are important elements in electrical circuits, although they tend to be more bulky than other elements, such as resistors, diodes and transistors, and their high-frequency performance remains an issue. The bulkiness is of particular concern when a large capacitance is required. Capacitors based on electrical double layers cannot operate at a high frequency, so capacitors based on dielectrics are most common for electronics. This paper is focused on capacitors based on dielectrics.

Capacitors based on dielectrics are conventionally parallel-plate capacitors in which a dielectric is sandwiched by electrically conducting plates. The dielectric material can be paper [1], polymer [1], a high dielectric constant ceramic such as barium titanate in thin film or thick film forms [2], or other electrically insulating materials. The conducting plates are commonly metals in foil, thick film or thin film forms [1,2]. To achieve a high capacitance, the conducting plates are large in area, the dielectric is low in thickness, and numerous layers of dielectric and conducting plates are alternatively stacked (and usually wound to save space).

This paper provides a new type of parallel-plate capacitor, namely carbon fiber polymer-matrix composites in which continuous carbon-fiber layers serve as the conducting plates and paper (placed between the fiber layers), together with the polymer matrix, serves as the dielectric. Carbon-fiber polymer-matrix composites are structural materials that are important for lightweight structures, such as aircrafts, automobiles, sporting goods, wheel chairs, etc. The ability of these composites to serve as capacitors and other circuit elements means that the structure is itself the electronics, so that the electronics 'vanish' into the structure. Electronics made from structural materials such as carbon-fiber polymer-matrix composites and concrete constitute a new field of electronics called structural electronics [3]. In the case of continuous carbon fiber polymer-matrix composites, the carbon fibers are the conductors (resistors) and they can be intercalated to become electron metals or hole metals [3]. By having the electronics vanish into the structure, space is saved. The space saving is particularly valuable for capacitors of large capacitance. As a result of the large area of a structure and the numerous fiber layers in a composite laminate, the capacitance in a composite structure can be very large. In addition to space saving, structural electronics have the advantage of being mechanically rugged and inexpensive, since structural materials are necessarily rugged and inexpensive. The use of a structure as a capacitor is particularly valuable in conjunction with structures that are powered by solar cells, as the structure (capacitor) can be used to store the electrical energy generated by the solar cells.

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PII: S0266-3538(00)00166-4

2. Experimental methods

Epoxy-matrix composites comprising four continuous unidirectional carbon-fiber layers were constructed from individual layers cut from a 12 inch wide unidirectional carbon fiber prepreg tape manufactured by ICI Fiberite (Tempe, AZ). The product used was Hy-E 1076E, which consisted of 976 epoxy matrix and 10E carbon fibers. The fiber and matrix properties are shown in Table 1. The matrix was electrically insulating, whereas the fibers were electrically conducting, with a resistivity of 2.2×10^{-3} Ω cm.

The composite laminates were laid up in a 0.5 inch (13 mm) diameter steel compression mold with laminate configuration $[0/90]_2$ (i.e. four unidirectional fiber layers stacked in the sequence [0/90/0/90]). The individual fiber layers were cut from the prepreg tape. A liquid mold release was used. The laminates were cured using a cycle based on the ICI Fiberite C-5 cure cycle. Curing occurred at 355±10°F (179±6°C) and 89 psi (0.61 MPa) for 120 min. Then the samples were sanded to a rectangular shape (about 8×8 mm; the exact dimensions were measured for each sample in order to calculate the area of the rectangle) for capacitance measurement.

Composites without interlayer (additive between the fiber layers) and with various types of dielectric interlayers were fabricated. In a composite with an interlayer, the interlayer was placed between the second and third fiber layers in the stack of four fiber layers. The interlayers were paper (Table 2) and barium titanate thick film (0.13–0.25 mm thick). The interlayer thickness after composite fabrication was determined by cross-sectional optical microscopy. The barium titanate thick film was applied as a paste, which was made by mixing barium titanate powder (1 μm size, from TAM Ceramics Inc., Niagara Falls, NY) and epoxy (Epon Resin 862 and EPI-Cure 3274 Curing Agent from Shell Chemical Co., Houston, TX). The barium titanate powder was in amounts ranging from 17 to 80 vol.% of the paste. The epoxy from the prepreg layers penetrated the paper (tissue, bond or writing) interlayer, which was porous, during the composite fabrication.

Capacitance (for the sample capacitance and sample resistance in parallel) measurement was made using a precision RLC capacitance meter (Model 7600, QuadTech, Inc., Marlborough, MA) at frequencies ranging from 10 Hz to 2 MHz. During the measurement, the rectangular sample (mechanically polished on the rectangular faces) was sandwiched by two copper disks (mechanically polished on the circular faces) of diameter 0.5 inch (13 mm). The sandwich was held together by pressure provided by a clip. The contact resistance of the interface between a copper disk and a sample was only a few ohms, so the interface contributed negligibly to the measured capacitance.

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}

Table 2
Thickness of paper (mm, ±0.01) before and after composite fabrication

	Before	After
Tissue paper	0.05	0.02
Writing paper	0.08	0.04
Bond paper	0.10	0.08

3. Results and discussion

As a consequence of the touching of the fibers of the two fiber layers, the through-thickness conductivity was too high (impedance too low; Table 3) for meaningful capacitance measurement for the composite without interlayer, the composite with tissue paper interlayer and the composite with barium titanate thick film (without paper) interlayer. The use of a writing paper or bond paper interlayer was sufficient for avoiding the fibers of one layer to touch those of the other layer, but the use of tissue paper or barium titanate thick film interlayer was not. The fibers were not able to go through the writing paper or bond paper, even though the epoxy matrix was able to penetrate the paper. (The penetration of epoxy is desirable for the mechanical integrity of the composite.) As a result, only composites with a writing paper or bond paper interlayer could serve as capacitors.

Table 3 shows the dielectric behavior of various composites in the through-thickness direction. The highest capacitance per unit area was attained by using writing paper alone as the interlayer. The use of bond paper as the interlayer gave lower capacitance per unit area because of the larger interlayer thickness. The combined use of writing paper and barium titanate as the interlayer also gave lower values of the capacitance per unit area than the use of writing paper alone. By using a paste with 80 vol.% barium titanate, the relative dielectric constant

Table 3
Dielectric behavior of carbon fiber epoxy-matrix composites in the through-thickness direction

Interlayer	Capacitance per unit area (μF/m^2)		Relative dielectric constant[a] at 2 MHz	Interlayer thickness (mm, ±0.01)	Impedance at 10 Hz (Ω)
	2 kHz	2 MHz			
None	–	–	–	0	33
33 vol.% $BaTiO_3$	–	–	–	0.11	30
Tissue paper	–	–	–	0.02	33
Bond paper	0.54	0.45	4.1	0.08	2.34×10^6
Writing paper	1.23	1.17	5.3	0.04	1.89×10^6
Writing paper + 17 vol.% $BaTiO_3$	0.90	0.78	8.9	0.10	2.84×10^6
Writing paper + 33 vol.% $BaTiO_3$	1.06	0.92	13.6	0.13	1.78×10^6
Writing paper + 65 vol.% $BaTiO_3$	0.99	0.87	14.2	0.14	1.59×10^6
Writing paper + 80 vol.% $BaTiO_3$	0.84	0.71	19.8	0.25	2.54×10^6

[a] Calculated from the capacitance, interlayer thickness, and the flat area of the laminate (the actual area of the conductor surface sandwiching the dielectric is much larger than the flat area as a result of the fact that a lamina consists of fibers of diameter 7 μm).

attained the highest value of 19.8, but the large thickness of the interlayer (resulting from the low workability of this paste) caused the capacitance per unit area to be low. Therefore, the use of a barium titanate interlayer is not attractive, whatever is the volume fraction of $BaTiO_3$ in the paste.

For all samples in Table 3 exhibiting dielectric (rather than conducting) behavior in the through-thickness direction, the capacitance decreased with increasing frequency. For example, for the composite with writing paper alone as the interlayer, the capacitance at 2 MHz was 95% of that at 2 kHz.

Table 4 shows the dielectric behavior of paper and epoxy-impregnated paper in the absence of fibers. The relative dielectric constant is higher for writing paper (used in some of the composites in Table 3) than bond paper. This is caused by contamination in the writing paper. The contamination is shown by dispersed dark spots on the optical micrograph. However, such contamination (spots) was not observed for the bond paper.

Table 4 shows that the relative dielectric constant of epoxy-impregnated writing paper (2.4) is below that of writing paper (2.7) and that of epoxy (3.0). The relative dielectric constant of epoxy-impregnated bond paper (2.1) is the same as that of bond paper, but less than that of epoxy. These effects are believed to be a result of the reactions between epoxy and paper.

Table 4
Dielectric behavior (at 2 MHz) of epoxy, paper and epoxy impregnated paper in the absence of fibers (all have thickness 0.10 mm)

	Capacitance per unit area (μF/m^2)	Relative dielectric constant
Writing paper	0.24	2.7
Writing paper + epoxy	0.21	2.4
Bond paper	0.19	2.1
Bond paper + epoxy	0.19	2.1
Epoxy	0.10	3.0

The relative dielectric constant of the writing paper interlayer (5.3) in Table 3 (in the presence of fibers) is much higher than that (2.4) of the epoxy-impregnated writing paper in Table 4 (in the absence of fibers). This is partly attributed to the fact that the surface of a fiber layer is not flat, so that the actual area of the surface is, from simple geometry, as much as $\pi/2$ or 1.6 times the flat area used in calculating the relative dielectric constant of Table 3. Since the relative dielectric constant is inversely related to the area, the ratio of the actual area to the flat area is $5.3/2.4 = 2.2$, if the dielectric behavior is assumed to be the same for the interlayer part of the composite and the epoxy-impregnated paper. In other words, the actual relative dielectric constant of the writing paper interlayer is only 2.4, but the large (actual) area makes the relative dielectric constant appear high (5.3). That this ratio exceeds 1.6 is probably because of the difference in dielectric behavior between the interlayer and the epoxy-impregnated paper resulting from the differences in the extent of reaction between epoxy and paper and in the extent of cure of the epoxy. Hence, the effectiveness of carbon-fiber epoxy-matrix composites for capacitors is partly caused by the large area provided by the lamina surface, which is not flat.

The capacitance per unit area is 1.17 μF/m^2 for the writing paper interlayer (Table 3), but just 0.21 μF/m^2 for the epoxy impregnated writing paper (Table 4). This is partly a result of the large actual area of the conducting surface sandwiching the writing paper interlayer, but is caused by the small thickness of the writing paper interlayer (0.04 mm) compared to the thickness of the epoxy-impregnated writing paper (0.10 mm). The pressure during composite fabrication probably resulted in the decrease of the thickness of the writing paper.

For a typical composite structure with a substantially large surface area and numerous fiber layers, the effective area is at least 1000 m^2, and the associated capacitance (for the case of the writing paper interlayer) is at least 1.2 mF. Hence a large capacitor is built in to the structure.

Although carbon-fiber polymer-matrix composites are much more conductive in the fiber direction than in the through-thickness direction, the conductivity in the through-thickness direction is substantial. The through-thickness conductivity is a consequence of the contact between fibers of adjacent fiber layers. The contact occurs in spite of the presence of the epoxy matrix because of the flow of the epoxy resin during composite fabrication and the waviness of the fibers. The contact cannot be stopped by the use of a tissue paper interlayer given the porosity of the tissue paper. For the same reason, the contact cannot be stopped by the use of a thick film interlayer. However, the contact can be effectively stopped by the use of a writing paper or bond paper interlayer, which has enough porosity for the epoxy resin to go through and has small enough a porosity for the fibers to be not able to go through.

4. Conclusion

Carbon-fiber epoxy-matrix composite was found to be a parallel-plate capacitor with capacitance 1.2 μF/m^2 at 2 MHz, if the composite contained a writing paper interlayer of thickness 0.04 mm. The further addition of a $BaTiO_3$ thick film to the interlayer decreased the capacitance as a results of the increase in interlayer thickness. Without an interlayer or with a more porous paper interlayer, the composite was conducting in the through-thickness direction. The capacitance of the epoxy-impregnated paper (0.10 mm thick) was 0.21 μF/m^2. The high capacitance for the composite with paper interlayer is partly a consequence of the large area of the surface of a fiber layer; this area is 2 times that of the flat area. The high capacitance is partly a result of the reduction in the paper thickness during composite fabrication.

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