



A method to produce binderless supercapacitor electrode monoliths from biomass carbon and carbon nanotubes



B.N.M. Dolah^a, M. Deraman^{a,*}, M.A.R. Othman^a, R. Farma^{a,b}, E. Taer^{a,b}, Awitdrus^{a,b}, N.H. Basri^a, I.A. Talib^a, R. Omar^a, N.S.M. Nor^a

^a School of Applied Physics, Faculty of Science and Technology, Universiti Kebangsaan Malaysia, Bangi, Selangor 43600, Malaysia

^b Department of Physics, Faculty of Mathematics and Natural Sciences, University of Riau, Pekanbaru, Riau 28293, Indonesia

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ABSTRACT

Binderless supercapacitor electrode monoliths (BSEM), prepared via the carbonization and activation of green monoliths from (a) self-adhesive carbon grains (SACG) from oil palm empty fruit bunch fibers, (b) SACG mixed with KOH, and (c) mixtures of SACG, KOH, and varying percentages of carbon nanotubes (CNTs), were characterized and evaluated in symmetrical supercapacitor cells. The porosity and the structural and microstructural characteristics of the electrodes are influenced by KOH and CNTs. The electrodes containing CNTs have a relatively lower specific capacitance but exhibit lower equivalent series resistance values and hence can sustain or improve the specific power of the cells, suggesting the need to optimize the quantity of CNTs used to sustain higher specific capacitance above 100 F/g. This innovative process uses inexpensive SACG with relatively small quantities of CNTs and KOH with no binder, and it directly combines both chemical (KOH) and physical (CO₂) activation during the production of BSEM.

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1. Introduction

Supercapacitors are electrochemical storage devices whose ranges of energy and power densities lie between those of batteries and conventional dielectric capacitors. Supercapacitors can deliver higher power than batteries and can store more energy, but they have lower power than conventional capacitors. These capabilities allow supercapacitors to be used as a complimentary or back-up power source in a wide range of electronic devices or systems. A supercapacitor stores its energy at the interface between the electrode and electrolyte. The commonly use electrodes are activated carbon [1,2], metal oxide [3,4], and electronically conducting polymer [5]. For activated-carbon-based supercapacitors, the energy is stored in electrostatic charges (ionic charges from the electrolyte and electronic charges from the electrode) formed at the activated carbon/electrolyte double layer interface, and high capacitance can be achieved because of the high specific surface area of the porous activated-carbon electrode.

Carbon-based supercapacitors are widely developed and well established because of their low cost, large capacitance, and long

life-cycles; however, active research to improve the deliverable power still continues because activated carbon itself does not have high enough electronic conductivity (or low resistance). These limitations are particularly applicable when the activated carbon has a very high surface area, thereby preventing the aim of producing supercapacitors with a very low equivalent series resistance (ESR). The power of a supercapacitor is inversely proportional to its ESR. CNTs have been widely used to produce composite CNT/activated carbon supercapacitor electrodes with improved ESR values because of their excellent electronic conductivity and other beneficial properties, such as hollow tube structure, nanometer dimensions, and reasonably high surface area [6,7]. The specific capacitance of CNTs was found to be lower than that of activated carbon; therefore, physical/chemical activation was used prior to or during composite electrode preparation to increase the resultant specific capacitance [8]. It has also been reported that the use of a binding agent with CNTs and activated carbon may obstruct pores, reducing the resulting specific capacitance of the composite electrode [9]. Therefore, a number of solutions have been proposed to overcome this drawback associated with binder usage [10,11].

The present study proposes a new method to prepare electrodes for supercapacitor application without using any binder. The activated carbon composite monoliths (ACM) were prepared from the carbonization (N₂) and activation (CO₂) of the binderless

* Corresponding author. Tel.: +60 3 8921 5902; fax: +60 3 8921 3777.

E-mail addresses: madra@ukm.edu.my, mderaman113@gmail.com (M. Deraman).

green monoliths containing (1) SACG from EFB fibers, (2) a mixture of SACG and KOH, and (3) a mixture of SACG, KOH, and a varying amount of CNTs. The binderless method offers advantages such as omission of binder, the use of inexpensive SACG, and a very small quantity of CNTs; this method also employs a direct combination of both chemical (KOH) and physical (CO₂) activation processes. The objective of the present study is to observe the effect of varying CNT content on the physical and electrochemical properties of the ACM electrodes, and, more specifically, the supercapacitive performance of the cells fabricated using these ACM electrodes. The physical properties of the ACM electrodes were investigated using field emission scanning electron microscope (FESEM), X-ray diffraction (XRD), and nitrogen adsorption–desorption isotherm methods. Symmetrical supercapacitor cells were fabricated using these ACM electrodes, and their electrochemical performance was evaluated using electrochemical impedance spectroscopy (EIS), cyclic voltammetry (CV), and galvanostatic charge–discharge (GCD) methods.

2. Experimental

2.1. Sample preparation

SACGs with particle size less than 106 microns were prepared from EFB fibers (Ecofibre Technology Sdn. Bhd.) by pre-carbonization (Furnace CTMSB46), milling for 36 h (ball mill AC Motor BS 500-110), and sieving (Matest 24030 Brembate Sopra (BG)) [12]. Three types (designated as 1, 2, and 3) of green monoliths (GMs) were prepared according to the composition shown in Table 1. For convenience, the carbon monoliths (CMs) and ACMs were labeled by their batch number, type, and CNT content, as shown in column 6 of Table 1. The first, second, third, fourth and fifth refer the experimental batches carried out on the respective samples. Type 1, type 2 and type 3 refer to the GMs with 100 wt% SACG, 95% SACG + 5 wt% KOH, 90 wt% SACG + 5 wt% KOH + 5 wt% CNT, respectively. It should be noted here that all type 2 samples in all batches are identical in terms of weight percentage composition. A similar rule was applied for all type 3 samples in all batches. The CNTs used in this study was multi-walled CNTs (MWCNTs), with 50–100 nm in diameter, 5–10 μm in length and a surface area of approximately 40 m²g⁻¹ (Nano Amor Nanostructured & Amorphous Materials, Inc. TX, USA, manufactured on February 19th, 2009). Fig. 1 shows a Raman spectra of MWCNTs recorded between 500 and 2000 cm⁻¹, which shows typical D band at 1344 cm⁻¹ and G band at 1568 cm⁻¹ for this material.

For the type 1 and type 2 GMs, 30 g of SACG and 28.5 g of SACG mixed with 1.5 g of KOH (5 wt%) were each poured into 300 ml of boiling water, stirred for 1 h, and dried in an oven at 100 °C for 48 h. For milling, 10 g of the dried SACG and 10 g of the SACG and 5 wt% KOH mixture, respectively, were milled for 20 min to obtain homogeneous powder samples. For the type 3 GMs, a dry mixture

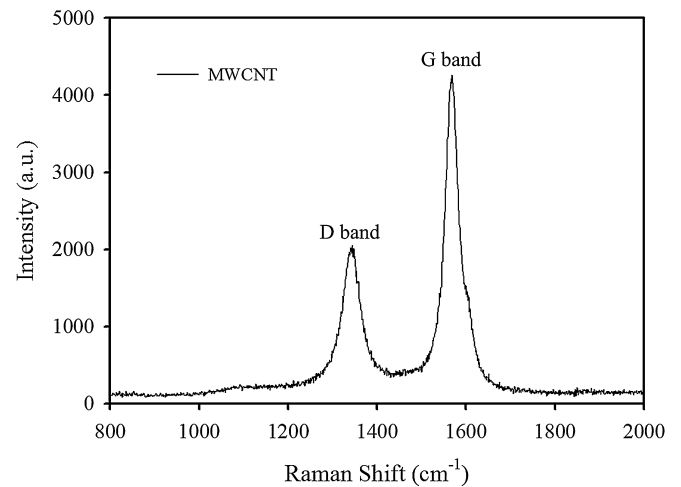


Fig. 1. Raman spectra of MWCNTs.

of SACG, KOH, and CNTs based on the weight-percentage in Table 1 was also prepared using similar steps. Finally, 0.75 g of the powder from each GM type was poured into a 20 mm-diameter mold and pressed using a pelletizing machine (VISITEC 2009-Malaysia) to convert the powders into GMs without using any binders.

Five sets of GMs were carbonized separately, according to the batches shown in Table 1, into their respective CMs in a carbonization furnace (Vulcan Box Furnace 3-1750) under a 1.5 l/min flow of N₂ gas and heated to 800 °C using our previous multi-step heating profile [13,14]. The samples were then activated with CO₂ (1.5 l/min) at 800 °C for 3 h with a heating rate of 5 °C per minute to produce five batches of ACMs. After being polished to a thickness of 0.4 mm, the ACMs were used as electrodes in symmetrical supercapacitor cells using 316L-stainless steel as the current collector, H₂SO₄ (1 Molar) as the electrolyte, and a Teflon ring (0.1 mm thick) as the separator. All five sets of prepared cells were labeled as Cell 110, Cell 120, etc., based on the label designated for the ACM precursor.

2.2. Characterization

The weight (measuring tool: Mettler Toledo AB204), thickness and diameter (measuring tool: Mitutoyo 193-253) of the monoliths were measured to determine the density of the GMs, CMs, and ACMs. FESEM (Zeiss SUPRA 55VP) was used to study the microstructures of the CMs and ACMs. XRD (Bruker AXS: model D8 Advance, 1.5406 Å wave length of incident beam, generated from a CuK_α of X-ray sources) was used to record the XRD patterns of the ACMs. The nitrogen adsorption–desorption isotherm

Table 1
Compositions of green monoliths and label for all the monoliths.

Batch	Type	Composition (% by weight)				Monoliths label		
		SACG	KOH	CNTs	GMs	CMs	ACMs	
1	1	100	0	0	GM110	CM110	ACM110	
	2	95	5	0	GM120	CM120	ACM120	
2	2	95	5	0	GM220	CM220	ACM220	
	3	92	5	3	GM233	CM233	ACM233	
3	2	95	5	0	GM320	CM320	ACM320	
	3	91	5	4	GM334	CM334	ACM334	
4	2	95	5	0	GM420	CM420	ACM420	
	3	90	5	5	GM435	CM435	ACM435	
5	2	95	5	0	GM520	CM520	ACM520	
	3	89	5	6	GM536	CM536	ACM536	

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