# A Review on Supercapacitors

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*Abstract--* **In this paper, a brief history of the emergence of supercapacitors is accounted for. Various types of materials used for fabrication are mentioned, classification of supercapacitors is done based on materials used and the performance of the various types of supercapacitors is reviewed. Activated carbon is the most widely used electrode material in supercapacitors. The mode of packaging of supercapacitors is highlighted. Most of the information presented is obtained from websites of companies that produce the supercapacitor itself or those that produce the materials used in producing it.**

*Keywords: Supercapacitor, Activated carbon, Equivalent series resistance, Packaging of supercapacitors.*

## I. INTRODUCTION

DISCOVERY of the electrochemical double layer capacitor (EDLC or supercapacitor) was done by General Electric engineers in 1957 while performing experiments with devices based on porous carbon electrodes. Then it was realized that energy is stored within the carbon pores. Though they did not make much effort towards its development, Standard Oil of Ohio made their own discovery accidentally while working on fuel cells in 1961. They designed it using activated charcoal as the electrode material in which the electrodes were separated by a thin insulator. This serves as the basis for the design of supercapacitors to date. Standard Oil did not commercialize their invention rather the technology was sold to NEC which then commercialized it in 1978.

In 1971 Tassati and Buzanca discovered that ruthenium dioxide films have almost the same electrochemical charging behavior with a capacitor. Continental Group Inc. contracted the author and his workers to carry out more researches on the use of ruthenium oxide type of supercapacitor. The research was carried in between 1975 and 1980 [1], [2].

During the earlier days of its discovery, it was used as a memory backup in computers. Production increased slowly and supercapacitors with improved performances and reduced cost were developed due to advances in materials science from the mid 90s to date [1].

Market analysis made by Lux Research showed that the supercapacitor market is expected to rise from $208 million in 2009 to $877 million by 2014 with a 27% compound annual growth. The forecast stated further that application in cell phones and digital camera will make up $550 million while large scale applications will make $320 million [3], [4]. Another research by Nano Markets revealed that its application in smart grid will approach $1.1 billion by year 2016. Thus in order to boost the supercapacitor production to meet up with its future demand, several R & D's on various materials that will enhance its performance and lower its cost are in progress. Moreover the technology of the ultra battery expected to be released within the next few years that integrate the lead acid battery and supercapacitor technology necessitates R & D in this field [5].

Supercapacitor as one of the components in power electronics for renewable energy devices aids in the usage of cheap sources of energy such as solar energy which reduces carbon emission. Another area which employs supercapacitors is in hybrid electric vehicles (HEV). Such vehicles make use of gasoline and power electronics devices in conjunction. By so means, fuel economy is enhanced thereby releasing lesser carbon into the atmosphere than in conventional vehicles. This together with many other advantages of the usage of supercapacitors necessitates its enhancement through R & D.

A supercapacitor is made up of two electrodes, a separator in between, an electrolyte and current collectors as shown in Figure 1 [2].
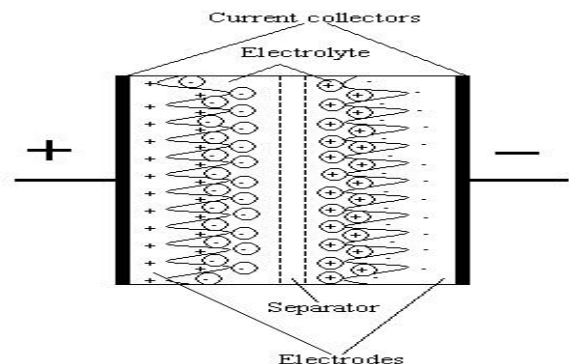


Figure 1: The Architecture of a supercapacitor [6].

## II. SUPERCAPACITOR ELECTRODE MATERIALS

There are three classes of supercapacitor electrode materials. The first one is based on carbon which offers a surface area as large as 3000m$^2$/g that can yield a capacitance of 145F/g. Supercapacitors formed with carbon are known as electrochemical double layer capacitors (EDLC).

The second class is for supercapacitors based on redox reactions (pseudocapacitors) in which electrodes are made up of electronically conductive polymers. Their capacitance values usually yields 300-400F/g but they have a problem of long term stability giving only thousands of cycles over a wide voltage range. Examples include polyaniline (pani), poly (3 – methylthiopene) (PMet), poly (dithieno [3, 4 –b: 3,'4'-d] thiopene) (PDTT1), poly (3-p-fluorophenylthipene) (PFPT) etc.

The third class is for materials with high redox capacitance. Examples include mesoporous metals, their oxides, hydroxides, oxyhydroxides, a combination of any two of them, and metal nitrides with very good electronic conductivity. The metal could be nickel, cobalt, titanium, tin, palladium, lead and ruthenium, in their pure form or as part of an alloy. Examples are nickel oxide (NiO), nickel hydroxide (Ni(OH)$_2$), nickel oxyhydroxide (NiOOH), lead dioxide (PbO$_2$), cobalt oxide (CoO$_2$), titanium dioxide, palladium oxide, ruthenium dioxide (RuO$_2$), molybdenum nitride (Mo$_2$N) and Iridium oxide (IrO$_2$) [2], [7], [8] and [9]. Its technology is similar to that of a battery.

The method of preparation of a supercapacitor electrode material determines its capacitance. For example, a capacitance of about 720F/g is obtained from a RuO$_2$ prepared at low temperatures by sol-gel process. The resulting material obtained is in amorphous form. Other methods of preparation give a typical value of 50F/g. Mo$_2$N with specific capacitance up to 40F/g is an alternative to RuO$_2$ [7].

Manganese oxide is also a low cost electrode material due to its multiple oxidation states and the structural ability to incorporate cations [10]. A lot of techniques have been employed in its preparation such as nanocasting, sonochemical method, thermal decomposition method, hydrothermal synthesis method, sol-gel route, the coprecipitation method and electrochemical deposition method. Each of the method gave a certain structure which resulted in several forms of manganese oxides such as hierarchically porous MnO$_2$, mesoporous MnO$_3$ with crystalline pore walls, discrete nanoporous MnO$_2$ and Mn$_2$O$_3$ particles, layered MnO$_2$ nanobelt etc. The maximum obtained specific capacitance from manganese oxide electrode was 350F/g [11].

Asymmetric hybrid capacitors have been developed by a combination of a double layer electrode and a pseudocapacitance electrode. The combination produces capacitances 3 to 10 times that of an EDLC. The thickness of the double layer electrode and that of the redox electrode ranges from 9:1 to 100:1, with the thickness of the redox electrode ranging from 10µm to 70µm. This gives an optimal energy density. In some cases the physical sizes of the electrodes is made to be the same in order to improve on the number of cycles. In essence the problem of sharper reductions in available capacity suffered by the conventional redox electrodes at high discharge rates is being tackled by reducing the extent to which it is discharged there by increasing its lifetime [10].

Based on the above discussion, the supercapacitor is categorized as shown in the block diagram below showing the different classes of supercapacitors down to their electrode sources.
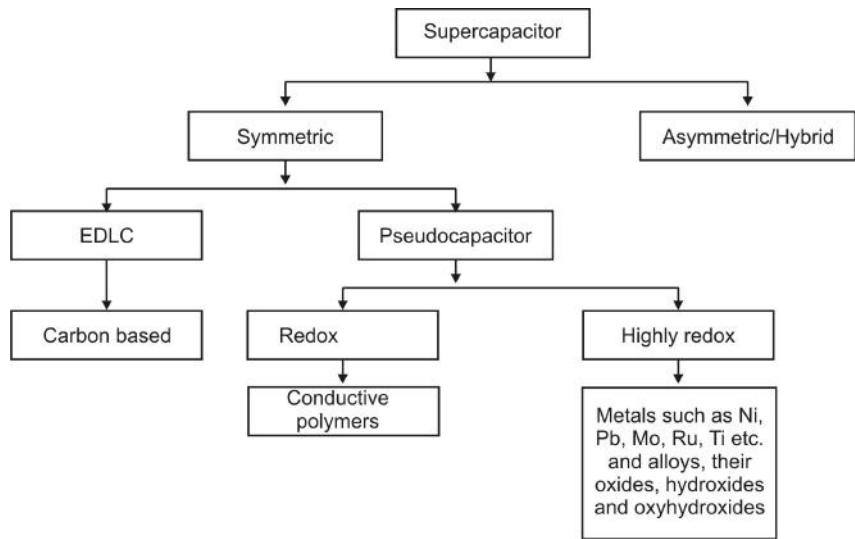


Figure 2. Block diagram of supercapacitor types and sources.

## III. ACTIVATED CARBON

Amongst all the supercapacitor electrodes materials, activated carbon is the most commonly used due to its low cost, high surface area, good electrical conductivity and thermal stability [11]. In the design of a supercapacitor using activated carbon, the issue of pore size is taken into consideration. This is because the pores in activated carbon have complex structure consisting of macro pores, meso pore, and micro pore. The importance of knowing the pore size is in order to select the electrolyte that will give a larger capacitive value. To obtain a large capacitance, the electrolyte's ions most penetrate and cover the surface of the electrode. This is only possible if the ion radius is less than the pore radius. It is known that micro pores have the least diameter. As such the ion radius must be smaller than the radius of the micro pores [12].



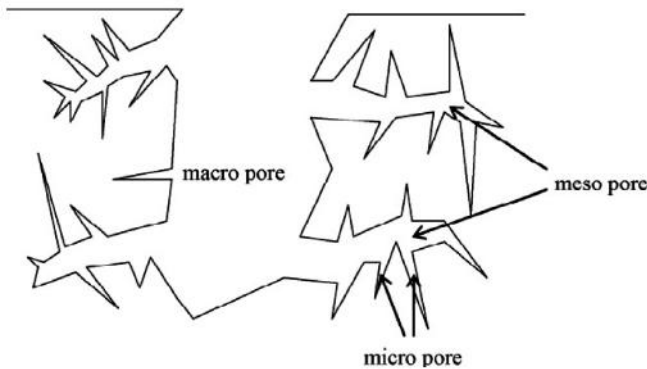Figure 3. Sample of activated carbon [13].



Figure 4. Conceptual scheme of pore of activated carbon [12].

The size of a macro pore is larger than 50nm, that of a micro pore less than 2nm and meso pores have sizes in between 2nm and 50 nm [14]. The pore is cylindrical. For the carbon scheme showed in figure 4, different sizes of cylindrical pores exist. It is called a branch pore electrode structure. In an instance where a single pore exists, the carbon electrode is called a single pore electrode [12].

The number of branches in the branch pore electrode structure is based on the number of a micro pores against a meso pore and meso pore against a micro pore. For example if there are 3 micro pores against a meso pore and 3 meso pores against a macro pore, the branch structure then resembles figure below:
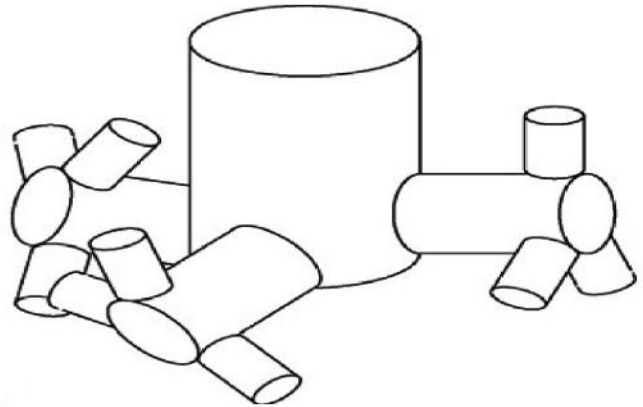


Figure 5. Branch pore Structure with 3 micro pores against a meso pore and 3 meso pores against a micro pore [12].

*Equivalent circuit model of a branch pore carbon:*
Analysis made in [12] based on theoretical formula of transmission line model impedance solved by Levie resulted in the following for the electrode model with repeated pore structure.
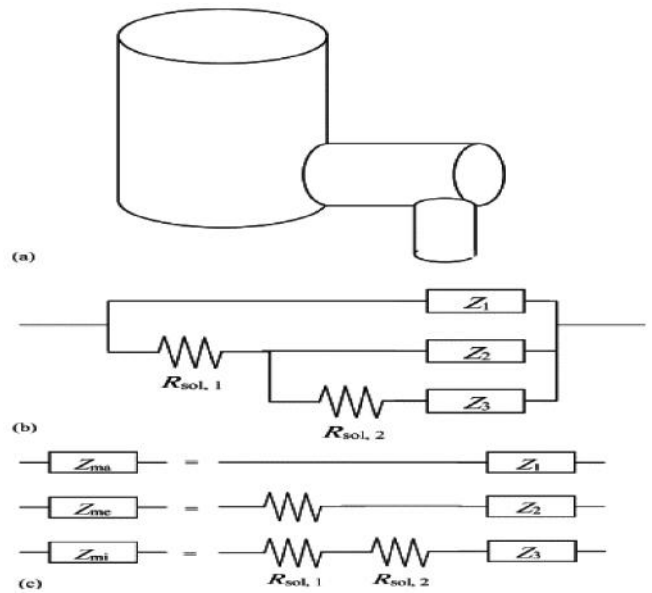


Figure 6. (a) Conceptual model of pore electrode with repeated structure, (b) equivalent circuit of pore electrode with repeated structure, and (c) simplified equivalent circuit of pore electrode with repeated structure [12].

$Z_m$ is the impedance of $m^{th}$ pore. The solution resistance of the $m^{th}$ pore designated $R_{sol,m}$ is given by:

$$R_{sol,m} = \sum_{n=1}^{N} R_{solseg,m} \tag{1}$$

The total impedance is then

$$\frac{1}{Z_{total}} = \frac{1}{Z_1} + \frac{1}{R_{sol,1} + 1/\left((1/Z_2) + \left(1/(R_{sol,2} + Z_3)\right)\right)} \tag{2}$$

$$Z_{ma} = Z_1 \tag{3}$$

$$Z_{me} = Z_2 + R_{sol,1} \tag{4}$$

$$Z_{mi} = Z_3 + R_{sol,1} + R_{sol,2} \tag{5}$$

Where $Z_{ma}$, $Z_{me}$ and $Z_{mi}$ represent the impedance for the macro pore, meso pore and micro pore respectively. In an instance where the carbon structure is composed of $b_1$ meso pores against a macro pore and $b_2$ micro pores against a meso pore, the total impedance is expressed as

$$\frac{1}{Z_{total}} = \frac{1}{Z_1} + \frac{1}{R_{sol,1} + 1/\left(b_1(1/Z_2) + \left(1/(R_{sol,2} + b_2 Z_3)\right)\right)} \tag{6}$$

In this case

$$Z_{ma} = Z_1 \tag{7}$$

$$Z_{me} = R_{sol,1 + \frac{Z_2}{b_1}} \tag{8}$$

$$Z_{mi} = R_{sol,1} + \frac{R_{sol,2} + (Z_3/b_2)}{b_1} \tag{9}$$

Figure 7. (a) Conceptual model of pore electrode with $b_1$ = 3 and $b_2$ = 3, (b) Equivalent circuit of the model and (c) Simplified equivalent circuit of the model [12].

*Types Of Activated Carbon:* Activated carbon is categorized based on source, physical form, the type of activation process and post treatment.

**I. Classification Based on Source**
Activated carbon can be from various sources such as bituminous coal, anthracite, coconut shells, wood, peat, lignite and a lot of other carbon containing materials. Activated carbon is sometimes designated or labeled with its source [15].

**II. Classification Based on Activation Process**
There are two types of activation processes
i)   Physical reactivation.
ii)  Chemical activation.

Physical reactivation is done in two stages. In the first stage, the material containing carbon is subjected to a temperature between the range of 600-900°C and then pyrolyzed under this condition. The word pyrolyzed is derived from pyrolysis meaning decomposition of organic material under heat in the absence of oxygen. This process is called carbonization. In the second stage the carbonized material is subjected to

a temperature above 250°C which usually ranges from 600-1200°C in an oxidizing atmosphere e.g. carbon dioxide, steam and oxygen. The second stage is called activation or oxidation. Activated carbon produced in this way has a fine pore structure which makes it suitable for the adsorption of liquids and vapor.

In chemical activation, the material is first influenced by a chemical which could be an acid, base or salt before being carbonized under lower ranges of temperatures typically 450-900°C. Examples of chemicals used include phosphoric acid ($P_2O_5$), zinc chloride ($ZnCl_2$), sodium hydroxide (NaOH) and potassium hydroxide (KOH). Activated carbon produced using chemical activation have larger pore structure and is more likely to adsorb large molecules. Chemical activation is mostly preferred because it involves the use of lower temperatures and is also not time consuming [13], [16].

The type of activated carbon suitable for EDLC is the one obtained by the pyrolysis of coconut shells, coal and wood in a nonoxidizing atmosphere or by pyrolysis of organic polymers [2]. KOH activated carbon nanotubes (CNTs) are also used for EDLC. It has been reported that supercapacitors produced using KOH activated CNTs produced a capacitance of $300Fg^{-1}$ with a specific capacitance of $9.9\mu F/cm^{-2}$. The activation was carried out on a highly volatile bituminous coal which resulted in a specific area of $3150m^2/g$ and a pore volume of $1.612cm^2/g$. Depending on the type of activation method used, the surface area of carbon can be increased by a factor up to 15 or more times its original surface area and since capacitance has a direct proportionality with the surface area of the electrode material used, activated carbon electrode is considered to be the best among the carbon based supercapacitors. The only thing that matters then is the carbon source and activation process [17].

**III. Classification based on physical form**
Based on physical form and particle size, activated carbon is classified as:

Powdered Activated Carbon (PAC): This type of carbon has a very small particle size. The particles are so small such that 95-100% of it will pass through a mesh sieve. It can be in the form of powder or fine granules with an average diameter ranging from 0.15 -0.25mm.

Granular Activated Carbon (GAC): This has a larger particle size when compared with PAC. Diameter ranges from 0.297-0.8mm.

**IV. Classification based on post treatment**
In this context activated carbon is classified as:
Extruded Activated carbon (EAC): This is a mixture of PAC and a binder which forms a cylindrical shaped activated carbon block with diameters ranging from 0.8-130mm.

Polymer Coated Carbon: This is obtained by coating a porous carbon with a biocompatible polymer without blocking its pores.

Impregnated Activated carbon: This is a type of porous carbon impregnated with an inorganic compound such as iodine, silver or cations of some selected metals such as Aluminum (Al), Manganese (Mn), Zinc (Zn), Iron (Fe), Lithium (Li) and Calcium (Ca) [13], [18].

## IV. SEPARATOR MATERIALS

The separator could either be cellulose based (special paper) or polypropylene in the form of self stranding films. The paper separator has the advantage of high porosity resulting in low equivalent series resistance. It has a disadvantage of poor mechanical properties resulting from low tensile and puncture strength. As a result the polypropylene film separator is commonly used due to its good mechanical properties, resistance against solvents and chemistry and low cost of production by means of biaxial stretching [19], [20]. Other separator materials include polyethylene membrane, porous glass fiber tissue or a combination of polypropylene and polyethylene [9].

## V. ELECTROLYTE MATERIALS

Electrolyte used in supercapacitors could either be organic or aqueous. Examples include sodium Chloride, hydrochloric acid, nitric acid, perchloric acid, sulphuric acid, sodium hydroxide and calcium hydroxide, acetic acid, ammonia, ligands and their associated metal ions and mercury chloride.

## VI. EQUIVALENT SERIES RESISTANCE

The capacitance of a supercapacitor is inversely proportional to the imaginary part of its impedance. The real part is called the equivalent series resistance (ESR) which is a combination of the electrode resistance, the electrolyte resistance and contact resistance. Thus the impedance is a summation of contributions in impedances of the materials that make up the supercapacitor. For example, the thickness of the separator (h) and porosity are directly proportional to its resistance as shown by equation (10) [2], [21].

$$R_{sep} = \frac{h.100}{x} \qquad (10)$$

$x$ is the conductivity of the separator.

The type of electrolyte used whether organic or aqueous also affects the impedance. Non-aqueous electrolytes have a higher resistance when compared with the aqueous type and are used to reduce decomposition and corrosion problems [22], [ 23].

The explanation for high resistance in non-aqueous electrolytes is that the resistance is dependent on the solubility of the solute of the electrolyte in its solvent which affects the concentration of dissociated ions. Organic electrolytes have a low solubility resulting in a few numbers of dissociated ions. A few number of dissociated ions results in a low conductivity leading to higher resistivity in turn. This makes the ESR of the supercapacitor to be high in circumstances where they are used. ESR is also affected by the viscosity of the electrolyte. An electrolyte that is less viscous has a higher conductance and vice versa. This is because of the relationship between conductance and the velocity of ions in the electrolyte. The higher the velocity, the higher the conductance will be [24], [25]. The diffusion of ions into the pores causes an additional resistance termed equivalent distributed (EDR) resistance which adds to ESR [24].

Electrolytes are classified as weak electrolytes and strong electrolytes depending on the rate of dissociation of ions in water (solubility). Weak electrolytes do not dissociate completely in water while the strong electrolytes dissociate completely in water. In general the strong electrolytes have higher conductivities which make their resistance to be very low. They are preferred for maximum power delivery since they have a very low internal resistance resulting in negligible internal voltage drops [22], [23]. Examples of strong electrolytes include sodium Chloride, hydrochloric acid, nitric acid, perchloric acid, sulphuric acid, sodium hydroxide and calcium hydroxide. Weak electrolytes include acetic acid, ammonia, ligands and their associated metal ions and mercury chloride [25].

## VII. PACKAGING OF SUPERCAPACITORS

When packaging a supercapacitor one has to take its size, type and use into consideration. Thin prismatic supercapacitors are used in mobile electronics such as phones cameras etc. coin-type cans are used for PC-board and so on. However, when packaging a supercapacitor the end plates must have good contact with the electrodes in order to minimize ESR. The packaging should be able to protect the electrochemical system against oxygen and water vapor in order to prevent oxidation transformations that can lead to the evolution of gas. The container used should be able to tolerate pressure generated by the electrochemical decomposition. In some cases a weak point is provided in the container for safety when there is hazard such as overcharge. This will enable the cell to open softly. The weight of the container is preferably taken to be one tenth the total weight of the supercapacitor cell.

There are mainly two types of supercapacitor packages:
1. Prismatic package.
2. Cylindrical package.

The cylindrical package comprises the coin type cell and the winded type. In the coin type cell an electrode is placed in the cell, a separator in between followed by the second electrode all of which most have been soaked in an electrolyte solution. The cell is then assembled with its cover and crimped [2], [26].

Figure 8: A coin type cell supercapacitor [27].
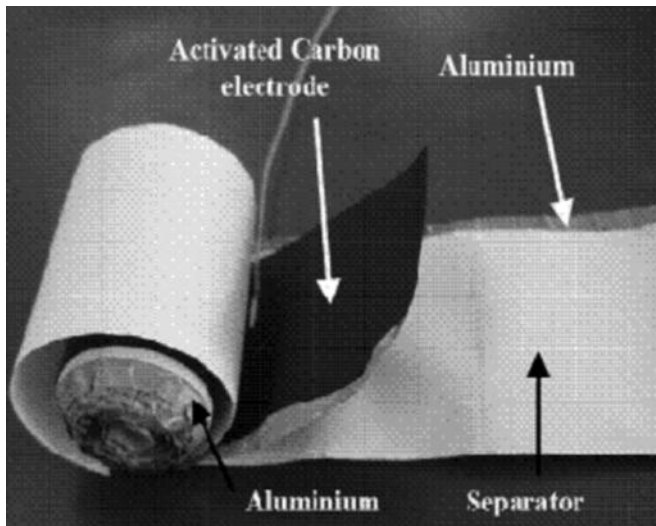
The winded type is as shown below:



Figure 9. A wound cylindrical cell supercapacitor [26].

Figure above showed the wound type with aluminum tabs inserted in between active layers. The active layers comprises of the electrodes and the separator. A good connection has to be established in between the aluminum and the protruding current collectors to minimize ESR as discussed earlier.

Figure 10 below shows a sample of a prismatic packaged supercapacitor internally composed of rectangular electrodes separated by a separator, an electrolyte and current collectors attached to each electrode which are further extended to form the external leads.
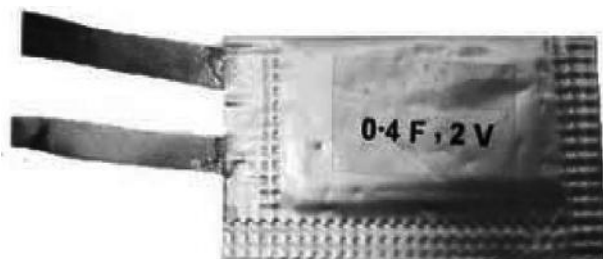


Figure 10. A prismatic supercapacitor [28].

Prismatic packaged supercapacitors have an advantage of low ESR when compared with the cylindrical type [27]. It is sometimes called the stacked type because several cells can be stacked together to form a single supercapacitor. The connection between the cells is dependent on which parameter is needed to be high. If capacitance is to be high, parallel connection is used. Series connection is used in a case where the voltage is needed to be higher. The increment in each parameter is at the expense of the other [29]. Thus several arrangements could be made to achieve a specific capacitance and voltage based on series and parallel connections which includes the use of several cells and may add to cost and size. This is an area that needs to be developed so that materials with high capacitance values could be achieved. By so doing the size of the supercapacitor will be reduced because a few numbers of cells would be required to compensate for an increased voltage rating, at the same time taking the material cost into consideration.

For testing applications in laboratories, cylindrical supercapacitors are packaged in a test cell as shown below:
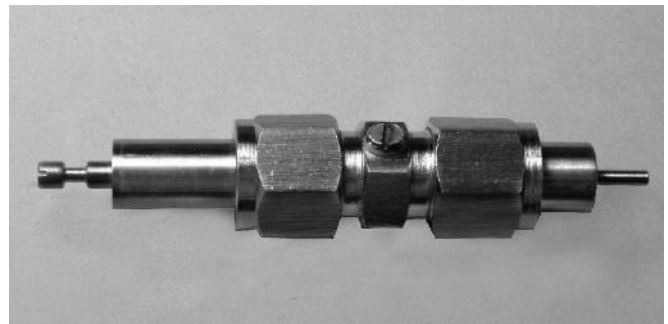


Figure 11: Sample of a supercapacitor test cell [29].

The method of assembly is the same with that of the coin cell type. The only difference is in the mechanical structure and how to assemble together. The test cell can take several other forms but still satisfying the same reason. There is also a plastic cell with metallic current collectors. It doesn't matter which type is used so far it satisfies the criteria needed in a supercapacitor cell.

VIII. CONCLUSION

The work showed the history of supercapacitors, its types and the various factors that affect the capacitance of a supercapacitor. It also shows the expected market value of supercapacitors in the coming years as well as highlighted some of the areas where it is applicable. Thus supercapacitor market is expected to continuously rise and this necessitates research and development in order to improve its performance as well as produce it at the least possible price.

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