

## Future scope and directions of nanotechnology in creating next-generation supercapacitors

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# **1 Future scope and directions of nanotechnology in creating next generation supercapacitors**

## **1.1 Nanotechnology and applications**

The first major worldwide research initiative of the 21<sup>st</sup> century is Nanotechnology. Anticipating the future, it seems that nanotechnologies' generalized diffusion will turn them into commodities, creating more space for dedicated, higher added value applications such as information technology, nanobiotechnologies, nanoenergy or nanomaterials (Mangematin and Walsh, 2012). Nanotechnology is the understanding and control of matter at dimensions of roughly 1-100 nm, where unique phenomena enable novel applications (Schilthuizen and Simonis, 2006). Nanotechnology covers the following application domains:

### **1.1.1 Information and communication technology (ICT)**

The ICT sector has undergone rapid expansion as work and social activities are transformed by new and varied technologies. This has required computers to become faster, enabled by the production of smaller transistors through advanced fabrication processes (Filipponi and Sutherland, 2010). There are now challenges to continue this miniaturisation path because as the materials are reduced to nano-size their properties begin to be determined and dominated by quantum effects. Miniaturised hardware (sensors, readers, displays and radio transmitters), personal secured access to equipment (biometric id) and information (digital id) are some application of Nanotechnology in ICT (Simonis and Schilthuizen, 2006).

### **1.1.2 Bio and life science**

The nano-bio fusion is a booming area with high expectations that major steps in health treatment, body repair and body improvement can be made. Nanomedicine, targeted drug delivery, programmed tissue engineering are some developments in this field (Simonis and Schilthuizen, 2006).

### **1.1.3 Materials and manufacturing**

Nanotechnology enables high strength, durable, sensoric and active materials. lightweight protective clothes, flexible antiballistic textiles, microsensors for body & brain sensing, wearable and/or flexible displays for visual feedback, exoskeletons and robotics are some developments of nanostructures and nanocomposites (Thilagavathi et al., 2008).

### **1.1.4 Energy or power**

With the increase in wearable functionalities and electronics, the need for lightweight wearable electric power is very critical. Developments in this field include flexible solar cells to recharge batteries, fuel cells and supercapacitor. One of the great challenges in the twenty first century is unquestionably energy storage. In response to the needs of modern society and emerging ecological concerns, it is now essential that new, low-cost and environmentally friendly energy conversion and storage systems are found; hence the rapid development of research in this field (Aricò et al., 2005). It is noteworthy to determine the opportunities and barriers relating to developing next-generation electrical energy storage products such as batteries and supercapacitors based on nanotechnology.

## **1.2 Historical background of supercapacitor**

Supercapacitor, also known as electrical double layer capacitor (EDLC), ultracapacitor, or electrochemical capacitor, is known as an electrical energy storage device. Consisting of two electrodes separating by membrane, electrochemical supercapacitors store electrical energy using reversible adsorption of ions from an electrolyte onto two porous electrodes to form an electric double layer at an electrode/electrolyte interface (**Error! Reference source not found.**). They are potentially important component of a future energy storage system. By combining the high energy/lower power characteristics of a battery with the high power/low energy characteristics of a supercapacitor (Lu et al., 2007), an energy storage device can be developed which allows rapid charge storage with endurance.

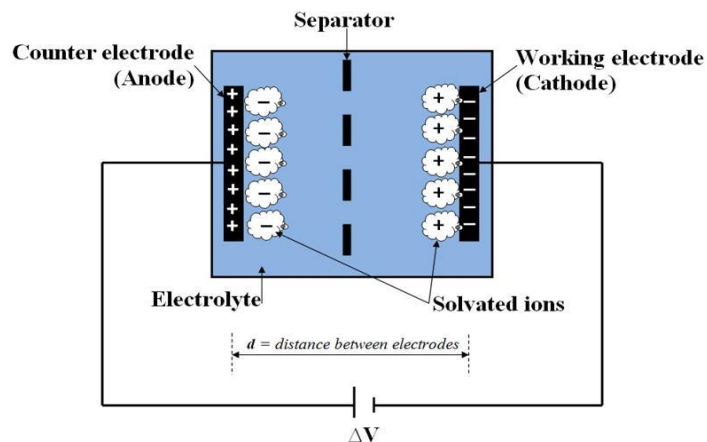


Figure 1. Schematic of an electrochemical double-layer capacitor (Kiamahalleh et al., 2012).

The other type of electrochemical supercapacitor is termed a pseudo-capacitor and is basically different from an EDLC in charge storing mechanism. Early researches concentrated on EDLCs which store charge electrostatically, or non-Faradaically, and there is no transfer of charge between electrode and electrolyte. EDLCs utilise an electrochemical double-layer of charge to store energy. As voltage is applied, charge

accumulates on the electrode surfaces (Halper and Ellenbogen, 2006). More recently, research activity has grown substantially; in particular with regard to the pseudo-capacitance behavior of transition metal oxides that store charge using redox-based Faradaic reactions. This pseudo-capacitance can be superimposed on any electric double-layer capacitance (Zhao et al., 2011).

The initial patent on supercapacitor was granted to Becker at General Electric Corp. in 1957 (Becker, 1957), in which he proposed a porous carbon based capacitor with high surface area. In 1969, SOHIO company for the first time attempted to market such energy storage device using high surface area carbon materials with tetra-alkylammonium salt electrolyte (Boos, 1970). In late 70's and 80's, Conway made a great research work contribution to the field of capacitor based on  $\text{RuO}_2$ , which showed high specific capacitance and low internal resistance (Conway, 1991b). In the 90's, supercapacitors received much attention in the term of hybrid electric vehicles. There are a large number of developers of capacitors/supercapacitors. The leading companies include SAFT (France), NESS (Korea), ESMA (Russia), PowerCache (Maxwell, USA), ELIT (Russia), PowerSystem Co. (Japan) and Chubu Electric Power (Japan), etc (Du et al., 2009). Large numbers of supercapacitor patents have been granted as cited by Sarangapani in 1996 (Sarangapani et al., 1996). All these researches triggered U.S. Department of Energy to initiate a 1998-2003 short-term and an after-2003 long-term supercapacitor development programs (Murphy et al., 1997).

### **1.3 Current commercial activity**

A number of companies around the world currently manufacture EDLCs in a commercial capacity. NEC and Panasonic in Japan have been producing EDLC

components since the 1980's. In the U.S.A Epcos, ELNA, AVX, and Cooper produce components, while Evans and Maxwell produce integrated modules that include voltage balancing circuitry. Kold Ban International markets a supercapacitor module designed specifically for starting internal combustion engines in cold weather. Cap-XX in Australia offers a range of components, as does Ness Capacitor Co. in Korea. In Canada, Tavrira manufactures a range of modules. ESMA in Russia sells a wide variety of EDLC modules for applications in power quality, electric vehicles, and for starting internal combustion engines (Rosa Palacin et al., 2011). Nowadays, commercial productions of electrochemical supercapacitors in markets are based on the high surface area porous carbon materials as well as based on noble metal oxides systems (Conway, 1999). For instance, Matsushita Electric Industrial (Panasonic, Japan) developed Gold capacitors (Conway, 1999), and Pinnacle Research (USA) (Conway, 1999) especially made high performance supercapacitors for military applications. The products currently available from present manufacturers as detailed on their websites are summarized in Table 1.



Table 1. Summary of current EDLC's commercially available (Namisnyk, 2003).

<b>Company name</b>	<b>Country</b>	<b>Device name</b>	<b>Capacitance range</b>	<b>Voltage range</b>	<b>Website</b>
AVX	USA	Bestcap	0.022-0.56	3.5-12	<a href="http://www.avxcorp.com">www.avxcorp.com</a>
Cap-XX	Australia	Supercapacitor	0.09-2.8	2.25-4.5	<a href="http://www.cap-xx.com">www.cap-xx.com</a>
Cooper	USA	Powerstore	0.47-50	2.3-5	<a href="http://www.powerstore.com">www.powerstore.com</a>
ELNA	USA	Dynacap	0.033-100	2.5-6.3	<a href="http://www.wlna-america.com">www.wlna-america.com</a>
ESMA	Russia	Capacitor modules	100-5000	12-52	<a href="http://www.esma-cap.com">www.esma-cap.com</a>
Epcos	USA	Ultracapacitor	5-5000	2.3-2.5	<a href="http://www.epcose.com">www.epcose.com</a>
Evans	USA	Capattery	0.01-1.5	5.5-11	<a href="http://www.evanscap.com">www.evanscap.com</a>
Kold Ban	USA	KAPower	1000	12	<a href="http://www.koldban.com">www.koldban.com</a>
Maxwell	USA	Bootcap	1.8-2000	2.5	<a href="http://www.maxwell.com">www.maxwell.com</a>
NEC	Japan	Supercapacitor	0.01-6.5	3.5-12	<a href="http://www.nec-tokin.net">www.nec-tokin.net</a>
Ness	Korea	EDLC	10-3500	3	<a href="http://www.nescap.com">www.nescap.com</a>
Panasonic	Japan	Gold capacitor	0.1-2000	2.3-5.6	<a href="http://www.macopanasonic.co.jp">www.macopanasonic.co.jp</a>
Tavrima	Canada	Supercapacitor	0.13-160	14-300	<a href="http://www.tavrima.com">www.tavrima.com</a>

The overall market size of all three areas of batteries, fuel cells and supercapacitors was estimated to have been 350 million USD and is forecast to reach 7,700 million USD (in 2012) (LUX Research 2008). Markets for nanotechnology-enabled energy-production and-storage are expected to take off from 2012. Robust sales growth opportunities are expected—in particular—for rechargeable batteries (Lithium-ion) and supercapacitors based on an early adoption of nanomaterials in the processes and productions of these devices. Supercapacitor technology is aimed at a market estimated to grow to over \$600 million by the year 2012, while the total market for battery and supercapacitor storage systems is forecast to grow from 1.5 billion USD in 2012 to 8.3 billion USD in 2016 (AZoNano 2009).

The commercial supercapacitors are widely used as power sources for activators (Conway, 1999), or as elements for long time constant circuits (Conway, 1999), or standby power for random access memory devices, and telephone equipments, etc (Liu et al., 2011). A comparison of the properties and performance between battery, capacitor, and supercapacitor is given in Table 2.

Table 2. Comparison of Capacitor, Supercapacitor and Battery (Nuintek, 2006)

<b>Parameters</b>	<b>Capacitor</b>	<b>Supercapacitor</b>	<b>Battery</b>
Charge Time	$10^{-6} \sim 10^{-3}$ sec	1 ~ 30 sec	0.3 ~ 3 hrs
Discharge Time	$10^{-6} \sim 10^{-3}$ sec	1 ~ 30 sec	1 ~ 5 hrs
Energy Density (Wh/kg)	< 0.1	1 ~ 10	20 ~ 100
Power Density (W/kg)	> 10,000	1,000 ~ 2,000	50 ~ 200
Cycle Life	> 500,000	> 100,000	500 ~ 2,000
Charge/Discharge Efficiency	~ 1.0	0.90 ~ 0.95	0.7 ~ 0.85

Batteries are typically low power devices compared to capacitors, which have power densities as high as  $10^4$  to  $10^6$  W/kg, but low energy densities. From this point of view, supercapacitors combine the properties of high power density and higher energy density, and also have long life cycles due to the absence of chemical reactions. A comprehensive review of the historical background, properties, and principles of capacitors has been given by Conway (Conway, 1999). The improvement in the performance of supercapacitor is shown in **Figure 2**; a graph termed a “Ragone plot.” This type of graph presents the power densities of various energy storage devices, measured along the vertical axis, versus their energy densities, measured along the horizontal axis. As shown in **Figure 2**, it is seen that supercapacitors occupy an area between conventional capacitors and batteries (Kötz and Carlen, 2000). Despite greater capacitances than conventional

capacitors, supercapacitors have yet to match the energy densities of mid to high-end batteries and fuel cells. Thus, much of the previous literature surveyed for this overview focuses on developing improved types or classes of supercapacitors to make their energy densities more comparable to those of batteries.

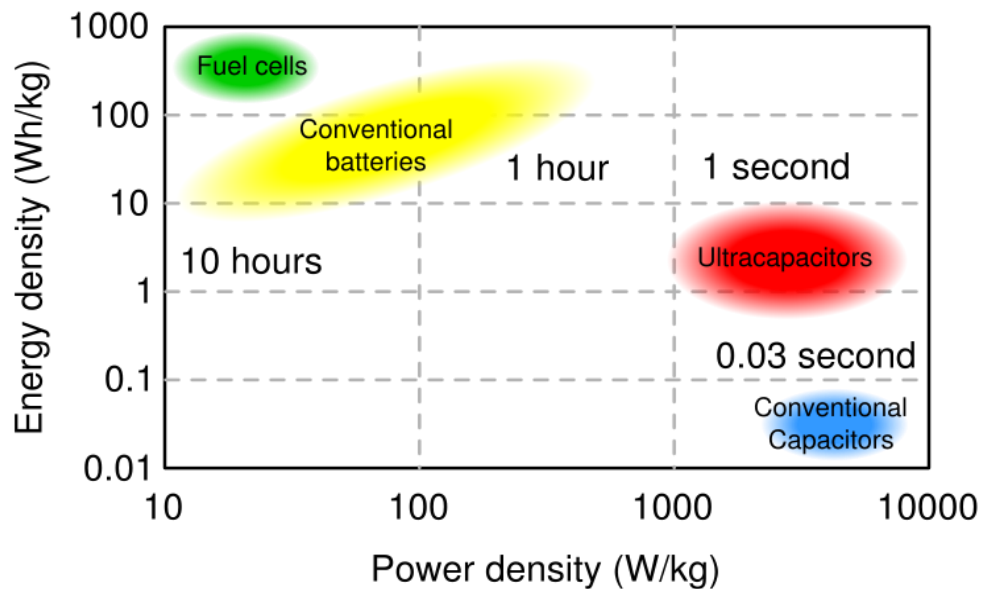


Figure 2. Ragone plot of energy storage devices (Schneuwly, 2006 ).

#### 1.4 Current research efforts

A critical aspect in nanotechnology for supercapacitors is to reach a compromise between specific surface area (to ensure high capacitance) and pore-size distribution (to permit easy access for the electrolyte) (Aricò et al., 2005). A number of institutions are currently conducting researches in the interests of improving both the energy and power densities of EDLC technology. Activated carbons are the most commonly used electrode material in commercial supercapacitors at present, and a good deal of research is interested in determining the factors that contribute to the specific capacitance and series resistance in such materials. Celzard and coworkers from the Université Henri Poincaré-Nancy in

France performed research on the correlation between the porous electrode structure and series resistance (Celzard et al., 2002). Other work done by another French laboratory at the Conservatoire National de Arts et Métiers confirms the impact of pore size distribution on specific capacitance (Gamby et al., 2001). Xie et al. (2012) and Feng and Cummings (2011) believed that electrode material is the key to supercapacitors and an ideal one should possess a large surface area and suitable pore size distribution. Xie et al. (2012) also stated that as a type of good candidate, carbonaceous materials have always attracted intense interest during the exploration of supercapacitor electrode materials, from the early activated carbon (AC) to mesoporous carbon (MC), carbon nanotubes (CNTs), and very recently to graphene. Yamada et al. (Yamada et al., 2002) compared different carbons of comparable specific surface areas and with different pore size distributions and showed that those that were mostly mesoporous had a higher capacitance.

Recently, CNTs have been explored and a number of academic institutions such as the Poznan University of Technology, Poland, and Sungkyunkwan University in Korea have constructed electrodes that demonstrate a higher specific capacitance than that achievable by activated carbons (An et al., 2001, Frackowiak et al., 2000). Researchers at Department of Applied Chemistry and Biotechnology, Hanbat National University in South Korea have taken this a step further and shown that activated CNTs have an even higher specific capacitance than normal CNTs (Ko and Kim, 2009). Because of their unique architecture, CNTs are now intensively studied as new electrode materials for supercapacitor structures although, as for batteries, cost may be an issue (Aricò et al., 2005). Considerable interest has also been shown in conducting polymer materials, and

research suggests that high specific capacitances should be attainable (Estaline Amitha et al., 2009).

Transition metal oxides have always been an attractive electrode material due to their low resistance and high specific capacitance, but their excessive cost has generally ruled them out as a commercially viable option (Namisnyk, 2003). Traditionally a strong sulfuric acid has been used as an electrolyte with metal oxide electrodes in order to increase the ion mobility, and hence the rate of charge and discharge. However, this limits the choice of electrode materials because most become unstable and corrode in a strongly acidic electrolyte. Research at the University of Texas, USA, has therefore focused on the possibility of using a milder, potassium chloride (KCl) aqueous electrolyte for use with metal-oxides. Their work suggested that the replacement is indeed possible, and should widen the availability of possible electrode materials (Lee and Goodenough, 1999). Manganese oxide ( $\text{MnO}_2$ ), a cheaper alternative to ruthenium-oxide, has been confirmed as a good electrode candidate by the researchers at Imperial College, London, (Jiang and Kucernak, 2002).

While ceramics with high oxide ion conductivity (especially zirconia ( $\text{ZrO}_2$ ) based) have been known for almost a century, protons were first shown to exist as minority charge carriers in oxides by Stotz and Wagner (Stotz and Wagner, 1966) in the 1960s. The systematic search of Takahashi and Iwahara (Takahashi and Iwahara, 1980) later demonstrated that acceptor doped perovskite-type oxides (e.g. doped Lanthanum Aluminate ( $\text{LaAlO}_3$ ), Lanthanum yttrium oxide ( $\text{LaYO}_3$ ), Strontium zirconate ( $\text{SrZrO}_3$ )), which were already known for their moderate oxide ion conductivity (Browall et al., 1976) could become proton conductors in water containing atmospheres. Unfortunately, the

observed conductivities were still far too low to compete with the high oxide ion conductivity of yttria ( $\text{Y}_2\text{O}_3$ ) stabilized  $\text{ZrO}_2$ , the standard electrolyte material at that time. But soon, related compounds (in particular acceptor doped strontium cerate ( $\text{SrCeO}_3$ ) (Iwahara et al., 1983) and barium cerate ( $\text{BaCeO}_3$ ) (Iwahara et al., 1988)) with higher proton conductivities were discovered and tested in different kind of electrochemical cells.

The most promising results seem to lie in the use of hybrid configurations, which consist of carbon materials and conducting polymers or transition metal-oxides. Research at the University of Bologna in Italy have resulted in a supercapacitor that has a positive activated carbon electrode and a negative polymer electrode which outperforms configurations of solely using activated carbon (Arbizzani et al., 2001). Studies at the National Cheng Kung University in Taiwan indicated that high specific capacitance can be achieved by the deposition of conducting polymers onto activated carbon (Chen et al., 2003). Frackowiak and coworkers at the Poznan University of Technology demonstrated an increase in specific capacitance of CNTs coated with a polymer (Frackowiak et al., 2002). The study of solid-state supercapacitors is also of interest being conducted at the University of Twente in The Netherlands, in which  $\text{Y}_2\text{O}_3$  stabilized  $\text{ZrO}_2$  is used instead of a liquid electrolyte (Hendriks et al., 2001).

Lee's group successfully fabricated a polypyrrole (PPy)–CNT composite electrode on a ceramic fabric by chemical vapor deposition (CVD) and chemical polymerization and it showed promising capacitive behavior and high stability (Lee et al., 2011). Moreover, the CNTs and porous structure of the ceramic fabrics provide the electrode with a high surface area.

## 1.5 Future research

Today electrochemical supercapacitors are the power sources of choice for hybrid vehicles and popular portable electronics, such as cellular phones and notebooks. However, despite their outstanding commercial success, these supercapacitors are still open to improvements. Active research is continuing on all aspects of supercapacitors, i.e., electrode materials (anodes, cathodes), electrolytes, and cell construction. The major factors limiting their wider application remain cost and safety. New developments are taking place to address these limitations.

High surface area activated carbon is the most common anode material used extensively in commercially established supercapacitors. Theoretically, the higher the surface area of the activated carbon, the higher the specific capacitance, but it has low electrochemical cycling stability. To overcome this problem, multiwalled CNTs (MWCNTs) have a good cycling stability, but a relatively low specific capacity, not exceeding than 80 F/g. Thus, if an improvement in energy content is desired, new, high-capacity alternative electrode materials have to be developed.

However, activated carbons currently dominate the market as an electrode material, but progress in the development of MWCNTs, conducting polymers and metal oxides is continuing at a steady rate. The exploitation of pseudocapacitive (conducting polymers and transition metal oxides) effects to enhance double-layer capacitance (carbon materials) seems to be a prevalent goal amongst current researchers, and offers a good chance of developing the next generation of high power, high-energy supercapacitors.

Several research teams have focused on the development of an alternative electrode material for electrochemical supercapacitors. Variety of transition metal oxides have been



introduced to MWCNTs and shown to be suitable as electrode materials for electrochemical capacitors. Among the oxide materials for application in electrochemical supercapacitors, ruthenium oxide ( $\text{RuO}_2$ ) and iridium oxide ( $\text{IrO}_2$ ) have achieved much attention.

$\text{RuO}_2$  has a high double-layer and pseudo-capacitance (up to  $\sim 493.9$  F/g (Yan et al., 2009)) and are stable in aqueous acid and alkaline electrolytes. The capacitance sensitively depends on the method of preparation. Unfortunately, disadvantage of  $\text{RuO}_2$  such as the high cost of the raw material and toxicity retarded its great application in supercapacitors electrode materials. Therefore, in recent years great efforts have been undertaken to find new and cheaper materials. Several metal oxides and hydroxides, for example, those of nickel (Ni), cobalt (Co), vanadium (V), and manganese (Mrunal et al.), are being studied extensively. Among all these cheap metal oxides,  $\text{MnO}_2$  (Ko and Kim, 2009) and nickel oxide (NiO) (Xie et al., 2008) are proposed as the most promising pseudocapacitors electrode materials with respect to both specific capacitance and cost effectiveness.

Recently, ceramic oxides (mixed metal oxides) have also attracted pretty much attention as electrode materials in supercapacitors. Gibson and Karthikeyan stated in their patents that improved supercapacitors can be developed to store charge by a combination of Faradaic and non-Faradaic mechanisms. They have invented ceramic materials having nominal (idealized) compositions that correspond to one of the following groups:  $\text{ABO}_3$  (e.g., perovskites),  $\text{A}_2\text{BO}_4$  (including the alternative form  $\text{AB}_2\text{O}_4$ ), and fluorites  $\text{AO}_2$ , where A and B are metals (A and B = La, Sr, Ca, Mn, Fe, Ni, Co, Ga, Ce, Gd, or other metal).

Generally, a supercapacitor uses a transition metal oxide and a conductive polymer as electroactive materials is termed as pseudocapacitor. Combination of pseudocapacitor with carbon based electrode material is termed as hybrid supercapacitor. In the next section application of supercapacitor electrode materials, their advantages and disadvantages against the hybrid supercapacitor will be investigated in details.

## **2 Scope of using transition metal oxides against carbon and polymer based supercapacitor materials: advantages and challenges**

The elementary structure of a supercapacitor consists on current collectors and electrodes impregnated in an organic or aqueous electrolyte. A separator is inserted between the two electrodes to insulate them. The assembly of the unit is carried out as for the traditional capacitors (Burke, 2000, Kötzt and Carlen, 2000, Rafik et al., 2007). The principle of operation of supercapacitor is based on their energy storage by distribution of the ions coming from the electrolyte in the vicinity of the surface of the two electrodes. Indeed, the electrode materials and their properties such as high surface area and electrical conductivity are the key components to improve the efficiency of the supercapacitors (Wu et al., 2010, Stoller and Ruoff, 2010). This section generally covers some reviews about the classification, charge storing mechanism as well as comparative studies to evaluate the performance of different electrode materials in supercapacitors. At the end, the advantages and the current challenges on how to enhance the performance of the supercapacitors is discussed in details.

### **2.1 Taxonomy of Supercapacitor**

Based upon current R&D trends, taxonomy of supercapacitors can be divided into three general classes: electrochemical double layer capacitors (EDLCs),

pseudocapacitors, and hybrid capacitors. Their charge storing mechanisms are non-Faradaic, Faradaic, and a combination of these two process, respectively. Faradaic processes, such as oxidation-reduction reactions, involve the transfer of charge between electrode and electrolyte. A non-Faradaic mechanism, by contrast, does not use a chemical mechanism. Rather, charges are distributed on surfaces by physical processes that do not involve either making or breaking of chemical bonds. This section will present an overview of each one of these three classes of supercapacitors and their subclasses, distinguished by electrode material. A graphical taxonomy of the different classes and subclasses of supercapacitors is presented in Figure 3.

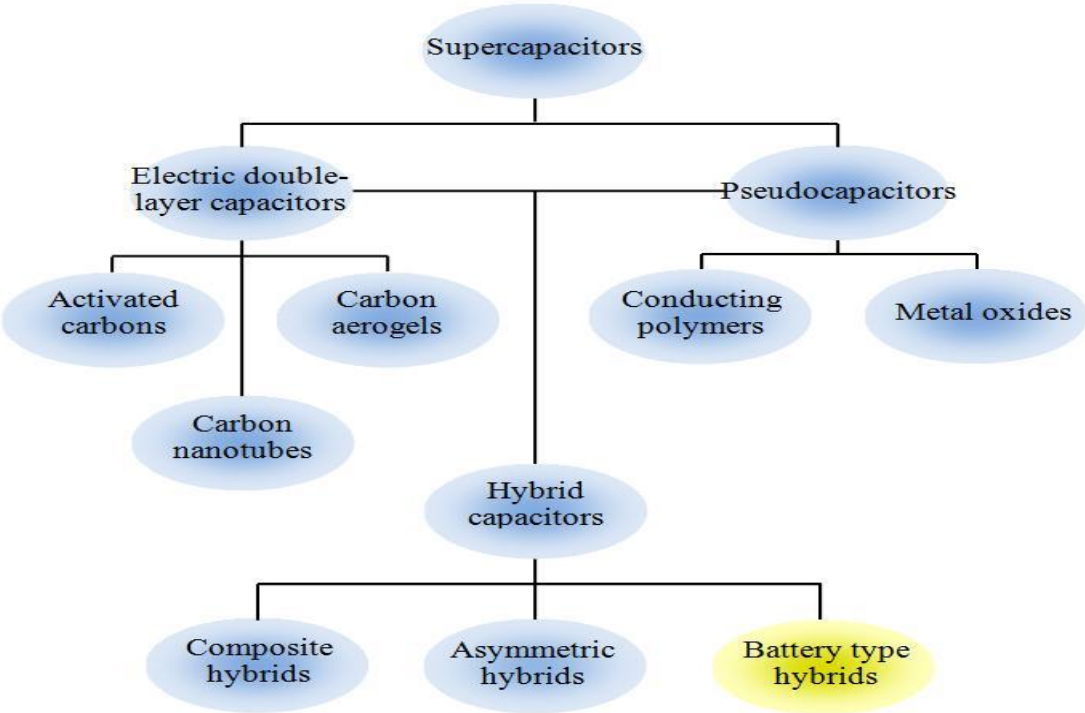


Figure 3. Taxonomy of supercapacitors (Kiamahalleh et al., 2012)

Recently several works yielded major developments and progresses in supercapacitors researches either in theoretical or practical (Zhang and Zhao, 2009, Pan et al., 2010, Inagaki et al., 2010, Burke, 2000, Wang et al., 2012). At the same time, the

disadvantages of super capacitors including low energy density and high production cost have been identified as major challenges for the furtherance of supercapacitor technologies. To overcome the drawbacks of low energy density, one of the most intensive approaches is the development of new materials for supercapacitor electrodes . To date carbon materials are the most popular, which have high surface areas for charge storage . In spite of having large specific surface areas, the carbon materials are unfortunately limited in charge storage. Supercapacitors based on carbon materials are called electrostatic or EDLS have a limited specific capacitance and a low energy density (Wang et al., 2012). Hybridizing the electrode materials by adding electrochemically active (pseudocapacitive) materials or to completely replace the carbon materials with electrochemically active materials are the new approaches to enhance the supercapacitors energy density. Supercapacitors with pseudocapacitive materials as electrodes are called Faradaic supercapacitors. Hence, it has been demonstrated that hybrid double-layer supercapacitors can yield much higher specific capacitance and energy density than EDLS (Wang et al., 2012).

It is also noteworthy to mention that hybrid supercapacitor with an asymmetrical electrode configuration (e.g. one electrode consists of carbon material while the other consists of pseudocapacitive material) have been extensively investigated recently to enhance overall cell voltage, energy, and power densities (Burke, 2000, Kisacikoglu et al., 2009, Ma et al., 2007). In this type of hybrid supercapacitor, both EDLC and Faradaic capacitance mechanisms take place simultaneously, but one of them plays a greater role (Wang et al., 2012).

## **2.2 Electrode Materials**

In general, the electrode materials of supercapacitor can be categorized into three types: (1) carbon materials with high specific surface area (Edwards) conducting polymers, and (3) metal oxides. In terms of the chemical composition, several types of supercapacitor electrode materials have been investigated intensively, which include electrically conducting metal oxides, e.g., RuO<sub>2</sub> (Gujar et al., 2007, Liu et al., 1997), IrO<sub>2</sub> (Michell et al., 1977, Liu et al., 2008), Fe<sub>2</sub>O<sub>3</sub> (Micka and Záborský, 1987, Nagarajan et al., 2006); Fe<sub>3</sub>O<sub>4</sub> (Du et al., 2009) MnO<sub>2</sub> (Goodenough, 1984, Deng, 2008, Ragupathy et al., 2009) NiO (Wang and Xia, 2006, Zheng et al., 2009, Patil et al., 2008b), conducting polymers, e.g., polythiophene (Laforgue et al., 1999, Mastragostino et al., 2001), PPy (Lee et al., 2011, Sharma et al., 2008), PANi (Bélanger et al., 2000, Yahya et al., 2009) and their derivatives, and different type of carbon materials, e.g., carbon aerogel (Frackowiak and Béguin, 2001, Fang and Binder, 2007), activated carbon (Shi, 1996, Obreja, 2008), and CNTs (Niu et al., 1997, Diederich et al., 1999, Maletin et al., 2006, He et al., 2008).

### **2.2.1 Carbon-Based Supercapacitor**

Carbon is the most common and economical material for supercapacitor electrodes. Different carbon material electrodes have been intensively studied (Shi, 1996, Qu and Shi, 1998). Carbon materials generally have high surface area, in the range of 1000 to 2000 m<sup>2</sup>/g, e.g., activated carbon, carbon cloth, carbon aerogels, carbon nanofibres, graphite, nanaocarbons and CNTs. The capacity of the basal plane and edge plane of graphite carbon are about 10-40 μF/cm<sup>2</sup> and 50-70 μF/cm<sup>2</sup>, respectively (Randin and Yeager, 1975, Randin and Yeager, 1972). High surface area and porosities can be achieved by carbonisation, physical or chemical activation (Torregrosa and Martín-Martínez, 1991,

Rodríguez-Reinoso and Molina-Sabio, 1992), phase separation (Hatori et al., 1992), gelation (Hatori et al., 1992), emulsification (Barby and Haq, 1982, Edwards, 1988), aerogel-xerogel formation (Pekala et al., 1990), replication (Ryoo and Joo, 2004), or burning carbon composites with controllable sizes and volume fractions of open pores which involves adding raw polymer particles, e.g. poly(methyl methacrylate) (PMMA) spheres (Conway, 1991a), silica sol and silica gel as templates (Jurewicz et al., 2004, Lee et al., 2004, Vix-Guterl et al., 2004). Generally, carbon based materials are in powder form and some processing is necessary to transform these materials into solid compact electrodes. These methods include permanent pressure (Shi, 1996), adding binders such as polytetrafluoroethylene (PTFE) (Frank Rose et al., 1994, Bonnefoi et al., 1999, Osaka et al., 1999, Mi et al., 2007, Reddy and Ramaprabhu, 2007), poly(vinylidene fluoride-hexafluoropropylene) (Osaka et al., 1999), methylcellulose (Bonnefoi et al., 1999) and aqueous dispersions of polystyrene, styrene/butadiene copolymer and ethylene/acrylic acid copolymer (Beck and Dolata, 2001), etc.. Theoretically, the SC value of carbon materials should increase with surface area. However, Qu and Shi (1998) and Shi et al. (1996) studied several activated carbon materials with different surface area, pore size, pore size distribution and pore volume, and correlated these parameters with electrochemical capacitance and found that the hypothesis is not necessarily true in practical cases.

Frackowiak and Béguin (2001) reported that porous texture of the carbon determines the ionic conductivity, which is related to the mobility of ions inside the pores. The rate of electrochemical accessibility is determined by the mobility of ions inside the pores, which is different from that in bulk electrolytes. Therefore, a resistor network was suggested to be considered in the equivalent circuit model instead of one resistor due to the uneven

resistance throughout the whole materials. Carbon aerogels are monolithic three-dimensional mesoporous network of carbon nanoparticles, which are obtained from the pyrolysis of organic aerogels based on resorcinol-formaldehyde or phenol-furfural precursors via a sol-gel process. While carbon xerogels (Lin and Ritter, 1997, Zanto et al., 2002) are obtained from a precursor prepared by conventional drying, and not by supercritical method in CO<sub>2</sub>. Carbon aerogel and xerogels are considered as promising materials for supercapacitors, because they have high surface area, low density, good electrical conductivity, and require no additional binding materials. In order to improve the capacitance of carbon materials, functional groups are introduced into the carbon materials because the functional groups are related to the pseudocapacitance, which is a very effective method of increasing the capacitance. Chu and Kinoshita (Chu and Kinoshita, 1996) investigated the effects of pre-treatment and surface modification on the electrical double-layer capacitance of various carbon electrodes, and the surface modification of carbon electrodes compromise electrochemical and chemical techniques such as oxidation/reduction and preadsorption of various surface functional groups.

### **2.2.2 Conducting Polymer-Based Supercapacitors**

Conducting polymers are the third group of candidate materials for supercapacitors due to their good electrical conductivity, large pseudocapacitance, and relatively low cost (Peng et al., 2008). Polymeric materials, such as p- and n-dopable poly(3-arylthiophene), p-doped poly(pyrrole), polyacetylene, poly[bis(phenylamino)disulfide], poly(3-methylthiophene), or poly(1,5-diaminoanthraquinone) have been suggested by several researchers (Arbizzani et al., 1996a, Ren et al., 1996, Balducci et al., 2005, Zhou et al.,

2005, Fan and Maier, 2006, Fonseca et al., 2006) as electrodes for electrochemical capacitors.

The most commonly used conducting polymers include PANi (Mengqiang Wu and Shuren, 2007), PPy (Wang et al., 2007) and poly[3,4-ethylenedioxythiophene] (PEDOT) (Chen et al., 2009b). The electrochemical capacitance and charge storage properties of conducting polymers have been studied by CV and CD techniques. The typical capacitive behaviour of a polymer electrode, however, is in general not similar to MWCNTs electrode, as is expected for a typical capacitor, but exhibits higher redox potential and pseudocapacitive behaviour than MWCNT electrode. In order to use the same electrode materials on both capacitor electrodes, polymers with a cathodic and an anodic redox process were utilised recently (Lao, 2006).

Conducting polymers, such as PANi, PPy, polythiophenes, polyacetylene, and poly[bis(phenylamino)disulfide] (Su et al., 2004), comprise a large degree of  $\pi$ -orbital conjugations that lead to electronic conductivity, and can be oxidised or reduced electrochemically by withdrawal or injection of electrons, respectively. Prasad and Munichandraiah (2002) coated the PANi on the stainless steel by a potentiodynamic method from an acidic electrolyte and obtained very high SC value, up to 450 F/g.

Conducting polymers have a very large SC value that is close to RuO<sub>2</sub>, e.g. 775 F/g for PANi (Gupta and Miura, 2006), 480 F/g for PPy (Fan and Maier, 2006), and 210 F/g for PEDOT (Xu et al., 2006). However, conducting polymers commonly have poor mechanical stability due to repeated intercalation and depletion of ions during charging and discharging.



### 2.2.3 Transition Metal Oxides-Based Supercapacitors

Besides the carbon material electrodes, transition metal oxide electrodes are very important in electrical storage devices. In general, transition metal oxides can provide higher energy density for supercapacitors than conventional carbon materials and better electrochemical stability than polymer materials (Wang et al., 2012). They not only store energy like electrostatic carbon materials but also exhibit electrochemical Faradaic reactions between electrode materials and ions within appropriate potential windows (Zhao et al., 2007).

Trasatti and Buzzanca (1971) first used  $\text{RuO}_2$  as supercapacitor electrodes. The  $\text{RuO}_2$  electrode, also called dimensionally stable anodes (DSA), is prepared thermochemically from  $\text{RuCl}_3$  or  $(\text{NH}_4)_3\text{RuCl}_6$  painted on Ti substrates between 350 and 550 °C, with the addition of titanium isopropoxide or  $\text{TiCl}_3$ . The  $\text{RuO}_2$ -aqueous system was extensively studied (Zheng et al., 1995, Conway, 1999) and the SC value was reported to be as high as 720 F/g with  $\text{H}_2\text{SO}_4$  electrolyte (Zheng et al., 1995).

The  $\text{RuO}_2$  electrodes have shown the best rectangular shape of cyclic voltammetry (CV) curve and exhibited the best capacitor behaviour (Kötz and Carlen, 2000) compare to other transition metal oxies. However, the shape of the CV is not a consequence of pure double-layer charging, but of a sequence of redox reactions (pseudocapacitance) occurring in the metallic oxide. The valence state of Ru may change from III to VI within a potential window of slightly above 1 V. The ratio of surface charging to bulk processes is nicely described by Ardizzone et al. (1990). In aqueous acid electrolytes the fundamental charge storage process is proton insertion into the bulk material.

Conducting metal oxides like  $\text{RuO}_2$  or  $\text{IrO}_2$  were the favoured electrode materials in early electrochemical capacitors used for space or military applications (Kötz and Carlen,

2000). The high SC value in combination with low resistance resulted in very high specific powers. These capacitors, however, turned out to be too expensive. A rough calculation of the capacitor cost showed that 90% of the costs reside in the electrode material. In addition, these capacitor materials are only suitable for aqueous electrolytes, thus limiting the nominal cell voltage to 1 V (Kötz and Carlen, 2000).

Thus, several attempts were undertaken to obtain the advantages of metal oxides at reduced cost for supercapacitor such as  $\text{Fe}_2\text{O}_3$  (Nagarajan and Zhitomirsky, 2006, Hang et al., 2008),  $\text{Fe}_3\text{O}_4$  (Du et al., 2009),  $\text{SnO}_2$  (Wu, 2002),  $\text{CuO}$  (Wu et al., 1998, Wu et al., 2002a)  $\text{MnO}_2$  (Goodenough, 1984, Toupin et al., 2002, Deng, 2008, Ragupathy et al., 2009, Xia et al., 2009)  $\text{NiO}$  (Wang et al., 2006, Zheng and Zhang, 2007, Patil et al., 2008a). The dilutions of the costly noble metals such as Ru, Sr, Ca and La by forming perovskites ( $\text{SrRuO}_3$ ,  $\text{CaRuO}_3$  and  $\text{Sr}_{0.8}\text{La}_{0.2}\text{RuO}_3$ ) were reported by Guther et al. (1997). Other forms of metal compounds such as nitrides were also investigated by Liu et al. (1998).

Among all, researchers recently have focused on searching for cheaper materials such as  $\text{MnO}_2$  (Malak et al., 2010, Yue et al., 2008),  $\text{NiO}$  (Lee et al., 2005, Nam et al., 2008),  $\text{Co}_3\text{O}_4$  (Wei et al., 2009, Xiong et al., 2009) and  $\text{CuO}$  (Patake et al., 2009) to replace  $\text{RuO}_2$ , but the selection has traditionally been limited by the use of concentrated sulphuric acid as an electrolyte (Namisnyk, 2003, Cottineau et al., 2006). It was believed that high capacitance and fast charging were largely the result of H sorption, so a strong acid was therefore necessary to provide good proton conductivity. This resulted in a narrow range of possible electrode materials, however, since most metal oxides break down quickly in acidic solutions.

Milder aqueous solutions such as KCl (Jiang and Kucernak, 2002) and KOH (Ko and Kim, 2009) have therefore been considered for use with metal oxides such as MnO<sub>2</sub>. The capacitance of Fe<sub>3</sub>O<sub>4</sub> in Na<sub>2</sub>SO<sub>3</sub> aqueous solution is also very encouraging, from a few tens to 510 F/g, and it depends on the state of dispersion of the oxide crystallites (Wu et al., 2002b). The SC value of approximate to 137.7 F /g could be achieved from NiO nanoflakes electrode in KOH electrolyte solution, due to higher surface area of NiO nanoflakes, which facilitates transport of electrolyte ions during rapid CD process (Zheng et al., 2009).

Porous CuO thin film electrode prepared by cathodic electrodeposition method has also shown a potential application in supercapacitor materials by giving maximum SC value of 36 F/g in Na<sub>2</sub>SO<sub>3</sub> aqueous electrolyte (Patake et al., 2009).

#### **2.2.4 Hybrid Nanocomposite-Based Supercapacitors**

As discussed above, different electrode materials have different strong points and drawbacks. Compared with the conventional capacitors, a higher energy density and a corresponding cycle life can be achieved by hybrid electrochemical supercapacitors which use two different electrode materials (Cheng et al., 2006, Park et al., 2009).

In order to take full advantage of different electrode materials, hybrid nanocomposite supercapacitor materials have been proposed by several researchers. Referring to Figure 1, Hybrid nanocomposites can be made from different double layer capacitor and pseudocapacitor materials such as conducting polymer/CNTs (Hughes et al., 2002, Lee et al., 2002, Xiao and Zhou, 2003, Konyushenko et al., 2006, Kumar and Chen, 2008, Peng et al., 2008), activated carbon/conducting polymer (Arbizzani et al., 1996b, Chen and Wen, 2003, Laforgue et al., 2003, Shekhar et al., 2006, Snook et al.,

2009), activated carbon/CNTs (Emmenegger et al., 2003, Wang et al., 2005, Taberna et al., 2006, Huang et al., 2008, Huang and Teng, 2008), metal oxide/activated carbon (Jang et al., 2003, Wu et al., 2004, Zhang et al., 2005, Liang et al., 2008), metal oxide/CNTs (Reddy and Ramaprabhu, 2007, Yan et al., 2008, Kiamahalleh et al., 2010b, Kiamahalleh et al., 2009), mixed metal oxide/CNTs (Fan et al., 2007, Li et al., 2008, NuLi et al., 2009) and metal oxide/CNTs/ conducting polymer (Kiamahalleh et al., 2011, Kiamahalleh et al., 2010a, Zamri et al., 2012, Sot et al., 2010, Sivakkumar et al., 2007, Hou et al., 2010).

When one compares electrochemical characteristics of the rest of the composites, there are some advantages of such electrode materials. For instance, Lota and coworkers (2011) showed that CV curves for active carbon and (NiO) composite material have a much more rectangular shape, which is a characteristic of a double-layer capacitive behaviour. They suggested that the capacity of pristine NiO is low and dramatically decreases with increasing current regimes. The bare NiO has only pseudocapacitive properties. In their work, composite with only 7wt.% of NiO showed the highest value of capacity. They believed that such amount of NiO was optimal to take advantage of both components of the composite. The active carbon played a role of excellent conducting support for NiO, which is characterized by quite big resistivity.

## **2.3 Advantages and challenges of current supercapacitors**

### **2.3.1 High power density**

Supercapacitors display a much higher power delivery (1-10 kW/kg) when compared to lithium ion batteries (150 W/kg). Since a supercapacitor stores electrical charges both at the electrode surface and in the bulk near the surface of the solid electrode, rather than within the entire electrode, the charge–discharge reaction will not necessarily

be limited by ionic conduction into the electrode bulk, so the charging and discharging rates are much faster than the electrochemical redox reactions inside batteries. These rapid rates lead to high power density in supercapacitor (Wang et al., 2012). For example, a supercapacitor can be fully charged or discharged in a time varies from fractions of a second to several minutes, and the energy can be taken from it very rapidly, within 0.1 s (Kusko and DeDad, 2007, Uzunoglu and Alam, 2008). However, the charging time for batteries is normally on the scale of hours.

Pasquier and co-workers (Pasquier et al., 2003) achieved high specific power of 800 W/kg from nonaqueous asymmetric hybrid electrochemical supercapacitors using a nanostructured  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  anode and an activated carbon cathode. However, before this work they used activated carbon (Laforgue et al., 2001) and nano-structured lithium titanate oxide (Du Pasquier et al., 2002) as anode with a poly(fluoro)phenylthiophene cathode to investigate the benefit of conducting polymer on the power density. But their polymer based system had two major drawbacks including lower cycle-life and higher cost than activated carbon. In 2004, they coupled a nano-structured  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  anode with poly(methyl)thiophene (PMeT) cathode. Using PMeT cathode prepared by electrochemical synthesis of the inexpensive monomer, they achieved a lower cost, higher purity polymer and enhanced to power density to 1000 w/kg (Du Pasquier et al., 2002).

### ***2.3.2 Long life expectancy***

On the contrary, with batteries, supercapacitors have no or negligibly small chemical charge transfer reactions and phase changes in charging and discharging process so that a supercapacitor can have almost unlimited sustainable cyclability. Moreover,

supercapacitor can operate at high rates for 500,000–1,000,000 cycles with only small changes in their characteristics, but such longevity is impossible for batteries (Wang et al., 2012). The life expectancy for supercapacitor is estimated to be up to 30 years, which is much longer than for lithium ion batteries which is only between 5 to 10 years (Wang et al., 2012). Although the redox reactions for Faradaic supercapacitors during their charge-recharge process is very quick, but their life expectancy is also much longer than that of batteries (Inagaki et al., 2010, Burke, 2000, Zhang et al., 2009).

Researches on the life expectancy for  $\text{MnO}_2$  based electrode was carried out by Chang et al. 2009 (Chang et al., 2009). Evaluation of the stability and cycle life of  $\text{MnO}_2$  was investigated for 600 times (at a rate of 5 mV/s) and there was no significant capacitance fading detected, which showed the excellent cyclic stability of  $\text{MnO}_2$ . Their experimental results have also introduced ionic liquid 1-ethyl-3-methylimidazolium–dicyanamide (EMI–DCA IL) as a good potential electrolyte for long-life  $\text{MnO}_2$  supercapacitors (Chang et al., 2009).

It is noteworthy to know that the specific capacitance of supercapacitor with any electrode materials will be faded in the long-term cycle performance. Pristine  $\text{MnO}_2$  and CNTs/ $\text{MnO}_2$  hybrid electrodes were undergone life testing for about 500 cycles by Chen and co-workers (Chen et al., 2009a). Noticeable decrease in the specific capacitances of both  $\text{MnO}_2$  and CNTs/ $\text{MnO}_2$  hybrid electrodes occurred as the cycle number increases, but fading was less observed for CNTs/ $\text{MnO}_2$  electrode. Hybrid electrode exhibited better cyclic efficiency than the  $\text{MnO}_2$  electrode without CNTs mix and it was attributed to the porous structure of CNTs/ $\text{MnO}_2$  electrode (Chen et al., 2009a). Hence, hybridizing pseudocapacitive materials with porous carbon structured materials may tend to provide

more space for dissolution and precipitation of pseudocapacitive material particles in the processes of repetitive CV reactions.

### ***2.3.3 Long shelf life***

Another advantage of supercapacitors is their long shelf life. Most rechargeable batteries will degrade and become noticeably useless if left unused for months and it is due to their gradual self-discharge and corrosion (Wang et al., 2012). On the contrary, supercapacitors retain their capacitance and thus are capable of being recharged to their original condition; even though self-discharge over a period of time can lead to a lower voltage. It is reported that supercapacitor can stay unused for several years, but still remain close to their original condition (Burke, 2000).

### ***2.3.4 Wide range of operating temperature***

The ability to operate efficiently of a wider range of temperatures is also an advantage of using supercapacitors. For some remote stations which are using battery energy storage system and located in cold climates, auxiliary heating system must be provided to maintain the temperature at close to room temperature. Thus, additional cost and energy consumption are required (Namisnyk, 2003). Supercapacitor can operate effectively at enormously high and low temperatures. The typical operating temperature for supercapacitors varies from  $-40$  to  $70$  °C. Militaries can benefit from this behavior, where reliable energy storage is required to run electronic devices under all temperature conditions during war (Wang et al., 2012). Using ionic liquids electrolyte such as ethylmethylimidazolium-bis(trifluoromethanesulfonyl)imide in supercapacitor was a

significant advantage due to its high temperature stability, even at 60°C (Largeot et al., 2008).

Balducci and coworkers, (Balducci et al., 2004) investigated the effect of different temperature for an activated carbon/poly(3-methylthiophene) hybrid supercapacitor using using ionic liquids based on 1-butyl-3-methyl-imidazolium as electrolytes and organic liquid electrolyte, propylene carbonate-tetraethylammonium tetrafluoroborate. The ionic liquid electrolytes exhibited conductivity values at room temperature significantly lower than that of organic liquid. However, since at temperatures increased to even higher than 60 °C, which are of interest for electric vehicle applications involving fuel cells, the conductivity values were almost the same for both ionic and organic electrolyte. This is also attributed to higher electrochemical stability of ionic electrolyte at high temperature (Largeot et al., 2008).

### ***2.3.5 Environmental friendliness***

Supercapacitors as alternative to batteries are much environmentally-friendly, lead free, and have no disposal issues at end of their life. They do not contain hazardous or toxic materials, and their waste materials are easily disposed (Wang et al., 2012). Using porous carbon materials (Laforgue et al., 2003, Yan et al., 2010, Hou et al., 2010, Kiamahalleh et al., 2011, Ruifeng et al., 2010), conducting polymers (Sot et al., Laforgue et al., 2003, Kiamahalleh et al., 2011, Sheng et al., 2012) and transition-metal oxides (Hou et al., 2010, Bose et al., 2012, Li et al., 2010, Lu et al., 2012, Kiamahalleh et al., 2011, Ruifeng et al., 2010) as single or hybrid electrodes and ionic based liquids (Laforgue et



al., 2003, Lu et al., 2012) as electrolyte suggested supercapacitor as the most environmental friendly and promising energy storage system.

## **2.4 Challenges for supercapacitor**

Although supercapacitors have many advantages over batteries and fuel cells, they also face some challenges at the current stage of technology.

### **2.4.1 Low energy density**

The main challenge for supercapacitors applications in the short and medium terms is their low energy density. Supercapacitors suffer from limited energy density (about 5 W h kg<sup>-1</sup>) when compared with batteries (>50 W h kg<sup>-1</sup>) (Wang et al., 2012). Commercially available supercapacitors can provide energy densities of only 3 to 4 W h kg<sup>-1</sup>. If a large energy capacity is necessary for an application, a larger supercapacitor must be assembled which increases the cost.

### **2.4.2 High cost**

The other important challenges for supercapacitors commercialization are the costs of raw materials and manufacturing. The major cost of supercapacitors arises from their electrode materials. Recently, highly porous carbon materials (Frackowiak and Béguin, 2001, Kötz and Carlen, 2000, Lu et al., 2011) and RuO<sub>2</sub> (Yuan et al., 2012, Zheng and Jow, 1995) are the most common electrode materials used practically in fabricating commercial supercapacitors. However, carbon materials, in particular those with a high surface area, are currently expensive (US\$50–100 per kg) (Burke, 2000), SWCNTs ~\$100,000/kg, MWCNTs ~\$5,000/kg, activated carbon ~\$15/kg (Ervin, 2010 #205), not to mention the cost of a rare metal oxide such as RuO<sub>2</sub>. Additionally, the separator and

the electrolyte can also boost the expense. For instance, if supercapacitors use organic electrolytes, their cost is very high and almost far from negligible (Wang et al., 2012).

### ***2.4.3 Low operating voltages***

Supercapacitors also have lower operating voltages (typically between 1–3.5 V per cell) compare to other type of capacitors and batteries in which the voltage is limited by the breakdown potential of the electrolyte (Wang et al., 2012). The solution to meet the expected voltage is that supercapacitors must be series connected, like batteries (Shukla et al., 2012). Commercial supercapacitor cells exhibit the potential window from 0 V to approximately 1 V for aqueous electrolytes and from 0 V to 2.5–2.7 V for organic electrolytes. Maximum voltages for hybrid supercapacitor cells depend upon the electrode materials and the electrolytes (Stoller and Ruoff, 2010). Matsumoto and coworkers (Matsumoto et al., 2004) showed that the Pt-deposited carbon nanotube electrode gave 10% higher voltages than Pt-deposited carbon black. However the amount of platinum deposited on CNT and carbon black were 12% and 29%, respectively. The reason of higher performance of the Pt/CNT electrodes compared to the Pt/CB electrodes were attributed to the well-dispersed Pt particles on the CNT surfaces and higher electric conductivity of CNTs (Matsumoto et al., 2004).

In summary, to develop new materials with optimal performance, two major research directions in supercapacitor electrode materials exploration are (i) Composite materials and (ii) Nanomaterials. In one hand and apart from of the type of electrode material for the supercapacitors, combining the advantages of different materials to make composites should be an important approach to optimize each component for increasing the supercapacitor performance (Jiang et al., 2012). It is worth

to point out that each component in the composites can have a synergistic effect on the performance of supercapacitor through minimizing particle size, enhancing specific surface area, inducing porosity, preventing particles from agglomerating, facilitating electron and proton conduction, expanding active sites, extending the potential window, protecting active materials from mechanical degradation, improving cycling stability, and providing extra pseudocapacitance (Wang et al., 2012). A high specific capacitance  $1809 \text{ F g}^{-1}$  has been reported on using mesoporous  $\text{Co}_x\text{Ni}_{1-x}$  layered double hydroxides composite (Hu et al., 2009). The key point to the excellent electrochemical capacitance performance of this composite was attributed to the synthesis of a novel mesoporous microstructure which can accommodate electrolyte and increase density of active sites for promoting fluid/solid reactions (Hu et al., 2009).

On the other hand, development of nanostructured materials, such as nanoaerogels, nanotubes /rods, nanoplates, nanospheres, etc. can also have significant enhancement in electrochemical performance by possessing high specific surface area. They can provide short transport /diffusion path lengths for ions and electrons, leading to faster kinetics, more efficient contact of electrolyte ions, and more electroactive sites for Faradaic energy storage, resulting in high charge/discharge capacities even at high current densities (Wang et al., 2012). An extremely large specific capacitance of  $3200 \text{ F/g}$  was reported by Zhang et al. (2010) by porous composite materials consisting of nanoflake-like nickel hydroxide and mesoporous carbon. They attributed the overall improved electrochemical behavior to the unique structure design in nickel hydroxide/mesoporous carbon composite in terms of its nanostructure, large specific surface area and good electrical conductance (Zhang et al., 2010).

As a result, material morphology is intimately related to the specific surface area and the electrolyte ions diffusion in the electrode . Hence, one-dimensional nanostructure materials are highly suggested to be promising for supercapacitor application due to their reduced diffusion paths and larger specific surface areas (Wang et al., 2012).

### **3 Current Applications: eg: Memory backup, electric vehicles Power quality, Battery improvement, electromechanical actuators, Adjustable-speed drive ‘ride-through, etc.**

Supercapacitors are operating the path into more and more applications where electrical energy requires being stored. These robust devices can be charged and discharged thousands of times and generally outlive a battery. Many supercapacitor manufacturers claim a life span of 10 years or more. It is also noteworthy that the supercapacitors and batteries are not in competition; rather they are different products having their unique applications.

Supercapacitors in terms of their power densities are divided into two segments: (i) large cans from 100F-5000F used for heavy duty applications such as automotive and utilities, (Endo et al.) and (ii) small devices in the range from 0.1F-10F used in general electronics (Mars, 2011). A practical way to compare supercapacitors is to employ a Ragone plot which plots power density versus energy density (as shown in figure 2, section 1.3).

#### **3.1 Memory backup**

Semiconductor memory back-up is the most common application of supercapacitor for electronic equipment which contains CMOS, RAM or microprocessor (Endo et al.,

2001). For several years Lithium batteries have served as backup power for volatile memory and real time clock, but they are not always an ideal solution due to their relatively short cycle life, limited operating temperature range. However, their end of life disposal issues is remained to be taken into account.

Supercapacitors, rather than operating as energy storage device, they also perform quit well as low maintenance memory backup in order to bridge short power interruptions. In 1978, the NEC Company marketed the supercapacitor as computer memory backup for the first time (Endo et al., 2001). They obtained the licence for commercializing the invention from the Japanese Standard Oil Company of OHIO which accidently rediscovered the effect of the electric double layer capacitor EDLC in developing fuel cell designs (Rightmire, 1966).

Pierre Mars (2008) reported the use of supercapacitor as backup power supply for the cache memory used in solid state drives (SSDs). The SSD types of hard disks are electrically, mechanically and software compatible with a conventional hard disk drive. (HDD) When cache is used in normal hard disks, noticeable data transfer speed in terms of reading and writing can be achieved. Pierre's power supply topology using CAP-XX supercapacitors also confirmed the simulation results. This supercapacitor showed to be highly supportive to the SSD when it flushes the cache and elegantly shuts down with no loss of data while the power fails (Mars, 2008).

A simplified backup systems having 2-cell series supercapacitor charger (LTC3226) designed with a PowerPath controller was by Linear Technology (Drew, 2011). This backup system includes a charge pump supercapacitor charger with programmable output

voltage and automatic cell voltage balancing, a low dropout regulator and a power-fail comparator for switching between normal and backup modes. Three different voltage points simplify and control the backup set up in this system. (i) High trigger point (3.6V): when the comparator encounters the power fail, (ii) standby mode point (3.15V) and (iii) backup mode point (3.10V): when the system initializes to hold up the power in the absence of battery power. The holdup power for the LTC3226 was reported to be for a time period of about 45 seconds (Drew, 2011).

### **3.2 Electric vehicles power quality**

The use of hybrid power system in vehicles eliminates the need of battery electrification as these systems are able to recover and restore the energy from several braking. The most important advantage of their usage in reduction of global CO<sub>2</sub>-emissions has been turned to a very critical topic nowadays (Lohner and Evers, 2004). However, upgrading the vehicles with hybrid power systems could be quite costly (approximately \$9,500 per kWh) (Zorpette, 2005), but their greater lifetime (10-20years), cycle life (0.5-1 million charge/discharge) and complete discharge with-out any life degradation are highly considerable in the economic evaluation (Smith and Sen, 2008). Supercapacitors have been served as power quality for direct current (DC) motor drives, uninterruptible power supply (UPS) systems, and hybrid and electric vehicles. The automotive industries are also very interested in this technology and currently are very involved in the use of supercapacitors for the hybrid electric vehicles. They have been used in many large scale application particularly transportation industry such as hybrid cars, autonomous rail-guided vehicles, support of substation voltage for trolleybuses, tramways, and subways (a proto-type being tested), busses and trains (Beale and Gerson,

2004, Zorpette, 2005, Lohner and Evers, 2004, Rufer et al., 2004, [On-line], 2008). These ultracapacitors due to their high power density can operate as supplementary power source to batteries in order to provide a burst of acceleration for the vehicles or can be used to absorb energy during regenerative braking schemes. Many of the hybrid electric vehicles coming to the market employ supercapacitors. For instance, Honda developed their market by employing the supercapacitor technology in many of their vehicles (Barker, 2002).

Smith and Sen (2008) also stated that large scale battery systems can benefit from setting up the supercapacitors in parallel with them in order to compensate for the momentary and temporary interruptions. Employing this technology may decrease the undue stress being put on the batteries by these short term interruptions. Lohner and Evers (Lohner and Evers, 2004) employed a double-layer capacitor (supercapacitor) based hybrid power train for light rail vehicles and run their first practical test on diesel-electric city bus.

Most of driving cycles of such as trams, light-rail vehicles and city busses are basically made of acceleration and deceleration phases. To date, the greater part of the kinetic energy generated by diesel engines deceleration has not been used as they are not able to recuperate energy. On the contrary the trams and light-rail vehicles require another accelerating vehicle as an energy consumer in their net part. Researchers have come out with primary energy save up to 30 %, if the power train could store the kinetic energy from the decelerating vehicle for the subsequent accelerating phase (Ackerman and Jefferson, 1998, Ehrhart, 1997).

The energy saving unit (Sharma et al.) includes a buck-boost inverter and a double-layer capacitor battery (DLC) which support the prime mover unit (PMU) during vehicle acceleration and deceleration phases. The supercapacitor can store the recovered kinetic energy from regenerative braking in order to use it used for the next acceleration phase (Lohner and Evers, 2004). The voltage required for the bus traction is in the range of the battery from 350 V to 720 V which can be provided by 288 supercapacitor cells with 2.5V, connected in series. The total storable energy of the battery is able to accelerate the vehicle **up** to 50 kmh (maximum speed of urban driving cycles) without help of the PMU. The supercapacitors installed on the urban buses benefit from several number of stops so that the whole braking energy can be recovered in the battery for several times (Lohner and Evers, 2004).

### **3.3 Electromechanical actuators**

Electromechanical actuators directly convert electrical energy to mechanical energy and high electric power is required for this conversion. Supercapacitors, having higher capacitances in comparison with that of ordinary conventional capacitors, are the better option to be used as electromechanical actuators. Artificial muscles in robots (Baughman et al., 2002, Agboola, 2005), biomimetic flyings, optical fiber switches, microsensors (Li et al., 2011), optical displays, prosthetic devices, sonar projectors, and microscopic pumps (Baughman et al., 1999) are some of great potential applications of electromechanical actuators. However, these applications are restricted into some extent by the maximum allowable operation temperature and the required voltages.

The electromechanical actuators and supercapacitors are both consist of two separate electrodes (anode and cathode) which are separated by an insulating ionically



conducting material in the electrochemical cell. Both Faradaic and non-Faradaic materials have been served as actuator electrode materials. For instance, over two decades ago, Baughman and co-workers became pioneer by proposing the conducting polymer actuators based on electrochemical dopant intercalation (Baughman et al., 1990). Since then, many researchers from different laboratories benefited from the idea of using conducting polymers with Faradaic properties such as polyaniline (Kaneto et al., 1995), Poly(2-acrylamido-2-methylpropanesulfonic acid) (PAMPS) (Osada et al., 1992) and polypyrrole (Otero and Angulo, 1992).

Transition metal oxides are other types of Faradaic materials which have also been used in actuators. A patent by Park's group from the University of Harvard reported their invention: nanoscale electromechanical actuator based on individual transition-metal-oxide nanowires (Park et al., 2005). They used different metal oxides from titanium and zirconium in actuator electrode. Both titanium and zirconium oxides showed high converse-piezoelectric properties which make them promising material to fabricate electromechanical actuators in order to position and move nanometer-sized objects. Recently, Mrunal's research group have showed that the piezoelectric and semiconducting properties of ZnO nanoparticles can effectively enhance the recorded motional current and lead to high integrated electrical actuation. However, Faradaic materials show the battery-like properties storing high energy, but they limit the power density, cycle life, and energy conversion efficiencies of the actuators due to their solid-state dopant diffusion and structural changes. Hence, new actuators were designed and fabricated having carbon electrode materials with high conductivity and non-Faradaic properties (Takeuchi et al.,

2009, Rogers and Liu, 2012) as they do not have any dopant intercalation (Baughman et al., 1999).

Generally, the capacitance of an electromechanical actuator similarly with supercapacitors directly depends on the separation between the charges on the electrode and the counter large in the electrolyte and this separation is in the scale of nanometer. Therefore, carbon nanomaterials, particularly CNTs (Baughman et al., 1999, Baughman et al., 2002, Li et al., 2011) and graphene (Liang et al., 2012, Stoller et al., 2008) are the promising candidates for electrode materials to store large capacitances as they possess high surface area accessible to the electrolyte. CNTs based electromechanical actuators was also reported by Terrones and co-workers. This actuator can function at low voltages and temperatures up to 350°C which indicates the high thermal stability of carbon nanotubes and industrial application of carbon electrodes (Terrones, 2003).

As earlier discussed, both Faradaic and non-Faradaic material have some advantages and drawbacks. In order to benefit from both of them, syntheses of their composites at nanoscale have been proposed by some researchers. Nanocomposite from incorporation of CNTs and polydiacetylene (PDA), a polymer with interesting electrical and optical properties, was made by Peng et al. (2009). This nanocomposite could reversibly change colour in response to electrical current and mechanical stress which made it promising for applications in many fields such as sensors, actuators. Recently, Liang and co-workers stated that the CNTs have poor processability and difficulty of forming standalone bulk film materials with pure PDA that hinder its practical use in the field of actuation (Liang et al., 2012). Hence, they proposed two-dimensional (2D) single layer of graphene (relatively smooth surface) as an alternative to CNTs, where the PDA could well-ordered

striped on its surface. Combining the outstanding intrinsic features of graphene with the properties of unique environmental perturbations-actuated deformational isomerisation of PDA caused the high conductivity for graphene-PDA nanocomposite and thermal-induced expansion of PDA. The actuation test showed that the actuator movement while applying an AC of high frequency was quite faster than that of in low frequency (Liang et al., 2012).

The most important parameter for actuators is the ratio of actuator strain to unit volume of charge in electrode materials. It is also noteworthy that every ion stored in the electrodes generates strain, and the larger the strain is, the better the actuator is as an electromechanical energy conversion device. Liu and co-workers showed that a relatively large strain can be obtained from ruthenium oxide when it is mixed with carbon material. The high surface area of the metal oxide combined with the high conductivity of carbon material assist to achieve rapid charging and fast motion of the transducer (Akle et al., 2006). The ion size is also a parameter which is proportionally influence on the electromechanical actuator efficiency. The larger ion size, the less mobility the greater ratio of volume strain/ion in the electrode materials, which will directly lead to higher actuator efficiency (Liu et al., 2010). Ionic liquids as the electrolytes depend on their vapour pressure and thermal stability have the potential to significantly influence on the performance of actuators. For instance, the near-zero vapor pressure and high thermal stability over a wide range of temperature can noticeably enhance the actuator lifetime and also the operational temperatures range (Wilkes, 2002).

### **3.4 Adjustable-speed drive ‘ride-through**

Several electric devices from home appliances to industrial plants face with a critical power disturbances problem caused by voltage sags and momentary interruptions. Voltage

sags normally do not cause equipment damage but can easily disrupt the operation of sensitive loads such as electronic adjustable speed drives (ASD's) (Dahiya et al., 2008). Voltage sags are the main cause for a momentary decrease voltage triggering an under voltage trip leading to nuisance tripping of ASD's employed in continuous-process industries which contributes to loss in revenue (Deswal et al., 2010.). For instance, statistical data by 1977 showed that Power quality disruptions costed U.S. companies more than \$25 billion annually (Jouanne and Enjeti, Dec. 1997, Sullivan et al., 1997). The nuisance of ASD's can be very disruptive to an industrial process when the voltage drops 15-20% below its nominal value (Conrad et al., 1991, Wagner et al., 1988). Thus, a practical ride-through scheme for an ASD's based on supercapacitor during voltage sag is necessarily required to maintain the ASD dc bus voltage under voltage sag condition.

A number of solutions have been raised to overcome this power tripping problem (van Zyl and Spee, 1998) including Lead-acid batteries, flywheels, and ultracapacitors. Among these, batteries and flywheels seemed to have profound maintenance, safety and cost issues (Schatz, 1998, Corley et al., 1999). The supercapacitors, however, are the best candidates as high power density and relatively low energy density is required. They provide a cost-effective solution at moderate power levels both in terms of initial and ownership costs (Hoffman and Smith, 1997, van Zyl and Spee, 1998). One thing which should too much be taken into account is about voltage decreases during supercapacitors discharge. To address this issue, Corley and co-workers (1999) designed and fabricated a 100 kW prototype ride-through system with a reliable, efficient, and cost-effective dc-dc convertor for Maxwell Technologies. A dc-dc conversion system can regulate the bus voltage to allow for ASD ride-through while the supercapacitor voltage decreases during

discharge. Their experimental results from using metal/carbon based supercapacitor proved the capability of the system to support ASDs and similar loads during the voltage sags and brief outages.

The patent from Enjeti and Duran-Gomez (1999) introduced a new method for a ride-through system for ASD. They claimed that their invention, method and system for ride-through of an ASD for voltage sags and short-term power interruptions substantially eliminates or reduces the disadvantages of any previous methods. They also stated that using an additional energy source diode like a supercapacitor, battery, fuel cells, photovoltaic cells and flywheel can maintain the dc-link voltage value in the event of voltage sag, or even a complete loss of input power, energy source. The difference of their system with existed systems was adding three diodes, an inductor and a control unit connected to dynamic braking circuit which keep the ride-through circuit activated when necessary.

In 2002, Duran-Gomez and co-workers simulated, designed and fabricated a ride-through with flyback converter modules powered by super capacitors (Duran-Gomez et al., 2002). Their simulation and experimental results from loaded and non-loaded ride-through system demonstrated the feasibility of their proposed approach. The voltage of both systems were reduced from 450V to 230V by a programmable ac power source facilitates the generation of a wide variety of transient and steady-state power quality disturbances. The non-loaded system showed the decay of 5 s for the dc-link voltage under short-term power interruptions, but the ride through system effectively maintained the voltage at its nominal value.

## 4 Conclusion

In Summary, the progress up to date on the supercapacitor electrode materials, including CNTs based, transition metal oxides and conductive polymers have been investigated. CNTs, because of their extraordinary mechanical properties, high conductivity and surface area, good corrosion resistance, high temperature stability and percolated pore structure are the most promising carbonaceous material for supercapacitor application. Compared with the conventional capacitors, a higher energy density and a corresponding cycle life can be achieved by hybrid electrochemical supercapacitors which benefits from both double layer and pseudocapacitive properties. It has been shown that developing an energy storage device by combining the transition metal oxides or conductive polymers with CNTs, the high electrical energy and power are achieved. Hence, fabricating of CNTs-based nanocomposite materials, and the use of such materials in supercapacitor electrodes provide significant higher SC value than for pure CNTs. RuO<sub>2</sub> having the best rectangular shape of cyclic voltammetry (CV) curve and exhibiting the best capacitor behaviour compare to other transition metal oxides is the most promising pseudocapacitive electrode material and it gives superior SC value. However, the toxicity and high cost of the precious metal (Ru) are major disadvantages for commercial production of RuO<sub>2</sub> based electrodes. MnO<sub>2</sub> and NiO are great alternatives for RuO<sub>2</sub> as they are the best electroactive materials among transition metal oxides with respect to both specific capacitance and cost effectiveness. Ceramics oxide (mixed metal oxide) from combination of the different transition metal using novel mixing methodologies will lead to superior pseudocapacitive behavior and high power-energy density. Ceramics are sometimes preferred to single metal oxides as adding one or more oxides help in augmenting the capacitances due to be complementary in their properties. The advantageous of using ceramics are in their high oxide ion conductivity and rapid charge transfer resulting in increase of the SC value of

supercapacitor. Conducting polymers due to their good electrical conductivity, large pseudocapacitance, and relatively low cost are the great candidate materials to be used with CNT and ceramics in supercapacitors electrode. Among the polymers, PEDOT seems to have a better cycleability than all other types of conducting polymers and it is recommended to be served in hybrid supercapacitor materials. Hybrid supercapacitor are beneficial as they possess high power density, long life expectance, long shelf life, wide range of operating temperature and they are environmental friendly. On the other hand, researches are still challenging with low energy density, high cost and low operating voltage of the supercapacitor materials. Current applications of supercapacitors are in memory backup, electric vehicles power quality, battery improvement, electromechanical actuators, Adjustable-speed drive ride-through, etc. Depending on the power density, supercapacitors are divided into two segments: (i) large cans from 100F-5000F used for heavy duty applications such as automotive and utilities, and (ii) small devices in the range from 0.1F-10F used in general electronics. Recently, quite large numbers of patents have reported the development and use of hybrid electrode materials in supercapacitors electrode materials and many companies fabricated hybrid supercapacitors in electronic devices. Active researches are still continuing on all aspects of hybrid supercapacitors, i.e., electrode materials (anodes, cathodes), electrolytes, and cell construction. The major factors limiting their wider application remain as their cost and safety and the recent developments from researchers have been taken place to address all of these limitations.

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