International Journal of Research in Engineering and Science (IJRES)
ISSN (Online): 2320-9364, ISSN (Print): 2320-9356
www.ijres.org Volume 10 Issue 10 ǁ October 2022 ǁ PP. 57-66

0 Dimensional Carbon-Based Nanomaterials for the Electrodes of Electrochemical Double Layer Capacitor: A Review

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Abstract
Since the discovery of carbonaceous materials, they have gained the spotlight in the research ramp owing to their numerous applications especially in the realm of electrochemical energy storage devices like supercapacitors. A lot of research has been dedicated for the enhancement of supercapacitor devices and its types. 0 dimensional (0D), 1D, 2D and 3D Carbonaceous materials mainly used in the electrodes of Electrochemical Double Layer capacitor (EDLC) have drawn much recognition. A brief understanding of working principle of EDLC has been discussed in this paper. Moreover, a short review on zero dimensional (0D) carbonaceous materials like Carbon Quantum Dots (CQDs), Activated Carbon (AC), Mesoporous Carbon, Carbon Nanospheres (CNSs) and Carbon Nano-onions (CNOs) has been presented in this paper. The versatility and advanced properties of 0D carbonaceous materials make them able for utilizing them in the fabrication of EDLC electrodes and have an ample capacity for further investigation and research.

Keywords: *Carbonaceous materials, EDLC,Carbon Quantum Dots (CQDs), Activated Carbon (AC), Mesoporous Carbon, Carbon Nanospheres (CNSs), Carbon Nano-onions (CNOs)*

Date of Submission: 20-09-2022 Date of acceptance: 04-10-2022

I. INTRODUCTION

Electrical energy has become a fundamental need of modern-day. The economic development of every country relies upon the electrical energy. From a common household to industries and factories that work using the complex heavy machineries all require electricity and other forms of energy. The world cannot function even for a day if power runs out. With every fleeting second the world burns inexplicable amount of fuels. Consequently, the global society is falling into a never-ending dark pit of energy crises. With such a problem at hand scientists are developing scores of energy storage electrochemical devices that majorly include supercapacitors, fuel cells and batteries that support sustainability of energy. Batteries and supercapacitors are at top in electrochemical industry among the other various energy storage technologies. Ultracapacitors or supercapacitors are amongst the best-known energy storage devices that work on the absorption and desorption of ions on the interface (EDLCs) or redox reactions (Pseudocapacitors) or both (Hybrid supercapacitors).

Batteries, on the other hand, store energy in chemical form through chemical bonds existing inside the active electrode where conversion of chemical energy into functioning electrical energy occurs as a consequence of redox reactions like in lead acid batteries or by intercalation phenomena for example in lithium ion batteries. Batteries and supercapacitors are principal electrochemical devices as shown in Ragone plot [1] in Fig 1 representing the correspondence between specific energy also known as energy density E_d having units $Whkg^{-1}$ and specific power also called power density P_d having units Wkg^{-1}. Batteries possess higher energy density in comparison to supercapacitors due to which they are in great demand in electronic industries. However, during high power operation there is a huge resistive loss owing to slow transport of ions and electrons resulting into thermal energy generation. Such problem leads to many safety issues. Regarding this, supercapacitors are popular in present era regarding energy storage technology on account of their unique electrochemical properties like larger capacitance, higher specific energy, higher power density, prolonged cycle life and swift charge-discharge feature [2, 3].

Undoubtedly, countless reviews on nano materials used for supercapacitor electrodes have been published in last decade. The electrode material plays a significant role in overall electrochemical performance of a supercapacitor so 0 dimensional carbonaceous nanomaterials that are being used for the fabrication of

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EDLC electrodes, will be in the spotlight of this review. In addition to this, we hereby present a brief understanding for EDLC devices and their performance based upon the electrode materials.

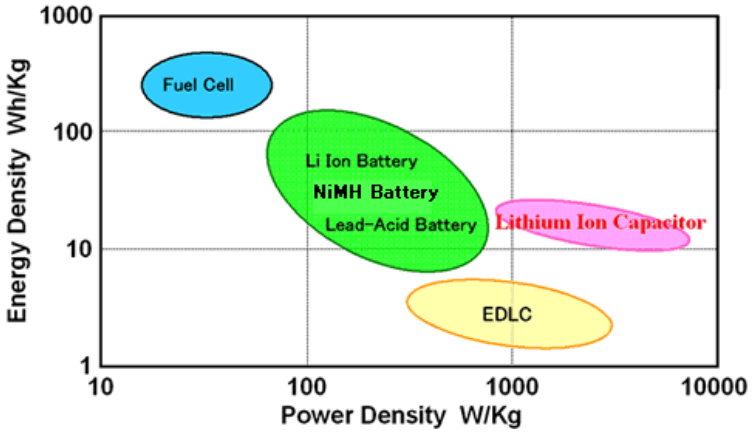

Figure1: Ragone Plot [1]

1.1. Supercapacitors

Supercapacitor falls under the category of polar capacitors. They consist of electrode material, an electrolyte, separator, binder, current collector and unlike conventional capacitors there is no dielectric. The feature of high capacitance, low voltage rating, extended cycle life, elevated power and energy density make supercapacitor superior to a capacitor. Supercapacitors have escalated power density in comparison to batteries however, they possess less energy density in comparison to batteries. Supercapacitors have energy density E_d up to maximum $5Whkg^{-1}$which is greater than solid state capacitor but still very less than the batteries that offer E_d up to $200Whkg^{-1}$. For this reason, researchers have been putting subtle efforts to improve E_d since E_d is a property that assesses the electrochemical behavior and performance of supercapacitors. The equation for E_d is:

$$E_d = \frac{1}{2} C_s V^2$$

Where C_s represents specific capacitance and V represents applied voltage window of the working cell. Hence, the value of energy density shown by a supercapacitor fundamentally depends upon C_s and the applied potential window. Various researches have been therefore, proposed to enhance the values of C_s and V [4]. Nanotechnology coupled with electrochemistry has been revamping supercapacitors in order to satiate the increasing energy demand of the world as fossil fuels are being drained at a very fast pace and will no longer be available in future. C_s depends on electrode and electrolyte. It primarily depends upon the material of the electrode. Therefore, the supercapacitor industry has been thriving mainly in the field of electrode material where there have been innumerable attempts to hoist the value of E_d by using the advanced nano materials and their harmonized composites that are compatible with the certain type of a supercapacitor [3].

1.2. Types of Supercapacitors

Following figure shows the types of supercapacitors along-with their working mechanism:

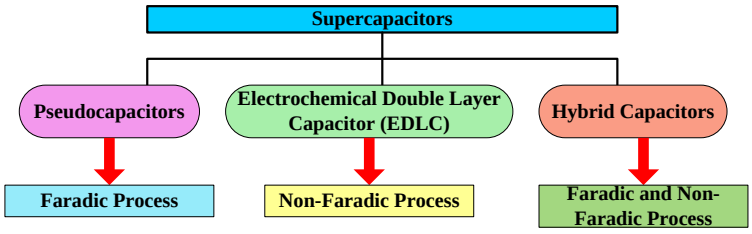

Figure2: Types of Supercapacitors and their Working Principles

There are three types of supercapacitor that are categorized in compliance with the mechanism and working principle on which they function as depicted in Figure 2. The first kind is recognized as Electrochemical Double Layer Capacitor which is abbreviated as EDLC and is the main topic around which our review revolves. EDLC works on the principle of non-Faradic process which alludes that there exists absolutely no transference of charge between the electrode and electrolyte. The charge is purely stored in electrostatic manner on the interface of electrode/electrolyte and on that account the value of capacitance is dependent on the

nature of electrode material and its chemical and physical properties. This review particularly discusses the EDLC electrode material manufactured by 0D carbon-based materials. The second category of supercapacitor is known as Pseudocapacitor which works on the principle of Faradic process which indicates that reversible redox reactions induce the storage of charge at the surface of a redox active medium by means of electrochemical mechanism called Intercalation process. The third type of supercapacitor is called hybrid supercapacitor because it involves the working principles of both EDLC and Pseudocapacitor. Hybrid supercapacitor works when both Faradic and non-Faradic processes coordinate and thus result into better capacitance and electrochemical properties [5].

The capacitance and electrochemical properties of any class of a supercapacitor predominantly depends upon the nature of electrode material possessing a particular set of physical and chemical properties. Therefore, the progression of supercapacitor technology is directly related to the advancement and evolution of electrode materials. The materials designed for the fabrication of electrode materials have generally been classified into four groups. The first group is carbon-based materials that are further divided into four categories based on their dimensions which are 0D, 1D, 2D and 3D e.g. Activated Carbon (AC), graphene, carbon nanotubes (CNTs), multi-walled carbon nanotubes (MWCNTs) etc. The second group of electrode materials are Metal sulfides and metal oxides like NiS, NiS_2, CuS, NiO, CuO, MoO etc. The third group consists of polymer-based electrode materials including polyaniline, polythiophene, polypyrrole etc. and their derivatives. EDLC have carbon-based electrode materials while polymer-based, metal oxides and metal sulfide-based electrodes are employed in Pseudocapacitors. The fusion of all these three material groups results in the formation of hybrid nanocomposites consequently, resulting into the enhanced electrochemical properties of a supercapacitor [6].

II. Electrochemical Double Layer Capacitor (EDLC)

In EDLC, the capacitance is obtained through electrostatic mechanism, by the aggregation of charges on the electrode/electrolyte interface without undergoing any sort of chemical change. This implies that there is absolutely no transference of charge within electrode/electrolyte when the supercapacitor is being charged or discharged. EDLC has a basic structure just like a battery. It contains polarizable electrodes, an ionic electrolyte, a separator and a current collector. When the voltage is applied, the electrolytic ions begin to diffuse into the porous electrode interface carrying an opposite charge. Subsequently, the accumulation of charge at the interface results into the emergence of charged double layer (Helmholtz Layer) leading to the charging of a supercapacitor. The separator has a crucial role to prevent the recombination of formed charged layers at the electrode interface by allowing only the transference of ions. EDLC discharges when the aggregated ions are freed and disengaged from the surface of active material and hence the ions disperse into the electrolyte solution breaking the charged double layer [6]. This phenomenon is shown in Figure 3.

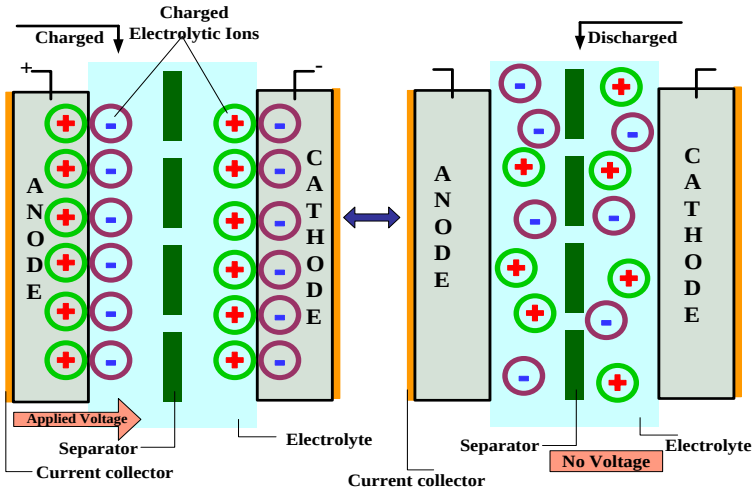

Figure 3: Basic Working Principle of Charging and Discharging of Electrochemical Double Layer Capacitor (EDLC)

Porous carbonaceous materials are being intensively used for the electrode material for EDLCs. Carbon-based materials are prospective materials for the electrodes of a supercapacitor because they are electrically conductive, abundant, non-toxic, manageable, chemically and thermally stable and they can process at wide temperature range. Over the past years, activated carbon, carbon nanotubes (CNTs), multi-walled carbon nanotubes (MWCNTs), graphene, carbon nano-horns, carbon fibers, carbon cloth and the derivatives of carbon have been used for EDLC electrodes because of their highly accessible surface area offering good conductivity

[6]. They have long-term cyclic stability however; specific capacitance is limited. Residual impurities on the surface of such electrodes may be responsible for limited capacitance as the mobility of charge carriers is hindered by the presence of impurities thereby affecting the overall performance of a supercapacitor. The porosity of active materials also decides the value of C_s. The researchers concluded that the pore size should be analogous to that of the ions. Their studies proposed that the C_s increases if the pore size is to a certain extent matches the ionic size [7]. There has been a vast research dedicated to uplift the less energy density of EDLCs by refinement of the electrochemical characteristics. Majorly, there are two ways and means for the pursuance of this aim. The first approach, as discussed earlier, is to increase adsorption capacity of ions by increasing SSA or specific surface area and entire pore volume [8, 9]. The second approach is to introduce metal oxides or surface functional groups in carbonaceous materials that ends up in improving the electrochemical features of EDLCs [10-12].

III. Carbon-based Materials

The electrochemical and capacitive properties of EDLC principally relies on size of pores, allocation or distribution of pores all over the electrode material and its specific surface area, hence carbonaceous materials are used for EDLCs [13, 14]. Becker fabricated EDLC for the very first time by using carbon-based materials in 1975 and opened the doors for further research [1]. Carbon-based materials have always been ruling on the billboard of the supercapacitor industry because of their premium mechanical, electrical and structural properties. However, the value of C_s (specific capacitance) is inferior, so in order to fabricate an electrode that gives improved value of specific capacitance and energy density; countless materials have been studied and proposed by researchers to make composites with carbon based materials and their derivatives in pursuance of getting the results beyond the limitations of each carbonaceous material [15].

There is a wide range of natural sources like crude oil, biomass or coal that are utilized as precursors for carbon-based materials. There is a radical change in properties of a material when it comes down on a nano scale. Referring to carbon-based materials, the properties change with the change in dimensions of nanostructure. There are basically four groups of carbonaceous materials based upon their dimensions namely 0D, 1D, 2D and 3D [16].

The following section reviews different 0 dimensional carbonaceous materials that are utilized in electrode fabrication of EDLCs along with their positive and negative aspects and their nanocomposites with various other nanomaterials that tend to balance and negate the limitations of each other and consequently result in enhancing the electrochemical performance of an electrode material by means of synergism.

IV. Zero-Dimensional Carbon Materials

Zero-Dimensional (0D) nanomaterials have been crowned as forerunner of nanotechnology. 0D carbon nanomaterials include Carbon quantum dots (CQDs), activated carbon (AC), carbon nanospheres, mesoporous carbon and carbon nano-onions (CNOs). Their spherical structure allows them to have more specific surface area ranging from hundreds to thousands m^2g^{-1}. Denser electronic structure, conductivity, tunable pore size and distribution are the factors that make 0D carbon nanomaterials suitable for their utilization in supercapacitor electrodes resulting into increased value of specific capacitance. They possess unique structural and physiochemical properties that make them potential candidates for the application in energy storage devices [17].

4.1. Carbon Quantum Dots

Carbon Quantum Dots (CQDs) that have size below 10nm have attracted great consideration in supercapacitor industry due to their promising properties like chemical stability, solubility in water, ability to get functionalized, excellent electrochemical and photochemical properties. They possess diverse functional groups and have photoluminescence properties. Their ability to combine with organic and inorganic materials consequently results into the strength enhancement of the bonds present between different materials. In other words, CQDs can be used as connectors or linkers to combine different materials to make a composite for the electrode enhancing the electrochemical properties of a supercapacitor. CQDs can not only strengthen the bonds between materials of the composites but also reduces the internal resistance along with charge transfer resistance [18]. CQDs can be synthesized by several techniques. Top-down routes like laser ablation, arc-discharge method, plasma reactor, ultrasonic, electrochemical and chemical oxidation methods have been used for synthesizing CQDs. Waste derived CQDs can be synthesized by hydrothermal heating of waste products, carbonization technique, plasma induced pyrolysis and microwave assisted heating process. Bottom-up approach that includes thermal pyrolysis, hydrothermal methods, solvothermal and metal-organic frameworks (MOF) template directed techniques can also be used for the synthesis of CQDs [18].

Haipeng et al. successfully proved the linking ability of CQDs to be effective by connecting MnO_2 to Graphene aerogel via CQDs that resulted into enhanced electrochemical properties like electrical conductivity,

cycle stability and specific capacitance of 721 Fg^{-1} at 1 Ag^{-1} current density [19]. The nanohybrid composites of CQDs with metal oxides and metal sulfides have been synthesized in order to uplift the supercapacitance and other electrochemical properties [20-22]. Srikant et al. improved the capacitive properties of NiS by incorporating CQDs of about 1.3nm. CQDs improved the charge transfer process that boosted the specific capacitance up to 880 Fg^{-1} at 2 Ag^{-1} current density and showed stability at about 2000 charge-discharge cycles [23]. Researchers have also studied the effects of numerous dopants like phosphorous [24], nitrogen [25], iodine and oxygen [26], phosphorous and boron [27] with CQDs and concluded the overall improvement in electrochemical properties of supercapacitor electrodes. Jingying et al. proposed novel approach by utilizing graphitic CQDs as intercalator for the synthesis of N, P-CQDs/rGO composite aerogel for the first time, showing the specific capacitance of 453.7 Fg^{-1} at 1 Ag^{-1} along with improved cycle performance over 10000 charge-discharge cycles in comparison to rGO, GO and N, P-rGO [28]. CQDs also capable of making composites with polymers. For instance, Wang et al. synthesized nitrogen-doped CQDs/PANI nanocomposite for supercapacitor electrode and presented that introduction of N doped CQDs in PANI enhanced the specific capacitance of PANI [29]. Customarily, CQDs when attached to other electrocatalysts, transition metal oxides and sulfides or heteroatoms to form nanocomposites, they behave as electron donors or acceptors and tend to improve the surface area, electrical conductivity, porosity and other electrochemical properties [18, 30-32].

Therefore, CQDs have a great potential to be more investigated and further research is expected in the fabrication of supercapacitor electrode by synthesizing nanocomposites with CQDs following heteroatom doping strategy because this approach shows promising results as compared to nanocomposite strategy.

4.2. Activated Carbon

Activated carbon (AC) has been broadly used for the electrode material for the commercial fabrication of EDLCs on account of its distinct advantages that include large specific surface area, excellent adsorption/desorption, conductivity, high packing density, low cost and commercial availability [33]. AC possesses a complex porous structure that consists of micropores of size smaller than 2nm, mesopores that range from 2nm to 50nm in size and macropores which are larger than 50nm in size contribute in AC having a large specific surface area [34]. Though, AC has not only low density but also shows low capacitance and operating voltage. Formerly, the low capacitive feature of AC was primarily improved by increasing specific surface area. However, the intricate and refined porous structure results into the breakage of conductive network which radically reduces the electrical conductivity of AC [35-37]. In fact, the electrical conductivity ranges from 1250-2500 Sm^{-1}[34]. Furthermore, the storage ability and transportation of electrolytic ions are critically inhibited in vast and branched microporous mesh. Such problems give rise to less utilization of surface area and hence lowers the specific capacitance along with slow electrochemical kinetics and unsatisfactory rate performance [36]. Therefore, to resolve such issues, other carbonaceous materials and nanomaterials have widely been combined with AC [38, 39]. Commercial supercapacitor electrodes are generally fabricated by the combination of AC or other nanocomposites with conductivity additive like Carbon Black (CB) and a binder like Polyvinylidene Fluoride (PVDF) or Nafion [40].

Nevertheless, the electrochemical properties and performance of such supercapacitors are still not good enough due to following reasons. Firstly, AC and CB particles are almost of the same size due to which there is a very less particle interaction mixing that gives rise to low packing fraction for electrodes and therefore affecting the volumetric performance. So, enhancement in packing and mass loading of electrode has been a major issue under consideration for the fabrication of commercial supercapacitors. Secondly, there is usually less availability of electrical conduction network in the electrode due to clustering of CB particles and loose contact between CB clusters and AC particles. Thirdly, during charge-discharge phenomenon AC suffers apparent contraction/expansion and CB undergoes extreme agglomeration and clustering; resulting into increased internal resistance in electrodes which obviously restricts the cycle life of a capacitor. Hence, scores of nanocomposites combined with AC have been prepared in past years [36, 37].

Yan et al. proposed a novel strategy to boost the supercapacitance by improving the overall conductive networks. They introduced highly crystalized Graphene Quantum Dots (GQDs) with AC that resulted into conductivity escalation within AC and hence improved capacitance of about 338 Fg^{-1} at 1 Ag^{-1}. They claimed that this technique increased the electrochemical reaction kinetics and provided a suitable path for ion transfer and hence the attained carbon delivered high energy density of 13.47$Whkg^{-1}$[36]. Tyagi et al. studied the performance of EDLC electrode that was fabricated by consuming mustard oil-derived Activated Carbon Soot (ACS) and concluded that ACS produced better results than raw Soot Carbon (SC) because ACS possessed higher carbon content along with spherical morphology and ordered layered structure. ACS showed better specific capacitance of 50 Fg^{-1} at 0.5 $mAcm^{-2}$, enhanced energy density of 20.25 $Whkg^{-1}$ and excellent power density of about 1620 Wkg^{-1} in comparison to SC that showed 28 Fg^{-1}[41].

Another technique to improve the capacitive behavior of AC based electrode for EDLCs was proposed by Cheng et al. This technique was to combine AC and CB with CNTs and CNFs. They proved that the

synergistic effects of these nanomaterials AC/CB/CNT/CNF all combined produced better results than AC alone, AC/CB and AC/CB/CNT nanocomposites. They reported that AC/CB/CNT/CNF generated the value of specific capacitance of 66.1 Fcm^{-3}, power density of about 101 kWL^{-1} and energy density of 29.6 WhL^{-1}[37]. Consecutive doping technique was used by Yang et al. to improve the performance of commercial AC for organic EDLC electrode [10]. Wang et al. prepared AC particles from waste-water purifier and attached these particles to the surface of reduced GO sheet which suppressed the aggregation of rGO boosting the specific surface area of the electrode material thus delivered the specific capacitance of 116.9 Fg^{-1} at 0.5 Ag^{-1} current density in 1M electrolyte of H_2SO_4[42]. In another study, Huang et al. fabricated nanocomposite of rambutan-like AC spheres/CNTs and studied its application as EDLC electrode. They reported the specific capacitance of ACS/CNT to be 180 Fg^{-1} at 2.5 Ag^{-1} which was three times better than pristine ACS [43]. Saini et al. compared and reviewed the electrochemical properties of various biowaste derived AC for EDLC electrode and deduced that AC derived from Jejubi fruit activated using NaOH and AC derived from orange peels by using KOH in activation method yielded highest values of capacitance of 460 Fg^{-1} at 1 Ag^{-1} and showed good cyclic stability [44].

Activated carbon, hence, has yet the potential to be investigated further with various other nanocomposites to boost the electrochemical properties of EDLCs.

4.3. Carbon Nanospheres

Carbon nanospheres (CNSs) and functional CNSs have attracted significant interest in carbon community due to their unique morphology, controllable size chemical composition and porosity. CNSs have numerous environmental and technological applications such as in catalyst support, adsorbents, lithium-ion batteries and supercapacitor electrodes. CNSs are basically formed by the pairing of heptagonal and pentagonal carbon rings. The graphitic sheets are spherically arranged such that the structure follows the spherical curvature giving rise to unclosed shells and thus open edges at the surface containing dangling bonds. These reactive bonds of carbon nanospheres are not only responsible for their high reactivity during chemical reactions but are also suitable for catalyst supports. The reactive bonds on the surface of CNSs also enables them to make various composites with other materials and play a vital role in energy storage and other electrochemical applications [45]. CNSs, therefore, are used as electrode material for supercapacitors as they are inexpensive, possess large surface area, easily processable and of course have large specific capacitance.

As explained earlier, increased porosity and the structural properties of a nanomaterial used in electrode is a major factor for good electrochemical properties. A reliable and ideal supercapacitor electrode bearing large value of specific capacitance and fast reaction kinetics is supposed to be made from a carbon material that offers significantly abundant and rich active sites accompanied by suitable electron-ion transport channels that assist in the formation of Helmholtz Layer actually responsible for energy storage in EDLCs. This led researchers to explore plenty of methodologies for the porosity engineering, functionalization and the hierarchical structure of CNSs suitable for EDLCs while investigating their electrochemical behavior in supercapacitor devices. Pore engineering endeavors to modify the material by designing and structuring a hierarchical arrangement of micropores, mesopores and macropores that not only provide rich active sites but also transport channels. Micropores are responsible for increased active sites for ions and electrons forming Helmholtz Layer for energy storage while mesopores and macropores serve as transfer pathways for the transference of ions to the active sites [46].

CNSs have been synthesized by numerous ways like Chemical Vapor Deposition (CVD), Self-assembly technique, Direct Pyrolysis, Templated processes, Sugar Carbonization and so on [47]. However, it is a complex and difficult process to tune active pore sites and engineer transport pathways keeping in view the delicate interior structure and outer shell and therefore, the synthesis of refined hierarchical porous carbon structure has been still a problem in synthetic methodology [48]. That being the case, some studies have suggested that the templated processes are better than other techniques since templated porous carbon materials carry microporous, mesoporous and microporous hierarchical structures that aid the material to show enhanced specific capacitance [47].

Liu et al. adopted oxygen-modulated porous activation strategy (OMPA) to activate CNSs for enhanced electrochemical features and prepared Activated Oxygen-functionalized Carbon Nanospheres (AOCN). Pre-oxidation of CNSs followed by activation process with KOH resulted in optimized hierarchical porosities and faster charge storage kinetics due to doped oxygen that increased active sites and facilitated ion and electron transportation. They claimed that doping oxygen produced better results than activated carbon nanospheres and reported specific capacitance of 282 Fg^{-1}at 0.5 Ag^{-1}[46]. Another approach of Pyrolysis technique was adopted by Yao et al. and synthesized porous hierarchical CNSs with a unique ball in ball structure and stated that their results showed ultralow LPPS (longest possible pore separation) of about only 10nm that led to excellent charge transfer and provided best known pore connectivity. They reported escalated specific capacitance of 405 Fg^{-1} at 1 Ag^{-1} current density [49]. Hollow carbon spheres (HCSs) show promising applications for EDLC electrode as

it has reduced transport path and high surface to volume ratios that leads to elevated capacitance. HCSs have a porous carbon shell which enhances the effective contact area between electrolyte and active area of the electrode. This results into reduction of ions diffusion path and improves the volume expansion and contraction during charge-discharge of supercapacitor [50]. Du et al suggested a template free method for the synthesis of nitrogen doped HCSs possessing tunable porous structure. They reported specific capacitance of 266.9 Fg^{-1} at $20Ag^{-1}$[51].

Therefore, unique properties and behavior of CNSs and their ability to make composites with other nanomaterials make them capable to be further investigated in perspective of improving the electrochemical features of EDLCs.

4.4. Mesoporous Carbon

Mesoporous carbon (MC) is a solid-based material that have pores in the range from 2nm to 50nm. There are two types of MC depending upon the distribution of the pores which are Ordered Mesoporous Carbon materials (OMC) and Disordered Mesoporous Carbon materials. OMC possess an orderly and periodic structure that makes it able to provide continuous and interconnected pathways for the diffusion of electroactive species like electrons and ions in electrochemical processes and systems. Disordered mesoporous carbon materials have no ordered structure and have random distribution of pores. Their ability of narrow pore size distribution, high surface area, conductivity, simple functionalization strategies and low toxicity make them a fine choice for EDLC electrode. However, capacitance value exhibited by mesoporous carbon is less than 2 dimensional CNTs because they are less conductive than CNTs although they both have uniform pore and pore size. Hence mesoporous carbon materials are further synthesized into 2 D or 3D structures and various researches show that the specific capacitance is improved when MC is doped with oxygen or nitrogen and utilizing it with other materials to make a composite.

Capacitance properties of self-ordered mesoporous carbon was studied by Zhou et al. and they calculated specific capacitance of about 60 to 95 Fg^{-1}[52]. Zhang et al. have followed multi-stage self-assembly technique in order to synthesize nitrogen doped mesoporous carbon having controlled and high content of Pyridine N for high surface area normalized capacitance. They reported specific capacitance of 15 to 25 μF cm^{-2}[53]. Maji et al. fabricated tube-like structure of mesoporous carbon derived from macaroni fullerene crystals. They reported a high capacitance value of 422 Fg^{-1} [54]. Another approach was followed by Bairi et al. in which they synthesized cube like mesoporous carbon derived from the crystals of fullerene and suggested this material as a good choice for supercapacitor electrodes as they reported specific capacitance of 286 Fg^{-1} at 1 Ag^{-1}[55].

Thirukumaran et al. synthesized rope-like mesoporous carbon doped with nitrogen derived from polybenzoxazine by simple calcination technique and reported the value of supercapacitance 60 Fg^{-1} at 1 Ag^{-1}[56]. Lei et al. used high temperature calcination process in order to synthesize mesoporous carbon by utilizing moss as the carbon precursor. Moss-derived biomass porous carbon (MPC) was activated by using NaOH in order to give it a mesoporous structure, high pore volume and high surface area that resulted into elevated value of capacitance of about 332 Fg^{-1} at 1 Ag^{-1}[57]. Fu et al. prepared a composite having a CNT core which was encapsulated by nitrogen doped mesoporous carbon particle making it a carbon nanotube@N-doped mesoporous carbon (CNT@N-PC) having increased active sites and thus this material was proposed to be a promising composite for supercapacitor electrode. Resorcinol-formaldehyde resin was used as their carbon source. They reported supercapacitance of about 244 Fg^{-1} at 1 Ag^{-1}[58].

Based on properties shown by mesoporous carbon investigated by numerous researchers, makes a room for further research related to its synthesis and nanocomposites consequently, resulting in the enhancement of supercapacitor electrodes and their efficiency.

4.5. Carbon Onions

Carbon onions or carbon nano-onions (CNOs) belong to fullerene family and they consist of closed spherical graphene shells possessing onion-like concentric hexagonal or pentagonal layered structure having size less than 100 nm. Substantial number of holes and defects are present in graphitic layers that make up a CNO. CNOs can be synthesized by different techniques like chemical vapor deposition (CVD), vacuum annealing, flame synthesis or pyrolysis, ion implantation, electron beam irradiation and arc discharge method. The synthesis routes play a pivotal role in deciding their chemical and physical properties because the properties are directly related to CNOs' morphology, inter-layer distances and chemistry. Inherent conductivity of CNOs can be controlled by annealing. Graphitization of carbon arrangement and higher degree of crystallinity of CNOs tend to improve by escalating the temperature. This annealing process enhances the graphitization consequently, leading to the improvement of transport channels for electrical conductivity. On behalf of physical properties CNOs are further categorized as spherical or polygonal shaped. Small sized CNOs are below 10nm in size. CNOs above 10nm in size are called big sized. CNOs that are either vacant or are stuffed with different metals are called hollow cored or densely cored CNOs. CNOs can be used as conductive additives as

well as active material for EDLCs owing to their higher power capabilities, good capacitance retention, small size, concentric layers having defects and holes, conductivity, good pore size and pore distribution. However, CNOs have low surface area and thus tend to have less energy density. Several methodologies have been adopted intending to improve the electrochemical properties of CNOs. CNOs can be combined with polyelectrolytes. Conducting polymers with CNOs also tend to increase the capacitance values and electrochemical behavior. Transition metal oxides and their hydro-oxides doped with CNOs result into uplifting the capacitance value. CNOs having the diameter in range of 5nm to 6nm are usually preferred over those having larger diameters because such CNOs disperse easily in liquid solutions during the process of polymerization. Consequently, the prepared composite is homogeneous and its porosity is regular [59-61].

Facile flame-pyrolysis technique was implemented by Mohapatra et al. to synthesize sulfur-doped CNOs and claimed that their methodology successfully prepared S-CNOs for supercapacitor electrodes having not only high capacitance of 305 Fg^{-1} at $2Ag^{-1}$ but also owning high surface wettability that enabled the composite to act as a conductive ink subsequently, limiting the use of binders while making the electrodes [62]. Mohapatra et al. applied in-situ flame-pyrolysis to prepare highly graphitic and mesoporous nitrogen doped CNOs. They reported capacitance of 113 Fg^{-1} at $4Ag^{-1}$[63]. Conducting polymers coupled with CNOs also tend to escalate the electrochemical properties of an electrode. Majumder et al. proved this fact by synthesizing a composite of polyaniline with CNOs by in-situ oxidative polymerization of monomer aniline. They discussed that CNOs reduced the agglomeration of PANI fibers, provided a large active surface area for electrolytic ion interaction, mesoporous structure increased the wettability and also resulted in shorter ion diffusion path. The synergistic effect of CNOs and PANI led to composite delivering specific capacitance of about 196 Fg^{-1} at $1Ag^{-1}$[64].

Proper functionalization of CNOs with correct structures by following reliable techniques also plays a crucial role in the enhancement of electrochemical properties of CNOs and their composites. Valasquez et al. have discussed the functionalization of CNOs by amidation reaction between oxidized CNOs and 4-(pyren-4-yl)butanehydrazide and proved this technique to be reliable [65]. CNOs have a perfect structure for electrochemical phenomenon as they have most of the pores which are equal in size and are easily approachable to the electrolytic ions. Their properties makes them a better candidate than multi-walled carbon nanotubes (MWCNTS) as CNOs deliver two times more capacitance than MWCNTs [60]. Jin et al. synthesized CNOs by facile nickel assisted graphitization and utilized rice husk as carbon precursor for activation process. They reported specific capacitance of about 350 Fg^{-1} at $0.1Ag^{-1}$ current density [66]. Bhaumik et al. prepared $Ni(OH)_2$@PANI nanocomposite with CNO which was synthesized from waste car wheels as a material of anode for supercapacitors and reported specific capacitance of 622 Fg^{-1} at 2 Ag^{-1}[67].

Carbon onions are the most versatile carbonaceous materials because they have exceptional structure as discussed above and therefore, possess unique properties. By making a good use of these properties, researchers have been synthesizing various nanocomposites to boost the electrochemical performance and overall efficiency of supercapacitors.

V. CONCLUSION

0D carbonaceous materials have emerged to represent rapidly expanding and developing field of research owing to their unique intrinsic properties, structural characteristics and their ability to couple with other nanomaterials consequently resulting into the enhancement of electrochemical properties of EDLCs by the synergistic effects. High packing density, tunable porosity, high surface to volume ratios and controllable particle size make 0D carbon materials to be deployed in supercapacitor electrodes. Although many successful attempts have been made to fabricate an EDLC electrodes with enhanced electrochemical properties but there is still a need to focus on some of the facts that are usually overlooked. Firstly, electrochemical mechanisms must also be given due attention while making a composite for electrode with enhanced electrochemical properties because at the end it is the electrochemical mechanism which also plays an effective part in giving out the results of the fabricated electrode. Secondly, the strategy of heteroatom doping indeed improves the electrochemical behavior however, in some cases, there are problems like decrease in electrical conductivity which surely decreases the efficiency of electrode. Thirdly, pore size and pore distribution must be under strict supervision while handling a sample as this directly affects the electrolyte ion transfer. Fourthly, carbonaceous materials derived from bio-mass lacks volume performance and needs to be improved nevertheless, they are inexpensive, available and possess excellent electrochemical properties.

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