FABRICATION OF GRAPHENE-BASED FLEXIBLE SUPERCAPACITORS IN PLANAR- AND FIBER-STRUCTURED CONFIGURATIONS

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By

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April 2019

Chair : Associate Professor Janet Lim Hong Ngee, PhD
Faculty : Science

With the emergence of flexible electronic devices, flexible supercapacitors have attracted widespread interest in developing lightweight, thin and efficient portable/wearable energy storage devices. Along with the general information about flexible supercapacitors, this thesis focuses on flexible supercapacitors including the planar-structured flexible supercapacitors as well as the new-type fiber supercapacitors. Thus, in this thesis, the construction of electroactive materials on the flexible substrates and feasible strategies to achieve high-performance flexible supercapacitors were discussed. In the planar-structured flexible supercapacitors, aluminium carbide was being employed as a current collector, where it is light, thin and highly flexible. The simultaneous exfoliation and reduction of graphene-based materials by rapid microwave irradiation were employed to generate a microwave graphene mix (MGM). To demonstrate the supercapacitors application, a supercapacitor device were constructed and yielded a specific capacitance value of 78.1 F g^{-1} using a solid-state electrolyte with excellent cycling stability of 93.8% after 1000 cycles of charge/discharge. Then, the as-prepared MGM was mixed with polypyrrole (PPy) to further enhance the electrochemical performance. A supercapacitor device using MGM-PPy as an electroactive material recorded a specific capacitance value of 137.2 F g^{-1} which is 1.8 times higher than that of MGM with cycling stability of 89.9% after 1000 cycles of charge/discharge.

Different from the planar-structured supercapacitors, the fiber-structured was fabricated through a simple electrochemical deposition process of polypyrrole/reduced graphene oxide onto the surface of carbon bundle fiber. The surface morphology revealed a high degree of porosity in the PPy-rGO-2 composite; facilitating the ionic penetration, leading to an excellent

electrochemical performance. The PPy-rGO-2 exhibits good electrochemical performance (96.2 F g^{-1}) with an energy density of 13.4 Wh kg^{-1} and a power density of 322.9 W kg^{-1}. However, after a series of charging-discharging cycles, the electrochemical performances of the PPy-rGO-2 deteriorated due to the changes in the structural properties such as the reduction in pore size, and transformation of the structure of rGO from amorphous to graphitic. To investigate the mechanical bendability/flexibility of the as-fabricated supercapacitor devices, both planar- and fiber-structured supercapacitor devices were bent at various angles and revealed that the bending had nearly no effect on the specific capacitance values. The combination of solid-state electrolyte and flexible current collector with flexible free-standing electroactive materials made up of graphene-based materials and PPy, capable of withstanding stress with no drastic changes in its electrochemical performance, demonstrating an excellent mechanical bendability. Overall, the sustainable electrochemical performance, mechanical flexibility, low-cost and lightweight, flexible supercapacitors are undoubtedly emerging as promising renewable energy technology for future energy storage systems.

.

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FABRIKASI SUPERKAPASITOR BOLEH LENTUR BERDASARKAN BAHAN *GRAFIN* DALAM KONFIGURASI BERBENTUK SATAH DAN GENTIAN

Oleh

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Dengan kemunculan alat-alat elektronik yang boleh lentur, superkapasitor boleh lentur telah menarik minat yang meluas dalam membangunkan peranti penyimpanan mudah alih yang nipis dan ringan. Melalui maklumat umum mengenai superkapasitor boleh lentur, tesis ini memfokuskan kepada superkapacitor boleh lentur, termasuk superkapasitor yang berstruktur satah serta jenis baru yang berstruktur gentian. Oleh itu, dalam tesis ini, pembinaan bahan-bahan elektroaktif pada substrat yang boleh lentur dan strategi yang boleh dilaksanakan untuk mencapai superkapasitor yang berprestasi tinggi telah dibincangkan. Dalam superkapasitor boleh lentur berstruktur satah, karbida aluminium telah digunakan sebagai pengumpul arus, di mana ianya ringan, nipis dan sangat lentur. Pengelupas dan pengurangan bahan *grafin* dan derivatifnya secara serentak dengan menggunakan penyinaran gelombang mikro yang mudah dan cepat telah digunakan untuk menghasilkan campuran gelombang mikro *grafin* (MGM). Untuk menunjukkan aplikasi superkapasitor, peranti superkapasitor telah dibina dan berjaya menghasilkan nilai kapasitif spesifik sebanyak 78.1 F g^{-1} dengan menggunakan elektrolit keadaan pepejal dan mengekalkan pengekalan kapasitif yang cemerlang iaitu sebanyak 93.8% selepas 1000 kitaran cas berterusan. Kemudian, MGM yang disediakan telah dicampurkan dengan *polipirrol* (PPy) untuk meningkatkan lagi prestasi elektrokimia. Peranti superkapasitor menggunakan MGM-PPy sebagai bahan elektroaktif mencatatkan nilai kapasitif spesifik 137.2 F g^{-1} iaitu 1.8 kali lebih tinggi daripada MGM dengan kestabilan kapasitif sebanyak 89.9% selepas 1000 kitaran caj/nyahcas.

Berbeza dari superkapasitor berstruktur satah, superkapasitor berstruktur gentian telah dibina melalui proses deposit bahan elektroaktif PPy and *grafin* ke atas permukaan gentian ikatan carbon. Morfologi permukaan mendedahkan tahap keliangan yang tinggi dalam komposit PPy-rGO-2; memudahkan penembusan ionik, membawa kepada prestasi elektrokimia yang sangat baik. PPy-rGO-2 mempamerkan prestasi elektrokimia yang baik (96.2 F g^{-1}) dengan ketumpatan tenaga 13.4 Wh kg^{-1} dan ketumpatan kuasa 322.9 W kg^{-1}. Walau bagaimanapun, selepas beberapa siri kitaran pengecasan, prestasi elektrokimia PPy-rGO merosot disebabkan oleh perubahan sifat-sifat struktur seperti pengurangan saiz liang, dan transformasi struktur rGO daripada amorfus kepada grafit. Untuk menyiasat kebolehan boleh lentur secara mekanikal peranti supercapacitor yang telah dibuat, kedua-dua superkapasitor yang berstruktur satah dan gentian telah dibengkokkan di pelbagai sudut dan ia mendedahkan bahawa lenturan itu tidak memberi kesan ke atas nilai kapasitapan spesifik. Gabungan elektrolit keadaan pepejal dan pengumpul arus dengan bahan elektroaktif yang boleh lentur yang terdiri daripada bahan berasaskan *grafin* dan PPy, mampu menahan tekanan tanpa memberikan perubahan drastik dalam prestasi elektrokimia, menunjukkan kebolehan mekanikal yang sangat baik. Secara keseluruhannya, prestasi elektrokimia yang mampan, kebolehan boleh lentur secara mekanikal, kos yang rendah dan ringan, superkapasitor boleh lentur tidak dapat dinafikan lagi sebagai teknologi tenaga boleh diperbaharui untuk sistem penyimpanan tenaga pada masa akan datang.

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I certify that a Thesis Examination Committee has met on 26 April 2019 to conduct the final examination of Hamra Assyaima Binti Abdul Bashid on her thesis entitled "Fabrication of Graphene-Based Flexible Supercapacitors in Planar- and Fiber-Structured Configurations" in accordance with the Universities and University Colleges Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) 106] 15 March 1998. The Committee recommends that the student be awarded the Doctor of Philosophy.

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LIST OF ABBREVIATIONS

AC	Activated carbon
ACF	Activated carbon fiber
ANS	6-amino-4-hydroxy-2-naphthalenesulfonic acid
CBF	Carbon bundle fiber
CNT	Carbon Nanotubes
CH_3COOK	Potassium acetate
Cm	specific capacitance
CPE	Contact phase elements
CV	Cyclic voltammetry
EDLC	Electrochemical double-layer capacitors
EIS	Electrochemical impedance spectra
EMIM-NTf_2	1-ethyl-3-methylimidazolium bis(trifluoromethylsulfonyl)amide
ESR	Equivalent series resistance
FESEM	Field emission scanning electron microscope
FG	Functionalized graphene
FT-IR	Fourier transform infrared
GCD	Galvanostatic charge/discharge
GNP	Graphene nanoplatelet
GO	Graphene oxide
HCl	Hydrogen chloride
HNO_3	Nitric acid
H_2O_2	hydrogen peroxide
H_2SO_4	Sulfuric acid
H_3PO_4	Phosphoric acid
I_D	D-band intensity

I_G	G-band intensity
IL	Ionic liquid
KOH	Potassium hydroxide
$KMnO_4$	Potassium permanganate
$Mg(OH)_2$	Magnesium hydroxide
MGM	Microwave graphene mix
MGM-PPy	Microwave graphene mix-Polypyrrole
MGO	Microwave graphene oxide
MGNP	Microwave graphene nanoplatelets
MnO_2	Manganese(IV) oxide
MWCNT	Multi-walled carbon nanotubes
N-CNT	Nitrogen doping carbon nanotubes
NaOH	Sodium hydroxide
NapTs	Sodium p-toluenesulfonate
$Ni(OH)_2$	Nickel hydroxide
NMP	N-methyl-2-pyrrolidione
PAA	Poly(amic acid)
PAN	Polyacrylonitrile
PANI	Polyaniline
PIL	Poly(ionic liquid)
PPy	Polypyrrole
PPy-rGO	Polypyrrole-Reduced graphene oxide
PPy-rGO-2	Polypyrrole-Reduced graphene oxide-2
Pt	Platinum
PVA	Polyvinyl alcohol
PVDF	Poly(vinylidene fluoride)
R_{ct}	Charge transfer resistance

R_c	Contact interface resistance
R_s	Bulk solution resistance
rGO	Reduced graphene oxide
$RuCl_3$	Ruthenium chloride
SCE	Saturated calomel electrode
SDS	Sodium dodecylsulfonate
SDBS	Sodium dodecylbenzene sulfonate
SEM	Scanning electron microscope
SWCNT	Single-walled carbon nanotubes
TBAOH	Tetrabutylammonium hydroxide
TGA	Thermogravimetric analysis
TiO_2	Titanium oxide
XPS	X-ray photoelectron spectroscopy
XRD	X-ray diffraction

CHAPTER 1

INTRODUCTION

1.1 Flexible Supercapacitors

The rapid growth of next-generation portable electronics has led to intensive efforts to develop supercapacitor with flexible, rigid, small, lightweight, eco-friendly, and high storage capacity (Lu, *et al*. 2014). Supercapacitor, which are also known as electrochemical capacitors, offer a promising alternative approach to energy storage devices because of their ability to store and deliver a high power density, and long life cycle with short charging time, simply by utilising the charge-separation of the electrochemical interface between the electrode and electrolyte (Sun, *et al*. 2016; Wang, *et al*. 2009; Zhang, *et al*. 2010). The conventional two-electrode system supercapacitor is planar-structured, consisting of two active electrodes kept apart by an electrolyte as an indispensable and electrically insulating separator (Shao, *et al*. 2015).

Fiber-structured supercapacitor are commonly built on fibrous or interwoven substrates, and can be directly integrated into a wearable and embedded device units in sensors, environmental monitoring, display, and implanted medical devices (Cai, *et al*. 2014). Metal-based fibers such as aluminium wires have previously been used as a current collector or core electrode because of its high conductivity and ease of availability. However, the performance is limited due to its heaviness and easily oxidised under ambient conditions (Le, *et al.* 2013; Nam, *et al.* 2011). Carbon-based fibers, like carbon microfibers and graphene fibers, have been used to replace metal-based fibers owing to its great flexibility, light weight, high mechanical strength, high conductivity, and stability under ambient conditions (Le, *et al*. 2013; Li, *et al*. 2011).

1.2 Graphene-based Electroactive Materials

The choice of electroactive materials also plays important roles in determining the electrochemical performances of supercapacitor devices. Graphene has been studied extensively as an electroactive material for supercapacitor and often suggested as a replacement for activated carbon in a supercapacitor, in part due to its promising properties such as large (theoretical) surface areas, high charge carrier mobility, excellent conductivity, high mechanical strength, and extremely high thermal conductivity (Antiohos, *et al.* 2013; Frackowiak and Beguin 2001; Wu, *et al.* 2011), with the ability to store and release energy through the separation of electronic and ionic charges in the electrode and electrolyte interface (Berger, *et al.* 2006; Lee, *et al*. 2008; Stankovich, *et al*. 2006; Stoller, *et al*. 2008). Also, being a material made up of one single atomic layer, it is lighter and graphene is an ecologically friendly, unlike most other forms of energy storage due to its essential form of carbon (Liu, *et al*. 2010).

Graphene was first obtained by mechanical exfoliation of graphite using scotch tape method (Yi and Shen 2015). Although scotch tape method has been reported to be successful for production of single-layer graphene with high structural and electronic quality, this method is generally unfavorable because of time-consuming and have low production yields (Geim 2009). Instead of cleaving graphite manually, other forms of graphene-based materials, including graphene oxide (GO) has been used as a precursor for the affordable and large-scale production of graphene (Huang, *et al.* 2011). Moreover, using GO as a starting material can provide good dispersion stability and prevent aggregation in the reaction solution (Lim, *et al.* 2013). The promising properties together with the ease of processibility and functionalization make graphene-based materials ideal candidates for incorporation into a variety of functional materials for supercapacitor applications.

1.3 Hybridization of Graphene-based Electroactive Materials

One of the most prominent electroactive materials for supercapacitor electrodes is formed by the hybridization of carbon-based materials (i.e., activated carbon, CNT, and graphene) with a conducting polymer (i.e., polyaniline, polypyrrole, or poly[3,4-ethylenedioxythiophene]) (Fusalba, *et al*. 2001; Futaba, *et al*. 2006; Laforgue 2011; Lota, *et al.* 2008; Meng, *et al.* 2009; Sharma, *et al.* 2008; Snook, *et al.* 2011; Wang, *et al.* 2014; Wu, *et al.* 2010; Zeng, *et al.* 2012). A hybrid composite is formed to utilize the relative advantages (i.e., the high specific surface area, good mechanical properties, and excellent electrochemical stability) and reduce the relative disadvantages (i.e., the limited energy storage, lower rate capability, and cycling stability) of electrochemical double-layer capacitors (EDLC) and pseudocapacitors to realize better electrochemical performances (Frackowiak, *et al.* 2001; Jin, *et al.* 2011; Jurewicz, *et al.* 2001; Ng, *et al.* 2014).

A hybrid capacitor has achieved high energy and power densities than EDLC and pseudocapacitors alone by utilizing both non-faradaic and faradaic processes to store a charge (Stoller, *et al.* 2008). The carbon-based materials increase the contact between the deposited materials and electrolyte through the high-surface-area backbone. Meanwhile, the conducting polymer provides the faradaic reactions to further enhance the capacitance by undergoing a redox reaction to store a charge in the bulk of the material and hence increase the energy stored and reduce self-discharge (Frackowiak, *et al.* 2006). Thus, the synergistic effect from the hybridization of carbon-based materials and conducting polymers is believed to be able to improve the capacitance and stability of a supercapacitor device (An, *et al.* 2010; Lee, *et al.* 2011).

1.4 Problem Statements

Though planar-structured supercapacitor is a conventional two-electrode system supercapacitor device, it is suffer from large, bulky, and heavy, which is not suitable to be used for portable electronic devices. Thus, to address this issue, much effort has been devoted to develop a fiber-structured supercapacitor that are flexible, lightweight, and easily shaped in portable electronic devices (Huang, *et al*. 2016; Le, *et al.* 2013; Liang, *et al*. 2015).

Although graphene-based materials including graphene oxide (GO) and graphene nanoplatelets (GNP) has re-emerged as an intensive research interest as active materials in a supercapacitor, due to the attached oxygen functional groups, GO is electrically insulating and various reduction methods have been developed to restore its electrical conductivity (Eda, *et al.* 2009; Hsiao, *et al.* 2013). While for GNP, due to the high inter-sheet van der Waals attractions between graphene layers, it tends to agglomerate and restacks together (Yan, *et al.* 2012). Consequently, minimized the specific surface area and electron mobility of that individual graphene layers as well as reduced the electrochemical performances in a supercapacitor (Sridhar, *et al.* 2013).

In addition, low cycling stability on the hybrid supercapacitor was found recently where the polypyrrole-graphene oxide-zinc oxide, only possible to retain up to ~77% of their capacitance after 500 cycles of charging-discharging at a current density of 1 A g^{-1} (Chee, *et al.* 2015). Moreover, other reported results for a polypyrrole/carbon aerogel showed that it only exhibited a ~55% of capacity retention after 500 cycles of charging-discharging (An, *et al.* 2010). An enormous amount of effort on the various spectroscopic and electrochemical studies of polypyrrole-based composites supercapacitor (Mosch, *et al.* 2015), however the understanding of their capacity retention after a series of charging-discharging cycles is still limited.

1.5 Scope of Research

In this research, I will focus on fabrication of flexible symmetrical solid-state supercapacitors in which both planar- and fiber-structured supercapacitors to be used as a flexible current collector together with polyvinyl alcohol-potassium acetate (PVA-CH_3COOK) served as an indispensable solid-state electrolyte. While the graphene-based materials together with the PPy acts as electroactive materials in the supercapacitor devices. Also, the behavior of solid-state supercapacitor devices after a series of charging-discharging cycles will be evaluated by performing the electrochemical performances and structural configuration of the electrode. The flexibility of the as-fabricated solid-state supercapacitor device will also be carried out as a proof-of-concept.

1.6 Research Objectives

This research is to fabricate a supercapacitor device that relies on the combination of graphene-based materials with polypyrrole. The main objective of this thesis is to investigate the flexible supercapacitor including the planar- and fiber-structured, and to use the graphene-based materials together with polypyrrole in the fabrication of supercapacitor device. Particular attention will be devoted to overcoming current problems and to improve the overall performances. Specifically, the objectives of the study involve:

i. To construct a flexible supercapacitors device using different types of current collectors, aluminium carbide (planar-structured) and carbon bundle fiber (fiber-structured) supercapacitors.
ii. To assemble a hybrid materials possessing electrical double layer capacitance like graphene-based materials, and pseudocapacitance like polypyrrole.
iii. To investigate the stability of electroactive materials after a series of charge/discharge cycles.

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