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Integration of supercapacitors into printed circuit boards

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ABSTRACT

Physically integrated energy storage devices are gaining increasing interest due to the rapid development of flexible, wearable and portable electronics technology. For the first time, supercapacitor components have been integrated into a printed circuit board (PCB) construct. This proof-of-concept study paves the way for integrating supercapacitors into power electronics devices and hybridising with PCB fuel cells. Commercial Norit activated carbon (NAC) was used as the electrode material and was tested in two types of electrolytes, sodium sulfate (Na₂SO₄) aqueous electrolyte, and Na₂SO₄-polyvinyl alcohol (Na₂SO₄-PVA) gel electrolyte. Electrochemical measurements compare the SC-PCBs to standard two-electrode button-cell supercapacitors. A volumetric energy density of 0.56 mW h cm⁻³ at a power density of 26 mW cm⁻³ was obtained in the solid-state SC-PCB system, which is over twice the values acquired in the standard cell configuration. This is due to the removal of bulky components in the standard cell, and/or decreased thickness of the overall device, and thus a decrease in the total volume of the SC-PCB configuration. The results show great potential for embedding supercapacitors into PCBs for a broad range of applications. In addition, further advantages can be realised through close physical integration with other PCB-based electrochemical power systems such as fuel cells.

1. Introduction

Supercapacitors (SC), also known as electrochemical capacitors or ultracapacitors, are charge-storage devices, consisting of two parallel electrodes, an electrolyte and usually a separator that electrically isolates the electrode compartments [1]. Supercapacitors have gained increasing interest in a range of applications including automotive and hybrid electrochemical systems due to their long cycle life, rapid charging-discharging and high power density compared to batteries [2,3]. However, the major drawback of supercapacitors is their low energy density, and hence the development of new electrode materials to meet the requirements of both high energy and power densities is gaining increasing interest [4]. Supercapacitors are classified into two main categories: i) electrical double-layer capacitors (EDLCs), mainly comprising of carbon materials, in which the charge is stored electrostatically at the electrode surface, and ii) pseudo-capacitors, including metal oxides and conductive polymers, whereby Faradaic reversible reactions between the electrode materials and the electrolyte ions, govern the charging and discharging mechanism [5]. Hybrid SCs with asymmetrical configurations have recently gained increasing interest, offering higher energy and power densities [6–8]. While extensive research has been, and continues to be, dedicated to developing improved materials for supercapacitors, much less attention has gone into looking at how they are packaged and physically integrated with other power sources to form unitised hybrid systems. This proof-of-concept study shows how supercapacitors can be integrated into printed circuit board (PCB) structures. The PCB construct offers a low-cost easily manufactured means of making supercapacitors and can directly integrate supercapacitors with PCB fuel cells to make for a highly flexible hybrid power source.

The advantages associated with integrating fuel cells into PCBs are well known. Polymer electrolyte membrane fuel cells (PEMFC), direct

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formic acid fuel cells (DFAFC) and direct ethanol fuel cells have been fabricated using the printed circuit board (PCB) technology [9–12], and the approach is being commercialized by the likes of Bramble Energy in the UK [13]. Printed circuit boards typically consist of fiberglass/epoxy composites, coated with a thin layer of conductive copper [14]. The flow-field can be constructed from the insulating composite and the conducting layer (typically protected with an anti-corrosive layer) can act as the current collector. The PCB approach to constructing fuel cells provides a range of advantages, including robustness, rapid manufacture times, low cost and design flexibility [10]. The scope of integrating fuel cell and supercapacitor technology into a single integrated construct offers the opportunity to improve fuel cell performance with only marginal increases in cost, weight and volume.

While the concept of PCB encapsulated capacitors has been asserted in the patent literature [15,16], to our knowledge this is the first time that a supercapacitor PCB has been demonstrated.

Supercapacitors using commercially available carbon electrodes have demonstrated good specific capacitances as high as 120 F g^{-1} in aqueous electrolyte [17–19], in addition to high power densities ranging between 10 and 20 kW kg⁻¹ [20,21]; such standard materials have therefore been used for integration into PCB composites using neutral electrolyte without a separator. In addition, an all-solid-state supercapacitor PCB has been fabricated using a gel electrolyte. Results are compared using the same materials in standard coin cells.

2. Experimental

2.1. Materials

Norit activated carbon (NAC) was provided by CABOT Corporation (Georgia, USA). Polyvinylidene fluoride (PVDF) was supplied by PI-KEM Ltd. (Staffordshire, UK) and Whatman glass microfiber filter papers, used as electrical separator materials, were purchased from Sigma-Aldrich Ltd (UK). Sodium sulphate (Na₂SO₄) and polyvinyl al-cohol (PVA) were used as electrolytes in the PCBs and were supplied by Fisher Scientific (UK) and Tokyo Chemical Industry Co. Ltd (Japan), respectively. All prepared electrodes were manually cast on an Arlon DiClad PCB configuration manufactured by Arlon Electronic Materials (UK) and supplied by ZOT Engineering Limited (UK). The pre-impregnated (prepreg) composite bonding fibers, (Arlon-47 N) were also manufactured by Arlon Electronic Materials (UK) and supplied by ZOT Engineering Limited (UK). Nickel foam (Suzhou JSD Co. Ltd., China) was used as the current collector in coin cell devices of CR2032 geometry (Hohsen Corporation, Japan).

2.2. Preparation of electrode materials and gel electrolyte

Commercial carbon, NAC, was used as the electrode material and was mixed with PVDF binder with 95:5 wt.% composition in N-methyl-2-pyrrolidine (NMP) solvent. The carbon pastes were manually cast onto the PCB, and on nickel foam current collector for the coin cell devices, to obtain a constant mass loading of 2 mg cm^2 of active electrode materials.

The flexible gel electrolyte was prepared by mixing 6 g PVA powder in 60 ml deionised water using magnetic stirring at 90 °C. 6 g of 0.5 M Na_2SO_4 was subsequently added to the mixture and stirred until the solution became clear.

2.3. PCB properties and assembly of the supercapacitor-PCBs

The 0.42 mm thick DiClad PCBs (85 mm \times 70 mm circuit size) were used as supplied. The DiClad laminates, composed of woven fiberglass/polytetrafluoroethylene (PTFE) composites were used as the substrate for the PCB devices. A thin copper film (38 μ m) was electrodeposited on one side of each of the laminates, to provide electrical conductivity, and a conductive carbon ink, which acts as a current collector for the



Fig. 1. PCB layout constituting the base of fiberglass/PTFE composite, a thin layer of copper coating along with another layer of carbon ink on which the activated carbon is deposited on.

supercapacitor PCBs (SC-PCBs), was coated on top of the copper with a thickness of $35.5 \,\mu\text{m}$ and 4 cm diameter circle. The prepared carbon electrode pastes were cast on the conductive ink area and vacuum dried overnight to give a final total mass of $2 \,\text{mg cm}^{-2}$ of physical surface area. Fig. 1 and S1 illustrate the PCB design used prior to assembly.

For the PCB device in aqueous electrolyte medium, the electrodecoated DiClads were separated by two A-47 N prepregs, each of $80 \,\mu\text{m}$ thickness, prior to hot pressing. The prepregs were laser-cut to the same dimensions of the PCBs using a CO₂ laser cutter/engraver LS3020 (HPC Laser Ltd, UK) and pre-vacuumed at room temperature for 2 h. The whole device, composed of the PCB and two prepregs, was then hot pressed at 140 °C (curing temperature of prepregs) at a pressure of 2 bar for 1 h in a multilayer press (RMP 210, Bungard, Germany). The final thickness of each of the prepregs ranged between 40 μm and 50 μm after the curing process in the hot press. The SC-PCB device was then irrigated with 0.5 M Na₂SO₄ solution via an access hole in the PCB which was subsequently taped to prevent any electrolyte loss.

The all-solid-state PCB was prepared using the same procedure as the aqueous one without using the prepregs material. The Na₂SO₄-PVA gel electrolyte was placed between both electrodes along with Whatman filter papers, acting as a separator and gently pressed. Fig. 2a and b demonstrates the layering of the two different configurations of the supercapacitor PCB assemblies, with gel and aqueous electrolytes, respectively.

2.4. Characterization

Suitable characterization of the electrode materials used in the SC-PCB was necessary in order to make a fair comparison with other systems reported in the literature. The surface morphology of the activated carbon was examined using scanning electron microscopy (SEM) operating at 10 kV (EVO MA10, ZEISS, Germany). Degassing of the carbon sample at 300 °C using a sample degas system (VacPrep 061 Sample Degas System, Micrometrics, USA) was followed by collection of nitrogen sorption isotherms (3Flex Surface and Catalyst Characterization System, Micromeritics, USA). The total pore volume of the activated carbon was calculated at a relative pressure (P/P_o) of 0.99 and specific surface areas (SSA) were obtained using Brunauer-Emmet-Teller (BET) method at relative pressure range between 0.001 and 0.2 [22]. Nonlocal density functional theory (NLDFT) was used to determine the micropore volume [23].

2.5. Electrochemical measurements

Electrochemical measurements were carried out in the aqueous and all-solid-state coin cell configurations and SC-PCBs using a potentiostat (Interface 1000, Gamry Instruments, USA). Cyclic voltammetry (CV) at scan rates of 1, 2, 5, 10, 50, 100 and 200 mV s⁻¹ were performed in the voltage range 0⁻² V.

The specific capacitance was evaluated from the CV curves using Eq. (1):

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