



A Critical Review on Design and Development of New Generation Energy Storage Devices

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Abstract

A next-generation technology, the Supercapacitor, has emerged with the potential to enable significant advances in energy storage. The fundamental phenomenon and equations for conventional capacitors and supercapacitors are the same. But the improved efficacy of supercapacitors over conventional capacitors is due to the larger surface area of the electrode and thinner dielectrics. The existing review addresses a brief overview of the history, principles, and theory of operation of supercapacitors, along with various models, and significantly in current energy saving and storage. Here, a concise description of technologies and working principles of different materials utilized for supercapacitors has been provided. The main focus has been on materials like carbon-based nanomaterials, metal oxides, and conducting polymers. The performance of nanocomposites has been analyzed by parameters like energy, capacitance, power, cyclic performance, and rate capability. The use of supercapacitors in various applications is cited, and their advantages over alternative technologies are considered. Also, it comprises efficient techniques to minimize multi-response problems such as the fabrication of supercapacitors and storage capacity size, *etc.* This review embedded the current progress achieved in innovative materials for supercapacitor electrodes.

Keywords: Supercapacitor; Hybrid energy storage; Voltage unbalance; Theoretical; Materials; Models; Supercapacitor construction. Received: 17 December 2021; Revised: 29 May 2022; Accepted: 24 June 2022.

Article type: Review article.

1. Introduction

Human civilization has achieved very high development, which consistently requires an expenditure of a high amount of energy.^[1] The rising global population and the global energy crisis have led to electricity generation and consumption concerns. With the rapid development of modern science and technology, the application of electronic products is also expanding at a very high speed. In the modern period, we are

living worldly life with the help of standard, intelligent, flexible, and miniaturized electronic devices and components.^[2] These electronic devices are widely used in industries such as mobile phones, health trackers, and microsensors. At present portable devices mainly rely on micro-batteries and micro-supercapacitors as power sources.^[3] Typically, they can be integrated with miniaturized electronic devices and provide the necessary power. Owing to the rapid development of the global economy and the consumption of fossil fuels like coal and natural gas, the problem of global climate change and environmental pollution is increasing to a significant level.^[4] Traditional energy source is limited; therefore, there is always a search for renewable energy sources. Thus, the solar, wind, tidal, hydroelectric, geothermal, and gravitational energy are gaining importance day by day.^[5] Also, global pollution is an important issue that should never be ruled out. Today's world is running fast against time and depends on energy-based sources. Still, there is insufficient energy to do new experiments and fulfill essential needs. Therefore, attempts have been made to store electrical energy in different devices such as batteries, capacitors, and supercapacitors.

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Supercapacitors have been described as a capacitor that offers high storage space, more significant than capacitors with low internal resistance, which viaducts the gap between rechargeable cells and conventional capacitors. In addition to high power capacity and longevity, low weight and extensive heat range of $-40\text{ }^{\circ}\text{C}$ to $70\text{ }^{\circ}\text{C}$, ease of packaging, and affordable maintenance are the main advantages supercapacitors have over other energy storage devices.^[6] However, each of these devices has its advantages and disadvantages.^[7-9] Electrochemical supercapacitors and fuel cells are the most important inventions among energy storage systems and conversion. The electrochemical Supercapacitor is also called an ultracapacitor or electrochemical double-layer capacitor. The global economy is highly affected by energy, its production, utilization, and the environment, *i.e.*, the ecosystem. The scarcity of energy is a burning problem for developing nations. Technological innovations can provide a possible solution.^[10,11] Today's energy requirement is fulfilled mainly by using fossil fuels with limited deposition. Also, of the excess use of fossil fuels environment is seriously affected, such as stress on the environment, global warming due to the greenhouse effect, *etc.* As a result, the scientific community started worldwide activity to develop more sustainable energy conservation and use advanced technologies to reduce dependence on fossil fuels. To overcome this scenario, efficient electrical energy storage in various devices is becoming important day by day becoming important.^[8,12,13]

As far as world development is concerned, the economy is entirely dependent upon imported crude oil, and a suitable example is INDIA, which imports 80% of its crude oil from other countries. Petroleum resources worldwide are depleting at a high rate due to the significant dependence of the transportation sector on petroleum as the primary fuel. Also, there is a vast greenhouse gas emission that is disturbing the quality of air and causing too many health problems to living things (life) and the environment. Therefore, it becomes essential to focus on unconventional sources of energy such as solar and wind energy, which are pure and renewable, easy to handle and store, and cost-effective too.^[14] Also, it is essential to store thermal and electrical energy for future uses. Every nation is seriously working on the burning energy issue and seeking something next-generation solutions through different research organizations. As far as India is concerned, the thermal conversion efficiency and utilization aspects are focused on government and non-government organizations like the Bureau of Energy (BE) and Petroleum Conversion Research Association, *etc.*^[14,15] Current energy-saving concerns with electric vehicles are attracting more and more consideration because of their clean, cost-effective, and eco-friendly features. Meanwhile, the usage of electric vehicles is facing two significant problems: battery life and charging time. However, new resources have been employed to lengthen the battery life, and also exhibited the growth in storage density to protect weight and space.

Some researchers are still working on forming hybrid energy storage systems to improve battery operation conditions using new kinds of simple or nanomaterials. Nevertheless, nowadays, some fast charging patterns and related devices are established to shorten the charging time. Associated with supercapacitors are energy-storing devices with the high power density and low energy density. Batteries have very low power density and high energy density compared to supercapacitors. The Supercapacitor is a next-generation technology that assurances to show an essential part in meeting the demands of electronic devices and schemes for both the present and future. The genuinely developed technology of Supercapacitors is creating a simple path for engineers to poise their use of both energy and power. Energy storage devices like supercapacitors are routinely employed along with batteries to compensate for the inadequate battery power capability.^[16] The proper control of the energy storage arrangements indicates both a challenge and an opportunity for the power and energy management system. To provide examples with which to outline practical implementation issues, systems incorporating supercapacitors as vital components. Because of the advantages of charging efficiency, long lifetime, fast response, and wide operating temperature range, it is tempting to try and apply super capacitors to any application that requires energy storage. The limitations of the current technology must be fully appreciated, however, and it is essential to realize that supercapacitors are only valid within a finite range of energy and power requirements. The most important thing to remember about supercapacitors technology is that it is a new and different technology in its right.^[17,18]

On the other hand, electric vehicles need high power density during acceleration or starting and high energy density to travel more distance on a single charge. Electric vehicles have regained a solid global presence, and no single source can supply energy and power demands. Supercapacitors are the best and most affordable to tackle this problem because of their perfectly small size, weight, and space compared to ordinary batteries. Most recently, the supercapacitor unit can be employed in blending with the batteries in a storage system of any electric vehicle to achieve the following advantages:^[19,20]

- 1) To improve vehicle acceleration.
- 2) To improve overall drive efficiency, thereby increasing the driving range.
- 3) To reduce life cycle costs by extending the battery life.
- 4) To reduce capital costs by direct replacement of some batteries.
- 5) Higher cycle life, making them suitable for automotive applications
- 6) Ability to operate at temperatures as low as $-40\text{ }^{\circ}\text{C}$
- 7) low equivalent series resistance (ESR).
- 8) High power density due to high discharge currents.

2. Energy storage devices

According to the first law of thermodynamics, energy is constant, *i.e.*, it is conserved that the energy can neither be created nor destroyed; it can be transformed from one form into another. Also, possible to store energy in a suitable form with minimum loss. Energy storage can be categorized in the following ways:

1. Thermally Storage of Energy (TSE).
2. Electrically Storage of Energy (EST).

Thermal energy storage is carried out with the support properties associated with the latent heat of various materials, while electrical energy is the majority resourceful form of energy. Depending upon the mechanism of electrical charge storage, it is classified as:

1. Capacitors (Storage employing Electrostatic)
2. Batteries and fuel cells (Storage employing Chemicals)
3. A supercapacitor (Storage employing Electrochemical)

Electricity is an enormous and flexible form of energy that is universally used. Storage of energy in chemicals is simple and long-lasting, but electrical energy storage is too crucial.^[21,22] The chronological development in energy storage devices is as batteries, capacitors, and supercapacitors. Batteries have a large energy density, but it requires more time for the charging process. The capacitors charge up instantly, but the charge storage capacity is smaller. The bridge between batteries and capacitors is nothing but Supercapacitor (SC), which is electric power-driven in the future.^[23,24] Supercapacitors can overcome the drawbacks described above and are available in a cost-effective too. Supercapacitors are high-energy electrochemical capacitors with ample higher capacitance than conventional capacitance of the same size. This emerging technology is also known by different titles such as ultra-capacitors, electric double-layer capacitors, gold capacitors, *etc.*^[25-27] They are valuable alternatives to conventional battery storage systems as they offer a much higher power density. Compared to the battery, the storage of energy involves electrochemical reactions, the supercapacitor stores energy in electrostatic fields by ion polarization. One of the significant challenges that supercapacitors face is that their energy density is considerably low compared to batteries.^[28,29]

There are mainly energy storage systems such as conventional capacitors, supercapacitors, and batteries. The plot explaining the performance comparison of various energy-storing devices is called the Ragone plot, as shown in Fig. 1. In this plot, the power density is plotted along the Y-axis measured in Watt per kilogram (W kg^{-1}), and the energy density along the X-axis, measured in Watt-hours per kilogram (Wh kg^{-1}). Electrical energy can be stored in batteries or capacitors differently. In the case of batteries, there are two electrical terminals known as electrodes separated by a chemical substance called an electrolyte. When potential is applied between the two terminals, a chemical reaction occurs between the electrodes and electrolyte solution. As a result, the chemical nature of the substance inside the battery is altered, releasing electrical energy. The Ragone plot illustrates storage

devices' specific power density and energy density.^[30] Also, the plot represents the power densities of various energy storage devices, measuring along the vertical axis versus their energy densities along the horizontal axis. Also, it was found that the supercapacitors occupied the region between conventional capacitors and batteries.^[31]

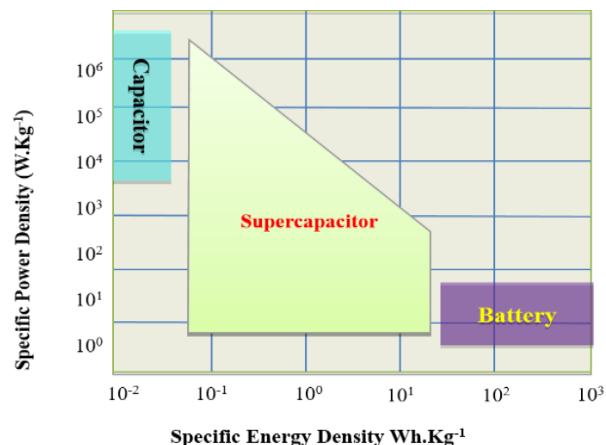


Fig. 1 Ragone plot-power density versus energy density for various electrical energy storage systems.

The specific energy density and power density are the most critical factors in determining the performance of electrochemical supercapacitors. The higher the densities, the better the device should perform. Research from around the globe employing their knowledge to increase the energy density of the supercapacitors while maintaining the power density is going on.^[31,32] Most of them focus on finding new electrode materials with a higher surface area, uniform porosity, and cost-effectiveness. Studies on pseudocapacitors, which utilize fast surface redox reactions, also gain momentum. Despite the wide variety of choices available for different materials and device architecture, electric double-layer capacitors having activated carbon or aerogel-based electrodes are the most widely used technology for transient power applications. Additionally, printed supercapacitors based on graphene and carbon nanotube-based electrode materials are famous for low-power applications.^[33,34]

Moreover, as they reversibly store charge, the number of charging-discharging cycles of a supercapacitor will be much higher than that of batteries. Currently, supercapacitors having a capacitance of thousands of Farads with a Voltage limited to 3 V are available in the market. They offer low serial resistance and are best suited for applications that demand high instantaneous power levels. Other advantages of electrochemical capacitors include extended shelf life, high efficiency, and the ability to charge and discharge without affecting the performance and lifetime entirely.^[35,36] In addition, they can work at shallow temperatures. Owing to these advantages, supercapacitors are widely employed for substituting/assisting batteries for various applications like electric vehicles, traction, microgrids, aeronautical systems, energy-harvesting systems in wireless nodes, and so on. While

appreciating all advantages of supercapacitors, the design of a Supercapacitor based system faces several challenges. Most of those challenges originate because of the porous structure of the supercapacitor electrodes, diffusion effects of the ions, and charge storage on the electrode-electrolyte double-layer interface.^[37-39]

Furthermore, Fig. 2 addresses the basic construction system of supercapacitors, which are embedded in two conductive plates separated by a small distance, and the gap is filled with dielectric materials. The nature of dielectric materials may vary, such as mica, air, oiled papers, glass, porcelain, titanate, *etc.*^[40-42] The conductive plates are generally metallic, and dielectric materials are always insulators. When an external potential difference is applied across the two plates, the charging of the capacitor takes place. Positive charges gradually accumulate on one plate during the charging process, while negative charges accumulate on another plate. When the external voltage difference is removed, both the positive and negative charges reside on their corresponding electrodes. Thus, the electrical charges are separated using capacitors. The capacitors are often better than batteries as they do not contain toxic chemicals. Their basic design prevents them from storing anything like the same amount of electrical energy as batteries^[42]

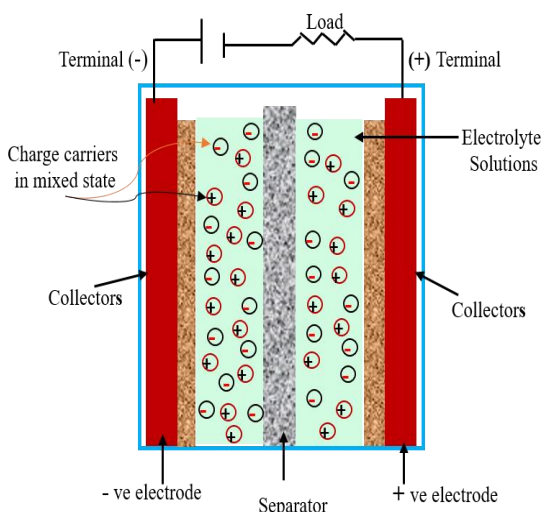


Fig. 2 General construction of Supercapacitor.

3. Construction of capacitance

The negative ions in the electrolyte experience a solid and attractive force from the positively charged electrode. As a result, negative ions move toward the positively charged electrode. Similarly, the positive ions in the electrolyte experience a solid and attractive force from the negatively charged electrode.^[43] As a result, positive ions move towards the negatively charged electrode, as shown in Fig. 2.

The Supercapacitor has the following parts, two Electrodes, two current collectors, a Separator, and Electrolyte Solution.

(a) Electrodes The electrodes of the Supercapacitor are made of Activated Carbon material. The electrodes have porous nature, which helps to store more charge carriers. Moreover,

we know that if the electrodes can store more charge carriers, the capacitance will be increased. There are two electrodes one is the positive electrode, and another is the negative electrode.

(b) Current collectors The current collectors are used to connecting the electrodes and the terminals of the capacitor. The current collectors are generally made up of foil metals. Mainly aluminum is used. There are two current collectors in the Supercapacitor, one for the positive electrode and another is the negative electrode.

(c) Separator The separator provides insulation or separates the electrodes to prevent short-circuit. The separator is mainly made up of Kapton material. The separator is very thin, like paper. The separator provides insulation between the electrodes, but it allows the flow of the charge carrier through it.

(d) Electrolyte solution The Acetonitrile or propylene carbonate as solvents are mainly used as electrolytic in the Supercapacitor. The electrolyte solution consists of the charge carriers like positive cations and negative anions. The explanation above confirmed that the Supercapacitor has the construction same as a lead-acid battery. The theory of the Supercapacitor is nearly the same as a lead-acid battery; in fact, the Supercapacitor is the combination of battery and capacitor. The Supercapacitor uses the charging property of a standard capacitor and the discharging property of the battery.

3.1 Working principle of supercapacitor

Understand the charging and discharging concepts of the Supercapacitor.

3.2 Charging of process

We already know that the positive and negative Ions are present in the electrolyte solution mixed in. So at first, when we apply a voltage across the electrodes of the Supercapacitor, then it starts charging. After that, the electrodes start to attract the ions of opposite polarity. That means the positive electrode attracts the negative Ions or charges, and the negative electrode attracts the positive Ions or charges. As a result, the positive ions or charges create a layer near the negative electrode, and the negative ions create a layer near the positive electrode. As the electrical charge carriers form these two layers, it is called an Electrical Double Layer Capacitor (EDLC). The process is illustrated in Fig. 3a.

3.3 Discharging process When we connect a load across the Supercapacitor, the electrodes cannot attract the ions, and the ions start distributing through the electrolyte solutions and go to the mixed state. The discharging of the Supercapacitor is shown in Fig. 3b.

Generally, the Supercapacitor is distinguished from an ordinary capacitor mainly in two ways: 1) Because of the porous nature of plates, SC has a much larger surface area, and 2) The distance between two plates is of the order in angstrom units which is much smaller, as compared to the separation between two electrodes in an ordinary capacitor.^[44] Due to the

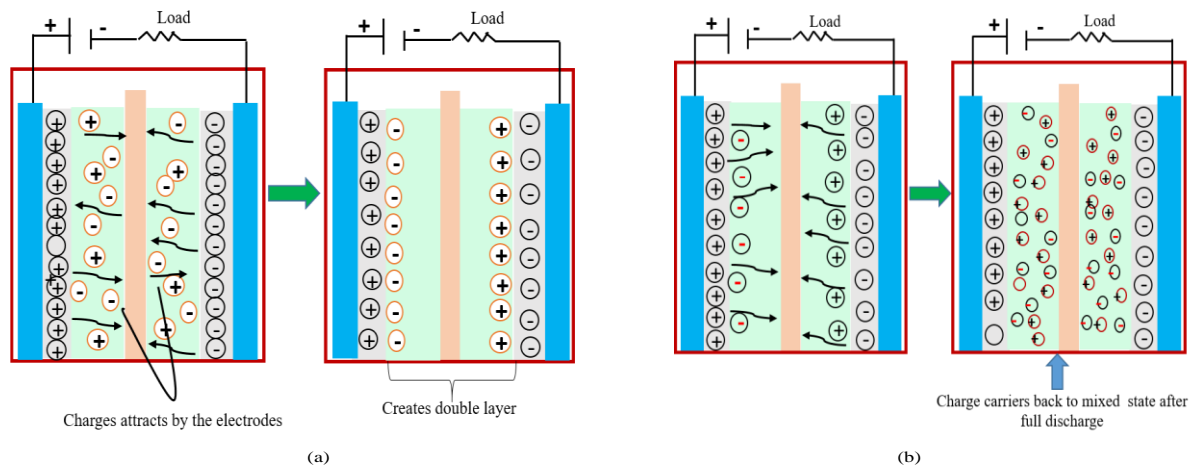


Fig. 3 (a) The charge process of the Supercapacitor, (b) the Discharge process of the Supercapacitor.

the large surface area obtained by coating powders, activated charcoal enhances the storage capacity. In an ordinary capacitor, the plates are separated by a comparatively thick insulator whose behavior can be modified in an electric field called a dielectric, e.g., mica (a ceramic), a thin plastic film, or even simply air. When we charge a capacitor, positive charges induce one plate and negative charges on the other, forming an electric field. Because of this, field polarization of the dielectric occurs, so its molecules align in the opposite direction in the applied field and reduce its strength which is responsible for the storage of charge on the plates in minimum Voltage.^[43,44]

The energy density and power density of the energy storage device can be calculated by the following equations:

$$E = \frac{1}{2} CV^2 \tag{1}$$

$$P = \frac{V^2}{4 \times ESR} \tag{2}$$

where, E - Energy density, P - Power density, C- Specific capacitance, V- Operating potential window, ESR-Equivalent series resistance. According to above Equations 1 and 2, it is seen that the energy density from the capacitor to the battery increases significantly, whereas the power density decreases (See Fig. 1). The conventional capacitor exhibits a high power density, while the battery and fuel cell shows a higher energy density. Supercapacitor bridges the gap between conventional capacitor and battery with intermediate power density and energy density.^[45,46]

4. Glimpses on the development of supercapacitors

The capacitor effect was discovered in 1745, consisting of two foil conductors separated by a dielectric region. Such a type of capacitor provides minimal energy, which is less than 360 joules per kilogram in energy density and with a unit of microfarad only. Conventional capacitors consist of two conducting electrodes separated by an insulating dielectric material. When a voltage is applied to a capacitor, an opposite charge accumulates on the surface of each electrode.^[47,48] The capacitance can be defined as below Equations 3:

$$C = A/t \tag{3}$$

The conventional capacitor C is directly proportional to the surface area A of each electrode, and t is the thickness of the electrical double layer. The storage of electrical charge at the interface between a metal and an electrolytic solution has been studied by chemists since the nineteenth century. However, for the first time, the practical use of double-layer capacitors only began in 1957, when a patent was placed by General Electric (GE) for an electrolytic capacitor using porous carbon electrodes. This patent admits its limitations that ‘it is not positively known precisely what takes place when the devices are used as energy-storing devices.’^[49] However, it was believed that carbon pores are responsible for energy storage, and the capacitor exhibited exceptionally high capacitance. In 1966, Standard Oil Company, Cleveland, Ohio, had patented a device that stored energy in the double-layer interface. Thus, it was assumed that the double layer at the interface behaves like a capacitor of relatively high specific capacity. Then, Standard Oil of Ohio (SOHIO) registered a patented device disc-shaped capacitor in which a carbon paste was soaked in an electrolyte.^[50]

SOHIO handed over the technology to NEC in the subsequent year, and this company finally marketed the device as supercapacitors. Later on, NEC started developing double-layer capacitors under the name of Supercapacitors. These devices could be operated at low Voltage and had high internal resistance and, therefore, could be used in memory backup applications.^[51] Later on, several companies developed an interest in fabricating supercapacitors, such as ELNA. ELNA had started to produce its Supercapacitor under the name of Dynacap. The standard oil company of Ohio or Sohio was an American oil company. Later on, this company was acquired by British Petroleum, founded by John D Rockefeller.^[50]

SOHIO acknowledged that "the 'double-layer' at the interface behaves like a relatively high specific capacity capacitor." SOHIO went on to patent a disc-shaped capacitor in 1970 utilizing a carbon paste soaked in an electrolyte. The capacitance of a device is mainly dependent on the characteristic of the electrode materials, particularly the surface area and pore size distribution. The electrolytic

stability usually determines the operating Voltage of supercapacitors. The electrolytes can be of two types, viz aqueous and non-aqueous. The aqueous electrolytes have the advantage of high ionic conductivity and low manufacturing costs. However, they suffer from specific problems such as relatively low decomposition voltages. Non-aqueous electrolytes allow cell operating Voltage above 2.5 V.^[50,51]

4.1 Need for supercapacitor

There is an ever-increasing pollution threat and vanishing oil layer deposition, which requires searching for supercapacitors as an alternative high-power source. In addition, there is tremendous growth in portable electronic devices such as laptops, cameras, *etc.* Thus, for high-power applications, supercapacitors are essentially required. Therefore, supercapacitors are claimed as an emerging technology that promises to play an essential role in meeting the requirements of electronic devices and systems. The scientific community is aware that supercapacitors are becoming attractive energy storage devices. Supercapacitors fill the gap between batteries and conventional capacitors. The superiority of supercapacitors over other energy storage devices is that they have higher power density and longer life cycles than batteries. Energy storage for portable electronic devices, which is becoming increasingly important to the present society, forms the largest mobile energy storage market today and is experiencing innovation for future applications such as flexible/printed electronics and displays. Flexible energy storage is required to operate flexible devices. Electrochemical supercapacitors with high energy and power capabilities can power flexible devices. In order to be integrated with flexible electronics, supercapacitors should be made flexible with sheet-like structures, which are light weight.^[52] The various designs and power needs of soft portable electronic equipment, such as roll-up displays, electric paper, and wearable systems for personal multimedia, require the development of flexible energy devices. Flexible supercapacitors have played an increasingly important role in power source applications since they combine the advantages of the high power of conventional capacitors and the high specific energy of batteries.^[53]

4.2 Electrolytes

The use of electrolytes as dielectric was initially developed by Charles Pollak in 1886. However, electrolytic capacitors have faced numerous difficulties in applications due to their low reliability. The capacitance of an EDLC is greatly influenced by the choice of electrolytes. The ability to store charge is dependent on the accessibility of the ions to the porous surface area, so ion size and pore size must be optimized. Therefore, the pore size distribution of the electrode is an essential factor. The pore size distribution of the electrode depends upon the size of the ions in the electrolyte.^[52,53] Therefore, the proper choice of electrode and electrolyte both are essential factors.

The breakdown voltage of electrolytes governs the

attainable cell voltage of a supercapacitor. Therefore, the energy density will be limited by the electrolytes. The power density is dependent on ESR, which ultimately depends upon the nature of electrolytes. The common electrolytes used can be broadly classified into two types (1) aqueous electrolytes and (2) organic electrolytes. Organic electrolytes are more prevalent in commercial devices due to their higher dissociation voltages. The cells using an organic electrolyte can usually achieve Voltage in the range of 2-2.5 V.

On the other hand, non-aqueous electrolytes suffer from disadvantages such as high cost, low conductivity compared with aqueous electrolytes, and low dielectric constant, resulting in smaller capacitance and complex purification procedures. Therefore, a large number of non-aqueous electrolytes were tested. In the current scenario, ionic liquid has attracted scientists' attention. Ionic liquids are suitable electrolytes because of their excellent chemical and physical properties, such as high thermal stability and high electrochemical stability.^[54]

5. Classification of supercapacitor

Based on the charge storage mechanism concept, supercapacitors can be classified into three types: 1) Electrochemical double-layer capacitors (EDLCs), 2) Pseudocapacitors, and 3) Hybrid types formed by a combination of EDLC and pseudocapacitors. Each capacitor class is recognized by its unique mechanism of charge storage.^[55] In the early period, the concept of an electrochemical Supercapacitor was based on an electrical double layer existing at the interface between a condenser and its containing electrolyte solution. Electrostatic and electrolytic capacitors are considered first and second-generation capacitors. With the rapid development in materials, the third generation, known as Supercapacitor, is developed. Fig. 4 addresses the classification of Supercapacitors. SC is mainly classified into two types based on the charge storage mechanism: electric double-layer capacitors (EDLCs) and pseudocapacitors.^[56] The details information on different kinds of SC based on different storage mechanism are given below:

5.1 Electrochemical double-layer capacitors (EDLCs)

Von Helmholtz first introduced the concept of EDLCs in the 19th century. The Helmholtz double-layer model has two layers of opposite charge formed at the electrode-electrolyte interface separated by an atomic distance. This model is almost similar to that of the two-plate capacitor.^[57] The Helmholtz EDLCs model was further modified by Gouy and Chapman,^[58] considering a continuous distribution of electrolyte ions in the electrolyte solution, driven by the thermal motion, which was referred to as the diffuse layer (as shown in Fig. 5). Later, Stern combined the Helmholtz model with the Gouy-Chapman model to explicitly recognize two regions of ion distribution—the inner region and the diffuse layer. In the inner layer, ions are strongly adsorbed to the electrode. The specifically adsorbed ions and non-specifically

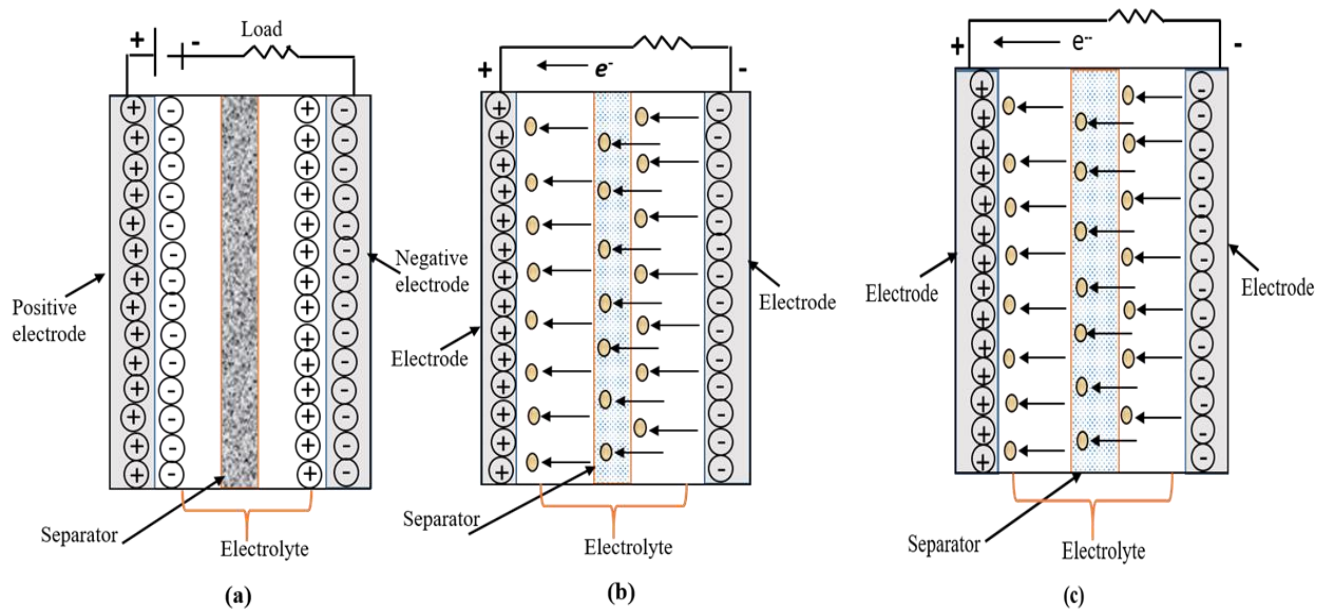


Fig. 4 Schematic representation of supercapacitor types: (a) EDLC type; (b) Pseudocapacitor type; (c) Hybrid capacitor type.

adsorbed counterions are distinguished as the inner Helmholtz plane (IHP) and the outer Helmholtz plane (OHP). The capacitance of two separated arrays of charges is directly proportional to the surface area of the electrode and inversely proportional to the distance between the two electrodes.^[59] The capacitance calculation for an EDL supercapacitor is similar to that of a parallel-plate capacitor, which can be defined as below **Equations 4**:

$$C = \frac{\epsilon_0 \epsilon_r A}{d} \tag{4}$$

where ϵ_r is the dielectric constant of the electrolyte, ϵ_0 is the permittivity of vacuum, A is the surface area of the electrode accessible to the electrolyte ions, and d is the effective thickness of the EDL (*i.e.*, the Debye length). Furthermore, the energy storage and release mechanism are based on nanoscale charge separation at the electrochemical interface formed

between an electrode and an electrolyte. The charge storage mechanism is nonfaradaic, and no chemical oxidation-reduction (redox) reactions are involved. EDLCs have relatively long cycle lives because only physical charge transferring occurs.^[60]

5.2 Pseudocapacitors

In the case of pseudocapacitors, the capacitance arises for thermodynamics reasons and mainly due to charge acceptance (ΔQ) and a change in potential (ΔV).^[61] The derivative $C = d(\Delta Q)/(d\Delta V)$ corresponds to a capacitance called pseudocapacitance.^[62] The primary difference between pseudocapacitance and EDL capacitance is that pseudocapacitance is faradic, involving redox reactions between the active material on the electrode and the electrolyte.^[63] The electrochemical characteristics of

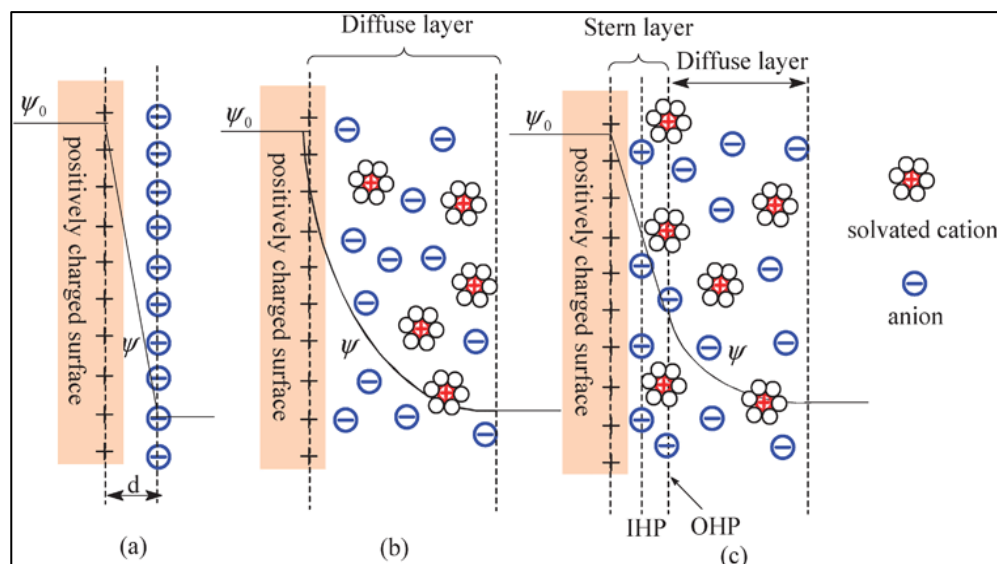


Fig. 5 Models of the electrical double layer at a positively charged surface: (a) the Helmholtz model, (b) the Gouy–Chapman model, and (c) the Stern model, showing the IHP and OHP.

pseudocapacitive materials are neither entirely capacitive nor bulk faradaic processes. Energy storage devices involving pseudocapacitive materials exist between EDLCs and batteries, which predominantly depend on the surface faradaic electron transfer by the intercalation or adsorption of charge-compensating ions.^[64] Pseudocapacitor can be considered a complementary form of EDLC since it is not purely electrostatic but exhibits similar electrochemical profiles (cyclic voltammetry and galvanostatic charge-discharge). The term "pseudo" is used to distinguish it from EDLCs. The surface or near-surface charge interactions leads to a surface faradaic reaction that exhibits swift reaction kinetics similar to EDLCs.^[65] As a result, pseudocapacitive electrodes have near rectangular cyclic voltammograms and almost linear charge-discharge curves. In recent years, the line between battery-like material and pseudocapacitive material has become very thin. To properly understand the charge storage mechanisms of the electrode material, *i.e.*, to accurately differentiate between the pseudocapacitive and battery materials, quantification of the kinetics is one of the most effective techniques.

5.3 Hybrid supercapacitors

Hybrid supercapacitors: are the third type of supercapacitors that consolidates the features of both EDLCs and pseudocapacitors. The electrodes of hybrid supercapacitors are made of composite materials that include EDLC materials and pseudocapacitive materials or can have asymmetric electrodes, one pseudocapacitive electrode, and another EDLC electrode.

On the other hand, the oxidation and reduction processes

simultaneously occur in supercapacitors. The process is based on the Nernst equation. The term redox is formed by 'Red,' which signifies reduction, and 'oxi,' which signifies oxidation. The Nernst equation that can calculate the emf of the cell is given below [Equations 5](#).

$$E = E^{\circ} - \frac{2.303RT}{nF} \log \frac{[Red]}{[Oxi]} \quad (5)$$

where, E° = the standard potential; R = universal gas constant; T = absolute temperature; F = Faraday constant; n = ion charge (moles of electrons)

Therefore, the amount of charge from the above equation is a function of potential E. The charge-discharge curve is the result of overlapping redox reactions.^[20] Adsorption of ions: The deposition of ions to form a monolayer on the electrode substrate is a reversible process that results in Faradic charge transfer and hence gives rise to pseudocapacitance in a similar manner as shown in redox reactions.^[66]

6. Materials for supercapacitor

For the construction of supercapacitors, mainly two types of materials are used such as (1) carbon-related materials such as carbon nanotubes (CNTs), graphene, and activated carbon, and (2) Based use of metal oxides such as Ruthenium, Manganese, and Copper, Cobalt, Nickel oxides and polymers like polyaniline (PANI) and polypyrrole (PPy), *etc.* The details of electrode materials related to their types are presented in [Fig. 6](#).

The other categories of supercapacitor materials are composites obtained by combining two or more constituent or

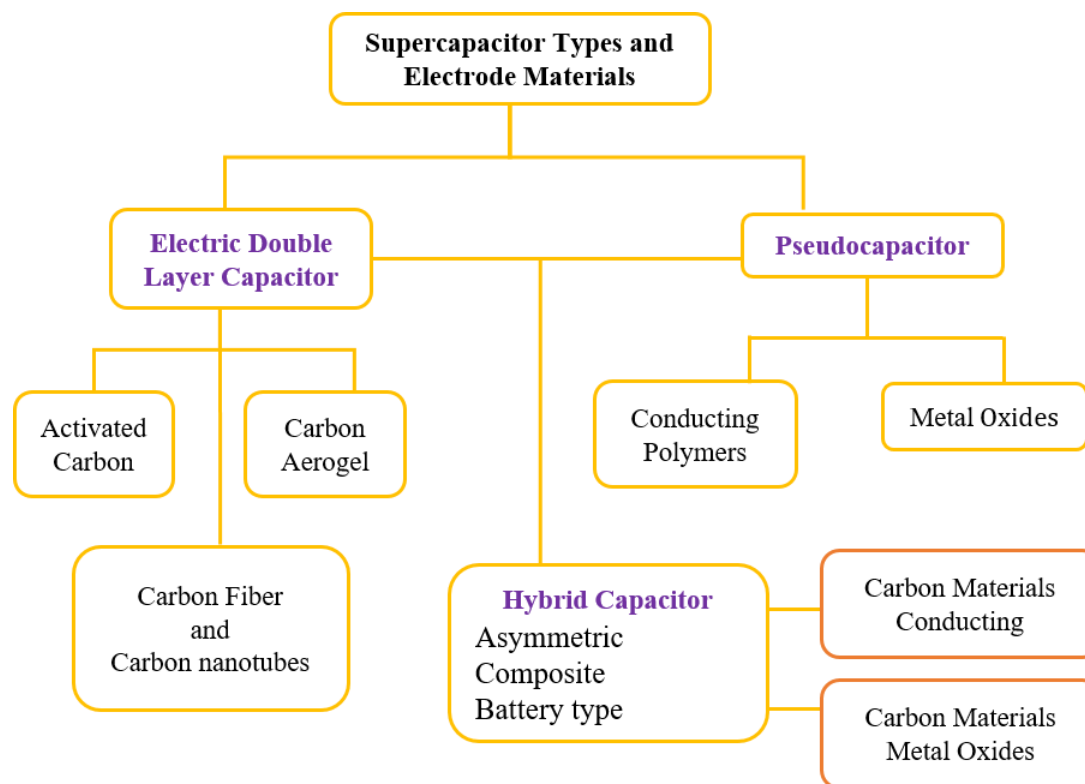


Fig. 6 Details of electrode materials based upon types of Supercapacitors.

nanomaterials. An example is a composite formed by mixing CNTs with conducting polymer or metal oxides. Combining CNTs with nanomaterials allows for a substantial increase in surface area and enhanced capacitive performance. As a result, the synthesis and engagement of nano-oriented resources are now recognized as a critical factor in developing the high performance of supercapacitor devices.^[67]

6.1 Carbon-based materials

Carbon is the 15th most abundant element in the earth's crust and fourth. It is a reactive non-metal. In nature, we observe several allotropes of carbon, such as diamond, graphite, lonsdaleite, Buckminsterfullerene, amorphous carbon, and carbon nanotubes. Carbon nanotubes are tube-shaped materials made up of carbon having a diameter measuring on the nanometer scale. Carbon nanotubes are formed from graphite sheets, and graphitic Nanotubes are a member of the fullerene structure family. Their name is derived from their long, hollow structure with the walls formed by one-atom-thick sheets of carbon called graphene. Carbon nanotubes were discovered by Japanese scientist Iijima in 1991 and are now considered a top-class subject in academic research and various industrial areas.^[68] The chronological developments in carbon nanotubes can be summarized in [Table 1](#).

Table 1. Evolution of Carbon Nanotubes and their Applications in Supercapacitors.

Year	Discovery
1952	Rudushkevich and Lukyanovich published a paper in the Soviet Journal of Physical Chemistry showing hollow graphite carbon fibers 50 nm in diameter. ^[69]
1979	John Abrahamson presented evidence of carbon nanotubes at the 14 th Biennial conference of carbon at Pennsylvania state university. ^[70]
1981	A group of Soviet scientists published the results of chemical and structural characterizations of carbon nanoparticles produced by a thermodynamical disproportionation of carbon monoxide. ^[71]
1991	Nanotubes were discovered in the soot of the discharge at NEC by Japanese researcher Sumio Iijima. ^[72]

These nanomaterials are allotropes of carbon, made of graphite, and constructed in cylindrical tubes with nanometer-scale in diameter and several millimeters in length. The carbon nanotubes have been constructed with a length-to-diameter ratio of 132,000,000:1. A carbon nanotube structure is formed by a layer of carbon atoms bonded together in a hexagonal mesh. This one-atom thick layer of carbon is called graphene, and it is wrapped in the shape of a cylindrical structure. Their impressive structural, mechanical, and electronic properties are due to their small size and mass, incredible mechanical strength, and high electrical and thermal conductivity. The

strength of the carbon-carbon bond gives rise to extreme interest in the mechanical properties of carbon nanotubes. Theoretically, carbon nanotubes should be stiffer and more robust than any known substance. Thus, carbon nanotubes have a higher tensile strength than steel and Kevlar. The extra strength in carbon nanotube comes from sp^2 bonds between the individual carbon atoms. Here the bonds are even more potent than the bonds observed in diamonds. Carbon nanotubes are strong, but they are also elastic. It is possible to press on the tip of a nanotube and cause it to bend without damaging the nanotubes. Once the deforming force is removed, the nanotube will regain its original dimensions.

The strength of nanotubes can be weakened by creating a defect in them. The defects can be generated from atomic vacancies or the rearrangement of carbon bonds. On the other hand, all nanotubes are expected to be excellent thermal conductors. It is expected that carbon nanotubes will be able to transmit up to 6000 watts per meter per kelvin at room temperature. Furthermore, graphene and carbon nanotubes are carbon-derived nanomaterials that have received significant attention in their potential application as efficient electrode materials in the design of supercapacitors owing to their high mechanical properties with great specific surface area and, most importantly, competent electrical properties. Other forms of carbon-based nanomaterials like xerogel, carbon fibers, activated carbon, and templet carbon have been used to design and develop supercapacitors. They also serve as the Supercapacitor's electrode. The carbon-based materials are comparatively cheap, readily available, and very common with characteristic permeability, which enables easy penetration of electrolytes in the electrode to boost the capacitance of the supercapacitors.^[73]

In order to enhance the performance of the supercapacitors, different forms of carbon materials used as electrodes for electric double-layer capacitors (EDLCs) are activated carbons, carbon aerogels, carbon fibers, and carbon nanotubes. Maximum-specific capacitance values of different materials are shown below in [Fig. 7](#).^[74]

6.1.1 Carbon nanotubes (CNTs)

Carbon nanotubes have excellent nano porosity properties 1D form of carbon, permitting minute spaces for the polymer to stand in the tube and act as a dielectric. Carbon nanotubes can store about the same charge as charcoal (almost pure carbon) per unit of surface area. However, nanotubes can be arranged in a regular pattern that exposes a better suitable surface area. Carbon nanotubes can be considered cylinders made of graphite sheets, mostly closed at the end. Thus, one considers CNTs the folding of graphite sheets, just like one rolls a piece of paper into cylindrical forms. These nanostructured materials were accidentally discovered by Iijima while observing under Transmission Electron Microscopes.^[75]

Moreover, electrodes of CNTs have been made as an intertwined material of CNTs, with an open and obtainable

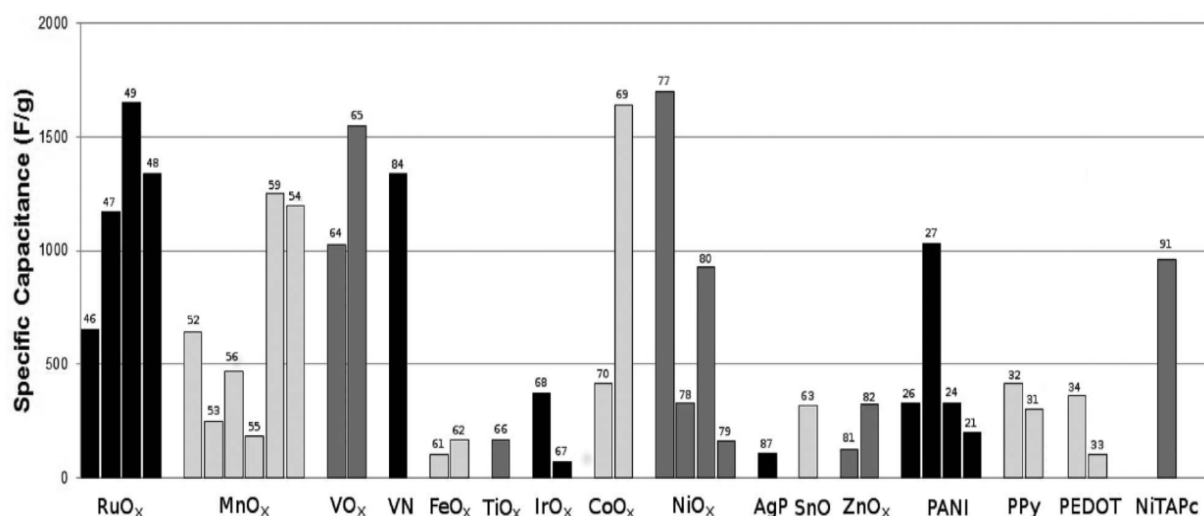


Fig. 7 Maximum specific capacitance values reported from the addition of different supercapacitive materials. Reproduced with permission from [74], Copyright 2016 American Chemical Society.

network of mesopores. The mesopores in CNTs are interconnected, forming a continuous distribution, thus developing the specific surface area more efficiently.^[76] CNT electrode has lower equivalent series resistance (ESR) than activated carbons, as the electrolyte ions can simply enter into the mesoporous network.^[77] Carbon nanotubes act as pillars for the uniform distribution of metal oxides, creating high pseudocapacitance and electric double-layer capacitance. A similar electrode type accomplishes higher specific capacitance (C_s) than specific carbon, metal oxide, or conducting polymer electrodes.^[78,79] Recently, PEDOT/PSS and CNTs composite materials have been developed^[80] and that could reach C_s varying from 85 to 150 Fg^{-1} , while the energy density (E_d) could exceed 0.92 Whkg^{-1} and P_d could range from 100 to 3000 Wkg^{-1} . Similarly, the same path has been approached to fabricate the $\text{Co}_3\text{O}_4/\text{CNTs}$, and $\text{Co}_3\text{O}_4/\text{CNFs}$ with enhanced electrical conductivity and a substantial surface area.^[81,82]

In addition, the carbon nanotubes are classified as (1) Single-walled carbon nanotubes (SWCNTs) and (2) Multiple walled carbon nanotubes (MWCNTs), respectively.

(a) Single-walled carbon nanotubes (SWCNTs)

Single-walled nanotubes are close to one nanometer and can be considerable enough. The structure of SWCNTs can be theorized by enfolding a one-atom-thick layer of graphite into a cylinder. SWCNTs are an essential variety of carbon nanotubes because most of the properties change significantly with the indices, and this dependence is non-monoatomic. Current research developments recommend growing attention to CNTs as an EDLC electrode material. Electrodes made from this material developed commonly as an entangled of CNTs, with an open and accessible network of mesopores. Unlike other carbon-based electrodes, the mesopores in CNT electrodes are interconnected, allowing a continuous charge distribution that uses almost all the available surface area.

Thus, the surface area is utilized more efficiently to achieve capacitances comparable to those in activated-carbon-based SCs, even though CNT electrodes have a modest surface area than activated carbon electrodes. The surface area is utilized more effectively to achieve capacitance comparable with activated carbon-based super capacitance. Due to the porous nature of carbon, the electrolyte ions can diffuse into the mesoporous network.^[83] Based on the nature of SWCNTs, some work has been done like freestanding PEDOT-PSS/SWCNTs, which exhibit the C_s of 104 at 0.2 Ag^{-1} , energy density (E_d) of 7 Wh kg^{-1} , power density (P_d) of 825 W kg^{-1} and 90% of the C_s retention after 1000 cycles^[84] shown C_s of 1715 Fg^{-1} for RuO_2 based SCs in which RuO_2 is electrodeposited on the SWCNTs film electrode. Also, they have fabricated superior-performance, self-standing composite electrodes with SWCNTs.^[85] The 3-D mesoporous SWCNT-based electrodes allowed unimpaired ionic carriage in thick films and brought enhanced results in an ac line frequency of 120 Hz. Measurements of $601 \mu\text{Fcm}^{-2}$ with a -81° phase angle and a time constant of 199 μs were obtained. As-fabricated electrodes were capable of cycling at higher than 200 V s^{-1} showing a parallelepiped cyclic voltammetry (CV) shape at one kV s^{-1} .

(b) Multiple walled carbon nanotubes (MWCNTs).

MWCNTs have mesopores in structure, which agree with the easy movement of ions across the electrode-electrolyte interface. Suppose the pore size becomes equivalent to the size of the ion, and the molecules of the solvent are moderately showing outcomes in the form of high ionic packing density. Nevertheless, their mechanical stability drops due to significant volume change because of repetitive intercalation and depletion.^[86] Some reported data shows that a C_s of 50 Fg^{-1} for MWCNTs and HPNCTs with willow catkins made by an easy carbonization process and exhibit SSA of 1775.7 $\text{m}^2 \text{g}^{-1}$, C_s of 292 Fg^{-1} at 1 Ag^{-1} and better rate capability with 83.5%

of the Cs retention at 10 Ag-1 for HPNCT-800.^[87] Applying asymmetric polymer electrolyte with NaOH-treated polyethylene oxide (PEO-NaOH) was prepared to produce a thermally-induced voltage in SCs. They found that Au and MWCNTs deposited on Au electrodes showed the thermopotential of 10 mV K⁻¹, the Cs of 1.03 mFcm⁻², and E_d of 1.35 mJcm⁻² at the temperature difference of 4.5 K.^[86] PANI nanowires have been deposition within the MWCNTs employing in situ electro-polymerization. It was found that aligned MWCNTs supported the organic polymers with a pathway for charge transfer. The films made with conducting polymers encapsulated in MWCNTs showed a Cs of 296 F g⁻¹ at 1.6 A g⁻¹. Also, found that MWCNT channels bound the structural changes in PANI chains while charging-discharging and increased the lifespan of the structure.^[88]

On the other hand, Different Π -conjugated sulfonate patterns and additional support of graphene and MWCNTs are used to investigate the polymerization activities of PEDOT.^[86] As fabricated PEDOT: MWCNT composite exposes an interconnected network due to the Π - Π interaction of PEDOT with non-covalent functionalized MWCNT and shows a specific capacitance (C_s) of 199 F g⁻¹ at 0.5 A g⁻¹. Meanwhile, the high-efficiency SCs with upgraded electrochemical characteristics can be made by chemical vapor deposition (CVD) and graphene hybridized with MWCNTs.^[89] Another work was done followed by CVD to prepare MWCNTs, and graphitic layers over MgO assisted metal catalyst, which designates that such materials are responsible for fast charge transport in the cell or SCs device.^[88]

6.1.2 Activated carbons

Due to its eco-friendly nature, less expensive, and high surface area, activated carbon (AC) has been the desired choice of electrode material for EDLC compared to other forms of carbon. Conventionally, AC was primarily obtained from coal, petroleum, and derived products.^[90] However, due to environmental pollution and the finite reserve of fossil fuels, academics are considering biomass waste as a possible resource for the fabrication of AC.^[91] Activated charcoal is a powder made up of tiny and very "rough" particles and a low-density heap with many holes that looks like a sponge. The surface area of even a thin layer of such a material is many times greater than a material like aluminum, letting many more charge carriers be deposited in any given volume. However, charcoal is not a better insulator; it still replaces the excellent insulators used in conventional devices, so in general, EDLCs can only use low potentials on the order of 2 to 3 V. Concerning the surface area of AC, the capacitance is directly proportional to the surface area, and calculation reveals that not all the high surface area contributes to the capacitance of the device. This discrepancy may be due to electrolyte ions that are very big that they cannot diffuse into smaller micropores. Large pore size correlates with higher power densities, and smaller pore correlates with higher energy densities.^[92] The specific capacitance values are directly

proportional to the surface area and published articles^[93-95] confirmed similar capacitive behavior of activated carbon. As of 2010, virtually all commercial supercapacitors use powdered activated carbon developed from coconut shells, and higher-performance devices are made based on synthetic carbon precursors activated with potassium hydroxide.^[96]

Taking account of activated carbon fiber material (ACFM), the composite of (ACFM)-Ni(OH)₂ has been reported and displayed the Cs of 370-380 Fg⁻¹. Also, the dependence of shape, structure, and volume of material on the composition and concentration of the solution was established.^[97] An asymmetric SC (with 1.9 V) was developed using AC as the -ve electrode and a silicon carbide-MnO₂ (SiC-NMnO₂) composite as the +ve electrode in Na₂SO₄ electrolyte solution.^[98,99] Also, porous activated carbon was extracted from rotten carrots at different values of activating temperature under an inert atmosphere, employing a chemical activation method and ZnCl₂ as an activation agent. Further, the electrochemical performance of synthesized AC was studied as an electrode in the aqueous, organic, and ionic liquid-based electrolyte. It was confirmed that the prepared electrode showed the highest specific capacitance in the aqueous electrolyte and the highest specific energy along with specific power in the ionic liquid-based electrolyte.^[100]

6.1.3 Carbon aerogels

The complete artificial ultralight solid, extracted from gel substance by supercritical extraction without a need for binder material, is called aerogel—the supercritical extraction outcomes in the formation of enormously low-density materials and low thermal conductivity. The electrodes of aerogel supercapacitors are a composite material usually made of non-woven paper made from carbon fibers and coated with organic aerogel, which undergoes pyrolysis. The carbon fibers provide structural integrity, and the aerogel provides the required large surface area. Carbon aerogels are nanoporous carbon consisting of a weighty class of carbon nanomaterials and three-dimensional hierarchical morphology.^[101] Due to these properties, carbon aerogels can be used as electrode material for EDLCs. Carbon aerogels are designed from the continuous net of carbon nanoparticles with a mesoporous structure with no binding agent. Usually, lower ESR was found in the binderless electrode of carbon aerogels. It was found that reduced ESR, which yields higher power, is of curiosity for investigators working in the SCs area.^[102] The small aerogel SCs are being used as backup electricity storage in microelectronics. However, Aerogel capacitors can only work at a few volts. At higher voltages, ionize the carbon and damage the capacitor. According to a literature survey, carbon aerogel capacitors have achieved 325 J/g (90 Wh/kg) energy density and 20 W/g power density. Some aerogel electrodes manufactured by pyrolysis of resorcinol-formaldehyde aerogels are better conductors than activated carbons. They provide thin and firm electrodes representing mechanical and vibrational constancy for SCs for their application in a high-

vibration environment. Recently, Ultrahigh-specific capacitance for supercapacitors has been achieved by nickel cobaltite/carbon aerogel composites.^[103]

6.1.4 Graphene

Graphene is nothing but a single layer of graphite material or crystal. The graphite crystal has a stack of carbon layers lightly bound with each other, and each layer embedded a hexagonally arranged carbon atoms. As a sole isolated unit, these layers are self-standing materials identified as graphene and widely act as superconducting hybrid materials. The expansion in capacitance depends on the high surface area, lattice density, and interlayer spacing of graphene. Also, combining graphene with other materials can bring many property alternations in super capacitance. This state that graphene has great potential as a supercapacitor electrode.^[104] Graphene-based ac line-filters on a large scale have been developed by Wu *et al.*^[105] Here, graphene oxide (GO) is reduced by patterned metal interdigitates and engaged as the electrode material. A quasi-solid state micro-SC with cellular graphene films as an active material with PVA/H₃PO₄ as the gel electrolyte shows as a new type of flexible energy storage device. Here 3D graphene films served as high-performance SC electrodes and an ion reservoir for the gel electrolyte. The 3D cellular graphene films are generally fabricated by a freeze-casting assisted filtration assembly method and show great mechanical strength and flexibility. The quasi-solid state device retained about 89% of its initial capacitance after 1000 cycles while tested under repeated bending from 0 to 120.^[106] Furthermore, Photoswitchable micro-SC based on the diaryl ethene-graphene film was prepared by Liu *et al.*^[107] to exhibit the C_s modulation up to 20% demonstrating photo switchable micro-supercapacitors. Meanwhile, Zhang *et al.*^[108] fabricated graphene-based Ni foam electrode, which shows a higher E_d and P_d and good cycling performance. Xu *et al.*^[109] have synthesized a nanocomposite of graphene/AC/PPy by vacuum filtration method.^[109] Gao^[110] developed a graphene/ polymer electrode on Ni foam using the vacuum pressure method. The distribution of graphene could not be tuned by applied vacuum pressure and its duration. In the end, Ramaprabhu has developed poly (phenylenediamine) (PpPD) and hydrogen exfoliated graphene (HEG) sheets which show a C_s of 248 F g⁻¹ at 2 A g⁻¹.^[111]

6.2 Conducting polymers (CPs)

The conducting polymers are pseudocapacitors with poor lifecycles compared with carbon-based materials. Conducting polymer has become a vital electrode material for pseudocapacitors due to the advantage of enormous capacity, good conductivity, facile synthesis, and cost-effectiveness. In recent years conducting polymers has attracted the attention of researchers due to their great potential in supercapacitors. Despite these properties, conducting polymers have not become so popular among researchers and industrialists due to drawbacks such as low stability and poor mechanical

properties, which restrict their applications in the fabrication of supercapacitor devices—conducting polymers, which are more correctly identified as intrinsically conducting polymers. CPs have attained considerable attention as they supply high specific capacitance (due to their redox behavior), rapid charge-discharge process, lesser cost than carbon-based material, and a low ESR value. Notably, the n/p type polymer configuration has enormous potential for high E_d and P_d.^[97] However, the dearth of proficient n-doped conducting material and less cycling stability has postponed the improvement of CPs pseudocapacitor. Conducting polymers are organic polymers that can conduct electricity. These compounds have metallic conductivity or may sometimes act as semiconductors, a desirable property for SCs electrode materials. Again, it was confirmed that the mechanical stress on the conducting polymers during the reduction-oxidation reaction bounds the stability of pseudocapacitors through many charge-discharge cycles. Basis of electrical conductivity: the conductivity of organic polymers is owed to many processes. Polymer-like polyethylene, the valence electrons are in sp³ hybridized, and sigma bonding electrons have low mobility and do not contribute to the electrical conductivity of the materials. The critical feature of bond structure available in conductive polymers can be easily calculated with the tight-binding model.^[112]

On the other hand, conducting PANI polymer is lightweight, highly conductive, mechanically flexible, eco-friendly, and highly theoretical capacitance. The problem is that, because of ion doping/dedoping, PANI shrinks and swells during the charge/discharge process. To reduce this problem, the PANI is coated on metal oxides (MOs)/carbon materials forming PANI/MOs/ nanocarbon ternary hybrid, which holds good cyclic stability and specific capacitance.^[97] Also, PANI obtained a wide range of colors due to their many protonation and oxidation forms. These electrochromic characteristics can be utilized for the fabrication of electrochromic SCs.

Moreover, Polyacetylene is the most crystalline CP, but it is easily oxidized in air. On the other hand, Polypyrrole (PPy) and polythiophene can be produced directly in doped form and are very stable.^[113] PPy has superior density and higher flexibility than other conducting polymers. It has high electrical conductivity (10-500 Scm⁻¹), and it undergoes a self-rapid redox reaction for charge storage.^[113] Composite like Lignin-PPy is synthesized by coating lignin with PPy by the polymerization of PPy with and without the presence of methyl orange, which conceded to the formation of PPy films of globular and nanotubular morphology. Stable PPy films with a surprising doping degree have been achieved by the pulse polymerization process.^[114] Pulse on time controls the chain size along with chain defects, and pulse off time controls the direction and conjugation of the polymer chain. A C_s of 400 F g⁻¹ has been visualized in these films with E_d of 250 Wh kg⁻¹ at 5 mA cm⁻² current density and long cycle life.

Another conducting polymer of Polythiophenes (PTs) was obtained by a chemical oxidative method using FeCl₃ as an

oxidant and the absence of numerous surfactants. It is found that surfactants change the PTs morphology, and PTs with TritonX-100 show a C_s of 117 F g^{-1} , whereas the C_s for surfactant-free PTs are 78 F g^{-1} .^[115] Polyindole (Pind) has gathered attention due to the mixed properties of both poly (p-phenylene) and PPy, such as high redox properties, good thermal stability, meager degradation, and better air stability compared to PPy and PANI.^[116] PANI-Solution (nanocrystal), PANI-emulsion (nano pedal), and PANI- interfacial (nanosphere) are reported.^[117] Furthermore, symmetric SCs based on alternate layers of different CPs prepared to develop SCs have been studied.^[118] The capacitive properties of a prepared multilayered system are better than individual CP, which may be related to the better porosity of multilayered material. Flexible worm-like SC electrodes are fabricated using cellulose nanofibres (CNFs) and graphite nanoplatelets (GNP), doped with PANI by in situ polymerizations.^[119] Also, an all-solid-state symmetric SC has been fabricated using PANI/CNF (20% loading)/ GNP electrodes, which shows good C_s retention at various bending angles. There are numerous good properties of conducting polymers, such as flexibility, conductivity, ease of synthesis, financial viability, and high pseudocapacitance. Conducting polymer materials such as polythiophene, polypyrrole and polyaniline have received significant attention in the potential supercapacitor applications. Despite these promising properties, polymer-

based supercapacitors exhibit poor cycling stability and low power density and energy density.

6.3 Transition metal oxide

Transition metal oxides (TMOs) of ruthenium, iron, vanadium, manganese, tin, and copper are highly used in research and applications of pseudocapacitances. Transition metal oxides exhibit multiple oxidation states at specific potentials, and the selection of materials with multiple stable states within an electrolyte's potential window allows maximum capacitance to develop. Another essential advantage of crystalline metal oxide is that its high conductivities allow charge propagation along with the lattice structures of thin surface layers. However, considerably lower conductivity is observed for amorphous hydrous metal oxides. The conductivity can be increased by crystallization. The process of crystallization needs the removal of water which eliminates pore spaces.^[120] For the super capacitance applications, firstly, ruthenium oxide was used. However, ruthenium is a rare earth mineral and a highly toxic metal. Also, ruthenium is very costly. Therefore, there was a need to search for alternative metal oxide that is less toxic and cost-effective. Thus, different transition metal oxides were tried, such as manganese oxide, vanadium oxide, *etc.* Fig. 8 depicts how a double layer is developed on the CP chain.

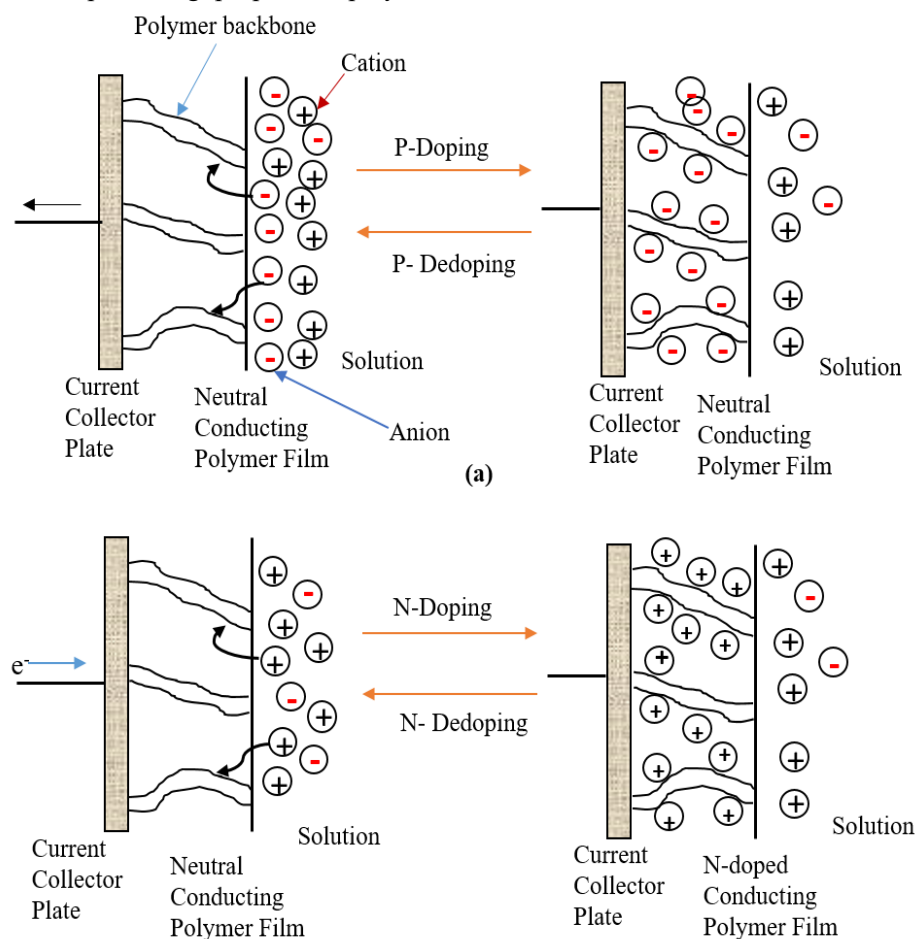


Fig. 8 (a) and (b) Development of double layer at CPs chain.

Here an attempt has been made to synthesize copper oxide nanoparticles dopping with different materials. Copper oxides exist in two stable forms, cuprous oxide Cu_2O and cupric oxide CuO . These two oxides have very different colors, crystal structures, and physical properties. The differences in such properties are due to differences in the oxidation state of transition metal copper. For example, Cu_2O is reddish in color, a cubic semiconductor, and displays interesting excitonic properties.

On the other hand, CuO is having different color-it is a dark, iron-gray color, and crystallizes in a more complicated monoclinic tenorite structure exhibiting interesting antiferromagnetic ordering.^[121] CuO is an intrinsic p-type semiconductor as some of the oxygen ions are lost from the crystals, which impart a positive charge on the entire crystalline structure. The capacitance of copper oxide is controlled by the structure, composition, type, and density of charge carriers. Metal oxides have been explored as an electrode material for SC due to their high conductivity, various oxidation states, and the possibility of constructing high-energy, high-power SCs.^[122] The majority of relevant research concerns ruthenium oxide. This is because other metal oxides have yet to obtain equivalent capacitances. The metal oxides like ruthenium oxide (RuO_2), nickel oxide (NiO), cobalt oxide (Co_3O_4), iridium oxide (IrO_2), and manganese oxide (MnO_2) have been extensively studied as electrodes for pseudocapacitors. The specific capacitance of RuO_2 is achieved through the insertion and extraction, or intercalation, of protons into its amorphous structure. In its hydrous form, the capacitance exceeds that of carbon-based and conducting polymer materials. Furthermore, the ESR of hydrous RuO_2 is lower than other electrode materials. As a result, ruthenium oxide pseudocapacitors may achieve higher energy and power densities than similar EDLCs and conducting polymer pseudocapacitors. Thus, a significant area of research is the development of fabrication methods and composite materials to replace an alternative electrode material to reduce its cost without reducing its performance. However, the prohibitive cost of ruthenium oxide makes it impractical.^[123]

Transition metal oxides/hydroxides are considered the best candidates as electrode materials for energy conversion and storage applications. In recent years, nanostructured materials have achieved great interest due to their large surface-to-volume ratio and exceptional mechanical, electrical, and optical properties endowed by confining the material dimensions. Some materials that are not stable in the bulk form and do not exist in the ambient atmosphere may exist in a more stable state at the nanoscale. Specially transition metal oxide (TMO) nanoparticles have received wide attention for energy applications.^[124] The extensive benefits of TMO nanoparticles are: (i) structural changes that allow the altering of lattice symmetry and cell parameters, (ii) a change in electrochemical behavior due to the quantum confinement effect, and (iii) a change in surface properties leading to a drastic increase in the bandgap that influences the conductivity and the chemical

activity of the nanoparticles (Immanuel *et al.*, 2019). Fabrication of desired quality of PDCU 38/180. Higher pseudocapacitance than the EDLC carbon electrode materials. It is well established that properties of nanomaterials, like composition, crystalline phase, structural and morphological properties, and the surface and interface properties between the electrode and electrolyte, greatly influence the electrochemical performance in device applications.^[125]

Different nanostructured metal oxides and hydroxides include RuO_2 , IrO_2 , MnO_2 , Co_3O_4 , NiO , $\text{Co}(\text{OH})_2$, and $\text{Ni}(\text{OH})_2$.^[126] TiO_2 , ZnO , WO_3 , SnO_2 , CuO , Cu_2O , Fe_2O_3 , and $\text{RuO}_2 \cdot \text{H}_2\text{O}$ show excellent electrochemical performance not only in electrochemical energy conversion, storage, sensors, and detector applications but also in terms of good electrical conductivity, UV absorption, photo-oxidizing capacity against chemical and biological species and antimicrobial/self-sterilization^[127]. Not restricted to energy applications, many metal oxide nanoparticles have been successfully employed in different biomedical applications such as efficient electrochemical detectors of biomolecules, including NiO , ZnO , MnO_2 , Fe_2O_3 , TiO_2 , and Co_3O_4 .^[128] Metal oxide nanoparticles such as MgO ,^[129] Al_2O_3 ,^[130] and ZnO ^[131] are also employed to decontaminate various nerve agents.^[132]

Copper oxide is one such transition metal oxide with exciting properties, which was also one of the first substances known to behave as a semiconductor. The study started in the 1920s and '30s. However, the interest in copper oxides was received after the 1970s. Man has used copper since prehistoric times; in fact, one of the significant stages of human history is named after a copper alloy, *i.e.*, Bronze Age. Humans have used it for about 10,000 years.^[133] As per records worldwide, copper resources are around 5.8 trillion pounds (out of which only 12% have been mined), and also, it is highly recyclable.^[133] It is a soft, flexible, ductile, and pliable metal that conducts heat and electricity very effectively, commonly making it suitable for electrical wiring. It is in group 11 of the periodic table; has one s-orbital electron on top of a filled d-shell. Copper oxide is a binary compound and has two PDCU 40/180 stable forms Copper(I) oxide (Cu_2O) and Copper(II) oxide (CuO). The physical properties, *i.e.*, color, crystal structure, and electrical and optical properties of Cu_2O and CuO , vary significantly. Intrinsically, CuO and Cu_2O are p-type semiconductors having band gap energies in the visible and near-infrared regions of 1.2 to 1.5 eV and 2.1 to 2.6 eV, respectively.^[134] Both are non-toxic and have abundant source material on earth. They have favorable bandgap and band edge positions for solar energy conversion applications, and the bandgap could be easily adjusted by altering the composition. They have a high absorption coefficient in the visible range and can be prepared at a low cost by adopting simple methods. There are several reasons for choosing Cu_2O and CuO as the absorber layers for solar cells, gas sensors, biosensors, magnetic media, enzyme-free glucose sensors, photodetectors, nanofluids for enhanced thermal conductivity, junction devices such as a p-n diode,^[135] the catalyst for water gas shift

reactions, cathode material in photoelectrochemical water splitting applications, heterogeneous catalysis in the complete conversion of hydrocarbons into carbon dioxide, nano-energetic materials, and as active materials in lithium-ion batteries (LIBs) owing to its peculiar properties.^[136,137] Copper oxide nanostructures also exhibit some exclusive magnetic and superhydrophobic properties. Along with the specific optical, electrical, and electrochemical properties, copper oxides have some general advantages, as environment friendly and abundantly available. However, copper oxides had not gained enough attention from researchers until recent years.^[138]

Thus, developing a reliable and cost-effective method for the large-scale production of Cu₂O and CuO is highly desirable.^[139] Manifold vacuum and non-vacuum techniques have been explored so far for the synthesis of copper oxides, including different nanostructures such as activated reactive evaporation, electrodeposition, spray pyrolysis, radical oxidation with nitrogen plasma treatment, sol-gel dip technique, reactive magnetron sputtering, oxidation of metallic Cu sheets, photochemical deposition, and thermal oxidation, wet chemical method, anodization, thermal transformation, electrospinning, solvothermal process, chemical bath deposition, and electrodeposition.^[136] Several studies on copper oxide-based systems have explored their efficacy in various applications. Based on the literature survey (few mentioned and some not mentioned due to scope of limitation), attempts have been made to develop copper oxide photocathodes and electrodes for photoelectrochemical cell (PEC) and supercapacitor applications, respectively, since nanostructured materials have a distinct feature compared to their bulk counterparts exhibiting enhanced properties which can be efficiently utilized for the PEC and supercapacitors.

Mahendra *et al.*^[140] have reported that the CuO thin films can be formed of 3 different thicknesses, *i.e.*, 20nm, 100nm, and 200 nm were deposited on glass substrates by magnetron sputtering techniques. They studied their structural, optical, and electrical properties. The study of nanostructured CuO thin film on stainless steel substrate is successfully done by Dubal *et al.* Here; they have synthesized the nanomaterials by simply changing the complexing agent. The result reveals maximum specific capacitance at 5mV s⁻¹. They also studied the long-term cycling performance of CuO is examined at different scan rates. Prasad *et al.*^[141] reported that the CuO thin film was synthesized by sol-gel route with carbon embedded. The specific capacitance of 300 Fg⁻¹ was reported. Shaikh *et al.*^[142] reported that they synthesized the hybrid CuO–PAA thin films using a facile spin coating technique at room temperature for supercapacitor application.

The coated films are annealed at different temperatures to study the annealing effect on Csp. They found that the Csp increases from 41 to 136 F g⁻¹ with an increase in annealing temperature. They also revealed that as annealing temperature increases with an increase in porosity. Patake *et al.*^[143] reported that the amorphous and porous copper oxide thin film electrodes were prepared by cathodic electrodeposition for

supercapacitor application. The cyclic voltammogram was obtained at 20 mV s⁻¹ scan rate of Csp is 36F g⁻¹ in 1M Na₂SO₄ electrolyte. The amorphous nature of oxide plays a vital role in improving Supercapacitors. The capacitance of the copper oxide electrode increases with an increase in film thickness. Dubal *et al.*^[144] reported that the chemical bath deposition (CBD) method prepared the complex hierarchical CuO nanostructures. The structures of CuO thin films are of woolen clumps, stacked nanosheets, and nanobuds fabricated on a stainless steel substrate. They also found high specific capacitance of nanobuds CuO sample is 396 F g⁻¹.

Shinde *et al.*^[145] synthesized nanoflowers-like CuO/Cu(OH)₂ hybrid thin films on stainless steel substrate by using simple and inexpensive successive ionic layer adsorption and reaction (SILAR) method. The CuO/Cu(OH)₂ nanoflower with diameters of about 4-5µm exhibits the maximum specific capacitance of 459 F g⁻¹. Hsu *et al.*^[146] prepared CuO/Cu(OH)₂ hybrid material using a liquid-solid reaction route, and Csp is found to be 278 Fg⁻¹. Shaikh *et al.*^[147] reported that nanostructured Ru-doped CuO thin films by colloidal solution method via a simple and cost-effective spin coating technique. They observed the highest Csp of 406 F g⁻¹. Wang *et al.*^[148] reported that CuO nanosheets with near-vertical orientation on nickel foam substrate are composed of interconnected nanoparticles by a template-free growth method. As a result, the CuO exhibits the highest Csp of 569 F g⁻¹. Dar *et al.*^[149] reported that Graphene nanosheets (GN) disseminate with macroporous copper oxide (macroCuO).

This was investigated on a glassy carbon electrode (GCE) as an electrode material synthesized by a modified Sol-Gel route for supercapacitor application. They reported a maximum specific capacitance of 417 F g⁻¹. Pendashteh *et al.*^[150] stated that the CuO nanoparticles had been synthesized on graphene oxide (GO) nanosheets by the electrostatic coprecipitation method. The composite CuO–GO material shows a better specific capacitance of 245 F g⁻¹ at a current density of 0.1 A g⁻¹. Dubal *et al.*^[144] reported that the different nanostructures of CuO thin films were directly grown onto stainless steel substrates with the help of surfactants such as Triton X-100 and polyvinyl alcohol (PVA) prepared by CBD. It shows the maximum specific capacitance of 411 F g⁻¹. Finally, Huang *et al.*^[151] reported that the electrode of Ag-doped CuO. Table 2 depicts the various types of materials used in the development of supercapacitors and their morphology and Csp.

Table 2. List of materials with their morphologies and Csp.

Material	Morphology	Csp
(in Fg-1)		
CuO		
CuO	micro-woolen	346
CuO	rodlike morphology	300
hybrid CuO–PAA	Island like structure	136
CuO	well-adherent and porous	

cauliflower structure	36				
CuO	Nanobuds	396	MnO ₂ / 2M KCl/ Pt	202 as-deposited charge-discharge method	
CuO/Cu(OH) ₂ hybrid (diameters-4-5 μm)	Nano flower			Thickness 1.0-1.2 mg cm ⁻²	
CuO in [HPMIM][Cl] IL	459 nanobuds with a bunch of nano pedals-like		MnO ₂ / 2M KCl/ Pt	MnO ₂ deposited at 0.5V/SCE	
Structure	60		MnO ₂ /0.5LiCl/Pt	45 Mf charge-discharge high stability	
CuO/Cu(OH) ₂ hybrid	Lotus like morphology	278	MnO ₂ /1.0 M Na ₂ SO ₄ /Pt	Area 1 cm ² with loading 0.5 mg	
Ru doped CuO	Nanocrystal structure	406	MnO ₂ / 1.0M Na ₂ SO ₄ /Pt	Thickness of 0.94 micrometer	
CuO multilayer nanosheets	43. Hierarchical		RuO ₂ hydrothermal		400 Fg ⁻¹
Hybride Cu-PAA	Circle-like structures	65	ZnO nanomembrane	Atomic layer deposition	846 Fg ⁻¹
CuO-PAA/CNT films	Circle-like structures	258			
CuO	Nanosheets	569			
GN/macroCuO	macro-structured porous	417			
CuO-GO	globular shaped nanoparticles	245			
CuO	Nanosheets arrays as well as flower-like				
Cluster	411				
NSA-Ag/CuO	Nanosheets nanowires and nanosheets and flower-like	689			
CuO	212				
Structure	Nanowire	305			
CuS	Nanoporous particles	136.3			
Cu ₂ O/CuO/RGO	unique particle-nanosheet structured	340			
CuO/N-RGO	Thickness				
MnO ₂ /NaCl/Pt	0.02mg/cm ⁻²				
MnO ₂ /2KCl/Pt	Specific conductance 212, 21%				
21% higher than plain Mn oxide					
MnO ₂ -MWCNT/3M KCl/Pt	Specific capacitance 160, Sc decreased				
from 160 to 80 Fg					
MnO ₂ /0.1M Na ₂ SO ₄ /Pt	230, potential window: 1V (for galvanostatic methods)				
MnO ₂ /0.1M Na ₂ SO ₄ /Pt	Thickness 0.4-0.5 mg cm ⁻²				

Nanosheet arrays were prepared by doping CuO nanosheet arrays using Ag by the silver mirror reaction. The specific capacitance of 689 F g⁻¹ at 1Ag-1 and 299 F g⁻¹ at 10 A g⁻¹, respectively. Yunhu Li *et al.*^[152] revealed that nanostructured CuO nanowires and nanosheets and flower-like different morphology grown on copper foam substrate were prepared by thermal treatments for supercapacitors application. CuO nanosheets exhibited higher specific capacitance than nanowires, with a flower-like nanostructure of 212 F g⁻¹. Yu-Kuei Hsu *et al.*^[153] revealed that hierarchical structures of wire-like CuS deposited on copper foil substrate using simple and cost-effective liquid-solid reactions. The high specific capacitance of CuS and NW nanoelectrodes is 305 F g⁻¹. Wang *et al.*^[154] reported that Cu₂O/CuO/RGO nanocomposite was synthesized using the hydrothermal method on RGO nanosheets for supercapacitor application. The nanocomposite exhibited supercapacitor performance of 173.4 F g⁻¹ at the current density of 1 Ag⁻¹ and remained at 136.3 F g⁻¹ at 10 A g⁻¹ with a retention of 78.6%.

Moreover, the capacity remains 133.8 F g⁻¹, almost unchanged (98.2%) after 100,000 cycles at 10 A g⁻¹. Li *et al.*^[155] revealed that the CuO/nitrogen-doped reduced graphene oxide (CuO/N-RGO) composites are prepared by refluxing in ammonia solution and low-temperature annealing. The CuO/N-RGO composite shows a high specific capacitance of 340 F g⁻¹.

7. Applications of supercapacitors^[156-161]

Supercapacitors have several applications, including:

1) Transportation: The transportation system requires the use of supercapacitors. The automobiles require supercapacitors with a compilation of other devices such as batteries. The fuel efficiency can be increased by storing the energy at braking and releasing the energy at acceleration. Also, the supercapacitors can perform well in a cold atmosphere at shallow temperatures of about – 40 °C temperature.

2) Consumer electronics: Supercapacitors are widely used in various electronic devices as backup energy sources for system memories, microcomputers, system boards, clocks, and toys. These are ideal for devices that require a quick charge. In addition, supercapacitors are cost-effective as they have an incredibly long life, and usually, their replacement is not required during the lifetime of the device they are used.

3) Uninterrupted power supply: UPS requires a continuous power supply. It is possible to maintain the continuous power supply of UPS by using supercapacitors. Also, supercapacitors are best suited to provide power for startup and peak-up buffering.

4) Other Application: Supercapacitors can be used in hospitals, cell phone towers, banking centers, transmission lines, in system frequency and stability control; supercapacitors are suited as an energy storage system for hardening sensitive equipment against voltage sag, in wind turbine systems supercapacitors can provide a simple, highly reliable solution, airport control towers. They are also used in many portable gadgets and devices.

5) Medical Applications of Supercapacitors: The internal organ of a human sometimes do not function properly, and even with the use of medicine, they can not be cured; in such a situation, transplantation is one possibility. However, transplantation of every organ is not possible, and even it is very costly. Therefore, some suitable devices are often planted into living organisms to perform the function of dysfunctional body parts. However, most of these devices require a power supply to function appropriately. The batteries can be used as a power supply source, but they have a life span that will never be long enough, especially if they are implanted. Thus, the device needs to be brought out whenever the batteries discharge, and the batteries need to be changed. Here, introducing a supercapacitor to the batteries or replacing the power is possible.

Biosupercapacitor comprises graphene layered with modified human protein as an electrode, a conductor through which electricity from the energy harvester can leave. The platform could eventually also be used to develop next-generation implantable devices to speed up bone growth, promotes healing, or stimulates the brain.

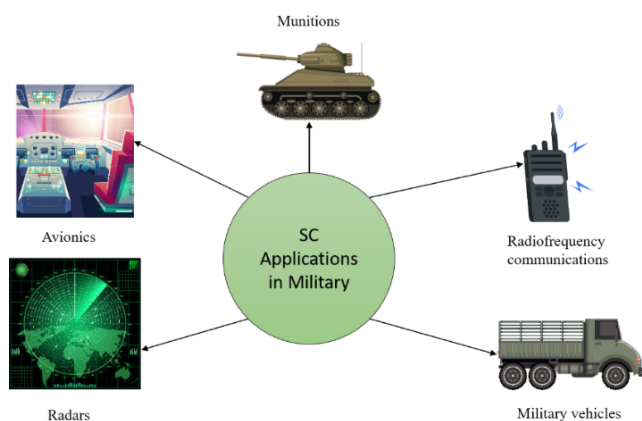


Fig. 9 Applications of Supercapacitors.

The biological Supercapacitor that the researchers invented uses charged particles of ions from bodily fluids to produce power. It uses electrolytes or ions present in body fluids such as blood, urine, and other extracellular biological fluids. It is possible that the supercapacitors can work with other devices known as energy harvesters. The biomedical and other types of applications of supercapacitors are depicted in Fig. 9.

8. Advantages of a supercapacitor:

1. Very high efficiency.
2. Fast-charging property
3. A supercapacitor has a longer life than a 10 to 15 years battery
4. The Supercapacitor can withstand temperatures between -30 to 65 degrees centigrade, whereas a battery can withstand -10 to 40 degrees centigrade.
5. The Supercapacitor is very light in weight.
6. Supercapacitors can be installed in a short area as they are significantly smaller.
7. The Supercapacitor has a swift transient response.

Acknowledgment

The authors pay sincere tribute to the Late Ms. Deepika Rai Dhirendra Prasad who suddenly left this world and lived a very short span of life, all the authors remember her on this occasion and pray to Almighty God for peace of her holy soul. Her sweet memories are in the heart of authors.

Conflict of Interest

There is no conflict of interest.

Supporting Information

Not applicable.

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