UNIVERSITI PUTRA MALAYSIA

STRUCTURAL AND ELECTROCHEMICAL PROPERTIES OF NICKELCOBALT OXIDE/ACTIVATED CARBON FOR SUPERCAPACITOR APPLICATION

CHANG SOOK KENG

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By

CHANG SOOK KENG

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirement for the Degree of Doctor of Philosophy

December 2012

Dedicated with much respect and gratefulness
to
my mentors,
who sparkled me with inspiring thoughts and undivided guidance,
to
my parents and brothers,
who showered me with their loving help, patience, understanding and support,
to
my loved one,
for his time, sacrifice and fascinating ideas that always be my great source of illumination

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Doctor of Philosophy

STRUCTURAL AND ELECTROCHEMICAL PROPERTIES OF NICKEL-COBALT OXIDE/ACTIVATED CARBON FOR SUPERCAPACITOR APPLICATION

By

CHANG SOOK KENG

December 2012

Chairman: Professor Zulkarnain Zainal, PhD

Faculty: Science

Co-precipitation method was adopted in the preparation of nickel-cobalt oxides for potential application in supercapacitors. The formation of spinel nickel-cobalt oxide, $NiCo_2O_4$ prepared by oxalate co-precipitation started below 400 °C as confirmed by X-ray diffraction (XRD) analysis. Single phase nickel-cobalt oxide with cation ratio of 1:2 (Ni:Co) was obtained at calcination temperature of 400 °C. The spinel phase decomposed gradually until 700 °C. The calcination time for the formation of $NiCo_2O_4$ was found to be between 2 to 4 hours. The particle size of the prepared sample studied by transmission electron microscopy (TEM) showed a value of 9.5 nm. Investigation on the compositional effect of $NiCo_2O_4$ revealed that the crystallinity of the synthesized oxides improved with the increment of Ni content. The entire range of Ni:Co compositions at 400 °C and 700 °C were investigated with respect to the formation of phases, lattice parameter and crystallite size. Nickel-cobalt oxide series was prepared through solid-state route as well. However, $NiCo_2O_4$ co-existed with NiO in this method preparation. Moreover, solid-state route produced metal oxides with larger crystallite

size than co-precipitation method. Therefore, co-precipitation served as a better method in synthesizing pure phase nanostructured $NiCo_2O_4$ compared to solid-state technique. The electrochemical properties of $NiCo_2O_4$ were measured in various acidic, neutral and alkaline electrolyte systems (1.0 M HCl, 1.0 M KCl and 1.0 M KOH) by employment of cyclic voltammetry (CV), galvanostatic charge-discharge test and electrochemical impedance spectroscopy (EIS). Ideal capacitor behaviour with the largest operating voltage of 1.0 V and good electrochemical stability were observed in $NiCo_2O_4$ using neutral KCl aqueous electrolyte. Meanwhile, the prepared sample displayed the highest surface redox activity in 1.0 M KOH alkaline electrolyte but showed the lowest electrochemical performance in acidic electrolyte.

Single phase $NiCo_2O_4$ and $NiMn_{0.5}Co_{1.5}O_4$ spinel powders have been synthesized by hydroxide co-precipitation method, and the effects of Mn substitution for Co have been studied. Electrodes of both materials exhibit nearly ideal electrochemical capacitor behaviour in neutral electrolyte solution (1.0 M KCl). Mn substitution greatly enhanced the specific capacitance of the spinel, giving a value of approximate 110 F g^{-1} due to the facile charge-transfer characteristic of the Mn ions, as revealed by in-situ X-ray absorption near-edge structure analysis.

Nickel-cobalt oxide/activated carbon composite was synthesised by adapting oxalate co-precipitation synthesis protocol followed by heat treatment under an open air atmosphere. X-ray diffraction analysis confirmed that nickel-cobalt oxide spinel phase was maintained in the pure and composite phases while transmission electron

microscopy revealed the nanostructured synthesis of nickel-cobalt oxide/activated carbon composite. The specific capacitance which was the sum of double-layer capacitance of the activated carbon and pseudocapacitance of the metal oxide increased with the composition of nickel-cobalt oxide before showing a decrement for heavily loaded electrodes. Utilisation of nickel-cobalt oxide component in the composite with 50 wt. % loading displayed a capacitance value of ~59 F g^{-1} in 1.0 M KCl. The prepared composite electrodes had good electrochemical stability upon cycling with tolerable variation in specific capacitance with increasing charge-discharge cycles.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia
sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

STRUKTUR DAN SIFAT-SIFAT ELEKTROKIMIA NIKEL-KOBALT OKSIDA/KARBON TERAKTIF UNTUK APPLIKASI SUPERKAPASITOR

Oleh

CHANG SOOK KENG

Disember 2012

Pengerusi: Profesor Zulkarnain Zainal, PhD

Fakulti: Sains

Kaedah ko-pemendakan telah digunakan bagi penyediaan nikel-kobalt oksida dalam applikasi yang berpotensi seperti superkapasitor. Pembentukan spinel nikel-kobalt oksida, $NiCo_2O_4$ yang disediakan melalui ko-pemendakan oxalat bermula pada suhu di bawah 400 °C seperti yang disahkan oleh analisis pembelauan sinar-X (XRD). Fasa tunggal nikel-kobalt oksida dengan nisbah kation 1:2 (Ni:Co) diperolehi pada suhu pemanasan 400 °C. Fasa spinel mengurai secara beransur-ansur sehingga 700 °C. Masa pemanasan untuk pembentukan $NiCo_2O_4$ adalah di antara 2 hingga 4 jam. Saiz partikel untuk sampel yang disediakan telah dikaji dengan menggunakan mikroskopi transmisi electron (TEM) yang memberikan nilai sebanyak 9.5 nm. Pengajian kesan komposisi atas $NiCo_2O_4$ mendedahkan peningkatan kehabluran oksida dengan penambahan kandungan Ni. Keseluruhan rangkaian komposisi Ni:Co pada 400 °C dan 700 °C yang dikaji adalah berkenaan dengan pembentukan fasa, parameter kekisi dan saiz hablur. Siri nikel-kobalt oksida juga disediakan melalui cara keadaan pepejal. Walau bagaimanapun, $NiCo_2O_4$ wujud bersama NiO dalam cara penyediaan ini. Tambahan

pula, cara penyediaan keadaan pepejal telah menghasilkan oksida logam dengan saiz hablur yang lebih besar berbanding ko-pemendakan. Oleh itu, ko-pemendakan merupakan cara yang lebih baik bagi sintesis fasa tulen $NiCo_2O_4$ yang bersaiz nano daripada teknik keadaan pepejal. Sifat-sifat elektrokimia $NiCo_2O_4$ dianalisis dalam pelbagai sistem elektrolit seperti asid, neutral dan alkali (1.0 M HCl, 1.0 M KCl dan 1.0 M KOH) melalui kitaran voltammetri (CV), ujian cas-nyahcas galvanostatik dan spektroskopi impedans elektrokimia (EIS). Kapasitor yang bersifat unggul ditunjukkan oleh $NiCo_2O_4$ dalam elektrolit akueus KCl dengan voltan operasi yang terluas sebanyak 1.0 V dan mempamerkan kestabilan elektrokimia. Sementara itu, sampel yang disediakan memaparkan aktiviti redoks yang tertinggi dalam elektrolit beralkali 1.0 M KOH tetapi menunjukkan prestasi elektrokimia yang terendah dalam elektrolit yang berasid.

Fasa tunggal serbuk spinel $NiCo_2O_4$ and $NiMn_{0.5}Co_{1.5}O_4$ telah disediakan dengan cara ko-pemendakan hidroksida dan kesan penggantian Mn ke atas Co juga telah dikaji. Elektrod daripada kedua-dua bahan itu memberikan sifat kapasitor elektrokimia yang hampir unggul dalam larutan elektrolit neutral (1.0 M KCl). Penggantian Mn telah mempertingkatkan kemuatan khusus spinel tersebut dengan memberikan nilai anggaran sebanyak 110 F g^{-1} disebabkan ciri-ciri cas pemindahan ion Mn yang sempurna seperti yang ditunjukkan dalam analisis in-situ sinar-X penyerapan struktur.

Komposit nikel-kobalt oksida/karbon teraktif telah disintesis melalui ko-pemendakan oxalat diikuti dengan rawatan haba di bawah atmosfera udara. Analisis pembelauan

sinar-X mengesahkan bahawa fasa spinel nikel-kobalt oksida dapat dikekalkan dalam fasa tulen dan komposit manakala mikroskopi transmisi elektron membuktikan sintesis komposit nikel-kobalt oksida/karbon teraktif dalam unit nanometer. Kemuatan khusus yang merupakan jumlah kemuatan dua-lapisan karbon teraktif dan pseudo-kemuatan oksida logam telah menujukkan peningkatan dengan komposisi nikel-kobalt oksida sebelum mempamerkan penyusutan dalam elektrod yang telah sarat dimuatkan. Penggunaan komponen nikel-kobalt oksida dalam 50 wt. % muatan komposit memaparkan nilai kemuatan ~59 F g^{-1} dalam 1.0 M KCl. Elektrod komposit yang disediakan mempunyai kestabilan elektrokimia yang baik di mana perubahan kemuatan khusus adalah tidak ketara walaupun bilangan kitaran cas-nyahcas diperbanyakkan.

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I certify that a Thesis Examination Committee has met on 21 December 2012 to conduct the final examination of Chang Sook Keng on her thesis entitled "Structural and Electrochemical Properties of Nickel-Cobalt Oxide/Activated Carbon for Supercapacitor Application" in accordance with the Universities and University College 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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Date: 26 February 2013

This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

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DECLARATION

I declare that the thesis is my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously, and is not concurrently, submitted for any other degree at Universiti Putra Malaysia or at any other institution.

CHANG SOOK KENG

Date: 21 December 2012

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LIST OF ABBREVIATIONS

ac	alternating current
AC	Activated Carbon
SC	Specific Capacitance
CNT	Carbon Nanotube
Co_3O_4	Cobalt Oxide
CV	Cyclic Voltammetry
dc	direct current
DSC	Differential Scanning Calorimetry
EDS	Energy Dispersive Spectroscopy
EIS	Electrochemical Impedance Spectroscopy
ESR	Equivalent Circuit Resistance
FESEM	Field Emission Scanning Electron Microscopy
HCl	Hydrochloric Acid
IHP	Inner Helmholtz Plane
KCl	Potassium Chloride
KOH	Potassium Hydroxide
Mn	Manganese
NMP	N-methyl-2-pyrrolidone
$NiCo_2O_4$	Nickel-Cobalt Oxide
$NiCo_2O_4$/AC	Nickel-Cobalt Oxide/Activated Carbon
$NiMn_{0.5}Co_{1.5}O_4$	Manganese Substituted Nickel-Cobalt Oxide

NiO	Nickel Oxide
OHP	Outer Helmholtz Plane
PTFE	Polytetrafluoroethylene
PVdF	Poly(vinylidene fluoride)
R_s	Cell-electrolyte Resistance
R_{ct}	Charge-transfer Resistance
SE	Specific Energy
SP	Specific Power
TEM	Transmission Electron Microscopy
TGA	Thermogravimetry
Ti	Titanium
XANES	X-ray Absorption Near-Edge Structure
XRD	X-ray Diffraction

CHAPTER 1

INTRODUCTION

1.1 Energy Storage and Conversion Systems

Depletion of fossil fuels has prompted the alarming situation in search for alternative energy storage and conversion systems. Moreover, the escalating power demand in worldwide nowadays, has ensured a lot of emphasis been placed on the development of devices with high power, high energy as well as robust in withstanding hundreds of thousands of charge/discharge cycles without degrading. Therefore, supercapacitor which is also referred to as 'electrochemical capacitor (EC)', 'double-layer capacitor' or 'ultracapacitor' is considered as an innovative technology due to its uniqueness that fills the gap between batteries and capacitors. This device possesses remarkable characteristics and is efficient and capable to combine the energy properties of batteries and the power discharge characteristics of capacitors.

There are three basic types of capacitors namely electrostatic, electrolytic and electrochemical capacitors. The charge storage system in electrostatic capacitors is through physical reaction and they possess the lowest energy density compared to the other two types of capacitors. Electrolytic capacitors involve the growth of insulator on a rough metal surface like etched aluminium and this type of capacitor has >10 times higher energy density than electrostatic capacitors. The formation of 'double-layer' at the electrode/electrolyte interface in electrochemical capacitors ensures impressive capacitance and energy performance are achieved (100 times greater than electrolytic capacitors) (Miller, 2009).

Electrochemical capacitors can be categorized into two groups namely double-layer capacitors and pseudocapacitors. Electrical energy storage in the former occurs at the phase boundary between an electrode (electronic conductor) and the electrolyte solution (liquid ionic conductor) (Kurzweil, 2009a) with no involvement of charge transfer. Moreover, the current generated in this type of capacitor is merely a displacement current due to charge rearrangement or better known as ideally polarized electrode (Shukla *et al.*, 2000). Fast faradaic redox reactions caused by redox-active species involving metal oxides and conducting polymers are responsible in the potential determining charge transfer reaction that induces the charge storage mechanism of pseudocapacitors (Kurzweil, 2009b).

1.2 The Supercapacitor Industry

Supercapacitors have a wide range of applications not only in electronics but in automobiles as well. These devices are designed with various capacitance values to cater for different industrial requirements. Electronics applications such as cell phones and digital cameras, medical devices and uninterruptible power supplies depend on supercapacitors ranging from 1 F to 150 F. Meanwhile, power back-up in industrial and Telecom based station, and renewable energy systems require supercapacitors with the capacitance value ranging from 300 F to 350 F. Higher capacitance supercapacitors (650 F to 3000 F) are designed for automotive subsystems, hybrid drive trains, rail system power, heavy transportation and many other applications. Besides, this technology has overcome the environmental issue whereby discarded batteries that may cause serious disposal waste are controllable. Moreover, the market interests in

supercapacitors are expected to grow due to their greater power and longer shelf life that may lead to greener environment. An impressive sales rate of this energy storage device in the last 5-10 years (~ 1 billion units in 2005 with total sales revenue of around $195 millions) has gained worldwide recognition on the promising capability of this device in the domination of the capacitor industry over the years (Burke, 2009).

1.3 Ways for Enhancement of Supercapacitors

At the present time, worldwide research and development focuses on enhancing the performance and ensuring the cost reduction of electrochemical capacitors. However, the crucial importance is to maintain the power capability, fulfilling the cycle life requirements as well as to increase the capacitance and energy density performance. Various ways have been developed to achieve these criteria such as mixing the metal oxides to become binary or ternary oxides, compositing the metal oxides with carbonaceous materials like activated carbons, fibers, aerogels, xerogels, fullerenes or nanostructures, as well as incorporation of the metal oxides with conducting polymers. Adopting nanotechnology in optimizing the microstructure of the electroactive materials in the electrodes has become a notable issue as the charge storage system in pseudocapacitors involve the first few nanometers from the surface (Simon and Gogotsi, 2008; Serrano *et al.*, 2009). Moreover, active material with smaller particle size may contribute to higher capacitive performance due to the larger contact area between the electrode/electrolyte in nanostructured oxides. Greater power delivery and better cycling stability can be achieved as well (Rajeswari *et al.*, 2009). Undoubtedly, nanodimensional materials possess high electrical conductivity that makes them as

promising energy storage systems in the current situation whereby energy demand is expanding (Wallace *et al.*, 2009). Therefore, much research effort has been poured into developing effective methods in the preparation of nanostructured metal oxides and hydroxides in various forms like nanoparticles, nanofibers, nanorods, nanowires, nanotubes, nanosheets and so on. Approaches that have been attempted by researchers are precipitation (Wang *et al.*, 2011a; Chen *et al.*, 2009e), solvothermal (Qing *et al.*, 2011), hydrothermal (Shakir *et al.*, 2010; Tang *et al.*, 2009a), sol-gel (Wang *et al.*, 2007b), room-temperature solid-reaction route (Gong and Liu, 2011), chemical bath deposition (Yuan *et al.*, 2011; Dubal *et al.*, 2010), anodization (Xie *et al.*, 2011), electrochemical deposition (Wu *et al.*, 2011b; Hu *et al.*, 2006), lithographically patterned nanowire electrodeposition (LPNE) (Yan *et al.*, 2011), electrospinning (Le Viet *et al.*, 2011) and others.

1.4 Synthesis of Spinel Nickel-Cobalt Oxide and Nickel-Cobalt Oxide/Activated Carbon Composite

Besides, researchers have developed different preparative methods in synthesizing binary nickel-cobalt oxide systems not only in nanosized but in micron sized particles as well to be utilised as electrocatalyst for oxygen evolution reaction in electrolysis of alkaline water (Wu et al., 2004b; Chi *et al.*, 2004; Castro *et al.*, 2004; Rashkova *et al.*, 2002; Hu *et al.*, 1997; Singh *et al.*, 1991; Gennero de Chialvo and Chialvo, 1991; Singh *et al.*, 1990; Haenen *et al.*, 1986; Mehandjiev *et al.*, 1985; Hibbert and Churchill, 1984; Davidson *et al.*, 1982; Rasiyah *et al.*, 1982), anode electrode to oxidise alcohols and benzyl chloride (Vijayabarathi *et al.*, 2007), application in molten carbonate fuel cells (Durairajan *et al.*, 2002; Fukui *et al.*, 2001), counter electrode within an electrochromic

device (ECD) (Monk and Ayub, 1997), anode material for sodium-ion batteries (Alcántara *et al.*, 2002), electrochemical capacitors (Wu *et al.*, 2011d; Salunkhe *et al.*, 2011; Wei *et al.*, 2010) and others. This binary oxide can be synthesized via co-precipitation (Tharayil *et al.*, 2007; Bo *et al.*, 2004; Lapham and Tseung, 2004), thermal decomposition (Lapham and Tseung, 2004; Marco *et al.*, 2001; Marco *et al.*, 2000; Kim *et al.*, 2000; Hu *et al.*, 1997), sol-gel (Wu *et al.*, 2011d; Wei *et al.*, 2010; Serebrennikova and Birss, 2001; Kim *et al.*, 2000; Marco *et al.*, 2000), hydrothermal (Zheng *et al.*, 2009c), chemical bath deposition (Salunkhe *et al.*, 2011), electrochemical deposition (Gupta *et al.*, 2009; Castro *et al.*, 2004; Wu *et al.*, 2004b; Hu and Cheng, 2002), electrophoretic deposition (Cui *et al.*, 2008), sputter deposition (Owings *et al.*, 2005), electrospinning (Guan *et al.*, 2004), combustion (Verma *et al.*, 2008), spray pyrolysis (Azurdia *et al.*, 2008; Lapham and Tseung, 2004), cryochemical (Lapham and Tseung, 2004) and so on.

However, cobaltite system with spinel structure has been the subject of intense research due to its established applications in electrochemistry. It is known that its fundamental and physicochemical properties are dependent on the methods of preparation, composition of oxides and temperatures of thermal decomposition. When the size of cobaltite nanoparticles is reduced to the nanometer range, some of their properties can be different compared to samples in micron range. It is believed that by mixing two or more individual oxides together will lead to synergistic effects that may help in the enhancement of the performance of the synthesized materials. This helps to densify the prepared oxides with lower sintering temperature as well as to enhance the grain growth

(Brito *et al.*, 2010). Researchers have given a considerable attention in synthesizing cobaltite system by exploring the precursors used, preparation methods, processing control and firing temperatures (Swathi and Buvaneswari, 2008; Lapham and Tseung, 2004; De Faria *et al.*, 1998; Roginskaya *et al.*, 1997; Tareen *et al.*, 1984).

Nickel-cobalt oxide, $NiCo_2O_4$ is one of the promising metal oxides in the family of cobaltite materials which has a spinel structure AB_2O_4, with the nickel ions reside at A-sites and cobalt ions at B-sites. The spinel structure has received much attention by numerous researchers as more than 30 ions with radii ranging from 0.5 to 1.0 Å can be incorporated in the spinel-like phases (Rao and Raveau, 1998). The metallic ions occupy one-eighth of the tetrahedral interstices and half of the octahedral interstices in this cubic structure ($a \approx 8$ Å). In our study, $NiCo_2O_4$ either with or without Mn substitution for Co was synthesized via a precipitation route. Synthesizing a compound either in the form of binary or ternary oxides without any traces of impurities is proven to be more challenging and feasible to be applied in electrochemical capacitor application compared to mixed oxides as the size and morphology of these compounds are controllable. Thermal treatment of co-precipitated precursors is proven to be the most promising method in preparing cobaltite spinels (Klissurki and Uzunova, 1994). The method is simple, cost effective, of low temperature and yet capable of producing homogeneous nanoparticles with controllable composition (Santhanam and Rambabu, 2010; Sharma *et al.*, 2009; Huber *et al.*, 2008; Cui *et al.*, 2008; Bo *et al.*, 2004; Pradhan *et al.*, 2001). Moreover, this method has the capability to synthesize nanostructured materials (Cui *et al.*, 2008; Kulkarni, 2007) that are chemically stable over a long period

of time frame (Kulkarni, 2007). Besides, we also investigated on the synthesis of nickel-cobalt oxide/activated carbon ($NiCo_2O_4$/AC) composite system via co-precipitation method in which activated carbon was used as support matrix to facilitate electrolyte penetration into the bulk film as well as for the improvement in electrochemical capacitance performance. Combination of pseudocapacitance from $NiCo_2O_4$ and electrochemical double-layer capacitance from activated carbon was proven to complement each other to overcome any unfavourable properties or drawback portrayed by these two systems.

1.5 Towards the Green Era

Supercapacitors are gaining huge attention as they are known as green technology. Designing electrode materials that can meet the present power demands with suitable electrolytes and less hazardous have become a global concern. The capability of organic electrolytes to withstand large potential window are overshadowed by their volatile, highly flammable and toxic properties. Although aqueous electrolytes have the disadvantage of water decomposition, they are relatively less harmful to the environment. However, concentrated acids and bases can be corrosive and deleterious as well. Thus, use of inert electrolytes derived from salts will be a desirable way in the development of supercapacitors with the hope of optimizing the electrochemical performance while conserving the nature.

1.6 Objectives

Several scopes were being highlighted in order to have a better understanding of the whole study. Experimental works have been planned accordingly in ensuring a smooth conduct towards the completion of the research. Therefore, the objectives of this study are listed as below:

1. To prepare nickel-cobalt oxides ($NiCo_2O_4$) via oxalate co-precipitation, hydroxide co-precipitation and solid-state routes.
2. To synthesize nickel-cobalt oxide/activated carbon ($NiCo_2O_4$/AC) composites via oxalate co-precipitation.
3. To investigate on the electrochemical properties of manganese ion substituted into nickel-cobalt oxide ($NiMn_{0.5}Co_{1.5}O_4$) by hydroxide co-precipitation method.
4. To study physico-chemical properties of $NiCo_2O_4$, $NiCo_2O_4$/AC and $NiMn_{0.5}Co_{1.5}O_4$.
5. To determine the double-layer behaviour of $NiCo_2O_4$, $NiCo_2O_4$/AC and $NiMn_{0.5}Co_{1.5}O_4$ electrodes from cyclic voltammetry.
6. To obtain values of specific capacitance, power density and energy density from galvanostatic charge-discharge tests.
7. To evaluate the electrochemical stability and reversibility of the systems through cycle stability tests.
8. To deduce frequency dependent capacitance from electrochemical impedance spectroscopy.

REFERENCES

Abe, Y., Meguro, T., Oyamatsu, S., Yokoyama, T. and Komeya, K. 1999. Formation region of monophase with cubic spinel-type oxides in Mn-Co-Ni ternary system. *Journal of Materials Science* 34: 4639-4644.

Ahmad, J., Awan, M.Q., Mazhar, M.E. and Ashiq, M.N. 2011. Effect of substitution of Co^{2+} ions on the structural and electrical properties of nanosized magnesium aluminate. *Physica B-Condensed Matter* 406: 254-258.

Ahmed, J., Ahmad, T., Ramanujachary, K.V., Lofland, S.E. and Ganguli, A.K. 2008. Development of a microemulsion-based process for synthesis of cobalt (Co) and cobalt oxide (Co_3O_4) nanoparticles from submicrometer rods of cobalt oxalate. *Journal of Colloid and Interface Science* 321: 434-441.

Alcántara, R., Jaraba, M., Lavela, P. and Tirado, J.L. 2002. $NiCo_2O_4$ spinel: first report on a transition metal oxide for the negative electrode of sodium-ion batteries. *Chemistry of Materials* 14: 2847-2448.

Alvi, F., Ram, M.K., Basnayaka, P.A., Stefanakos, E., Goswami, Y. and Kumar, A. 2011. Graphene-polyethylenedioxythiophene conducting polymer nanocomposite based supercapacitor. *Electrochimica Acta* 56: 9406-9412.

Ammundsen, B., Jones, D.J. and Rozière, J. 1998. X-ray absorption fine structure spectroscopy as a probe of local structure in lithium manganese oxides. *Journal of Solid State Chemistry* 141: 294-297.

Amuda, O.S., Giwa, A.A. and Bello, I.A. 2007. Removal of heavy metal from industrial wastewater using modified activated coconut shell carbon. *Biochemical Engineering Journal* 36: 174-181.

An, K.H., Kim, W.S., Park, Y.S., Choi, Y.C., Lee, S.M., Chung, D.C., Bae, D.J., Lim, S.C. and Lee, Y.H. 2001a. Supercapacitors using single-walled carbon nanotube electrodes. *Advanced Materials* 13: 497-500.

An, K.H., Kim, W.S., Park, Y.S., Moon, J.-M., Bae, D.J., Lim, S.C., Lee, Y.S. and Lee, Y.H. 2001b. Electrochemical properties of high-power supercapacitors using single-walled carbon nanotube electrodes. *Advanced Functional Materials* 11: 387-392.

Ando, M., Kobayashi, T. and Haruta, M. 1996. Humidity-sensitive optical absorption of Co_3O_4 film. *Sensors and Actuators B-Chemical* 32: 157-160.

Andreas, H.A. and Conway, B.E. 2006. Examination of the double-layer capacitance of an high specific-area C-cloth electrode as titrated from acidic to alkaline pHs. *Electrochimica Acta* 51: 6510-6520.

Arabale, G., Wagh, D., Kulkarni, M., Mulla, I.S., Vernekar, S.P., Vijayamohanan, K. and Rao, A.M. 2003. Enhanced supercapacitance of multiwalled carbon nanotubes functionalized with ruthenium oxide. *Chemical Physics Letters* 376: 207-213.

Arbizzani, C., Beninati, S., Lazzari, M., Soavi, F. and Mastragostino, M. 2007. Electrode materials for ionic liquid-based supercapacitors. *Journal of Power Sources* 174: 648-652.

Arbizzani, C., Biso, M., Cericola, D., Lazzari, M., Soavi, F. and Mastragostino, M. 2008. Safe, high-energy supercapacitors based on solvent-free ionic liquid electrolytes. *Journal of Power Sources* 185: 1575-1579.

Ataherian, F., Lee, K.-T. and Wu, N.-L. 2010. Long-term electrochemical behaviors of manganese oxide aqueous electrochemical capacitor under reducing potentials. *Electrochimica Acta* 55: 7429-7435.

Attia, A.A., Girgis, B.S. and Fathy, N.A. 2008. Removal of methylene blue by carbons derived from peach stones by H_3PO_4 activation: batch and column studies. *Dyes and Pigments* 76: 282-289.

Ayad, M.Y., Becherif, M. and Henni, A. 2011. Vehicle hybridization with fuel cell, supercapacitors and batteries by sliding mode control. *Renewable Energy* 36: 2627-2634.

Azurdia, J.A., McCrum, A. and Laine, R.M. 2008. Systematic synthesis of mixed-metal oxides in NiO-Co_3O_4, NiO-MoO_3, and NiO-CuO systems via liquid-feed flame spray pyrolysis. *Journal of Materials Chemistry* 18: 3249-3258.

Babakhani, B. and Ivey, D.G. 2010. Anodic deposition of manganese oxide electrode with rod-like structures for application as electrochemical capacitors. *Journal of Power Sources* 195: 2110-2117.

Bard, A.J. and Faulkner, L.R. 2001. Electrochemical Methods: Fundamentals and Applications 2nd edition, John Wiley, New York.

Barranco, V., Pico, F., Ibañez, J., Lillo-Rodenas, M.A., Linares-Solano, A., Kimura, M., Oya, A., Rojas, R.M., Amarilla, J.M. and Rojo, J.M. 2009. Amorphous carbon nanofibres inducing high specific capacitance of deposited hydrous ruthenium oxide. *Electrochimica Acta* 54: 7452-7457.

Battisha, I.K., Hamad, A.B.A. and Mahani, R.M. 2009. Structure and dielectric studies of nano-composite Fe_2O_3: $BaTiO_3$ prepared by sol-gel method. *Physica B-Condensed Matter* 404: 2274-2279.

Belli, M., Scafati, A., Bianconi, A., Mobilio, S., Palladino, L., Reale, A. and Burattini, E. 1980. X-ray absorption near edge structures (XANES) in simple and complex Mn compounds. *Solid State Communications* 35: 355-361.

Bo, C., Li, J.-B., Han, Y.-S. and Dai, J.-H. 2004. Effect of precipitant on preparation of Ni-Co spinel oxide by coprecipitation method. *Materials Letters* 58: 1415-1418.

Bordjiba, T. and Bélanger, D. 2010. Development of new nanocomposite based on nanosized-manganese oxide and carbon nanotubes for high performance electrochemical capacitors. *Electrochimica Acta* 55: 3428-3433.

Bouchelta, C., Medjram, M.S., Bertrand, O. and Bellat, J.-P. 2008. Preparation and characterization of activated carbon from date stones by physical activation with steam. *Journal of Analytical and Applied Pyrolysis* 82: 70-77.

Brito, P.C.A., Santos, D.A.A., Duque, J.G.S. and Macêdo, M.A. 2010. Structural and magnetic study of Fe-doped CeO_2. *Physica B-Condensed Matter* 405: 1821-1825.

Brownson, D.A.C., Kampouris, D.K. and Banks, C.E. 2011. An overview of graphene in energy production and storage applications. *Journal of Power Sources* 196: 4873-4885.

Brownson, J.R.S. and Lévy-Clément, C. 2009. Nanostructured α- and β-cobalt hydroxide thin films. *Electrochimica Acta* 54: 6637-6644.

Brousse, T. and Bélanger, D. 2003. A hybrid Fe_3O_4-MnO_2 capacitor in mild aqueous electrolyte. *Electrochemical and Solid-State Letters* 6: A244-A248.

Brumbach, M.T., Alam, T.M., Nilson, R.H., Kotula, P.G., McKenzie, B.B., Tissot, R.G. and Bunker, B.C. 2010. Ruthenium oxide–niobium hydroxide composites for pseudocapacitor electrodes. *Materials Chemistry and Physics* 124: 359-370.

Burke, A. 2007. R&D considerations for the performance and application of electrochemical capacitors. *Electrochimica Acta* 53: 1083-1091.

Burke, A. 2009. Capacitors: Application. In *Encyclopedia of Electrochemical Power Sources*, ed. Garche, J., Dyer, C.K., Moseley, P.T., Ogumi, Z., Rand, D.A.J. and Scrosati, B., pp. 685-694. Amsterdam: Elsevier.

Cao, A.-M., Hu, J.-S., Liang, H.-P., Song, W.-G., Wan, L.-J., He, X.-L., Gao, X.-G. and Xia, S.-H. 2006. Hierarchically structured cobalt oxide (Co_3O_4): the morphology control and its potential in sensors. *Journal of Physical Chemistry B* 110: 15858-15863.

Cao, L., Lu, M. and Li, H.-L. 2005. Preparation of mesoporous nanocrystalline Co_3O_4 and its applicability of porosity to the formation of electrochemical capacitance. *Journal of The Electrochemical Society* 152: A871-A875.

Castro, E.B., Real, S.G. and Pinhero Dick, L.F. 2004. Electrochemical characterization of porous nickel-cobalt oxide electrodes. *International Journal of Hydrogen Energy* 29: 255-261.

Cericola, D., Kötz, R. and Wokaun, A. 2011. Effect of electrode mass ratio on aging of activated carbon based supercapacitors utilizing organic electrolytes. *Journal of Power Sources* 196: 3114-3118.

Chang, J.-K. and Tsai, W.-T. 2003. Material characterization and electrochemical performance of hydrous manganese oxide electrodes for use in electrochemical pseudocapacitors. *Journal of The Electrochemical Society* 150: A1333-A1338.

Chang, J.K., Hsieh, W.-C. and Tsai, W.-T. 2008. Effects of the Co content in the material characteristics and supercapacitive performance of binary Mn-Co oxide electrodes. *Journal of Alloys and Compounds* 461: 667-674.

Chang, J.-K., Lee, M.-T., Tsai, W.-T., Deng, M.-J., Cheng, H.-F. and Sun, I.-W. 2009. Pseudocapacitive mechanism of manganese oxide in 1-ethyl-3-methylimidazolium thiocyanate ionic liquid electrolyte studied using X-ray photoelectron spectroscopy. *Langmuir* 25: 11955-11960.

Chang, K.-H., Hu, C.-C. and Chou, C.-Y. 2007. Textural and capacitive characteristics of hydrothermally derived $RuO_2 \cdot xH_2O$ nanocrystallites: independent control of crystal size and water content. *Chemistry of Materials* 19: 2112-2119.

Chen, C.-Y., Chien, T.-C., Chan, Y.-C., Lin, C.-K. and Wang, S.-C. 2009a. Pseudocapacitive properties of carbon nanotube/manganese oxide electrode deposited by electrophoretic deposition. *Diamond and Related Materials* 18: 482-485.

Chen, C.-Y., Wang, S.-C., Lin, C.-Y., Chen, F.-S. and Lin, C.-K. 2009b. Electrophoretically deposited manganese oxide coatings for supercapacitor application. *Ceramics International* 35: 3469-3474.

Chen, C.Y., Wang, S.-C., Tien, Y.-H., Tsai, W.-T. and Lin, C.-K. 2009c. Hybrid manganese oxide films for supercapacitor application prepared by sol-gel technique. *Thin Solid Films* 518: 1557-1560.

Chen, H.-R., H.-H. Lai and J.-J. Jow 2011a. Annealing effect on the performance of RuO_2–Ta_2O_5/Ti electrodes for use in supercapacitors. *Materials Chemistry and Physics* 125: 652-655.

Chen, L.-M., Lai, Q.-Y., Hao, Y.-J., Zhao, Y. and Ji, X.-Y. 2009d. Investigations on capacitive properties of the AC/V_2O_5 hybrid supercapacitor in various aqueous electrolytes. *Journal of Alloys and Compounds* 467: 465-471.

Chen, Q.-L., Xue, K.-H., Shen, W., Tao, F.-F., Yin, S.-Y. and Xu, W. 2004. Fabrication and electrochemical properties of carbon nanotube array electrode for supercapacitors. *Electrochimica Acta* 49: 4157-4161.

Chen, S., Chu, J. and Wang, X. 2011b. An *in situ* oxidation route to fabricate graphene nanoplate-metal oxide composites. *Journal of Solid State Chemistry* 184: 1393-1399.

Chen, S., Zhu, J., Han, Q., Zheng, Z., Yang, Y. and Wang, X. 2009e. Shape-controlled synthesis of one-dimensional MnO_2 via a facile quick-precipitation procedure and its electrochemical properties. *Crystal Growth & Design* 9: 4356-4361.

Cheng, J., Cao, G.-P. and Yang, Y.-S. 2006. Characterization of sol–gel-derived NiO_x xerogels as supercapacitors. *Journal of Power Sources* 159: 734-741.

Cheng, J.P., Chen, X., Ma, R., Liu, F. and Zhang, X.B. 2011a. A facile method to fabricate porous Co_3O_4 hierarchical microspheres. *Materials Characterization* 62: 775-780.

Cheng, Q., Tang, J., Ma, J., Zhang, H., Shinya, N. and Qin, L.-C. 2011b. Graphene and nanostructured MnO_2 composite electrodes for supercapacitors. *Carbon* 49: 2917-2925.

Chi, B., Li, J., Han, Y. and Chen, Y. 2004. Effect of temperature on the preparation and electrocatalytic properties of a spinel $NiCo_2O_4$/Ni electrode. *International Journal of Hydrogen Energy* 29: 605-610.

Chuang, P.-Y. and Hu, C.-C. 2005. The electrochemical characteristics of binary manganese-cobalt oxides prepared by anodic deposition. *Materials Chemistry and Physics* 92: 138-145.

Conesa, J.A., Sakurai, M. and Antal Jr., M.J. 2000. Synthesis of a high-yield activated carbon by oxygen gasification of macadamia nut shell charcoal in hot, liquid water. *Carbon* 38: 839-848.

Conway, B.E. 1981. Ionic Hydration in Chemistry and Biophysics, Elsevier Scientific Publishing Company, Amsterdam.

Conway, B.E. 1999. Electrochemical Supercapacitors: Scientific Fundamentals and Technological Applications, Kluwer Academic/ Plenum Publishers, New York.

Conway, B.E., Birss, V. and Wojtowicz, J. 1997. The role and utilization of pseudocapacitance for energy storage by supercapacitors. *Journal of Power Sources* 66: 1-14.

Cormie, A., Cross, A., Hollenkamp, A.F. and Donne, S.W. 2010. Cycle stability of birnessite manganese dioxide for electrochemical capacitors. *Electrochimica Acta* 55: 7470-7478.

Cui, B., Lin, H., Li, J.-B., Li, X., Yang, J. and Tao, J. 2008. Core-ring structured $NiCo_2O_4$ nanoplatelets: synthesis, characterization, and electrocatalytic applications. *Advanced Functional Materials* 18: 1440-1447.

Cui, G.-L., Zhou, X.-H., Zhi, L.-J., Thomas, A. and Müllen, K. 2007. Carbon/nanostructured Ru composites as electrodes for supercapacitors. *New Carbon Materials* 22: 302-306.

Cui, L., Li, J. and Zhang, X.-G. 2009. Preparation and properties of Co_3O_4 nanorods as supercapacitor material. *Journal of Applied Electrochemistry* 39: 1871-1876.

Cui, X., Hu, F., Wei, W. and Chen, W. 2011. Dense and long carbon nanotube arrays decorated with Mn_3O_4 nanoparticles for electrodes of electrochemical supercapacitors. *Carbon* 49: 1225-1234.

Dandekar, M.S., Arabale, G. and Vijayamohanan, K. 2005. Preparation and characterization of composite electrodes of coconut-shell-based activated carbon and hydrous ruthenium oxide for supercapacitors. *Journal of Power Sources* 141: 198-203.

Davidson, C.R., Kissel, G. and Srinivasan, S. 1982. Electrode kinetics of the oxygen evolution reaction at $NiCo_2O_4$ from 30% KOH: dependence on temperature. *Journal of Electroanalytical Chemistry* 132: 129-135.

De Faria, L.A., Prestat, M., Koenig, J.-F., Chartier, P. and Trasatti, S. 1998. Surface properties of Ni + Co mixed oxides: a study by X-rays, XPS, BET and PZC. *Electrochimica Acta* 44: 1481-1489.

Deng, J.-J., Deng, J.-C., Liu, Z.-L., Deng, H.-R. and Liu B. 2009a. Influence of addition of cobalt oxide on microstructure and electrochemical capacitive performance of nickel oxide. *Journal of Solid State Electrochemistry* 13: 1387-1394.

Deng, J.-J., Deng, J.-C., Liu, Z.-L., Deng, H.-R. and Liu B. 2009b. Capacitive characteristics of Ni-Co oxide nano-composite via coordination homogeneous co-precipitation method. *Journal of Materials Science* 44: 2828-2835.

Deng, L., Zhu, G., Wang, J., Kang, L., Liu, Z.-H., Yang, Z. and Wang, Z. 2011. Graphene-MnO_2 and graphene asymmetrical electrochemical capacitor with a high energy density in aqueous electrolyte. *Journal of Power Sources* 196: 10782-10787.

Devaraj, S. and Munichandraiah, N. 2008. Effect of crystallographic structure of MnO_2 on its electrochemical capacitance properties. *Journal of Physical Chemistry C* 112: 4406-4417.

Dong, X., Shen, W., Gu, J., Xiong, L., Zhu, Y., Li, H. and Shi, J. 2006. MnO_2-embedded-in-mesoporous-carbon-wall structure for use as electrochemical capacitors. *Journal of Physical Chemistry B* 110: 6015-6019.

Dong, Z., Kennedy, S.J. and Wu, Y. 2011. Electrospinning materials for energy-related applications and devices. *Journal of Power Sources* 196: 4886-4904.

Du, C. and Pan, N. 2006. Supercapacitors using carbon nanotubes films by electrophoretic deposition. *Journal of Power Sources* 160: 1487-1494.

Dubal, D.P., Dhawale, D.S., Salunkhe, R.R., Jamdade, V.S. and Lokhande, C.D. 2010. Fabrication of copper oxide multilayer nanosheets for supercapacitor application. *Journal of Alloys and Compounds* 492: 26-30.

Duque, J.G.S., Souza, E.A., Meneses, C.T. and Kubota, L. 2007. Magnetic properties of $NiFe_2O_4$ nanoparticles produced by a new chemical method. *Physica B-Condensed Matter* 398: 287-290.

Durairajan, A., Colon-Mercado, H., Haran, B., White, R. and Popov, B. 2002. Electrochemical characterization of cobalt-encapsulated nickel as cathodes for MCFC. *Journal of Power Sources* 104: 157-168.

Durán, P., Tartaj, J., Rubio, F., Moure, C. and Peña, O. 2005a. Synthesis and sintering behavior of spinel-type $Co_xNiMn_{2-x}O_4$ ($0.2 \leq x \leq 1.2$) prepared by the ethylene glycol-metal nitrate polymerized complex process. *Ceramics International* 31: 599-610.

Durán, P., Tartaj, J., Rubio, F., Peña, O. and Moure, C. 2005b. Preparation and sintering behaviour of spinel-type $Co_xNiMn_{2-x}O_4$ ($0.2 \leq x \leq 1.2$) by the ethylene glycol-metal nitrate polymerized complex process. *Journal of the European Ceramic Society* 25: 3021-3025.

El-Shobaky, G.A., El-Nabarawy, T., Morsi, I.M. and Ghoneim, N.M. 1983. Physicochemical properties of NiO-Al_2O_3 mixed oxides and their relationship with thermal treatment and chemical composition. *Surface Technology* 19: 109-118.

Fan, Z., Chen, J., Cui, K., Sun, F., Xu, Y. and Kuang, Y. 2007. Preparation and capacitive properties of cobalt-nickel oxides/carbon nanotube composites. *Electrochimica Acta* 52: 2959-2965.

Fan, Z., Chen, J., Wang, M., Cui, K., Zhou, H. and Kuang, Y. 2006. Preparation and characterization of manganese oxide/CNT composites as supercapacitive materials. *Diamond and Related Materials* 15: 1478-1483.

Fan, Z., Xie, M., Jin, X., Yan, J. and Wei, T. 2011. Characteristics and electrochemical performances of supercapacitors using double-walled carbon nanotube/δ-MnO_2 hybrid material electrodes. *Journal of Electroanalytical Chemistry* 659: 191-195.

Fang, D.-L., Chen, Z.-D., Wu, B.-C., Yan, Y. and Zheng, C.-H. 2011. Preparation and electrochemical properties of ultra-fine Mn-Ni-Cu oxides for supercapacitors. *Materials Chemistry and Physics* 128: 311-316.

Fang, W.-C. 2008. Synthesis and electrochemical characterization of vanadium oxide/carbon nanotube composites for supercapacitors. *Journal of Physical Chemistry C* 112: 11552-11555.

Farbod, M., Shoushtari, M.Z. and Parhoodeh, S. 2011. Fabrication and characterization of $Zn_{1-x}Al_xO$ nanoparticles by DC arc plasma. *Physica B-Condensed Matter* 406: 205-210.

Fernandes, D.M., Winkler Hechenleitner, A.A., Silva, M.F., Lima, M.K., Stival Bittencourt, P.R., Silva, R., Custódio Melo, M.A. and Gómez Pineda, E.A. 2009. Preparation and characterization of NiO, Fe_2O_3, $Ni_{0.04}Zn_{0.96}O$ and $Fe_{0.03}Zn_{0.97}O$ nanoparticles. *Materials Chemistry and Physics* 118: 447-452.

Frackowiak, E., Lota, G., Machnikowski, J., Vix-Guterl, C. and Béguin, F. 2006. Optimisation of supercapacitors using carbons with controlled nanotexture and nitrogen content. *Electrochimica Acta* 51: 2209-2214.

Fuertes, A. B., Lota, G., Centeno, T. A. and Frackowiak, E. 2005. Templated mesoporous carbons for supercapacitor application. *Electrochimica Acta* 50: 2799-2805.

Fukui, T., Okawa, H., Hotta, T., Naito, M. and Yokohama, T. 2001. Synthesis of CoO/Ni composite powders for molten carbonate fuel cells. *Journal of the American Ceramic Society* 84: 233-235.

Gamby, J., Taberna, P.L., Simon, P., Fauvarque, J.F. and Chesneau, M. 2001. Studies and characterisations of various activated carbons used for carbon/carbon supercapacitors. *Journal of Power Sources* 101: 109-116.

Ganesh, V., Pitchumani, S. and Lakshminarayanan, V. 2006. New symmetric and asymmetric supercapacitors based on high surface area porous nickel and activated carbon. *Journal of Power Sources* 158: 1523-1532.

Gao, L., Peng, A., Wang, Z.Y., Zhang, H., Shi, Z., Gu, Z., Cao, G. and Ding, B. 2008. Growth of aligned carbon nanotube arrays on metallic substrate and its application to supercapacitors. *Solid State Communications* 146: 380-383.

Gao, Y., Chen, S., Cao, D., Wang, G. and Yin, J. 2010. Electrochemical capacitance of Co_3O_4 nanowire arrays supported on nickel foam. *Journal of Power Sources* 195: 1757-1760.

Garcia, B.B., Candelaria, S.L., Liu, D., Sepheri, S., Cruz, J.A. and Cao, G. 2011. High performance high-purity sol-gel derived carbon supercapacitors from renewable sources. *Renewable Energy* 36: 1788-1794.

Gennero de Chialvo, M.R. and Chialvo, A.C. 1991. Development of thick oxide layers on nickel-cobalt (40%/60%) alloy by fast square wave potential perturbations. *Electrochimica Acta* 36: 1963-1969.

Girija, T. C. and Sangaranarayanan, M. V. 2006. Polyaniline-based nickel electrodes for electrochemical supercapacitors—Influence of TRITON X-100. *Journal of Power Sources* 159: 1519-1526.

Gómez, H., Ram, M.K., Alvi, F., Villalba, P., Stefanakos, E. and Kumar, A. 2011. Graphene-conducting polymer nanocomposite as novel electrode for supercapacitors. *Journal of Power Sources* 196: 4102-4108.

Gong, L. and Liu, X. 2011. Facile synthesis and capacitive characteristics of $Co(OH)_2$ nanoflakes via a solid-reaction route at room temperature. *Materials Letters* 65: 2025-2028.

Guan, H., Shao, C., Liu, Y., Yu, N. and Yang, X. 2004. Fabrication of $NiCo_2O_4$ nanofibers by electrospinning. *Solid State Communications* 131: 107-109.

Gujar, T.P., Shinde, V.R., Lokhande, C.D. and Han, S.-H. 2006. Electrosynthesis of Bi_2O_3 thin films and their use in electrochemical supercapacitors. *Journal of Power Sources* 161: 1479-1485.

Gunjakar, J.L., More, A.M., Shinde, V.R. and Lokhande, C.D. 2008. Synthesis of nanocrystalline nickel ferrite ($NiFe_2O_4$) thin films using low temperature modified chemical method. *Journal of Alloys and Compounds* 465: 468-473.

Guo, B., Li, C. and Yuan, Z.-Y. 2010a. Nanostructured Co_3O_4 materials: synthesis, characterization, and electrochemical behaviors as anode reactants in rechargeable lithium ion batteries. *Journal of Physical Chemistry C* 114: 12805-12817.

Guo, J. and Lua, A.C. 2001. Experimental and kinetic studies on pore development during CO_2 activation of oil-palm-shell char. *Journal of Porous Materials* 8: 149-157.

Guo, J. and Lua, A.C. 2003. Textural and chemical properties of adsorbent prepared from palm shell by phosphoric acid activation. *Materials Chemistry and Physics* 80: 114-119.

Guo, J., Luo, Y., Lua, A.C., Chi, R.-A., Chen, Y.-L., Bao, X.-.T. and Xiang, S.-X. 2007. Adsorption of hydrogen sulphide (H_2S) by activated carbons derived from oil-palm shell. *Carbon* 45: 330-336.

Guo, R.-S., Wang, C.-W., Chen, J.-B., Wang, J., Wang, L.-Q. and Liu, W.-M. 2010b. Field emission from TiO_2/Ti nanotube arrays with different morphologies. *Physica B-Condensed Matter* 405: 4682-4696.

Gupta, V., Gupta, S. and Miura, N. 2008. Potentiostatically deposited nanostructured Co_xNi_{1-x} layered double hydroxides as electrode materials for redox-supercapacitors. *Journal of Power Sources* 175: 680-685.

Gupta, V., Kawaguchi, T. and Miura, N. 2009. Synthesis and electrochemical behavior of nanostructured cauliflower-shape Co-Ni/Co-Ni oxides composites. *Materials Research Bulletin* 44: 202-206.

Haenen, J., Visscher, W. and Barendrecht, E. 1986. Characterization of $NiCo_2O_4$ electrodes for O_2 evolution. Part 1. Electrochemical characterization of freshly prepared $NiCo_2O_4$ electrodes. *Journal of Electroanalytical Chemistry* 208: 273-296.

Hakim, M.A., Manjurul Haque, M., Huq, M. and Nordblad, P. 2011. Spin-glass-like ordering in the spinel $ZnFe_2O_4$ ferrite. *Physica B-Condensed Matter* 406: 48-51.

Hassler, J. W. 1974. Purification with Activated Carbon: Industrial, Commercial, Environmental, Chemical Publishing Company, New York.

Hayashi, J., Horikawa, T., Takeda, I., Muroyama, K. and Ani, F. N. 2002. Preparing activated carbon from various nutshells by chemical activation with K_2CO_3. *Carbon* 40: 2381-2386.

He, K.-X., Wu, Q.-F., Zhang, X.-G. and Wang, X.-L. 2006. Electrodeposition of nickel and cobalt mixed oxide/carbon nanotube thin films and their charge storage properties. *Journal of The Electrochemical Society* 153: A1568-A1574.

He, T., Chen, D. and Jiao, X. 2004a. Controlled synthesis of Co_3O_4 nanoparticles through oriented aggregation. *Chemistry of Materials* 16: 737-743.

He, T., Chen, D., Jiao, X., Xu, Y. and Gu, Y. 2004b. Surfactant-assisted solvothermal synthesis of Co_3O_4 hollow spheres with oriented-aggregation nanostructures and tunable particle size. *Langmuir* 20: 8404-8408.

He, W.L., Zhang, Y.C., Zhang, X.X., Wang, H. and Yan, H. 2003. Low temperature preparation of nanocrystalline Mn_2O_3 via ethanol-thermal reduction of MnO_2. *Journal of Crystal Growth* 252: 285-288.

He, X.J., Geng, Y.J., Oke, S., Higashi, K., Yamamoto, M. and Takikawa, H. 2009. Electrochemical performance of RuO_x/activated carbon black composite for supercapacitors. *Synthetic Metals* 159: 7-12.

Heli, H. and Yadegari, H. 2010. Nanoflakes of the cobaltous oxide, CoO: synthesis and characterization. *Electrochimica Acta* 55: 2139-2148.

Hibbert, D.B. and Churchill, C.R. 1984. Kinetics of the electrochemical evolution of isotopically enriched gases: part 2.—$^{18}O^{16}O$ evolution on $NiCo_2O_4$ and $Li_xCo_{3-x}O_4$ in alkaline solution. *Journal of the Chemical Society, Faraday Transactions 1: Physical Chemistry in Condensed Phases* 80: 1965-1975.

Hong, M.S., Lee, S.H. and Kim, S.W. 2002. Use of KCl aqueous electrolyte for 2 V manganese oxide/activated carbon hybrid capacitor. *Electrochemical and Solid-State Letters* 5: A227-A230.

Hosseini, M. 2000. The effect of cation composition on the electrical properties and aging of Mn-Co-Ni thermistors. *Ceramics International* 26: 245-249.

Hsiang, H.-I. and Yao, R.-Q. 2008. Formation mechanism of $3BaO{\cdot}2CoO{\cdot}12Fe_2O_3$ powder synthesized using chemical precipitation. *Journal of Alloys and Compounds* 453: 366-370.

Hsieh, Y.-C., Lee, K.-T., Lin, Y.-P., N.-L. Wu and Donne, S.W. 2008. Investigation on capacity fading of aqueous $MnO_2{\cdot}nH_2O$ electrochemical capacitor. *Journal of Power Sources* 177: 660-664.

Hu, C.-C. and Chang, K.-H. 2002. Cyclic voltammetric deposition of hydrous ruthenium oxide for electrochemical supercapacitors: effects of the chloride precursor transformation. *Journal of Power Sources* 112: 401-409.

Hu, C.-C., Chang, K-H., Lin, M.-C. and Wu, Y.-T. 2006. Design and tailoring of the nanotubular arrayed architecture of hydrous RuO_2 for next generation supercapacitors. *Nano Letters* 6: 2690-2695.

Hu, C.-C. and Cheng, C.-Y. 2002. Ideally pseudocapacitive behavior of amorphous hydrous cobalt-nickel oxide prepared by anodic deposition. *Electrochemical and Solid-State Letters* 5: A43-A46.

Hu, C.-C., Guo, H.-Y., Chang, K.-H. and Huang, C.-C. 2009a. Anodic composite deposition of $RuO_2.xH_2O$–TiO_2 for electrochemical supercapacitors. *Electrochemistry Communications* 11: 1631-1634.

Hu, C.-C. and Hsu, T.-Y. 2008. Effects of complex agents on the anodic deposition and electrochemical characteristics of cobalt oxides. *Electrochimica Acta* 53: 2386-2395.

Hu, C.-C., Hung, C.-Y., Chang, K.-H. and Yang, Y.-L. 2011. A hierarchical nanostructure consisting of amorphous MnO_2, Mn_3O_4 nanocrystallites, and single-crystalline MnOOH nanowires for supercapacitors. *Journal of Power Sources* 196: 847-850.

Hu, C.-C., Lee, Y.-S. and Wen, T.-C. 1997. The physicochemical/electrochemical properties of binary Ni-Co oxides. *Materials Chemistry and Physics* 48: 246-254.

Hu, C.-C. and Tsou, T.-W. 2003. The optimization of specific capacitance of amorphous manganese oxide for electrochemical supercapacitors using experimental strategies. *Journal of Power Sources* 115: 179-186.

Hu, C.-C. and Tsou, T.-W. 2002. Ideal capacitive behavior of hydrous manganese oxide prepared by anodic deposition. *Electrochemistry Communications* 4: 105-109.

Hu, C.-C. and Wang, C.-C. 2004. Effects of electrolytes and electrochemical pretreatments on the capacitive characteristics of activated carbon fabrics for supercapacitors. *Journal of Power Sources* 125: 299-308.

Hu, J., Wang, H., Gao, Q. and Guo, H. 2010. Porous carbons prepared by using metal–organic framework as the precursor for supercapacitors. *Carbon* 48: 3599-3606.

Hu, Z., Mo, L., Feng, X., Shi, J., Wang, Y. and Xie, Y. 2009b. Synthesis and electrochemical capacitance of sheet-like cobalt hydroxide. *Materials Chemistry and Physics* 114: 53-57.

Hu, Z.-A., Xie, Y.-L., Wang, Y.-X., Wu, H.-Y., Yang, Y.-Y. and Zhang, Z.-Y. 2009c. Synthesis and electrochemical characterization of mesoporous Co_xNi_{1-x} layered double hydroxides as electrode materials for supercapacitors. *Electrochimica Acta* 54: 2737-2741.

Huai, Y., Hu, X., Lin, Z., Deng, Z. and Suo, J. 2009. Preparation of nano-TiO_2/activated carbon composite and its electrochemical characteristics in non-aqueous electrolyte. *Materials Chemistry and Physics* 113: 962-966.

Huang, C.-C., Xing, W. and Zhuo, S.-P. 2009. Capacitive performances of amorphous tungsten oxide prepared by microwave irradiation. *Scripta Materialia* 61: 985-987.

Huang, C.-W., Wu, Y.-T., Hu, C.-C. and Li, Y.-Y. 2007a. Textural and electrochemical characterization of porous carbon nanofibers as electrodes for supercapacitors. *Journal of Power Sources* 172: 460-467.

Huang, Q., Wang, X., Li, J., Dai, C., Gamboa, S. and Sebastian, P.J. 2007b. Nickel hydroxide/activated carbon composite electrodes for electrochemical capacitors. *Journal of Power Sources* 164: 425-429.

Huber, F., Venvik, H., Rønning, M., Walmsley, J. and Holmen, A. 2008. Preparation and characterization of nanocrystalline, high surface area Cu-Ce-Zr mixed oxide catalysts from homogeneous co-precipitation. *Chemical Engineering Journal* 137: 686-702.

Hussein, M.Z., Tarmizi, R.S.H., Zainal, Z., Ibrahim, R. and Badri, M. 1996. Preparation and characterization of active carbons from oil palm shells. *Carbon* 34: 1447-1454.

Hussein, M.Z., Abdul Rahman, M.B., Yahaya, A., Taufiq-Yap, Y.H. and Ahmad, N. 2001. Oil palm trunk as a raw material for activated carbon production. *Journal of Porous Materials* 8: 327-334.

Ibarra-Palos, A., Strobel, P., Proux, O., Hazemann, J.L., Anne, M. and Morcrette, M. 2002. *In situ* X-ray absorption spectroscopy study of lithium insertion in a new disordered manganese oxi-iodide. *Electrochimica Acta* 47: 3171-3178.

Inamdar, A.I., Kim, Y., Pawar, S.M., Kim, J.H., Im, H. and Kim, H. 2011. Chemically grown, porous, nickel oxide thin-film for electrochemical supercapacitors. *Journal of Power Sources* 196: 2393-2397.

Jamadade, V.S., Fulari, V.J. and Lokhande, C.D. 2011. Supercapacitive behavior of electrosynthesized marygold-like structured nickel doped iron hydroxide thin film. *Journal of Alloys and Compounds* 509: 6257-6261.

Jang, J.H., Han, S., Hyeon, T. and Oh, S.M. 2003. Electrochemical capacitor performance of hydrous ruthenium oxide/mesoporous carbon composite electrodes. *Journal of Power Sources* 123: 79-85.

Jankowska, H., Świątkowski, A. and Choma, J. 1991. Active Carbon, Ellis Horwood Limited, New York.

Jayalakshmi, M. and Balasubramanian, K. 2009. Hydrothermal synthesis of CuO-SnO_2 and CuO-SnO_2-Fe_2O_3 mixed oxides and their electrochemical characterization in neutral electrolyte. *International Journal of Electrochemical Science* 4: 571-581.

Jayalakshmi, M., Venugopal, N., Phani Raja, K. and Mohan Rao, M. 2006. Nano SnO_2-Al_2O_3 mixed oxide and SnO_2-Al_2O_3-carbon composite oxides as new and novel electrodes for supercapacitor applications. *Journal of Power Sources* 158: 1538-1543.

Jeong, Y.U. and Manthiram, A. 2002. Nanocrystalline manganese oxides for electrochemical capacitors with neutral electrolytes. *Journal of The Electrochemical Society* 149: A1419-A1422.

Ji, Z., Shen, X., Song, Y. and Zhu, G. 2011. *In situ* synthesis of graphene/cobalt nanocomposites and their magnetic properties. *Materials Science and Engineering B-Advanced Functional Solid-State Materials* 176: 711-715.

Jiang, H., Yang, L., Li, C., Yan, C., Lee, P.S. and Ma, J. 2011. High–rate electrochemical capacitors from highly graphitic carbon-tipped manganese oxide/mesoporous carbon/manganese oxide hybrid nanowires. *Energy and Environmental Science* 4: 1813-1819.

Jiang, J. and Kucernak, A. 2002. Electrochemical supercapacitor material based on manganese oxide: preparation and characterization. *Electrochimica Acta* 47: 2381-2386.

Jiménez-Mateos, J.M., Morales, J. and Tirado, J.L. 1989. Preparation and sintering behaviour of ultrafine Mn/Fe and Fe/Co mixed oxides. *Journal of the European Ceramic Society* 5: 357-363.

Jisha, M.R., Hwang, Y.J., Shin, J.S., Nahm, K.S., Prem Kumar, T., Karthikeyan, K., Dhanikaivelu, N., Kalpana, D., Renganathan, N.G. and Manuel Stephan, A. 2009. Electrochemical characterization of supercapacitors based on carbons derived from coffee shells. *Materials Chemistry and Physics* 115: 33-39.

Jow, J.-J., Lai, H.-H., Chen, H.-R., Wang, C.-C., Wu, M.-S. and Ling, T.-R. 2010. Effect of hydrothermal treatment on the performance of RuO_2–Ta_2O_5/Ti electrodes for use in supercapacitors. *Electrochimica Acta* 55: 2793-2798.

Jurewicz, K., Delpeux, S., Bertagna, V., Béguin, F. and Frackowiak, E. 2001. Supercapacitors from nanotubes/polypyrrole composites. *Chemical Physics Letters* 347: 36-40.

Kalpana, D., Karhikeyan, K., Renganathan, N.G. and Lee, Y.S. 2008. Camphoric carbon nanobeads - a new electrode material for supercapacitors. *Electrochemistry Communications* 10: 977-979.

Kambale, R.C., Shaikh, P.A., Kamble, S.S. and Kolekar, Y.D. 2009. Effect of cobalt substitution on structural, magnetic and electric properties of nickel ferrite. *Journal of Alloys and Compounds* 478: 599-603.

Kandalkar, S.G., Dhawale, D.S., Kim, C.-K. and Lokhande, C.D. 2010. Chemical synthesis of cobalt oxide thin film electrode for supercapacitor application. *Synthetic Metals* 160: 1299-1302.

Kandalkar, S.G., Gunjakar, J.L., Lokhande, C.D. and Joo, O.-S. 2009. Synthesis of cobalt oxide interconnected flacks and nano-worms structures using low temperature chemical bath deposition. *Journal of Alloys and Compounds* 478: 594-598.

Kandalkar, S.G., Lee, H.-M., Chae, H. and Kim, C.-K. 2011. Structural, morphological, and electrical characteristics of the electrodeposited cobalt oxide electrode for supercapacitor applications. *Materials Research Bulletin* 46: 48-51.

Kang, Y.J., Kim, B., Chung, H. and Kim, W. 2010. Fabrication and characterization of flexible and high capacitance supercapacitors based on MnO_2/CNT/papers. *Synthetic Metals* 160: 2510-2514.

Kang, Y.-M., Kim, K.-T., Kim, J.-H., Kim, H.-S., Lee, P.S., Lee, J.-Y., Liu, H.K. and Dou, S.X. 2004. Electrochemical properties of Co_3O_4, Ni-Co_3O_4 mixture and Ni-Co_3O_4 composite as anode materials for Li ion secondary batteries. *Journal of Power Sources* 133: 252-259.

Karolus, M. and Łągiewka, E. 2004. Crystallite size and lattice strain in nanocrystalline Ni–Mo alloys studied by Rietveld refinement. *Journal of Alloys and Compounds* 367: 235-238.

Karthikeyan, K., Kalpana, D. and Renganathan, N.G. 2009. Synthesis and characterization of $ZnCo_2O_4$ nanomaterial for symmetric supercapacitor applications. *Ionics* 15: 107-110.

Kim, B.C., Wallace, G.G., Yoon, Y.I., Ko, J.M. and Too, C.O. 2009a. Capacitive properties of RuO_2 and Ru-Co mixed oxide deposited on single-walled carbon nanotubes for high-performance supercapacitors. *Synthetic Metals* 159: 1389-1392.

Kim, C. 2005. Electrochemical characterization of electrospun activated carbon nanofibres as an electrode in supercapacitors. *Journal of Power Sources* 142: 382-388.

Kim, H. and Popov, B.N. 2002. Characterisation of hydrous ruthenium oxide/carbon nanocomposite supercapacitors prepared by a colloidal method. *Journal of Power Sources* 104: 52-61.

Kim, H.-K., Seong, T.-Y., Lim, J.-H., Cho, W.I. and Yoon, Y.S. 2001. Electrochemical and structural properties of radio frequency sputtered cobalt oxide electrodes for thin-film supercapacitors. *Journal of Power Sources* 102: 167-171.

Kim, J.-G., Pugmire, D.L., Battaglia, D. and Langell, M.A. 2000. Analysis of the $NiCo_2O_4$ spinel surface with Auger and X-ray photoelectron spectroscopy. *Applied Surface Science* 165: 70-84.

Kim, J.-Y., Kim, K.-H., Park, S.-H. and Kim, K.-B. 2010. Microwave-polyol synthesis of nanocrystalline ruthenium oxide nanoparticles on carbon nanotubes for electrochemical capacitors. *Electrochimica Acta* 55: 8056-8061.

Kim, S.H., Kim, Y.I., Park, J.H. and Ko, J.M. 2009b. Cobalt-manganese oxide/carbon-nanofiber composite electrodes for supercapacitors. *International Journal of Electrochemical Science* 4: 1489-1496.

Klissurski, D. and Uzunova, E. 1994. Review synthesis and features of binary cobaltite spinels. *Journal of Materials Science* 29: 285-293.

Knop, O., Reid, K.I.G., Sutarno and Nakagawa, Y. 1968. Chalkogenides of the transition elements. VI. X-Ray, neutron, and magnetic investigation of the spinels Co_3O_4, $NiCo_2O_4$, Co_3S_4, and $NiCo_2S_4$. *Canadian Journal of Chemistry* 46: 3463-3476.

Ko, J.M. and Kim, K.M. 2009. Electrochemical properties of MnO_2/activated carbon nanotube composite as an electrode material for supercapacitor. *Materials Chemistry and Physics* 114: 837-841.

Kong, L.-B., Lang, J.-W., Liu, M., Luo, Y.-C. and Kang, L. 2009. Facile approach to prepare loose-packed cobalt hydroxide nano-flakes materials for electrochemical capacitors. *Journal of Power Sources* 194: 1194-1201.

Kuilla, T., Bhadra, S., Yao, D., Kim, N.H., Bose, S. and Lee, J.H. 2010. Recent advances in graphene based polymer composites. *Progress in Polymer Science* 35: 1350-1375.

Kula, I., Uğurlu, M., Karaoğlu, H. and Çelik, A. 2008. Adsorption of Cd(II) ions from aqueous solutions using activated carbon prepared from olive stone by $ZnCl_2$ activation. *Bioresource Technology* 99: 492-501.

Kulal, P.M., Dubal, D.P., Lokhande, C.D. and Fulari, V.J. 2011. Chemical synthesis of Fe_2O_3 thin films for supercapacitor application. *Journal of Alloys and Compounds* 509: 2567-2571.

Kulkarni, S.K. 2007. Nanotechnology: Principles and Practices, Capital Publishing Company, New Delhi.

Kumar, A. and Yadav, K.L. 2010. The effect of Ni substitution on magnetic, dielectric and magnetoelectric properties in $BiFe_{1-x}Ni_xO_3$ system. *Physica B-Condensed Matter* 405: 4650-4654.

Kuo, S.-L. and Wu, N.-L. 2005. Electrochemical capacitor of $MnFe_2O_4$ with NaCl electrolyte. *Electrochemical and Solid-State Letters* 8: A495-A499.

Kuo, S.-L. and Wu, N.-L. 2006. Electrochemical characterization on $MnFe_2O_4$/carbon black composite aqueous supercapacitors. *Journal of Power Sources* 162: 1437-1443.

Kurzweil, P. 2009a. Capacitors: Electrochemical double-layer capacitors. In *Encyclopedia of Electrochemical Power Sources*, ed. Garche, J., Dyer, C.K., Moseley, P.T., Ogumi, Z., Rand, D.A.J. and Scrosati, B., pp. 607-633. Amsterdam: Elsevier.

Kurzweil, P. 2009b. Capacitors: Electrochemical metal oxides capacitors. In *Encyclopedia of Electrochemical Power Sources*, ed. Garche, J., Dyer, C.K., Moseley, P.T., Ogumi, Z., Rand, D.A.J. and Scrosati, B., pp. 665-687. Amsterdam: Elsevier.

Kurzweil, P. 2009c. History: Electrochemical capacitors. In *Encyclopedia of Electrochemical Power Sources*, ed. Garche, J., Dyer, C.K., Moseley, P.T., Ogumi, Z., Rand, D.A.J. and Scrosati, B., pp. 596-606. Amsterdam: Elsevier.

Kurzweil, P. 2009d. Precious metal oxides for electrochemical energy converters: Pseudocapacitance and pH dependence of redox processes. *Journal of Power Sources* 190: 189-200.

Lapham, D.P. and Tseung, A.C.C. 2004. The effect of firing temperature, preparation technique and composition on the electrical properties of the nickel cobalt series $Ni_xCo_{1-x}O_y$. *Journal of Materials Science* 39: 251-264.

Le Viet, A., Reddy, M.V., Jose, R., Chowdari, B.V.R. and Ramakrishna, S. 2011. Electrochemical properties of bare and Ta-substituted Nb_2O_5 nanostructures. *Electrochimica Acta* 56: 1518-1528.

Lee, B.J., Sivakkumar, S.R., Ko, J.M., Kim, J.H., Jo, S.M. and Kim, D.Y. 2007. Carbon nanofibre/hydrous RuO_2 nanocomposite electrodes for supercapacitors. *Journal of Power Sources* 168: 546-552.

Lee, H.Y. and Goodenough, J.B. 1999. Supercapacitor behavior with KCl electrolyte. *Journal of Solid State Chemistry* 144: 220-223.

Lee, J.-B., Jeong, S.-Y., Moon, W.-J., Seong, T.-Y. and Ahn, H.-J. 2011. Preparation and characterization of electron-spun RuO_2-Ag_2O composite nanowires for electrochemical capacitors. *Journal of Alloys and Compounds* 509: 4336-4340.

Lee, J.Y., Liang, K., An, K.H. and Lee, Y.H. 2005. Nickel oxide/carbon nanotubes nanocomposite for electrochemical capacitance. *Synthetic Metals* 150: 153-157.

Lee, K.-T., Lee, J.-F. and Wu, N.-L. 2009. Electrochemical characterizations on MnO_2 supercapacitors with potassium polyacrylate and potassium polyacrylate-co-polyacrylamide gel polymer electrolytes. *Electrochimica Acta* 54: 6148-6153.

Lee, K.-T. and Wu, N.-L. 2008a. Manganese oxide electrochemical capacitor with potassium poly(acrylate) hydrogel electrolyte. *Journal of Power Sources* 179: 430-434.

Lee, M.-T., Chang, J.-K., Hsieh, Y.-T. and Tsai, W.-T. 2008b. Annealed Mn-Fe binary oxides for supercapacitor applications. *Journal of Power Sources* 185: 1550-1556.

Lee, Y.-H., Oh, J.-G., Oh, H.-S. and Kim, H. 2008c. Novel method for the preparation of carbon supported nano-sized amorphous ruthenium oxides for supercapacitors. *Electrochemistry Communications* 10: 1035-1037.

Lee, Y.J., Jung, J.C., Park, S., Seo, J.G., Baeck, S.-H., Yoon, J.R., Yi, J. and Song, I.K. 2010. Preparation and characterization of metal-doped carbon aerogel for supercapacitor. *Current Applied Physics* 10: 947-951.

Leitner, K., Lerf, A., Winter, M., Besenhard, J. O., Villaer-Rodil, S., Suárez-García, F., Martínez-Alonso, A. and Tascón, J. M. D. 2006. Nomex-derived activated carbon fibers as electrode materials in carbon based supercapacitors. *Journal of Power Sources* 153: 419-423.

Lenglet, M., Guillamet, R., Dürr, J., Gryffroy, D. and Vandenberghe, R.E. 1990. Electronic structure of $NiCo_2O_4$ by XANES, EXAFS and ^{61}Ni Mössbauer studies. *Solid State Communications* 74: 1035-1039.

Lewandowski, A., Olejniczak, A., Galinski, M. and Stepniak, I. 2010. Performance of carbon–carbon supercapacitors based on organic, aqueous and ionic liquid electrolytes. *Journal of Power Sources* 195: 5814-5819.

Li, H., Li, Y., Wang, R. and Cao, R. 2009. Synthesis and electrochemical capacitor performance of mesostructured nickel oxide/carbon composites by a co-casting method. *Journal of Alloys and Compounds* 481: 100-105.

Li, H., Wang, R. and Cao, R. 2008a. Physical and electrochemical characterization of hydrous ruthenium oxide/ordered mesoporous carbon composites as supercapacitor. *Microporous and Mesoporous Materials* 111: 32-38.

Li, H., Zhu, S., Xi, H. and Wang, R. 2006. Nickel oxide nanocrystallites within the wall of ordered mesoporous carbon CMK-3: synthesis and characterization. *Microporous and Mesoporous Materials* 89: 196-203.

Li, H.-Q., Wang, Y.-G., Wang, C.-X. and Xia, Y.-Y. 2008b. A competitive candidate material for aqueous supercapacitors: high surface-area graphite. *Journal of Power Sources* 185: 1557-1562.

Li, J. and Zhitomirsky, I. 2009. Electrophoretic deposition of manganese dioxide–carbon nanotube composites. *Journal of Materials Processing Technology* 209: 3452-3459.

Li, Q., Li, K.-X., Gu, J.-Y. and Fan, H. 2008c. Preparation and electrochemical characterization of cobalt-manganese oxide as electrode materials for electrochemical capacitors. *Journal of Physics and Chemistry of Solids* 69: 1733-1739.

Li, Q., Liu, J., Zou, J., Chunder, A., Chen, Y. and Zhai, L. 2011. Synthesis and electrochemical performance of multi-walled carbon nanotube/polyaniline/MnO_2 ternary coaxial nanostructures for supercapacitors. *Journal of Power Sources* 196: 565-572.

Li, Q.-Y., Li, Z.-S., Lin, L., Wang, X.Y., Wang, Y.-F., Zhang, C.-H. and Wang, H.-Q. 2010a. Facile synthesis of activated carbon/carbon nanotubes compound for supercapacitor application. *Chemical Engineering Journal* 156: 500-504.

Li, Q.-Y., Wang, H.-Q., Dai, Q.-F., Yang, J.-H. and Zhong, Y.-L. 2008d. Novel activated carbons as electrode materials for electrochemical capacitors from a series of starch. *Solid State Ionics* 179: 269-273.

Li, X., Han, C., Chen, X. and Shi, C. 2010b. Preparation and performance of straw based activated carbon for supercapacitor in non-aqueous electrolytes. *Microporous and Mesoporous Materials* 131: 303-309.

Li, Y., Tan, B. and Wu, Y. 2008e. Mesoporous Co_3O_4 nanowire arrays for lithium ion batteries with high capacity and rate capability. *Nano Letters* 8: 265-270.

Li, Z.-S., Wang, H.-Q., Huang, Y.-G., Li, Q.-Y. and Wang, X.-Y. 2010c. Manganese dioxide-coated activated mesocarbon microbeads for supercapacitors in organic electrolyte. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* 366: 104-109.

Lian, K.K. and Birss, V.I. 1991. Properties of hydrous oxide films formed at amorphous Ni-Co alloys. *Journal of The Electrochemical Society* 138: 2885-2890.

Liang, H., Chen, F., Li, R., Wang, L. and Deng, Z. 2004. Electrochemical study of activated carbon-semiconducting oxide composites as electrode materials of double-layer capacitors. *Electrochimica Acta* 49: 3463-3467.

Liang, Y.-Y., Li, H.L. and Zhang, X.-G. 2007. Solid state synthesis of hydrous ruthenium oxide for supercapacitors. *Journal of Power Sources* 173: 599-605.

Lide, D.R. 2006-2007. CRC Handbook of Chemistry and Physics 87th edition, Taylor and Francis Group, Boca Raton.

Lim, K.O. and Lim, K.S. 1992. Carbonisation of oil palm trunks at moderate temperatures. *Bioresource Technology* 40: 215-219.

Lin, C., Ritter, J.A. and Popov, B.N. 1998. Characterization of sol-gel derived cobalt oxide xerogels as electrochemical capacitors. *Journal of The Electrochemical Society* 145: 4097-4103.

Lin, C., Ritter, J.A. and Popov, B.N. 1999. Development of carbon-metal oxide supercapacitors from sol-gel derived carbon-ruthenium xerogels. *Journal of The Electrochemical Society* 146: 3155-3160.

Lin, C.-C. and Chen, H.-W. 2009. Coating manganese oxide onto graphite electrodes by immersion for electrochemical capacitors. *Electrochimica Acta* 54: 3073-3077.

Lin, C.-K., Chuang, K.-H., Lin, C.-Y., Tsay, C.-Y. and Chen, C.-Y. 2007. Manganese oxide films prepared by sol-gel process for supercapacitor application. *Surface & Coatings Technology* 202: 1272-1276.

Lin, C.K., Wu, C.H., Tsai, C.Y., Chen, C.Y. and Wang, S.C. 2010. Pseudocapacitive performance of hybrid manganese oxide films with multiwalled-CNT additions. *Surface & Coatings Technology* 205: 1595-1598.

Liou, T.-H. and Wu, S.-J. 2009. Characteristics of microporous/mesoporous carbons prepared from rice husk under base- and acid-treated conditions. *Journal of Hazardous Materials* 171: 693-703.

Liu, C.-C., Tsai, D.-S., Chung, W.-H., Li, K.-W., Lee, K.-Y. and Huang, Y.-S. 2011. Electrochemical micro-capacitors of patterned electrodes loaded with manganese oxide and carbon nanotubes. *Journal of Power Sources* 196: 5761-5768.

Liu, H., Mao, C., Lu, J. and Wang, D. 2009a. Electronic power transformer with supercapacitors storage energy system. *Electric Power Systems Research* 79: 1200-1208.

Liu, J., Cao, G., Yang, Z., Wang, D., Dubois, D., Zhou, X., Graff, G.L., Pederson, L.R. and Zhang, J.-G. 2008a. Oriented nanostructures for energy conversion and storage. *ChemSusChem* 1: 676-697.

Liu, C.-G., Lee, Y.-S., Kim, Y.-J., Song, I.-C. and Kim, J.-H. 2009b. Electrochemical characteristics of hydrothermally deposited nickel hydroxide on multi-walled carbon nanotube for supercapacitor electrode. *Synthetic Metals* 159: 2009-2012.

Liu, E.-H., Li, W., Li, J., Meng, X.-Y., Ding, R. and Tan, S.-T. 2009c. Preparation and characterization of nanostructured NiO/MnO_2 composite electrode for electrochemical supercapacitors. *Materials Research Bulletin* 44: 1122-1126.

Liu, E.-H., Meng, X.-Y., Ding, R., Zhou, J.-C. and Tan, S.-T. 2007a. Potentiodynamical co-deposited manganese oxide/carbon composite for high capacitance electrochemical capacitors. *Materials Letters* 61: 3486-3489.

Liu, H., He, P., Li, Z., Liu, Y. and Li, J. 2006. A novel nickel-based mixed rare-earth oxide/activated carbon supercapacitor using room temperature ionic liquid electrolyte. *Electrochimica Acta* 51: 1925-1931.

Liu, X., Huber, T.A., Kopac, M.C. and Pickup P.G. 2009d. Ru oxide/carbon nanotube composites for supercapacitors prepared by spontaneous reduction of Ru(VI) and Ru(VII). *Electrochimica Acta* 54: 7141-7147.

Liu, X. and Pickup, P.G. 2008b. Ru oxide supercapacitors with high loadings and high power and energy densities. *Journal of Power Sources* 176: 410-416.

Liu, X.-M., Fu, S.-Y., Xiao, H.-M. and Huang, C.-J. 2005. Synthesis of nanocrystalline spinel $CoFe_2O_4$ via a polymer-pyrolysis route. *Physica B-Condensed Matter* 370: 14-21.

Liu, X.-M., Zhang, R., Zhan, L., Long, D.-H., Qiao, W.-M., Yang, J.-H. and Ling, L.-C. 2007b. Impedance of carbon aerogel/activated carbon composites as electrodes of electrochemical capacitors in aprotic electrolyte. *New Carbon Materials* 22: 153-158.

Lokhande, C.D., Dubal, D.P. and Joo, O.-S. 2011. Metal oxide thin film based supercapacitors. *Current Applied Physics* 11: 255-270.

Long, J.W., Swider, K.E., Merzbacher, C.I. and Rolison, D.R. 1999. Voltammetric characterization of ruthenium oxide-based aerogels and other RuO_2 solids: the nature of capacitance in nanostructured materials. *Langmuir* 15: 780-785.

Long, J.W., Young, A.L. and Rolison, D.R. 2003. Spectrochemical characterization of nanostructured, mesoporous manganese oxide in aqueous electrolytes. *Journal of The Electrochemical Society* 150: A1161-A1165.

López-González, J.D., Martínez-Vilchez, F. and Rodríguez-Reinoso, F. 1980. Preparation and characterization of active carbons from olive stones. *Carbon* 18: 413-418.

Lota, K., Khomenko, V. and Frackowiak, E. 2004. Capacitance properties of poly(3,4-ethylenedioxythiophene)/carbon nanotubes composites. *Journal of Physics and Chemistry of Solids* 65: 295-301.

Lu, T., Pan, L., Li, H., Zhu, G., Lv, T., Liu, X., Sun, Z., Chen, T. and Chua, D.H.C. 2011a. Microwave-assisted synthesis of graphene–ZnO nanocomposite for electrochemical supercapacitors. *Journal of Alloys and Compounds* 509: 5488-5492.

Lu, X., Dou, H., Gao, B., Yuan, C., Yang, S., Hao, L., Shen, L. and Zhang, X. 2011b. A flexible graphene/multiwalled carbon nanotube film as a high performance electrode material for supercapacitors. *Electrochimica Acta* 56: 5115-5121.

Lu, T., Zhang, Y., Li, H., Pan, L., Li, Y. and Sun, Z. 2010a. Electrochemical behaviors of graphene-ZnO and graphene-SnO_2 composite films for supercapacitors. *Electrochimica Acta* 55: 4170-4173.

Lu, W., Qu, L., Henry, K. and Dai, L. 2009. High performance electrochemical capacitors from aligned carbon nanotube electrodes and ionic liquid electrolytes. *Journal of Power Sources* 189: 1270-1277.

Lu, Y., Wang, Y., Zou, Y., Jiao, Z., Zhao, B., He, Y. and Wu, M. 2010b. Macroporous Co_3O_4 platelets with excellent rate capability as anodes for lithium ion batteries. *Electrochemistry Communications* 12: 101-105.

Lua, A.C., Lau, F.W. and Guo, J. 2006. Influence of pyrolysis conditions on pore development of oil-palm-shell activated carbons. *Journal of Analytical and Applied Pyrolysis* 76: 96-102.

Lua, A.C. and Guo, J. 1999. Chars pyrolyzed from oil palm wastes for activated carbon preparation. *Journal of Environmental Engineering* 125: 72-76.

Luo, J.-M., Gao, B. and Zhang, X.-G. 2008. High capacitive performance of nanostructured Mn-Ni-Co oxide composites for supercapacitor. *Materials Research Bulletin* 43: 1119-1125.

Maaz, K., Karim, S., Mashiatullah, A., Liu, J., Hou, M.D., Sun, Y.M., Duan, J.L., Yao, H.J., Mo, D. and Chen, Y.F. 2009. Structural analysis of nickel doped cobalt ferrite nanoparticles prepared by coprecipitation route. *Physica B-Condensed Matter* 404: 3947-3951.

Machefaux, E., Brousse, T., Bélanger, D. and Guyomard, D. 2007. Supercapacitor behavior of new substituted manganese dioxides. *Journal of Power Sources* 165: 651-655.

Mallouki, M., Tran-Van, F., Sarrazin, C., Chevrot, C. and Fauvarque, J.F. 2009. Electrochemical storage of polypyrrole-Fe_2O_3 nanocomposites in ionic liquids. *Electrochimica Acta* 54: 2992-2997.

Marco, J.F., Gancedo, J.R., Gracia, M., Gautier, J.L., Ríos, E. and Berry, F.J. 2000. Characterization of the nickel cobaltite, $NiCo_2O_4$, prepared by several methods: an XRD, XANES, EXAFS, and XPS study. *Journal of Solid State Chemistry* 153: 74-81.

Marco, J.F., Gancedo, J.R., Gracia, M., Gautier, J.L., Ríos, E.I., Palmer, H.M., Greaves, C. and Berry, F.J. 2001. Cation distribution and magnetic structure of the ferromagnetic spinel $NiCo_2O_4$. *Journal of Materials Chemistry* 11: 3087-3093.

Martín De Vidales, J.L., García-Chain, P., Rojas, R.M., Vila, E. and García-Martínez, O. 1998. Preparation and characterization of spinel-type Mn-Ni-Co-O negative temperature coefficient ceramic thermistors. *Journal of Materials Science* 33: 1491-1496.

Mathe, V.L. and Sheikh, A.D. 2010. Magnetostrictive properties of nanocrystalline Co-Ni ferrites. *Physica B-Condensed Matter* 405: 3594-3598.

Mecerreyes, D. 2011. Polymeric ionic liquids: broadening the properties and applications of polyelectrolytes. *Progress in Polymer Science* 36: 1629-1648.

Mehandjiev, D., Zhecheva, E. and Angelov, S. 1985. On the possibility of formation of 3*d*-transition metal mixed oxides with spinel structure. *Thermochimica Acta* 95: 155-158.

Meher, S.K., Justin, P. and Ranga Rao, G. 2010. Pine-cone morphology and pseudocapacitive behavior of nanoporous nickel oxide. *Electrochimica Acta* 55: 8388-8396.

Merino, C., Soto, P., Vilaplana-Ortego, E., Gomez de Salazar, J.M., Pico, F. and Rojo, J.M. 2005. Carbon nanofibres and activated carbon nanofibres as electrodes in supercapacitors. *Carbon* 43: 551-557.

Miller, J.M. and Dunn, B. 1999. Morphology and electrochemistry of ruthenium/carbon aerogel nanostructures. *Langmuir* 15: 799-806.

Miller, J.R. 2009. Capacitors: Overview. In *Encyclopedia of Electrochemical Power Sources*, ed. Garche, J., Dyer, C.K., Moseley, P.T., Ogumi, Z., Rand, D.A.J. and Scrosati, B., pp. 587-599. Amsterdam: Elsevier.

Min, C.-K., Wu, T.-B., Yang, W.-T. and Li, C.-L. 2009. Nanocrystalline ruthenium oxide of fine mesoporosity prepared by templating for electrochemical capacitor applications. *Materials Chemistry and Physics* 117: 70-73.

Mitra, S., Lokesh, K.S. and Sampath, S. 2008. Exfoliated graphite-ruthenium oxide composite electrodes for electrochemical supercapacitors. *Journal of Power Sources* 185: 1544-1549.

Mohanty, P., Rath, C., Mallick, P., Biswal, R. and Mishra, N.C. 2010. UV–visible studies of nickel oxide thin film grown by thermal oxidation of nickel. *Physica B-Condensed Matter* 405: 2711-2714.

Monk, P.M.S. and Ayub, S. 1997. Solid-state properties of thin film electrochromic cobalt-nickel oxide. *Solid State Ionics* 99: 115-124.

Mora, E., Ruiz, V., Santamaría, R., Blanco, C., Granda, M., Menéndez, R., Juarez-Galán, J.M. and Rodríguez-Reinoso, F. 2006. Influence of mesophase activation conditions on the specific capacitance of the resulting oxides. *Journal of Power Sources* 156: 719-724.

Nam, H.-S., Yoon, J.-K., Ko, J.M. and Kim, J.-D. 2010. Electrochemical capacitors of flower-like and nanowire structured MnO_2 by a sonochemical method. *Materials Chemistry and Physics* 123: 331-336.

Nam, K.-W. and Kim, K.-B. 2002. A study of the preparation of NiO_x electrode via electrochemical route for supercapacitor applications and their charge storage mechanism. *Journal of The Electrochemical Society* 149: A346-A354.

Nam, K.-W., Kim, K.-H., Lee, E.-S., Yoon, W.-S., Yang, X.-Q. and Kim, K.-B. 2008. Pseudocapacitive properties of electrochemically prepared nickel oxides on 3-dimensional carbon nanotube film substrates. *Journal of Power Sources* 182: 642-652.

Nam, K.-W., Yoon, W.-S. and Kim, K.-B. 2002. X-ray absorption spectroscopy studies of nickel oxide thin film electrodes for supercapacitors. *Electrochimica Acta* 47: 3201-3209.

Nathan, T., Aziz, A., Noor, A.F. and Prabaharan, S.R.S. 2008. Nanostructured NiO for electrochemical capacitors: synthesis and electrochemical properties. *Journal of Solid State Electrochemistry* 12: 1003-1009.

Ni, J., Lu, W., Zhang, L., Yue, B., Shang, X. and Lv, Y. 2009. Low-temperature synthesis of monodisperse 3D manganese oxide nanoflowers and their pseudocapacitance properties. *Journal of Physical Chemistry C* 113: 54-60.

NuLi, Y., Zhang, P., Guo, Z., Liu, H and Yang, J. 2008. $NiCo_2O_4$/C nanocomposite as a highly reversible anode material for lithium-ion batteries. *Electrochemical and Solid-State Letters* 11: A64-A67.

Obreja, V.V.N. 2008. On the performance of supercapacitors with electrodes based on carbon nanotubes and carbon activated material-a review. *Physica E-Low-Dimensional Systems & Nanostructures* 40: 2596-2605.

Oke, S., Yamamoto, M., Shinohara, K., Takikawa, H., Xiaojun, H., Itoh, S., Yamaura, T., Miura, K., Yoshikawa, K., Okawa, T. and Aoyagi, N. 2009. Specific capacitance of electrochemical capacitor using RuO_2 loading arc-soot/activated carbon composite electrode. *Chemical Engineering Journal* 146: 434-438.

Olivares-Marín, M., Fernández, J.A., Lázaro, M.J., Fernández-González, C., Macías-García, A., Gómez-Serrano, V., Stoeckli, F. and Centeno, T.A. 2009. Cherry stones as precursor of activated carbons for supercapacitors. *Materials Chemistry and Physics* 114: 323-327.

Orazem, M.E. and Tribollet, B. 2008. Electrochemical Impedance Spectroscopy, John Wiley, New York.

Owings, R.R., Exarhos, G.J., Windisch, C.F., Holloway, P.H. and Wen, J.G. 2005. Process enhanced polaron conductivity of infrared transparent nickel-cobalt oxide. *Thin Solid Films* 483: 175-184.

Pan, S.-J., Shih, Y.-J., Chen, J.-R., Chang, J.-K. and Tsai, W.-T. 2009. Selective micro-etching of duplex stainless steel for preparing manganese oxide supercapacitor electrode. *Journal of Power Sources* 187: 261-267.

Pang, S.-C., Anderson, M.A. and Chapman, T.W. 2000. Novel electrode materials for thin-film ultracapacitors: comparison of electrochemical properties of sol–gel-derived and electrodeposited manganese dioxide. *Journal of The Electrochemical Society* 147: 444-450.

Panić, V.V., Dekanski, A.B. and Stevanović, R.M. 2010. Sol-gel processed thin-layer ruthenium oxide/carbon black supercapacitors: a revelation of the energy storage issues. *Journal of Power Sources* 195: 3969-3976.

Park, B.-O., Lokhande, C.D., Park, H.-S., Jung, K.-D. and Joo, O.-S. 2004. Electrodeposited ruthenium oxide (RuO_2) films for electrochemical supercapacitors. *Journal of Materials Science* 39: 4313-4317.

Park, J.H., Park, O.O., Shin, K.H., Chang, S.J. and Kim, J.H. 2002. An electrochemical capacitor based in a $Ni(OH)_2$/activated carbon composite electrode. *Electrochemical and Solid-State Letters* 5: H7-H10.

Patake, V.D., Joshi, S.S., Lokhande, C.D. and Joo, O.-S. 2009. Electrodeposited porous and amorphous copper oxide film for application in supercapacitor. *Materials Chemistry and Physics* 114: 6-9.

Patake, V.D., Pawar, S.M., Shinde, V.R., Gujar, T.P. and Lokhande, C.D. 2010. The growth mechanism and supercapacitor study of anodically deposited amorphous ruthenium oxide films. *Current Applied Physics* 10: 99-103.

Pawar, D.K., Pawar, S.M., Patil, P.S. and Kolekar, S.S. 2011. Synthesis of nanocrystalline nickel-zinc ferrite ($Ni_{0.8}Zn_{0.2}Fe_2O_4$) thin films by chemical bath deposition method. *Journal of Alloys and Compounds* 509: 3587-3591.

Pietrzak, R., Jurewicz, K., Nowicki, P., Babeł, K. and Wachowska, H. 2007. Microporous activated carbons from ammoxidised anthracite and their capacitance behaviours. *Fuel* 86: 1086-1092.

Pietrzak, R., Jurewicz, K., Nowicki, P., Babeł, K. and Wachowska, H. 2010. Nitrogen-enriched bituminous coal-based active carbons as materials for supercapacitors. *Fuel* 89:3457-3467.

Prabaharan, S.R.S., Vimala, R. and Zainal, Z. 2006. Nanostructured mesoporous carbon as electrodes for supercapacitors. *Journal of Power Sources* 161: 730-736.

Pradhan, S.D., Sathaye, S.D., Patil, K.R. and Mitra, A. 2001. Low temperature synthesis of stoichiometric lead zirconate by coprecipitation in non-aqueous medium. *Materials Letters* 48l: 351-355.

Peña, O., Ma, Y., Barahona, P., Bahout, M., Durán, P., Moure, C., Baibich, M.N. and Martinez, G. 2005. Magnetic properties of $NiMn_{2-x}Co_xO_4$ spinel oxides. *Journal of the European Ceramic Society* 25: 3051-3054.

Peshev, P., Toshev, A. and Gyurov, G. 1989. Preparation of high-dispersity MCo_2O_4 (M = Mg, Ni, Zn) spinels by thermal dissociation of coprecipitated oxalates. *Materials Research Bulletin* 24: 33-40.

Pico, F., Ibañez, J., Lillo-Rodenas, M.A., Linares-Solano, A., Rojas, R.M., Amarilla, J.M. and Rojo, J.M. 2008. Understanding $RuO_2 \cdot xH_2O$/carbon nanofibre composites as supercapacitor electrodes. *Journal of Power Sources* 176: 417-425.

Pico, F., Morales, E., Fernandez, J.A., Centeno, T.A., Ibañez, J., Rojas, R.M., Amarilla, J.M. and Rojo, J.M. 2009. Ruthenium oxide/carbon composites with microporous or mesoporous carbon as support and prepared by two procedures. A comparative study as supercapacitor electrodes. *Electrochimica Acta* 54: 2239-2245.

Pico, F., Pecharroman, C., Ansón, A., Martínez, M.T. and Rojo, J.M. 2007. Understanding carbon-carbon composites as electrodes of supercapacitors: a study by AC and DC measurements. *Journal of The Electrochemical Society* 154: A579-A586.

Pradhan, S.D., Sathaye, S.D., Patil, K.R. and Mitra, A. 2001. Low temperature synthesis of stoichiometric lead zirconate by coprecipitation in non-aqueous medium. *Materials Letters* 48: 351-355.

Prasad, K.R., Koga, K. and Miura, N. 2004. Electrochemical deposition of nanostructured indium oxide: high-performance electrode material for redox supercapacitors. *Chemistry of Materials* 16: 1845-1847.

Prasad, K.R. and Miura, N. 2004a. Potentiodynamically deposited nanostructured manganese dioxide as electrode material for electrochemical redox supercapacitors. *Journal of Power Sources* 135: 354-360.

Prasad, K.R. and Miura, N. 2004b. Electrochemical synthesis and characterization of nanostructured tin oxide for electrochemical redox supercapacitors. *Electrochemistry Communications* 6: 849-852.

Qin, C., Lu, X., Yin, G., Jin, Z., Tan, Q. and Bai, X. 2011. Study of activated nitrogen-enriched carbon and nitrogen-enriched carbon/carbon aerogel composite as cathode materials for supercapacitors. *Materials Chemistry and Physics* 126: 453-458.

Qing, X., Liu, S., Huang, K., Lv, K., Yang, Y., Lu, Z., Fang, D. and Liang, X. 2011. Facile synthesis of Co_3O_4 nanoflowers grown on Ni foam with superior electrochemical performance. *Electrochimica Acta* 56: 4985-4991.

Qu, D. 2002. Studies of the activated carbons used in double-layer supercapacitors. *Journal of Power Sources* 109: 403-411.

Ragupathy, P., Park, D.H., Campet, G.C., Vasan, H.N., Hwang, S.-J., Choy, J.-H. and Munichandraiah, N. 2009. Remarkable capacity retention of nanostructured manganese oxide upon cycling as an electrode material for supercapacitor. *Journal of Physical Chemistry C* 113: 6303-6309.

Rajeswari, J., Kishore, P.S., Viswanathan, B. and Varadarajan, T.K. 2009. One-dimensional MoO_2 nanorods for supercapacitor applications. *Electrochemistry Communications* 11: 572-575.

Ramani, M., Haran, B.S., White, R.E., Popov, B.N. and Arsov, L. 2001. Studies on activated carbon capacitor materials loaded with different amounts of ruthenium oxide. *Journal of Power Sources* 93: 209-214.

Rao, C.N.R. and Raveau, B. 1998. Transition Metal Oxides: Structure, Properties, and Synthesis of Ceramic Oxides (second edition), John Wiley, New York.

Rashkova, V., Kitova, S., Konstantinov, I. and Vitanov, T. 2002. Vacuum evaporated thin films of mixed cobalt and nickel oxides as electrocatalyst for oxygen evolution and reduction. *Electrochimica Acta* 47: 1555-1560.

Rasiyah, P., Tseung, A.C.C. and Hibbert, D.B. 1982. A mechanistic study of oxygen evolution on $NiCo_2O_4$. *Journal of The Electrochemical Society* 129: 1724-1727.

Reddy, R.N. and Reddy, R.G. 2003. Sol-gel MnO_2 as an electrode material for electrochemical capacitors. *Journal of Power Sources* 124: 330-337.

Rodrigues, E.C., Carvalhob, C.T., de Siqueirac, A.B., Bannach, G. and Ionashiro, M. 2009. Synthesis, characterization and thermal behaviour on solid tartrates of some bivalent metal ions. *Thermochimica Acta* 496: 156-160.

Roginskaya, Y.E., Morozova, O.V., Lubnin, E.N., Ulitina, Y.E., Lopukhova, G.V. and Trasatti, S. 1997. Characterization of bulk and surface composition of $Co_xNi_{1-x}O_y$ mixed oxides for electrocatalysis. *Langmuir* 13: 4621-4627.

Rondinini, S., Ardizzone, S., Cappelletti, G., Minguzzi, A. and Vertova, A. 2009. Sol-gel synthesis. In *Encyclopedia of Electrochemical Power Sources*, ed. Garche, J., Dyer, C.K., Moseley, P.T., Ogumi, Z., Rand, D.A.J. and Scrosati, B., pp. 613-624. Amsterdam: Elsevier.

Rudge, A., Davey, J., Raistrick, I., Gottesfeld, S. and Ferraris, J.P. 1994. Conducting polymers as active materials in electrochemical capacitors. *Journal of Power Sources* 47: 89-107.

Rufford, T.E., Hulicova-Jurcakova, D., Zhu, Z. and Lu, G.Q. 2008. Nanoporous carbon electrode from waste coffee beans for high performance supercapacitors. *Electrochemistry Communications* 10: 1594-1597.

Ruiz, V., Blanco, C., Granda, M., Menéndez, R. and Santamaría, R. 2008a. Effect of the thermal treatment of carbon-based electrodes on the electrochemical performance of supercapacitors. *Journal of Electroanalytical Chemistry* 618: 17-23.

Ruiz, V., Blanco, C., Granda, M. and Santamaría, R. 2008b. Enhanced life-cycle supercapacitors by thermal treatment of mesophase-derived activated carbons. *Electrochimica Acta* 54: 305-310.

Ruiz, V., Blanco, C., Raymundo-Piñero, E., Khomenko, V., Béguin, F. and Santamaría, R. 2007. Effects of thermal treatment of activated carbon on the electrochemical behaviour in supercapacitors. *Electrochimica Acta* 52: 4969-4973.

Ruiz, V., Blanco, C., Santamaría, R., Ramos-Fernández, J.M., Martínez-Escandell, M., Sepúlveda-Escribano, A. and Rodríguez-Reinoso, F. 2009a. An activated carbon monolith as an electrode material for supercapacitors. *Carbon* 47: 195-200.

Ruiz, V., Santamaría, R., Granda, M. and Blanco, C. 2009b. Long term cycling of carbon based supercapacitors in aqueous media. *Electrochimica Acta* 54: 4481-4486.

Saliger, R., Fischer, U., Herta, C and Fricke, J. 1998. High surface area carbon aerogels for supercapacitors. *Journal of Non-Crystalline Solids* 225: 81-85.

Salunkhe, R.R., Jang, K., Yu, H., Yu, S., Ganesh, T., Han, S.-H. and Ahn, H. 2011. Chemical synthesis and electrochemical analysis of nickel cobaltite nanostructures for supercapacitor applications. *Journal of Alloys and Compounds* 509: 6677-6682.

Salavati-Niasari, M., Davar, F. and Fereshteh, Z. 2010. Synthesis of nickel and nickel oxide nanoparticles via heat-treatment of simple octanoate precursor. *Journal of Alloys and Compounds* 494: 410-414.

Sanjinés, R., Abad, M.D., Vâju, Cr., Smajda, R., Mionić, M. and Magrez, A. 2011. Electrical properties and applications of carbon based nanocomposite materials: an overview. *Surface & Coatings Technology* 206: 727-733.

Santhanam, R and Rambabu, B. 2010. High rate cycling performance of $Li_{1.05}Ni_{1/3}Co_{1/3}Mn_{1/3}O_2$ materials prepared by sol-gel and co-precipitation methods for lithium-ion batteries. *Journal of Power Sources* 195: 4313-4317.

Sawaby, A., Selim, M.S., Marzouk, S.Y., Mostafa, M.A. and Hosny, A. 2010. Structure, optical and electrochromic properties of NiO thin films. *Physica B-Condensed Matter* 405: 3412-3420.

Selvakumar, M., Krishna Bhat, D., Aggarwal, A.M., Iyer, S.P. and Sravani, G. 2010. Nano ZnO-activated carbon composite electrodes for supercapacitors. *Physica B-Condensed Matter* 405: 2286-2289.

Seo, M.-K. and Park, S.-J. 2009. Electrochemical characteristics of activated carbon nanofiber electrodes for supercapacitors. *Materials Science and Engineering B-Advanced Functional Solid-State Materials* 164: 106-111.

Serebrennikova, I. and Birss, V.I. 2001. Structural and compositional properties of sol-gel formed Ni, Co and Ni-Co oxide films. *Journal of Materials Science* 36: 4331-4343.

Serrano, E., Rus, G. and García-Martínez, J. 2009. Nanotechnology for sustainable energy. *Renewable and Sustainable Energy Reviews* 13: 2373-2384.

Sevilla, M., Álvarez, S., Centeno, T.A., Fuertes, A.B. and Stoeckli, F. 2007. Performance of templated mesoporous carbons in supercapacitors. *Electrochimica Acta* 52: 3207-3215.

Shakir, I., Shahid, M., Yang, H.W. and Kang, D.J. 2010. Structural and electrochemical characterization of α-MoO_3 nanorod-based electrochemical energy storage devices. *Electrochimica Acta* 56: 376-380.

Shan, Y. and Gao, L. 2007. Formation and characterization of multi-walled carbon nanotubes/Co_3O_4 nanocomposites for supercapacitors. *Materials Chemistry and Physics* 103: 206-210.

Sharma, P. and Bhatti, T.S. 2010. A review on electrochemical double-layer capacitors. *Energy Conversion and Management* 51: 2901-2912.

Sharma, Y.K., Kharkwal, M., Uma, S. and Nagarajan, R. 2009. Synthesis and characterization of titanates of the formula $MTiO_3$ (M = Mn, Fe, Co, Ni and Cd) by co-precipitation of mixed metal oxalates. *Polyhedron* 28: 579-585.

Shi, H. 1996. Activated carbons and double layer capacitance. *Electrochimica Acta* 41: 1633-1639.

Shinde, V.R., Mahadik, S.B., Gujar, T.P. and Lokhande, C.D. 2006. Supercapacitive cobalt oxide (Co_3O_4) thin films by spray pyrolysis. *Applied Surface Science* 252: 7487-7492.

Shinomiya, T., Gupta, V. and Miura, N. 2006. Effects of electrochemical-deposition method and microstructure on the capacitive characteristics of nano-sized manganese oxide. *Electrochimica Acta* 51: 4412-4419.

Shukla, A.K., Sampath, S. and Vijayamohanan, K. 2000. Electrochemical supercapacitors: energy storage beyond batteries. *Current Science* 79: 1656-1661.

Simon, P. and Gogotsi, Y. 2008. Materials for electrochemical capacitors. *Nature Materials* 7: 845-854.

Singh, R.-N., Hamdani, M., Koenig, J.-F., Poillerat, G., Gautier, J.L. and Chartier, P. 1990. Thin films of Co_3O_4 and $NiCo_2O_4$ obtained by the method of chemical spray pyrolysis for electrocatalysis. III. The electrocatalysis of oxygen evolution. *Journal of Applied Electrochemistry* 20: 442-446.

Singh, R.-N., Koenig, J.-F., Poillerat, G. and Chartier, P. 1991. Thin films of Co_3O_4 and $NiCo_2O_4$ prepared by the method of chemical spray pyrolysis for electrocatalysis. Part IV. The electrocatalysis of oxygen reduction. *Journal of Electroanalytical Chemistry* 314: 241-257.

Sivakkumar, S.R., Ko, J.M., Kim, D.Y., Kim, B.C. and Wallace, G.G. 2007. Performance evaluation of CNT/polypyrrole/MnO_2 composite electrodes for electrochemical capacitors. *Electrochimica Acta* 52: 7377-7385.

Snook, G.A., Kao, P. and Best, A.S. 2011. Conducting-polymer-based supercapacitor devices and electrodes. *Journal of Power Sources* 196: 1-12.

Song, F., Huang, L., Chen, D. and Tang, W. 2008. Preparation and characterization of nanosized Zn-Co spinel oxide by solid state reaction method. *Materials Letters* 62: 543-547.

Sopčić, S., Mandić, Z., Inzelt, G., Roković, M.K. and Meštrović, E. 2011. Ion dynamics in the pseudocapacitive reaction of hydrous ruthenium oxide. Effect of the temperature pre-treatment. *Journal of Power Sources* 196: 4849-4858.

Soudan, P., Gaudet, J., Guay, D., Bélanger, D. and Schulz, R. 2002. Electrochemical properties of ruthenium-based nanocrystalline materials as electrodes for supercapacitors. *Chemistry of Materials* 14: 1210-1215.

Sreekantan, S., Gee, L.R. and Lockman, Z. 2009. Room temperature anodic deposition and shape control of one-dimensional nanostructured zinc oxide. *Journal of Alloys and Compounds* 476: 513-518.

Srinivasan, R., Yogamalar, R. and Bose A.C. 2010. Structural and optical studies of yttrium oxide nanoparticles synthesized by co-precipitation method. *Materials Research Bulletin* 45: 1165-1170.

Srinivasan, V. and Weidner, J.W. 1997. An electrochemical route for making porous nickel oxide electrochemical capacitors. *Journal of The Electrochemical Society* 144: L210-L213.

Srinivasan, V. and Weidner, J.W. 2000. Studies on the capacitance of nickel oxide films: effect of heating temperature and electrolyte concentration. *Journal of The Electrochemical Society* 147: 880-885.

Srivastava, D.N., Perkas, N., Seisenbaeva, G.A., Koltypin, Y., Kessler, V.G. and Gedanken, A. 2003. Preparation of porous cobalt and nickel oxides from corresponding alkoxides using a sonochemical technique and its application as a catalyst in the oxidation of hydrocarbons. *Ultrasonics Sonochemistry* 10: 1-9.

Su, Y.C., Chen, C.A., Chen, Y.M., Huang, Y.S., Lee, K.Y. and Tiong, K.K. 2011. Characterization of RuO_2 nanocrystals deposited on carbon nanotubes by reactive sputtering. *Journal of Alloys and Compounds* 509: 2011-2015.

Subramanian, V., Zhu, H., Vajtai, R., Ajayan, P.M. and Wei, B. 2005. Hydrothermal synthesis and pseudocapacitance properties of MnO_2 nanostructures. *Journal of Physical Chemistry B* 109: 20207-20214.

Swathi, T. and Buvaneswari, G. 2008. Application of $NiCo_2O_4$ as a catalyst in the conversion of *p*-nitrophenol to *p*-aminophenol. *Materials Letters* 62: 3900-3902.

Taberna, P.L., Chevallier, G., Simon, P., Plée, D. and Aubert, T. 2006. Activated carbon-carbon nanotube composite porous film for supercapacitor applications. *Materials Research Bulletin* 41: 48-484.

Tai, Y.-L. and Teng, H. 2004. Modification of porous carbon with nickel oxide impregnation to enhance the electrochemical capacitance and conductivity. *Carbon* 42: 2329-2366.

Tan, I.A.W., Hameed, B.H. and Ahmad, A.L. 2007. Equilibrium and kinetic studies on basic dye adsorption by oil palm fibre activated carbon. *Chemical Engineering Journal* 127: 111-119.

Tan, I.A.W., Ahmad, A.L. and Hameed, B.H. 2008. Enhancement of basic dye adsorption uptake from aqueous solutions using chemically modified oil palm shell activated carbon. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* 318: 88-96.

Tang, X., Li, H., Liu, Z.-H., Yang, Z. and Wang, Z. 2011. Preparation and capacitive property of manganese oxide nanobelt bundles with birnessite-type structure. *Journal of Power Sources* 196: 855-859.

Tang, N., Tian, X., Yang, C. and Pi, Z. 2009a. Facile synthesis of α-MnO_2 nanostructures for supercapacitors. *Materials Research Bulletin* 44: 2062-2067.

Tang, X., Liu, Z.-H., Zhang, C., Yang, Z. and Wang, Z. 2009b. Synthesis and capacitive property of hierarchical hollow manganese oxide nanospheres with large specific surface area. *Journal of Power Sources* 193: 939-943.

Tareen, J.A.K., Małecki, A., Doumerc, J.P., Launay, J.C., Dordor, P., Pouchard, M. and Hagenmuller, P. 1984. Growth and electrical properties of pure and Ni-doped Co_3O_4 single crystals. *Materials Research Bulletin* 19: 989-997.

Teng, F., Santhanagopalan, S., Wang, Y. and Meng, D.D. 2010. *In-situ* hydrothermal synthesis of three-dimensional MnO_2-CNT nanocomposites and their electrochemical properties. *Journal of Alloys and Compounds* 499: 259-264.

Teng, H., Yeh, T.-S. and Hsu, L.-Y. 1998. Preparation of activated carbon from bituminous coal with phosphoric acid activation. *Carbon* 36: 1387-1395.

Tharayil, N.J., Sagar, S., Raveendran, R. and Vaidyan, A.V. 2007. Dielectric studies of nanocrystalline nickel-cobalt oxide. *Physica B-Condensed Matter* 399: 1-8.

Thota, S., Kumar, A. and Kumar J. 2009. Optical, electrical and magnetic properties of Co_3O_4 nanocrystallites obtained by thermal decomposition of sol-gel derived oxalates. *Materials Science and Engineering B-Advanced Functional Solid-State Materials* 164: 30-37.

Thounthong, P., Raël, S. and Davat, B. 2006. Control strategy of fuel cell/supercapacitors hybrid power sources for electric vehicle. *Journal of Power Sources* 158: 806-814.

Tian, Y., Song, Y., Tang, Z., Guo, Q. and Liu, L. 2008. Influence of high temperature treatment of porous carbon on the electrochemical performance in supercapacitor. *Journal of Power Sources* 184: 675-681.

Timperman, L., Galiano, H., Lemordant, D. and Anouti, M. 2011. Phosphonium-based protic ionic liquid as electrolyte for carbon-based supercapacitors. *Electrochemistry Communications* 13: 1112-1115.

Toniolo, J.C., Takimi, A.S. and Bergmann, C.P. 2010. Nanostructured cobalt oxides (Co_3O_4 and CoO) and metallic Co powders synthesized by the solution combustion method. *Materials Research Bulletin* 45: 672-676.

Toupin, M., Bélanger, D., Hill, I.R. and Quinn, D. 2005. Performance of experimental carbon blacks in aqueous supercapacitors. *Journal of Power Sources* 140: 203-210.

Trasatti, S. 1981a. Electrodes of Conductive Metallic Oxides (Part A), Elsevier Scientific Publishing Company, Amsterdam.

Trasatti, S. 1981b. Electrodes of Conductive Metallic Oxides (Part B), Elsevier Scientific Publishing Company, Amsterdam.

Verma, S., Joshi, H.M., Jagadale, T., Chawla, A., Chandra, R. and Ogale, S. 2008. Nearly monodispersed multifunctional $NiCo_2O_4$ spinel nanoparticles: magnetism, infrared transparency, and radiofrequency absorption. *Journal of Physical Chemistry C* 112: 15106-15112.

Vijayabarathi, T., Muzhumathi, S. and Noel, M. 2007. The use of hydrated nickel-cobalt mixed oxide electrodes for oxidation of aliphatic and aromatic alcohols. *Journal of Applied Electrochemistry* 37: 297-301.

Vioux, A., Viau, L., Volland, S. and Le Bideau, J. 2010. Use of ionic liquids in sol-gel; ionogels and applications. *Comptes Rendus Chimie* 13: 242-255.

Wallace, G.G., Chen, J., Mozer, A.J., Forsyth, M., MacFarlane, D.R. and Wang, C. 2009. Nanoelectrodes: energy storage and conversion. *Materials Today* 12: 20-27.

Wan Daud, W.M.A. and Wan Ali, W.S. 2004. Comparison on pore development of activated carbon produced from palm shell and coconut shell. *Bioresource Technology* 93: 63-69.

Wang, C.-C. and Hu, C.-C. 2005. Electrochemical and textural characterization of binary Ru-Sn oxides synthesized under mild hydrothermal conditions for supercapacitors. *Electrochimica Acta* 50: 2573-2581.

Wang, D., Ni, W., Pang, H., Lu, Q., Huang, Z. and Zhao, J. 2010a. Preparation of mesoporous NiO with a bimodal pore size distribution and application in electrochemical capacitors. *Electrochimica Acta* 55: 6830-6835.

Wang, D., Wang, Q. and Wang, T. 2011a. Morphology-controllable synthesis of cobalt oxalates and their conversion to mesoporous Co_3O_4 nanostructures for application in supercapacitors. *Inorganic Chemistry* 50: 6482-6492.

Wang, D.-W., Li, F. and Cheng, H.-M. 2008a. Hierarchical porous nickel oxide and carbon as electrode materials for asymmetric supercapacitor. *Journal of Power Sources* 185: 1563-1568.

Wang. G., Liu, T., Luo, Y., Zhao, Y., Ren, Z., Bai, J. and Wang, H. 2011b. Preparation of Fe_2O_3/graphene composite and its electrochemical performance as an anode material for lithium ion batteries. *Journal of Alloys and Compounds* 509: L216-L220.

Wang, G., Shen, X., Horvat, J., Wang, B., Liu, H., Wexler, D. and Yao, J. 2009a. Hydrothermal synthesis and optical, magnetic, and supercapacitance properties of nanoporous cobalt oxide nanorods. *Journal of Physical Chemistry C* 113: 4357-4361.

Wang, H.-W., Hu, Z.-A., Chang, Y.-Q., Chen, Y.-L., Lei, Z.-Q., Zhang, Z.-Y. and Yang, Y.-Y. 2010b. Facile solvothermal synthesis of a graphene nanosheet-bismuth oxide composite and its electrochemical characteristics. *Electrochimica Acta* 55: 8974-8980.

Wang, J. 2006. Analytical Electrochemistry, VCH Publishers, New York.

Wang, L., Zhao, Y., Lai, Q. and Hao, Y. 2010c. Preparation of 3D rose-like NiO complex structure and its electrochemical property. *Journal of Alloys and Compounds* 495: 82-87.

Wang, L., Liu, X., Wang, X., Yang, X. and Lu, L. 2010d. Preparation and electrochemical properties of mesoporous Co_3O_4 crater-like microspheres as supercapacitor electrode materials. *Current Applied Physics* 10: 1422-1426.

Wang, W., Liu, X., Gao, F. and Tian, C. 2007a. Synthesis of nanocrystalline $Ni_1Co_{0.2}Mn_{1.8}O_4$ powders for NTC thermistor by a gel auto-combustion process. *Ceramics International* 33: 459-462.

Wang, X., Chen, X., Gao, L., Zheng, H., Zhang, Z. and Qian, Y. 2004. One-dimensional arrays of Co_3O_4 nanoparticles: synthesis, characterization, and optical and electrochemical properties. *Journal of Physical Chemistry B* 108: 16401-16404.

Wang, X., Li, Y., Wang, G., Xiang, R., Jiang, D., Fu, S., Wu. K., Yang, X., DuanMu, Q., Tian, J. and Fu, L. 2009b. Characterization of NiO thin film grown by two-step processes. *Physica B-Condensed Matter* 404: 1058-1060.

Wang, X., Yuan, A. and Wang, Y. 2007b. Supercapacitive behaviors and their temperature dependence of sol-gel synthesized nanostructured manganese dioxide in lithium hydroxide electrolyte. *Journal of Power Sources* 172: 1007-1011.

Wang, Y., Shi, Z., Huang, Y., Ma, Y., Wang, C., Chen, M. and Chen, Y. 2009c. Supercapacitor devices based on graphene materials. *Journal of Physical Chemistry C* 113: 13103-13107.

Wang, X., Ruan, D., Wang, P. and Lu, Y. 2008b. Pseudo-capacitance of ruthenium oxide/carbon black composites for electrochemical capacitors. *Materials* 15: 816-821.

Wang, Y., Yang, W., Chen, C. and Evans, D.G. 2008c. Fabrication and electrochemical characterization of cobalt-based layered double hydroxide nanosheet thin-film electrodes. *Journal of Power Sources* 184: 682-690.

Wei, D. and Ng, T.W. 2009. Application of novel room temperature ionic liquids in flexible supercapacitors. *Electrochemistry Communications* 11: 1996-1999.

Wei, L. and Yushin, G. 2011a. Electrical double layer capacitors with sucrose derived carbon electrodes in ionic liquid electrolytes. *Journal of Power Sources* 196: 4072-4079.

Wei, T.-Y., Chen, C.H., Chien, H.-C., Lu, S.-Y. and Hu, C.C. 2010. A cost-effective supercapacitor material of ultrahigh specific capacitances: spinel nickel cobaltite aerogels from an epoxide-driven sol-gel process. *Advanced Materials* 22: 347-351.

Wei, W., Cui, X., Chen, W. and Ivey, D.G. 2009a. Electrochemical cyclability mechanism for MnO_2 electrodes utilized as electrochemical supercapacitors. *Journal of Power Sources* 186: 543-540.

Wei, W., Cui, X., Chen, W. and Ivey, D.G. 2009b. Improved electrochemical impedance response induced by morphological and structural evolution in nanocrystalline MnO_2 electrodes. *Electrochimica Acta* 54: 2271-2275.

Wei, W., Cui, X., Mao, X., Chen, W. and Ivey, D.G. 2011b. Morphology evolution in anodically electrodeposited manganese oxide nanostructures for electrochemical supercapacitor applications-effect of supersaturation ratio. *Electrochimica Acta* 56: 1619-1628.

Wei, X. and Chen, D. 2006. Synthesis and characterization of nanosized zinc aluminate spinel by sol-gel technique. *Materials Letters* 60: 823-827.

Wei, X., Chen, D. and Tang, W. 2007. Preparation and characterization of the spinel oxide $ZnCo_2O_4$ obtained by sol-gel method. *Materials Chemistry and Physics* 103: 54-58.

Wen, J., Ruan, X. and Zhou, Z. 2009. Preparation and electrochemical performance of novel ruthenium–manganese oxide electrode materials for electrochemical capacitors. *Journal of Physics and Chemistry of Solids* 70: 816-820.

Wen, S., Lee, J.-W., Yeo, I.-H., Park, J. and Mho, S.I. 2004. The role of cations of the electrolyte for the pseudocapacitive behavior of metal oxide electrodes, MnO_2 and RuO_2. *Electrochimica Acta* 50: 849-855.

Wen, T.-C. and Kang, H.-M. 1998. Co-Ni-Cu ternary spinel oxide-coated electrodes for oxygen evolution in alkaline solution. *Electrochimica Acta* 43: 1729-1745.

Winter, M. and Brodd, R.J. 2004. What are batteries, fuel cells, and supercapacitors?. *Chemical Reviews* 104: 4245-4269.

Wu, C.-M., Fan, C.-Y., Sun, I.-W., Tsai, W.-T. and Chang, J.-K. 2011a. Improved pseudocapacitive performance and cycle life of cobalt hydroxide on an electrochemically derived nano-porous Ni framework. *Journal of Power Sources* 196: 7828-7834.

Wu, F.-C., Tseng, R.-L., Hu, C.-C. and Wang, C.-C. 2005. Effects of pore structure and electrolyte on the capacitive characteristics of steam- and KOH-activated carbons for supercapacitors. *Journal of Power Sources* 144: 302-309.

Wu, F. C., Tseng, R. L., Hu, C. C. and Wang, C. C. 2004a. Physical and electrochemical characterization of activated carbons prepared from firwoods for supercapacitors. *Journal of Power Sources* 138: 351-359.

Wu, G., Li, N., Zhou, D.-R., Mitsuo, K. and Xu, B.-Q. 2004b. Anodically electrodeposited Co+Ni mixed oxide electrode: preparation and electrocatalytic activity for oxygen evolution in alkaline media. *Journal of Solid State Chemistry* 177: 3682-3692.

Wu, H.-M., Hsu, P.-F. and Hung, W.-T. 2009a. Investigation of redox reaction of Ru on carbon nanotubes by pulse potential electrochemical deposition. *Diamond and Related Materials* 18: 337-340.

Wu, J.B., Lin, Y., Xia, X.H., Xu, J.Y. and Shi, Q.Y. 2011b. Pseudocapacitive properties of electrodeposited porous nanowall Co_3O_4 film. *Electrochimica Acta* 56: 7163-7170.

Wu, M., Snook, G.A., Chen, G.Z. and Fray, D.J. 2004c. Redox deposition of manganese oxide on graphite for supercapacitors. *Electrochemistry Communications* 6: 499-504.

Wu, M.-S., Chan, D.-S., Lin, K.-H. and Jow, J.-J. 2011c. A simple route to electrophoretic deposition of transition metal-coated nickel oxide films for electrochemical capacitors. *Materials Chemistry and Physics* 130: 1239-1245.

Wu, M.-S., Huang, C.-Y. and Jow, J.-J. 2009b. Electrophoretic deposition of network-like carbon nanofiber as a conducting substrate for nanostructured nickel oxide electrode. *Electrochemistry Communications* 11: 779-782.

Wu, M.-S., Huang, Y.-A., Jow, J.-J., Yang, W.-D., Hsieh, C.-Y. and Tsai, H.-M. 2008. Anodically potentiostatic deposition of flaky nickel oxide nanostructures and their electrochemical performances. *International Journal of Hydrogen Energy* 33: 2921-2926.

Wu, M.-S., Lee, J.-T., Wang, Y.-Y. and Wan, C.-C. 2004d. Field emission from manganese oxide nanotubes synthesized by cyclic voltammetric electrodeposition. *Journal of Physical Chemistry B* 108: 16331-16333.

Wu, N.-L., Kuo, S.L. and Lee, M.-H. 2002. Preparation and optimization of RuO_2-impregnated SnO_2 xerogel supercapacitor. *Journal of Power Sources* 104: 62-65.

Wu, Y.Q., Chen, X.Y., Ji, P.T. and Zhou, Q.Q. 2011d. Sol-gel approach for controllable synthesis and electrochemical properties of $NiCo_2O_4$ crystals as electrode materials for application in supercapacitors. *Electrochimica Acta* 56: 7517-7522.

Xi, Y.Y., Li, D., Djurišić, A.B., Xie, M.H., Man, K.Y.K. and Chan, W.K. 2008. Hydrothermal synthesis vs electrodeposition for high specific capacitance nanostructured NiO films. *Electrochemical and Solid-State Letters* 11: D56-D59.

Xie, K., Li, J., Lai, Y., Lu, W., Zhang, Z., Liu, Y., Zhou, L. and Huang, H. 2011. Highly ordered iron oxide nanotube arrays as electrodes for electrochemical energy storage. *Electrochemistry Communications* 13: 657-660.

Xie, X. and Gao, L. 2007. Characterization of a manganese dioxide/carbon nanotube composite fabricated using an in situ coating method. *Carbon* 45: 2365-2373.

Xie, Y. and Fu, D. 2010. Supercapacitance of ruthenium oxide deposited on titania and titanium substrates. *Materials Chemistry and Physics* 122: 23-29.

Xie, Y., Huang, C., Zhou, L., Liu, Y. and Huang, H. 2009. Supercapacitor application of nickel oxide-titania nanocomposites. *Composites Science and Technology* 69: 2108-2114.

Xiong, S., Yuan, C., Zhang, X., Xi, B. and Qian, Y. 2009. Controllable synthesis of mesoporous Co_3O_4 nanostructures with tunable morphology for application in supercapacitors. *Chemistry-A European Journal* 15: 5320-5326.

Xing, W., Li, F., Yan, Z.-F. and Lu, G.Q. 2004. Synthesis and electrochemical properties of mesoporous nickel oxide. *Journal of Power Sources* 134: 324-330.

Xu, B., Chen, Y., Wei, G., Cao, G., Zhang, H. and Yang, Y. 2010a. Activated carbon with high capacitance prepared by NaOH activation for supercapacitors. *Materials Chemistry and Physics* 124: 504-509.

Xu, B., Wu, F., Chen, S., Zhang, C., Cao, G. and Yang, Y. 2007a. Activated carbon fiber cloths as electrodes for high performance electric double layer capacitors. *Electrochimica Acta* 52: 4595-4598.

Xu, C., Sun, J. and Gao, L. 2011. Large scale synthesis of nickel oxide/multiwalled carbon nanotube composites by direct thermal decomposition and their lithium storage properties. *Journal of Power Sources* 196: 5138-5142.

Xu, J., Gao, L., Cao, J., Wang, W. and Chen, Z. 2010b. Preparation and electrochemical capacitance of cobalt oxide (Co_3O_4) nanotubes as supercapacitor material. *Electrochimica Acta* 56: 732-736.

Xu, M., Kong, L., Zhou, W. and Li, H. 2007b. Hydrothermal synthesis and pseudocapacitance properties of α-MnO_2 hollow spheres and hollow urchins. *Journal of Physical Chemistry C* 111: 19141-19147.

Xu, M.-W., Bao, S.-J. and Li, H.-L. 2007c. Synthesis and characterization of mesoporous nickel oxide for electrochemical capacitor. *Journal of Solid State Electrochemistry* 11: 372-377.

Xu, Y., Wang, C., Sun, Y., Zhang, G. and Gao, D. 2010c. Fabrication and characterization of nearly monodisperse Co_3O_4 nanospheres. *Materials Letters* 64: 1275-1278.

Yan, J., Fan, Z., Wei, T., Cheng, J., Shao, B., Wang, K., Song, L. and Zhang, M. 2009a. Carbon nanotube/MnO_2 composites synthesized by microwave-assisted method for supercapacitors with high power and energy densities. *Journal of Power Sources* 194: 1202-1207.

Yan, J., Fan, Z., Wei, T., Qian, W., Zhang, M. and Wie, F. 2010a. Fast and reversible surface redox reaction of graphene-MnO_2 composites as supercapacitor electrodes. *Carbon* 48: 3825-3833.

Yan, J., Wei. T., Fan, Z., Qian, W., Zhang, M., Shen, X. and Wei, F. 2010b. Preparation of graphene nanosheet/carbon nanotube/polyaniline composite as electrode material for supercapacitors. *Journal of Power Sources* 195: 3041-3045.

Yan, J., Wei, T., Qiao, W., Shao, B., Zhao, Q., Zhang, L. and Fan, Z. 2010c. Rapid microwave-assisted synthesis of graphene nanosheet/Co_3O_4 composite for supercapacitors. *Electrochimica Acta* 55: 6973-6978.

Yan, J., Wei, T., Shao, B., Ma, F., Fan, Z., Zhang, M., Zheng, C., Shang, Y., Qian, W. and Wei, F. 2010d. Electrochemical properties of graphene nanosheet/carbon black composites as electrodes for supercapacitors. *Carbon* 48: 1731-1737.

Yan, S., Wang, H., Qu, P., Zhang, Y. and Xiao, Z. 2009b. RuO_2/carbon nanotubes composites synthesized by microwave-assisted method for electrochemical supercapacitor. *Synthetic Metals* 159: 158-161.

Yan, W., Ayvazian, T., Kim. J., Liu, Y., Donavan, K.C., Xing, W., Yang, Y., Hemminger, J.C. and Penner, R.M. 2011. Mesoporous manganese oxide nanowires for high-capacity, high-rate, hybrid electrical energy storage. *ACS Nano* 5: 8275-8287.

Yang, H. and Zhang, Y. 2011. Self-discharge analysis and characterization of supercapacitors for environmentally powered wireless sensor network applications. *Journal of Power Sources* 196: 8866-8873.

Yang, J., Liu, Y., Chen, X., Hu, Z. and Zhao, G. 2008. Carbon electrode material with high densities of energy and power. *Acta Physico-Chimica Sinica* 24: 13-19.

Yang, Y.-J., Liu, E.-H., Li, L.-M., Huang, Z.-Z., Shen, H.-J. and Xiang, X.-X. 2009. Nanostructured MnO_2/exfoliated graphite composite electrode as supercapacitors. *Journal of Alloys and Compounds* 487: 564-567.

Yokoshima, K., Shibutani, T., Hirota, M., Sugimoto, W., Murakami, Y. and Takasu, Y. 2006. Electrochemical supercapacitor behavior of nanoparticulate rutile-type $Ru_{1-x}V_xO_2$. *Journal of Power Sources* 160: 1480-1486.

Yokoyama, T., Kondou, K., Komeya, K., Meguro, T., Abe, Y. and Sasamoto, T. 1995. Preparation and electrical properties of monophase cubic spinel, $Mn_{1.5}Co_{0.95}Ni_{0.55}O_4$, derived from rock salt type oxide. *Journal of Materials Science* 30: 1845-1848.

Yoon, Y.I. and Ko, J.M. 2008. CoNi oxide/carbon-nanofiber composite electrodes for supercapacitors. *International Journal of Electrochemical Science* 3: 1340-1347.

Yu, P., Zhang, X., Chen, Y. and Ma, Y. 2010. Self-template route to MnO_2 hollow structures for supercapacitors. *Materials Letters* 64: 1480-1482.

Yuan, A., Wang, X., Wang, Y. and Hu, J. 2010. Comparison of nano-MnO_2 derived from different manganese sources and influence of active material weight ratio on performance of nano-MnO_2/activated carbon supercapacitor. *Energy Conversion and Management* 51: 2588-2594.

Yuan, D., Zeng, J., Kristian, N., Wang, Y. and Wang, X. 2009a. Bi_2O_3 deposited on highly ordered mesoporous carbon for supercapacitors. *Electrochemistry Communications* 11: 313-317.

Yuan, J., Liu, Z.-H., Qiao, S., Ma, X. and Xu, N. 2009b. Fabrication of MnO_2-pillared layered manganese oxide through an exfoliation/reassembling and oxidation process. *Journal of Power Sources* 189: 1278-1283.

Yuan, Y.F., Xia, X.H., Wu, J.B., Yang, J.L., Chen, Y.B. and Guo, S.Y. 2011. Nickel foam-supported porous $Ni(OH)_2$/NiOOH composite film as advanced pseudocapacitor material. *Electrochimica Acta* 56: 2627-2632.

Zeng, H.M., Zhao, Y., Hao, Y.J., Lai, Q.Y., Huang, J.H. and Ji, X.Y. 2009. Preparation and capacitive properties of sheet V_6O_{13} for electrochemical supercapacitor. *Journal of Alloys and Compounds* 477: 800-804.

Zhang, D., Zhang, X., Chen, Y., Yu, P., Wang, C. and Ma, Y. 2011a. Enhanced capacitance and rate capability of graphene/polypyrrole composite as electrode material for supercapacitors. *Journal of Power Sources* 196: 5990-5996.

Zhang, F.-B., Zhou, Y.-K. and Li, H.-L. 2004. Nanocrystalline NiO as an electrode material for electrochemical capacitor. *Materials Chemistry and Physics* 83: 260-264.

Zhang, J., Jiang, D., Chen, B., Zhu, J., Jiang, L. and Fang, H. 2001. Preparation and electrochemistry of hydrous ruthenium oxide/active carbon electrode materials for supercapacitor. *Journal of The Electrochemical Society* 148: A1362-A1367.

Zhang, J., Kong, L.-B., Wang, B., Luo, Y.-C. and Kang, L. 2009a. In-situ electrochemical polymerization of multi-walled carbon nanotube/polyaniline composite films for electrochemical supercapacitors. *Synthetic Metals* 159: 260-266.

Zhang, L.L., Wei, T., Wang, W. and Zhao, X.S. 2009b. Manganese oxide–carbon composite as supercapacitor electrode materials. *Microporous and Mesoporous Materials* 123: 260-267.

Zhang, L.L. and Zhao X.S. 2009. Carbon-based materials as supercapacitor electrodes. *Chemical Society Reviews* 38: 2520-2531.

Zhang, S., Peng, C., Ng, K.C. and Chen, G.Z. 2010a. Nanocomposites of manganese oxides and carbon nanotubes for aqueous supercapacitor stacks. *Electrochimica Acta* 55: 7447-7453.

Zhang, S.S., Xu, K. and Jow, T.R. 2004. Electrochemical impedance study on the low temperature of Li-ion batteries. *Electrochimica Acta* 49: 1057-1061.

Zhang, W.-D., Xu, B. and Jiang, L.-C. 2010b. Functional hybrid materials based on carbon nanotubes and metal oxides. *Journal of Materials Chemistry* 20: 6383-6391.

Zhang, Y., Feng, H., Wu, X., Wang, L., Zhang, A., Xia, T., Dong, H., Li, X. and Zhang, L. 2009c. Progress of electrochemical capacitor electrode materials: a review. *International Journal of Hydrogen Energy* 34: 4889-4899.

Zhang, Y., Sun, X., Pan, L., Li, H., Sun, Z., Sun, C. and Tay, B.K. 2009d. Carbon nanotube-zinc oxide electrode and gel polymer electrolyte for electrochemical supercapacitors. *Journal of Alloys and Compounds* 480: L17-L19.

Zhang, Y., Sun, X., Pan, L., Li, H., Sun, Z., Sun, C. and Tay, B.K. 2009e. Carbon nanotube-ZnO nanocomposite electrodes for supercapacitors. *Solid State Ionics* 180: 1525-1528.

Zhang, Y. and Yang, H. 2011b. Modeling and characterization of supercapacitors for wireless sensor network applications. *Journal of Power Sources* 196: 4128-4135.

Zhao, T., Jiang, H. and Ma, J. 2011. Surfactant-assisted electrochemical deposition of α-cobalt hydroxide for supercapacitors. *Journal of Power Sources* 196: 860-864.

Zheng, H., Tang, F., Jia, Y., Wang, L., Chen, Y., Lim, M., Zhang, L., Max Lu, G. 2009a. Layer-by-layer assembly and electrochemical properties of sandwiched film of manganese oxide nanosheet and carbon nanotube. *Carbon* 47: 1534-1542.

Zheng, H., Tang, F., Lim, M., Mukerji, A., Yan, X., Wang, L. and Max Lu, G.Q. 2010. Multilayered films of cobalt oxyhydroxide nanowires/manganese oxide nanosheets for electrochemical capacitor. *Journal of Power Sources* 195: 680-683.

Zheng, M.-B., Cao, J., Liao, S.-T., Liu, J.-S., Chen, H.-Q., Zhao, Y., Dai, W.-J., Ji, G.-B., Cao, J.-M. and Tao, J. 2009b. Preparation of mesoporous Co_3O_4 nanoparticles via solid-liquid route and effects of calcination temperature and textural parameters on their electrochemical capacitive behaviors. *Journal of Physical Chemistry C* 113: 3887-3894.

Zheng, Y., Zhang, M. and Gao, P. 2007. Preparation and electrochemical properties of multiwalled carbon nanotubes-nickel oxide porous composite for supercapacitors. *Materials Research Bulletin* 42: 1740-1747.

Zheng, Y.-Z., Ding, H.-Y. and Zhang, M.-L. 2008. Hydrous-ruthenium-oxide thin film electrodes prepared by cathodic electrodeposition for supercapacitors. *Thin Solid Films* 516: 7381-7385.

Zheng, Z. and Gao, Q. 2011. Hierarchical porous carbons prepared by an easy one-step carbonization and activation of phenol-formaldehyde resins with high performance for supercapacitors. *Journal of Power Sources* 196: 1615-1619.

Zheng, Z., Huang, L., Zhou, Y., Hu, X. and Ni, X. 2009c. Large scale synthesis of mesoporous CoO-doped NiO hexagonal nanoplatelets with improved electrochemical performance. *Solid State Sciences* 11: 1439-1443.

Zhou, S.-Y., Li, X.-H., Wang, Z.-X., Guo, H.-J. and Peng, W.-J. 2007. Effect of activated carbon and electrolyte on properties of supercapacitor. *Transactions of Nonferrous Metals Society of China* 17: 1328-1333.

Zhou, W.-J., Xu, M.-W., Zhao, D.-D., Xu, C.-L. and Li, H.-L. 2009. Electrodeposition and characterization of ordered mesoporous cobalt hydroxide films on different substrates for supercapacitors. *Microporous and Mesoporous Materials* 117: 55-60.

Zhou, W.-J., Zhao, D.-D., Xu, M.-W., Xu, C.-L. and Li, H.-L. 2008a. Effects of the electrodeposition potential and temperature on the electrochemical capacitance behavior of ordered mesoporous cobalt hydroxide films. *Electrochimica Acta* 53: 7210-7219.

Zhou, X., Ge, S., Yao, D., Zuo, Y. and Xiao, Y. 2008b. Effect of thermal treatment on room-temperature ferromagnetism in Co-doped ZnO powders. *Physica B-Condensed Matter* 403: 3336-3339.

Ziemniak, S.E., Anovitza, L.M., Castelli, R.A. and Porter, W.D. 2007a. Magnetic contribution to heat capacity and entropy of nickel ferrite ($NiFe_2O_4$). *Journal of Physics and Chemistry of Solids* 68: 10-21.

Ziemniak, S.E., Anovitza, L.M., Castelli, R.A. and Porter, W.D. 2007b. Thermodynamics of Cr_2O_3, $FeCr_2O_4$, $ZnCr_2O_4$, and $CoCr_2O_4$. *The Journal of Chemical Thermodynamics* 39: 1474-1492.

Zolfaghari, A., Ataherian, F., Ghaemi, M. and Gholami, A. 2007. Capacitive behavior of nanostructured MnO_2 prepared by sonochemistry method. *Electrochimica Acta* 52: 2806-2814.

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Chang Sook Keng was born and bred in Ipoh, Perak. She is the eldest in the family and has two younger brothers. After receiving her primary and secondary education from Main Convent Ipoh, she left her alma mater for Saint Michael's Institution Ipoh to attend her Form Six education. Then, she was enrolled in Universiti Putra Malaysia and obtained her first degree in Bachelor of Science (Hons) Industrial Chemistry in 2004. Upon her graduation, she pursued her Master of Science in Materials Chemistry in the same university and graduated in 2007. Later on, she furthered to her third degree in the same field in Universiti Putra Malaysia as well.

LIST OF PUBLICATIONS

Chang, S.-K., Zainal, Z., Tan, K.B., Yusof, N.A., Yusoff, W.M.D.W. and Prabaharan, S.R.S. 2009. Phase formation and surface morphology of nanosized nickel-cobalt oxide prepared by co-precipitation method. *Solid State Science and Technology Letters* 16: 187-194.

Chang, S.-K., Zainal, Z., Tan, K.B., Yusof, N.A., Yusoff, W.M.D.W. and Prabaharan, S.R.S. 2012. Surface morphology and crystallinity of metal oxides in nickel-cobalt binary system. *Sains Malaysiana* 41: 465-470.

Chang, S.-K., Lee, K.-T., Zainal, Z., Tan, K.-B., Yusof, N.A., Yusoff, W.M.D.W., Lee, J.-F. and Wu, N.-L. 2012. Structural and electrochemical properties of manganese substituted nickel cobaltite for supercapacitor application. *Electrochimica Acta* 67: 67-72.

Chang, S.-K., Zainal, Z., Tan, K.B., Yusof, N.A., Yusoff, W.M.D.W. and Prabaharan, S.R.S. 2012. Nickel-cobalt oxide/activated carbon composite electrodes for electrochemical capacitors. *Current Applied Physics* 12: 1421-1428.