Composites: Part A 68 (2015) 47–55

Contents lists available at ScienceDirect

Composites: Part A

journal homepage: www.elsevier.com/locate/compositesa

Effect of processing conditions and electrode characteristics on the electrical properties of structural composite capacitors

D.J. O'Brien *, D.M. Baechle, O.B. Yurchak [1], E.D. Wetzel

U.S. Army Research Laboratory, Weapons and Materials Research Directorate, Aberdeen Proving Ground, MD 21005, United States

ARTICLE INFO

Article history:
Received 30 May 2014
Received in revised form 2 September 2014
Accepted 4 September 2014
Available online 16 September 2014

Keywords:
A. Smart materials
A. Hybrid
B. Electrical properties
E. Cure

ABSTRACT

Structural capacitors are manufactured from glass fabric/epoxy prepreg dielectrics and metalized polymer film electrodes. The electrical breakdown strengths of these multifunctional materials are investigated across a wide range of electrode constructions and processing parameters. The results show that electrode selection and materials processing have a significant impact on the energy that the device can store. Also, this careful consideration of processing parameters and electrode construction has led to the development of a structural capacitor with an energy density exceeding 0.90 J/cm^3, the highest yet reported.

Published by Elsevier Ltd.

1. Introduction

There has been significant interest in developing multifunctional materials that both store energy and support mechanical load, referred to as structural power devices [1–8]. By combining energy storage and structural functions, such materials offer the possibility to reduce platform weight and volume, resulting in performance improvements such as vehicles with increased fuel economy and range, satellites with reduced launch masses, and electronics with more compact form factors. Several researchers have pursued a variety of structural power concepts including structural capacitors [8,9], batteries [2–4,10], supercapacitors [11,12] and fuel cells [13,14]. Regardless of the energy storage mechanism, combining energy storage and structure often involves a reduction in per-mass or per-volume performance for the structural and/or energy storage functions when compared to their conventional, monofunctional counterparts. Despite this compromise, the multifunctional approach can lead to overall volume or mass savings because the same mass and volume elements are performing energy storage and mechanical functions simultaneously. The performance thresholds necessary to achieve mass or volume savings have been derived and expressed in terms of a minimum multifunctional performance metric [15].

In the present work we study how processing and environmental conditioning affect the performance of structural capacitors, multifunctional materials that could enable mobilizing pulsed power-based technologies such as electromagnetic armor, rail guns, and high energy lasers. Earlier contributions demonstrated the feasibility of achieving system-level weight [15] and volume [16] savings with a structural capacitor made from a continuous glass fiber reinforced epoxy dielectric, sandwiched between metallized polymer film electrodes (Fig. 1). The results showed that the choice of dielectric material has a significant effect on structural capacitor energy density. It was also found that the breakdown strength decreases with increasing fiber volume fraction, a result consistent with another investigation [17]. Unfortunately, the mechanism causing this decrease was unclear. It is possible that the electric field concentration caused by the dielectric mismatch between the glass and polymer constituents results in an increased probability of a critical flaw at higher fiber volume fractions. However, an increase in void content accompanying the increased fiber volume fraction confounded the experimental results. Like reinforcing fibers, voids would also act as electric field stress raisers that could precipitate breakdown in the surrounding matrix, or directly initiate breakdown due to the relatively low dielectric strength of the gas-filled void relative to the solid polymer matrix. Also, specimens cured at higher pressures had higher breakdown strengths, possibly as a result of decreased void content. Several researchers have shown that voids can play a significant role in the performance of capacitors [18,19]. In other work Flandin et al. [20] showed that the extent of reaction plays an important role in breakdown strength of polymer composite dielectrics. Together, these results suggest that processing plays a significant role in the performance of structural capacitors.

* Corresponding author. Tel.: +1 410 306 0843.
E-mail address: daniel.j.obrien58.civ@mail.mil (D.J. O'Brien).
[1] Current address: Carleton Technologies Inc., Westminster, MD 21157, United States.

http://dx.doi.org/10.1016/j.compositesa.2014.09.003
1359-835X/Published by Elsevier Ltd.

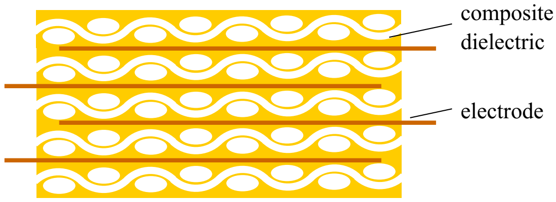


Fig. 1. Schematic of a structural capacitor. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

In addition to processing and environmental conditions, we study the effects of electrode characteristics (metallization type, thickness, and carrier material) on the energy density of structural polymer composite capacitors. Previous work has shown that these parameters can affect performance of conventional capacitors. For example, researchers have shown that breakdown strength in solid dielectrics has a positive correlation with the electrode metal's work function, suggesting that a greater work function necessitates more energy to liberate a surface electron [21,22]. If such electrons participate in the cascade that causes the breakdown event, selecting an electrode material with a high work function (thus limiting the source of electrons) might increase breakdown strength. In another work considering electrode properties, Sato et al. studied the effect of electrode roughness and found that the breakdown strength decreased as the electrode roughness increased [23].

The sections that follow describe a series of experiments conducted on single dielectric layer capacitors. These experiments investigate the effect of a range of parameters on the breakdown strength, including processing method, processing pressure, cure cycle, humidity, electrode metallization type, metallization thickness, and electrode carrier material. The experimental techniques for manufacturing and testing the capacitor specimens are described first, followed by a presentation and interpretation of the results.

2. Experimental

2.1. Materials

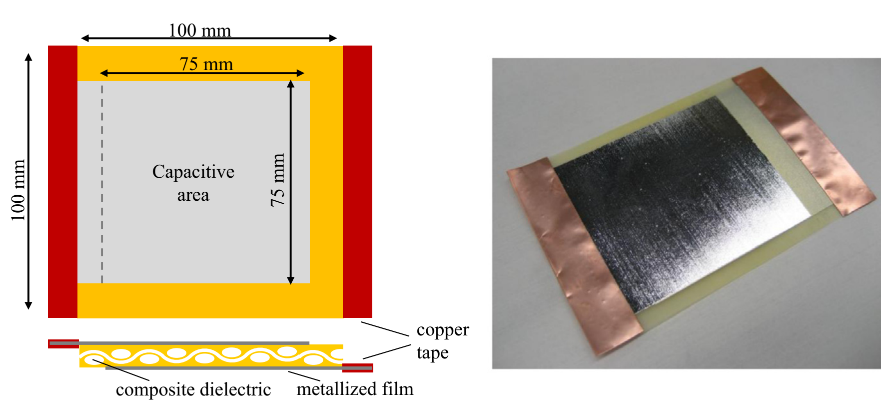
For this study, sixteen sets each of at least five capacitor specimens were constructed using geometries similar to a previous study, Figs. 2a and 3a [15]. A range of processing conditions and laminate constructions, outlined in Table 1, were used in order to study their effects on capacitor performance. All capacitors consisted of layers of glass fabric-reinforced prepreg interleaved with thin metallized films of paper, polyimide (PI), or biaxially oriented polypropylene (BOPP). The prepreg material used for most specimens consisted of an FR4-type epoxy reinforced with a style 2116 woven E-glass (N4205-6FC, Nelcote, Northfield MN). Two capacitor sets, used to study the effect of pressure on breakdown strength, were constructed from a structural epoxy prepreg reinforced with a style 120 woven E-glass (120–765, Nelcote). Both prepregs were approximately 100 μm thick. The PI electrode carrier material was 12.7 μm thick with a 100 nm layer of aluminum on one side (146454-004, Sheldahl Inc., Northfield MN). The BOPP films (Bollore Inc. Dayville, CT) had metallizations of either Al, Zn, or a Zn–Al alloy in thickness ranges from 4.3 to 12 nm (10–4 Ω/sq). While the precise composition of the Zn–Al alloy was not specified, the manufacturer indicated that the metallization is mostly Zn with a small amount of Al added to improve stability against atmospheric corrosion [24]. Rutherford backscattering spectroscopy indicated an atomic ratio of 9:1 Zn for Zn to Al. All paper carriers (Winter-Wolff International, Inc., Jericho, NY) were 8 μm thick, 1.3 g/cm^3 density, with Al metallizations ranging from 7 nm to 54 nm (4–0.5 Ω/sq). All electrode metal resistivity values were provided by the electrode manufacturers, and electrode metal thickness was calculated using the manufacturer-provided sheet resistivities along with the bulk resistivities of aluminum and zinc, 2.65 μΩ-cm and 5.92 μΩ-cm, respectively [25]. Copper tape with an electrically conductive pressure-sensitive adhesive (05012D-AB, 3 M via SPI Supplies, West Chester, PA) and conductive silver paint (05002-GA, SPI Supplies) were used to enhance connectorization, as described below.

2.2. Manufacturing

2.2.1. Layup and connectorization

Prior to layup, prepregs were wiped with technical wipes (TX612, Texwipe, Mahwah NJ) that were partially soaked in 100% ethanol. Compressed nitrogen was then blown on prepregs and electrodes to help remove any remaining surface dust. A previous study showed that cleaning the constituent materials greatly improves their dielectric breakdown strength [26].

Two types of capacitors were fabricated, "unencapsulated" and "encapsulated". All polyimide electrode capacitors, sets 1–7, were unencapsulated and consisted of one prepreg ply sandwiched between two electrode layers, as seen in Fig. 2. The metallized faces of the electrodes both faced the same direction, as this is the configuration that would be used in a capacitor with multiple dielectric layers. As such, one layer of the electrode support film was present in the dielectric layer of all capacitors. Encapsulated



(a) (b)

Fig. 2. (a) Schematic and (b) photograph of unencapsulated capacitors, sets 1–7. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)