

CFRP structural capacitor materials for automotive applications

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In this paper, an approach towards realising novel multifunctional polymer composites is presented. A series of structural capacitor materials made from carbon fibre reinforced polymers have been developed, manufactured and tested. The structural capacitor materials were made from carbon fibre epoxy prepreg woven lamina separated by a polymer film dielectric separator. The structural capacitor multifunctional performance was characterised measuring capacitance, dielectric strength and tearing force. The developed structural carbon fibre reinforced polymer (CFRP) capacitor designs employing polymer film dielectrics (PA, PC and PET) offer remarkable multifunctional potential.

Keywords: Multifunctional, Electrical properties, Mechanical properties, Carbon fibre

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Introduction

The use of lightweight materials in structural applications is ever increasing. Today, lightweight engineering materials are needed to realise greener, safer and more competitive products in all transportation modes. To facilitate development of such products, a step change towards electrification to urban mobility and transport is imminent, further requiring yet lighter vehicles. The immediate need for electrical vehicles is driven by the forecast shortage of crude oil based energy carriers together with the necessity to reduce greenhouse gas emissions.

To realise electrical vehicles, and to keep up with the power requirements of new and emerging technologies, the mobile platforms must carry increasingly larger masses and volume of energy storage components such as capacitors, supercapacitors and batteries. This development works against realisation of efficient electrical vehicles, for which low weight is essential. A decade ago, Chung and Wang¹ presented the idea of using carbon fibre reinforced polymers (CFRP) in 'structural electronics'. They suggested that the semiconductive nature of carbon fibre composites could be used to make electrical devices, e.g. diodes, detectors, transistors, etc. In this spirit, they were first to propose use of a high dielectric constant material as an interface between CFRP laminas to provide a capacitor material, i.e. a structural parallel plate capacitor. By this approach, truly multifunctional material, i.e. a material that can perform more than one function, emerges. In the case of a structural capacitor, the material is stiff and strong to sustain mechanical loading and at the same time, is able to store electrical energy. In a follow-up study, Luo and Chung²

demonstrated structural capacitor materials for the first time. Lou and Chang made thin structural capacitors from single unidirectional carbon fibre epoxy prepreg layers separated by different paper dielectrics. For these materials, dynamic capacitance up to 1200 nF m^{-2} at 2 MHz was demonstrated; however, no mechanical characterisation was performed. More recently, another approach for making structural capacitors was suggested by Baechle *et al.*³ To achieve high energy density of the capacitor for maximised multifunctional efficiency, Baechle and co-workers made structural capacitors from dielectric glass/epoxy prepreg with thin metal electrodes. By this approach, Baechle and colleagues utilise the dielectric layer for structural performance.

Since Chung and Wang¹ first suggested the use of structural electronics, development of such materials has been reported in the open literature and a new research area has emerged, that of 'composite structural power storage materials'. Recently, concepts for structural polymer composite batteries^{4,5} and supercapacitors⁶ have been presented. All these materials are developed with a desire to reduce vehicle weight permitting replacement of structural components (e.g. car floor panels) and energy storage devices (e.g. batteries). The work presented here was enthused by the ambition to develop a truly multifunctional composite material that may boost the development of future ultralight electrical vehicles.

The objective of this study is to develop high performance multifunctional polymer composite capacitor materials. These capacitor materials are developed in the spirit of Luo and Chung,² employing carbon fibre prepreg lamina separated by a dielectric material. In this study, polymer films are utilised as dielectric separator layers. The electrical and mechanical performance is characterised for each dielectric material employed and its overall multifunctional performance is assessed.

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Experimental

Materials

Structural capacitors were made from carbon fibre epoxy composites to facilitate high performance mechanical electrodes. The electrode layers (laminae) were made from 0.125 mm thick prepreg weaves. The prepreg was a 245 g m^{-2} 2×2 Twill HS (3 K) $0/90^\circ$ configuration, MTM57/CF3200-42% RW, supplied by the Advanced Composite Group (Heanor, UK). The resulting CFRP composite had a fibre volume fraction of 52%. A dielectric layer in a composite laminate separated the electrode layers. A selection of materials was employed as separator. The dielectric materials employed in this study, and their nominal thicknesses, are listed in Table 1. The polyamide film brand was Airtech Wrightlon 5400. The polyester film was DuPont Teijin Films Mylinex 301 Bondable Film. Finally, the polycarbonate film was a 0.17 mm thick PC quality supplied by Andren & Söner (Göteborg, Sweden). Stacking two electrode layers separated by the dielectric layer made the structural capacitors. To facilitate mechanical testing, thicker laminates were manufactured. Here, the specimens were made from six layers of prepreg weaves alternated with five dielectric layers, resulting in a laminate with a nominal thickness of 2 mm.

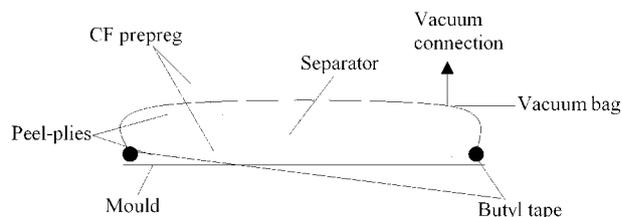
In addition to the dielectrics reported in Table 1, dielectrics were employed to study effects of surface treatment of the polymer dielectric films on structural performance. For this purpose, composite material capacitors utilising plasma treated (using a Technics Plasma 440G equipment) PA and PET films were manufactured. In the plasma treatment, the surface of a substrate is exposed to plasma, i.e. excited in a gas, e.g. N_2 , as used in these experiments. The treatment is carried out to increase the adhesion between the dielectric film and matrix of a composite as chemical bonds in the film surface are broken and reactive positions are generated.⁷ These reactive sites react with nitrogen creating polar groups at the surface. The advantage of this process is that no thermal or mechanical strains are introduced into the specimen. The treatment was performed during 15 s.

Composite manufacture and characterisation

During manufacture, prepreg layers were stacked in a release agent coated mould. To achieve equal surface properties on both sides of the laminate, the structural capacitor laminates were manufactured using peel plies on both top and bottom surfaces. The mould was sealed with butyl tape and a vacuum bag. A schematic of the bagged lay-up is shown in Fig. 1. Vacuum was applied and debulking without heat was performed. The mould was then placed in an oven and heated according to the supplier's recommendations (120°C for 30 min) to achieve fully cured laminates. The vacuum was necessary to achieve void free, high quality composite

Table 1 Dielectrics and their nominal thickness

Dielectric material	Thickness/mm
PA film	0.05
PET film	0.02
PC film	0.17



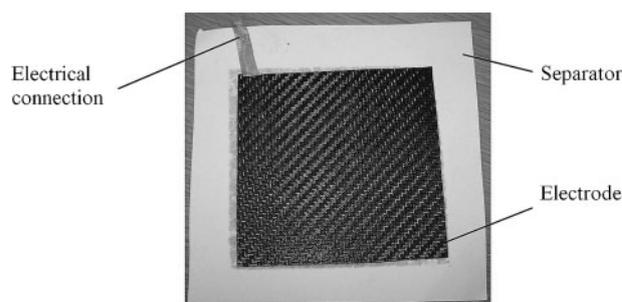
1 Manufacture of structural capacitor laminates

laminates. Voids must be kept to a minimum. Air has a lower dielectric constant than the tested dielectrics. The presence of voids will therefore locally reduce the isolative properties of the dielectric, and mechanical properties are also lowered.

In total three types of structural capacitor laminates were manufactured for electrical characterisation. For each type of material, a set of five specimens was manufactured. The specimen designed for electrical (capacitance) characterisation is depicted in Fig. 2. The specimen had a total area of 0.010 m^2 and a nominal thickness between 0.27 and 0.42 mm, depending on the dielectric material employed. The square shape was chosen because it is easy to control size and placement, though a circular shape would be better from an electrical point of view, since corners concentrate charge. The dielectric material has an excess of $\sim 10 \text{ mm}$ around carbon fibre plies to avoid edge effects. A copper mesh was used as electrical connection due to good electrical conductivity and compatibility to most matrices, as well as flexibility.

Two structural capacitor materials were manufactured for mechanical characterisation. In addition to the three capacitor materials made for electrical characterisation capacitors employing the plasma treated PA and PET separator films, PA-PT and PET-PT respectively were manufactured. Note that no capacitance measurements were performed on capacitors using plasma treated dielectrics. The reason for this is that plasma treatment alters only the surface properties of the polymer film and not its intrinsic properties, nor its thickness. It is therefore assumed that plasma treatment of the dielectric will have negligible influence on the capacitance of the structural capacitor.

Specimens for tearing were manufactured; specimens were cut and grinded to the nominal dimensions: 2 mm thickness, 30 mm width and 150 mm length, and a hole of 5 mm diameter was drilled.



2 Single layer structural capacitor for electrical characterisation

Experimental characterisation

Capacitance

Capacitance measurements were carried out on in total 15 specimens, five for each type of dielectric. Measurements were performed as follows. The capacitor was charged repeatedly to an increasing voltage V until the structural capacitor was short circuited. For each voltage, the structural capacitor was discharged and the charge Q , in coulomb, gave the capacitance as

$$C = \frac{Q}{V} \quad (1)$$

The voltage to which the capacitor was charged was measured using a voltmeter, Eltex EMF 58. The charge during discharge was measured employing a Coulomb meter, Keithley Instruments 602 Solid State Electrometer.

Dielectric breakdown voltage

Dielectric breakdown voltage (dielectric strength) of the capacitors was measured using the ASTM standard for direct current measurement of dielectric breakdown⁸ as carried out by O'Brien *et al.*⁹ Voltage was applied with a rate of 100 V s^{-1} with the specimen submerged in mineral oil. Breakdown tests were performed on three structural capacitors of each type, in total 18 measurements. The equipment used was a Spellman SR6 High Voltage Supply controlled by PCI GPIB and a HP 33120A Function Waveform Generator, with a maximum voltage of 28.3 kV. A PCI NI-6023E (DAQ) with a BNC-2111 connector block was used to record the results from the tests. Voltage was applied until failure was evident by a large drop in voltage.

Tearing

A tearing test was chosen to evaluate the structural capacitors used in a crash situation that could be found in e.g. an automotive application. The tearing force was evaluated using a tearing test developed for simulating a tearing failure.¹⁰ The test is easily performed and requires very little specimen preparation making it very fast and robust. The test was performed in an ordinary tensile tester utilising a purpose made fixture. The specimen was clamped at end and a bolt was put through the predrilled hole.

When the test was performed, a fixed crosshead speed of 50 mm min^{-1} and a tearing distance of 70 mm were used. The force was measured continuously throughout the test. The structural capacitor materials have different thicknesses. Consequently, to allow comparison of tearing force, the measured force is normalised with bolt diameter and laminate thickness, expressed in equation (2) as

$$F_{\text{norm}} = \frac{\bar{F}}{td} \quad (2)$$

where \bar{F} is the average tearing force, t is the thickness of the sample and d is the diameter of the bolt (4.88 mm for the test equipment used here).

Owing to the nature of this test being a process measuring over a bulk of material, it was considered sufficient to use only two specimens of each type.

Electrical energy density

Evaluation of electrical energy density allows comparison between the different capacitor devices. Use of thin film dielectric separators usually results in capacitors with high capacitance but low breakdown voltage.⁹ The energy density is given by

$$\bar{\Gamma}_{\text{sc}} = \frac{\frac{1}{2} CV^2}{m_{\text{sc}}} \quad (3)$$

where $\bar{\Gamma}_{\text{sc}}$ is the energy density of the structural capacitor, C is the capacitance, V is the voltage at dielectric breakdown and m_{sc} is the mass of the structural capacitor.

Results and discussion

Electrical properties

The results from the discharge and dielectric strength experiments are presented in Table 2. The results are measured average capacitance and dielectric strength with standard deviations, for respective dielectric.

Capacitance

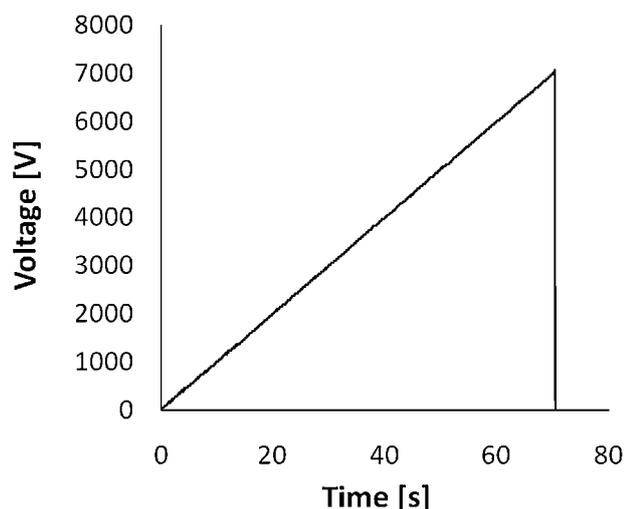
For the polymer films, thickness is the main variable controlling capacitance. This is evident for the PC film, which has the largest thickness and consequently the lowest capacitance. An explanation for the noticeably high standard deviation for the PET film capacitor could be test equipment limitations. The capacitors with high capacitance required low voltage in order to be measured, affecting accuracy of measurements.

The reported capacitances for all structural CFRP electrode capacitor designs studied here were significantly higher than those found for capacitors employing structural dielectric layers.^{3,9} The highest capacitance reported⁹ for those materials was $\sim 150 \text{ nF m}^{-2}$, cf. Table 2. No experimental comparison with capacitance of a conventional capacitor material was possible. Nevertheless, a comparison with theoretical capacitance of conventional film capacitors (20 μm thick PET film covered by 4 μm copper on both sides) suggests that the structural capacitors developed in this study match the conventional capacitors considering capacitance. The theoretical capacitance for such a conventional film capacitor is 1460 nF m^{-2} to be compared with the 1860 nF m^{-2} for the PET film structural capacitor developed here. As discussed by Luo and Chung,² the carbon fibres in the structural capacitor electrodes

Table 2 Summary of results for various structural capacitors

Dielectric	Thickness/ μm	Capacitance/ nF m^{-2}	Dielectric strength/ kV	Average electrical energy density/ J g^{-1}
PA film	50 ± 3	868 ± 198	7.80 ± 1.03	0.034
PET film	19 ± 1	1860 ± 1024	6.40 ± 0.66	0.052
PC film	155 ± 8	206 ± 11	28.3*	0.089*

*The capacitors had not failed at maximum voltage in the breakdown voltage tests.



3 Voltage history of typical dielectric strength test

provide an increased surface area, and therefore promote an increase in capacity compared to conventional film electrode capacitors.

Dielectric strength

In Fig. 3, the voltage history for a typical dielectric strength test series is depicted. Note the absence of clearing reported in tests on capacitors employing structural dielectric separators.⁹ Results from the dielectric breakdown voltage measurements are presented in Table 2. The performances of PA and PET films are in the same region, but the lower thickness of the PET film makes it favourable as an insulator in a structural capacitor. The performance of the PC film capacitor was outstanding, and did not break down before maximum voltage was reached. Hence it should be recognised that maximum electrical energy density for the PC film capacitor could not be measured with the current test set-up.

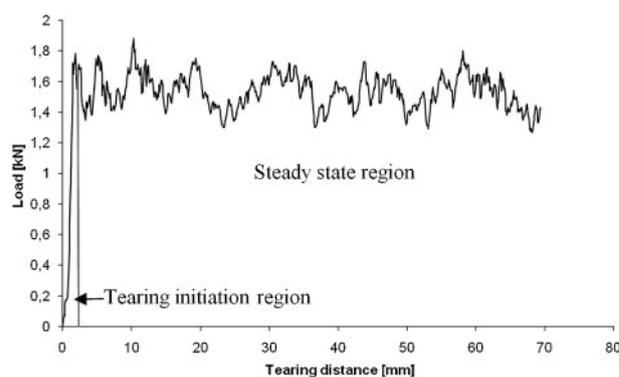
The average static capacitance, average breakdown voltage and average weight were used to calculate the electrical energy density according to equation (3). The calculated energy densities are presented in Table 2.

The electrical energy density for PC film capacitor was particularly good. All PC film capacitors exceeded an electrical energy density of 0.089 J g^{-1} . This value is lower than the 0.28 J g^{-1} reported for structural dielectric separator capacitors developed by O'Brien *et al.*⁹ However, the breakdown voltage was not reached for the PD film dielectric capacitor studied here. As the energy density is proportional to the breakdown voltage squared, even a moderate increase in measured dielectric strength will result in a significant increase in electrical energy density.

Mechanical properties: tearing

A typical force measurement is depicted in Fig. 4. The force used in the calculations is the average force in the steady state region of the tearing.

The normalised tearing forces (equation (2)) are presented in Table 3. As seen in the table, no structural capacitor can compete with the pure CFRP laminate. Worth noting, however, is the great performance for the plasma treated PA film capacitor material compared to the non-treated PA film material. No such improvements from treatment were noted for the PET film, for



4 Typical tearing force measurement

which the plasma treated film shows slightly lower performance than that of the non-treated film material. The variation is quite small and could be due to natural variations within the samples.

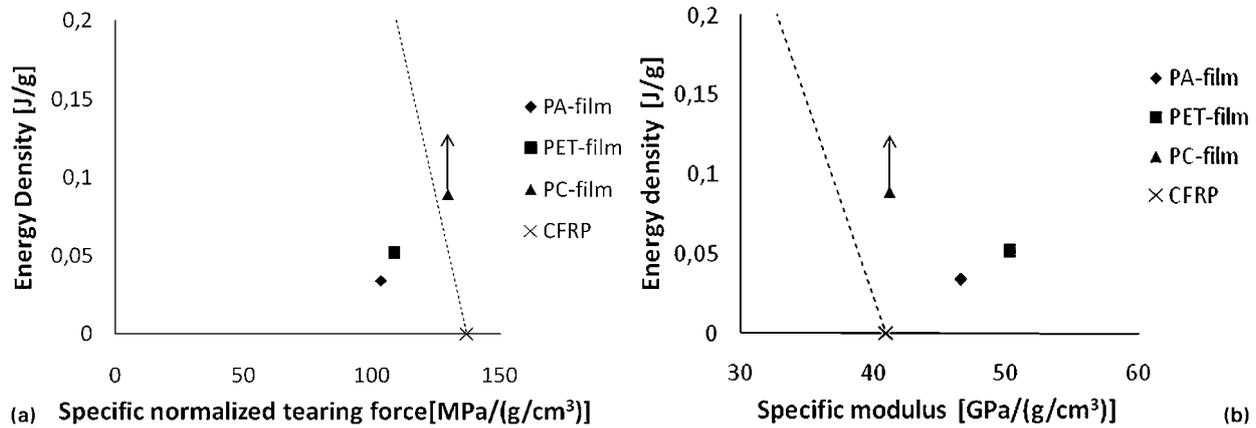
Multifunctional performance

Multifunctional performance is evaluated by assessment of measured energy density versus specific tearing force and calculated specific in plane stiffness. Here, the in-plane stiffness was calculated by rule of mixture using the stiffness data for the CFRP and the polymer films provided by the material suppliers. Although capacitance and dielectric strength are important properties for the performance of a capacitor, use of these properties individually to assess multifunctional performance of a structural capacitor will produce contradicting results. For example, if one was to assess multifunctional performance with respect to capacitance alone, PET film would be found most suitable to achieve structural capacitor materials, whereas the PC film capacitor would be found worse. In contrast, if only tearing energy was to be considered, none could match the pure CFRP, but the best choice would be the PC film capacitor.

Employing energy density as the parameter to assess multifunctional performance allows us to compare the different structural capacitor designs for their applicability in a structural system, where energy needs to be stored, with respect to their potential to reduce system weight. Consequently, this approach allows us to evaluate the structural capacitors influence on system weight, as described by O'Brien *et al.*⁹ This is important as even though the multifunctional element exhibits energy density and strength and/or stiffness that usually are lower than the best monofunctional materials, at a system level the multifunctional material enables an overall mass saving. In the paper by O'Brien *et al.*,⁹ a procedure to evaluate multifunctional capacitor designs, following an approach suggested by Wetzel,¹¹ is

Table 3 Average tearing force normalised by sample thickness and bolt diameter (SD within brackets)

Dielectric	Normalised tearing force/MPa
PA film	124.5 (11.4)
PA film (plasma treated)	171.0 (12.4)
PET film	127.4 (16.0)
PET film (plasma treated)	121.9 (15.8)
PC film	154.2 (14.8)
CFRP	202.8 (15.6)



5 **a** energy density versus specific normalised tearing force for structural capacitors and **b** energy density versus calculated specific stiffness for plastic film structural capacitors

presented. O'Brien and co-workers⁹ define a total system mass M equal to the sum of the mass of the capacitors m_c and the mass of the structure m_s . The design metric for capacitor performance is energy density $\bar{\Gamma}$ (J kg^{-1}) with overall system energy storage defined as $\Gamma = \bar{\Gamma}m_c$. Similarly, the mechanical performance, e.g. specific modulus or ILSS (J kg^{-1}), can be defined as \bar{E} and $\bar{\tau}$. From these, the energy density and specific mechanical properties of the structural capacitors can be found as $\sigma^e \bar{\Gamma}$, $\sigma^s \bar{E}$ and $\sigma^s \bar{\tau}$. σ^e and σ^s are the structural capacitor's energy and structural efficiency respectively. An improved multifunctional design would maintain the same overall system energy and mechanical performance, but reduce the total system weight. However, a structural capacitor will only enable such system level mass savings if

$$\sigma^{\text{mf}} \equiv \sigma^e + \sigma^s > 1 \quad (4)$$

Multifunctional performance is depicted in Fig. 5. In Fig. 5a, electrical energy density is plotted as a function of specific normalized tearing force. In this figure, a dashed line, according to equation (4), is introduced to indicate multifunctional efficiency σ^{mf} of the structural capacitors, with respect to tearing force. Data points located to the right of the dashed line exhibit a multifunctional efficiency greater than unity. Here, the reference electrical energy density for a state of the art capacitor material has been assumed to 1 J g^{-1} ,⁹ whereas the tearing force for the CFRP is used as benchmark for the strength. From the graph, it is evident that only the PC film capacitor exhibits multifunctional efficiencies exceeding unity. Note that these data refer to the tests on untreated films. When plasma was treated, the tearing force increased substantially for the PA film and would therefore achieve multifunctional efficiency exceeding unity by a large margin.

Results for multifunctional efficiency with respect to specific in-plane stiffness are plotted in Fig. 5b. Again a dashed line indicating multifunctional efficiency of unity, according to equation (4), is introduced, where electrical energy density of 1 J g^{-1} and specific modulus of $40.93 \text{ GPa g}^{-1} \text{ cm}^{-3}$ are used for the monofunctional materials. Consequently, the stiffness offered by the CFRP structural capacitor electrodes greatly exceeds that resulting from use of GFRP dielectrics developed by O'Brien *et al.*⁹ In all cases, a multifunctional efficiency higher than unity is achieved for all polymer film capacitor materials. Note that in Fig. 5, the energy

density plotted for the PC film capacitor is a lower than the maximum energy density, which remains unknown at this stage.

Conclusions

A series of structural capacitor materials made from carbon fibre reinforced polymers electrodes have been manufactured and evaluated for their mechanical, electrical and multifunctional performance. The structural capacitor materials were made from carbon fibre epoxy prepreg woven lamina as electrodes separated by a dielectric material. The dielectric materials employed in this study were three different polymer films (PA, PET and PC).

Multifunctional efficiency of the developed structural capacitors was evaluated on the basis of achieved electrical energy density and tearing force as well as in plane stiffness. All capacitors investigated indicate potential for high multifunctional efficiency. Depending on film thickness and surface plasma treatment, significantly improved multifunctional designs with overall weight savings can be achieved. In particular, use of CFRP in the capacitor electrodes will result in significantly higher in-plane stiffness of the multifunctional component than use of GFRP structural dielectric separators. Nevertheless, further research is needed to identify the best choice of polymer film and film thickness as well as the best practice for surface treatment.

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