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Electrode Materials for Electrochemical Capacitors: An Overview

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ABSTACT: This review, mainly paid attention to how supercapacitor electrodes are made in order to increase energy transfer and storage, so that, superior performance energy and power density can be gained as excellent operating conditions plus long cycling life is attained. Review shows that specific capacitance of supercapacitor is influenced by electrolyte accessibility, pore size distribution, and structure, electrolyte type, pore shape plus electrical conductivity also affects it. Instrumental analysis of performance of electrode materials was looked into by means of supercapacitor evaluation techniques like CV, EIS and galvanostatic charge/discharge. We understood from review that solvent used in making of electrolytes has enormous effect on it working voltage; the electrolyte–electrode relations with ionic conductivity of electrolyte were shown to have participated very much in internal resistance of SCs. In conclusion, studies on electrode materials for SCs in general, have significantly progressed and have brought lots of innovative improvements in supercapacitors proficiency and application; but, further studies on optimization of energy density and power density of EC need to be carried out while their weight, volume and production expenditure require reduction.

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Contemporary economy as we know today is propelled by the ease of use of reliable energy sources (Mensah-Darkwa *et al.*, 2019). These renewable energy sources are of these forms: solar energy, wind energy, and nuclear power plants, etc. These energy sources often possess characters such as clean, renewable, efficient and cost effective ways by which energy is stored. As these renewable energy technologies are utilized, a surplus of energy is sometimes made available in some areas, and deficient in some other areas, balancing this energy gap becomes a difficult task. Consequently, the surplus energy generated by these renewable sources when available has to be captured, and a competent storage system which can preserve such energy is provided (Barzegar *et al.*, 2017). To overcome this constraint of excess energy wastage, researchers have developed sustainable energy power sources such as batteries (Jäckel, 2018), fuel cells (Fahimi and Moradlou, 2020), electrochemical capacitors (ECs) (Tasnin and Saikia, 2018) etc. These devices that store power as electric energy, have found use in areas which include; electric vehicles, and consumer appliances, etc (Lee *et al.*, 2018). Amongst all these sustainable energy power sources, electrochemical capacitor (ECs) has demonstrated more promising features in energy storage technology with ability to store and discharge energy rapidly and effectively. Electrochemical capacitors (ECs) also known as Supercapacitor, is a device that is designed to build up and accumulate

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large amounts of energy as electrochemical double-layer field, which is likened to spatial charge separation, and electrochemical change in two nano engineered interfacial layers, that can rapidly discharge their stored energy effectively (Hepel, 2016). Electrochemical capacitors (ECs) comprise two conductive electrodes, divider (separator), in addition to an ionic electrolyte, their operation is governed by the physical separation of charges by means of ion diffusion brought about by an electric dipole.(King *et al.*, 2018). They comprise of these, electric double layer capacitor (Jian *et al.*, 2016) (made of carbon forms, carbon aerogels, carbid-derived carbon, carbon nanotubes and activated carbon), hybrid supercapacitors whose charge storage is both electrostatic and electrochemical (made of Asymmetric Pseudo/EDLC, Composites and rechargeable battery-type) and pseudo-capacitors (made of conducting polymer and metal oxides). Non-faradaic processes is used by Double-layer capacitors to store charges while pseudo-capacitors are through faradaic process (Jiang and Liu, 2019). Electrochemical capacitors, is seen as storage device of tomorrow (Xing *et al.*, 2019),because of high specific power, good cycle stability, quick charge-discharge performance, long cycle life, flexible operating temperature, and cost effectiveness as no cost of maintenance is needed for the ECs and due to the fact they are not often charged neither do they have special control systems for operation makes them of abundant importance (Burke and Zhao, 2022). Major disadvantages of ECs are low-energy density, high self-discharge rate in addition to expensive cost of fabrication. These weaknesses, limits the use and application of the major types of ECs available (Ike *et al.*, 2015). In order for electrochemical capacitors to become an everyday use product in the energy sector, these short falls must be overcome devoid of lose to power density and long cycle life. High cost associated with production of ECs must also be overcome for it to have wider acceptability as an alternative energy storage pack that is more efficient and cost effective than battery. Electrochemical capacitors (ECs), can serve as back-up to batteries(Da Silva *et al.*, 2020; Simon and Gogotsi, 2008). Nevertheless, even with the advantages associated with ECs, their energy density cannot still compare to that of batteries. But, batteries have been known to be non-eco-friendly as they are toxic and harmful to both man and nature since they most often contain hazardous, corrosive and toxic materials such as mercury, cadmium, lithium, lead etc (Mosa *et al.*, 2017), consequently, there is need to design an efficient ECs with high energy density which can serve as an effective substitute for battery. Researchers in the field of electrochemical capacitors are investigating new and effective materials for electrochemical capacitor electrodes and electrolytes having in mind aim of making better the energy density of supercapacitors. Two methods were suggested, these include; enhancing specific capacitance otherwise extending voltage window of device (Hwang *et al.*, 2017). As earlier stated, electrochemical capacitors (ECs) are exceptional energy-storage appliance because of extended life span, eco-friendliness (case of aqueous electrolytes (Da Tiea *et al.*, 2019; Gou *et al.*, 2020; Zhao and Zheng, 2015) and bio-sourced binders(Xiang *et al.*, 2019) great charge-discharge rate (Deshmukh, *et al.*,. 2013; Shinde *et al.*, 2018). However, like batteries, they are also associated with the problem of voltage drop (self-discharge). Voltage drop also known as self-discharge in dielectric or electrolytic capacitors takes place without any chemical processes owing to leakage of current between electrodes, *(*Jakobczy*k and* Biegun, *2013;* Menzel *et al.*, 2020). Self-discharge and its associated drawbacks, such as reduced life circle, has limited the use of ECs and have attracted the attention of researchers (Ike and Iyuke, 2015). Given that self-discharge is a significant pointer to measure performances of an electrochemical capacitor (aging, energetic efficiency, etc.), effects of self-discharge need to be drastically minimized. Above all, it is important that cost-friendly and eco-friendly materials be used when making supercapacitor electrodes.

This work, presents an overview of study efforts on electrode materials for ECs, with intent on better understanding of carbon based electrode material in order to overcome the shortfalls experienced in the production of supercapacitor electrode as it concerns energy density with focus on EDLCs.

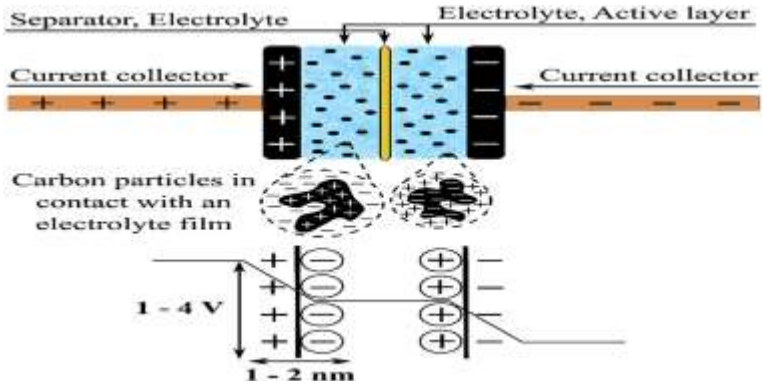

Fig 1. *Schematic diagram showing the voltage drop at interface region, between the electrode and electrolyte copied from* (Da Silva *et al.*, 2020)

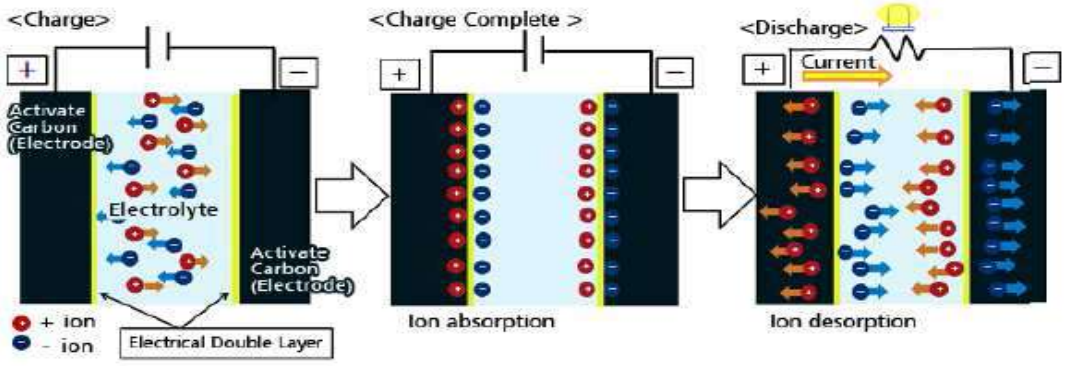

Fig 2. *Schematic diagram of charging of an uncharged EDLC, its completely charged state and its discharging copied from* (Venkataraman, 2015)

Mechanisms of energy storage: Electrochemical supercapacitors store energy through the separation of charge between two electrodes using an electrolyte. The separator is typically a material that allows for ionic charge transfer while preventing the electrodes from coming into direct contact. Organic electrolytes are commonly used for polymer or paper separators, while aqueous electrolytes are preferred for ceramic or glass-fiber separators. Capacitive energy storage materials and battery-type energy storage materials can be distinguished based on their ion transport kinetics. Capacitive materials show surface control charge storage kinetics, while battery-type materials exhibit diffusion-controlled kinetics. To evaluate the performance of supercapacitors, different techniques such as cyclic voltammetry, galvanostatic charge/discharge, and electrochemical impedance spectroscopy are used. The materials used to build supercapacitors can be categorized into three types: electric double layer capacitors, pseudocapacitors, and materials with battery-like behavior (Zhao and Zheng, 2015).

EDLC Mechanism: When mechanism is considered, EDLC is likened to conventional capacitor. But the disparity between them depends on their charge/energy storage mechanism. EDLC owe its mechanism to electric double layer on and around electrode surface. That is, adsorption/desorption of opposite charges on electrode (electron holes) and electrolyte (i.e. solution side of electrode (ions)) interface.(Ike *et al.*, 2015; Zhang *et al.*, 2019). As shown in Figure 3. As a result of good potential-difference across EDLC, electrode micro pores with different polarities are generated and induced, this results in high affinity of electrolyte ions towards electrode micro pores. EDLCs specific double layer capacitance is estimated using the formula in equation (1):

$$C = \frac{\varepsilon_r \varepsilon_0 A}{d} \quad or \quad {}^{C}/_{A} = \frac{\varepsilon_r \varepsilon_0}{d} \qquad (1)$$

Where $\varepsilon_{r,}\varepsilon_0$ is denoted as relative static permittivity of medium in double- layer region, dielectric constant vacuum respectively. A is surface area of electrode (m^2), and d, given as thickness of electrical double-layer (m), (that is, charge separation distance between charges in the metal part of electrode and charges passing through centers of ions adsorbed chemically or physically on the electrode). Based on (eqn1), high capacitance can be attained in double layer capacitors

if given an excellent double layer thickness and a large surface area. An electrolyte with good electrical conductivity can greatly reduce internal resistance of EDLC electrodes. Mobility of ions in pores of electrodes is increased by excellent wettability of electrolytes which results in superior conductivity(Ambare *et al.*, 2016). Numerous literatures have reported the use of aqueous electrolytes, such as (H_2SO_4) (Misnon *et al.*, 2015; Qin *et al.*, 2020), and neutral electrolytes such as (NaOH, KCl, Na_2SO_3, Na_2SO_4 and KOH, etc.) (Taer *et al.*, 2018; Wu *et al.*, 2017).Their great ionic conductivity, cost effectiveness and extensive reception nonetheless; they are beset with some disadvantages like relative low decomposition voltage of 1.23V (Kim *et al.*, 2015a; Qin *et al.*, 2020).

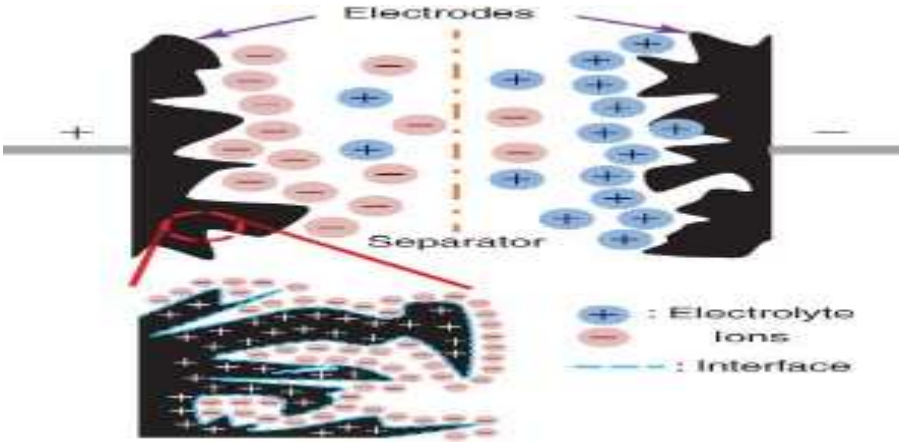

Fig 3. A schematic of charged EDLCs copied from (Kim *et al.*, 2015)

Mechanism of pseudocapacitors: The buildup of electric double layer adsorption of ions from electrolyte on electrode and ensuing redox reaction constitute charge storage mechanism of pseudocapacitors. Chemical reactions involved aid to improve capacitance values of pseudocapacitors. Transition metal oxides demonstrate pseudocapacitive performance accompanied by high specific capacitance. The multiple oxidation states possessed by transition metals gives them advantage of been used for capacitive purpose (Iro *et al.*, 2016; Li *et al.*, 2018; Venkataraman, 2015). Theoretical capacitance is represented as:

$$C = \frac{nF}{M\Delta V} \qquad (2)$$

Where n, F, M and ΔV embody number of electrons migrating in redox reaction, Faraday constant, molar mass of electrode material and operating voltage window respectively (Da Silva *et al.*, 2020; Ike *et al.*, 2015).

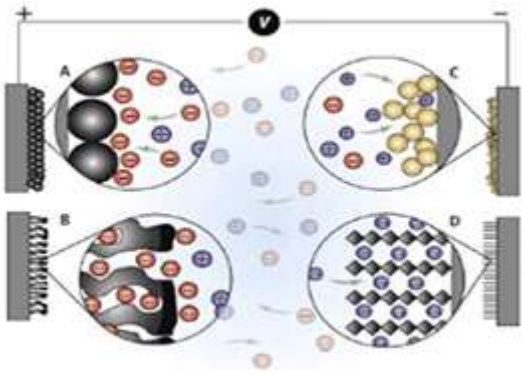

Fig 4. A diagram depicting Double-layer charge storage mechanism (Aken, 2017)

Maximum energy is usually measured in $\frac{Wh}{kg}$ and power densities in$\frac{W}{kg}$. Energy density (E) and power density (P) of supercapacitor is obtained by Equations (3) and (4) below, correspondingly (Ike and Iyuke, 2015; Melchior *et al.*, 2018).

$$E = \frac{CV^2}{2} \qquad (3)$$

$$P = \frac{V^2}{4R} \qquad (4)$$

Where overall capacitance of cell is given as C and V, operating voltage, which may be determined by examining electrolyte's stability window and is

typically influenced by electroactive components of electrode material (Yan *et al.*, 2014). The equivalent series resistance abbreviated ESR and expressed in Ohms (Ω) (Yan *et al.*, 2014).
Researchers in the field of ECs are still searching for the best ways/ methods of improving the energy density of supercapacitors using eco-friendly materials in a cost effective manner. To realize these, crucial necessities, like high specific capacity, good cell voltage and least amount of ESR values needs be achieved. It is imperative to select good electrode materials with corresponding fitting electrolyte solutions in order to optimize overall performance of supercapacitors.(Saswata *et al.*, 2012).

Elements that affects how well supercapacitors performs: In terms of customer satisfaction and energy storage/output, whole-cell capacitance (C), operating voltage (V), and equivalent series resistance (ESR) are crucial factors that affect the performance of electrochemical capacitors (ECs). C is determined by the electrodes and electrolyte used and provides a measure of the capacitance of the entire EC. V is the voltage applied to the EC, which drives the electrochemical reaction and can affect response times and noise. ESR is a measure of the resistance of the EC in parallel with the electrodes, and a low ESR indicates a more stable EC with faster response times. These parameters are influenced by various factors that affect customer satisfaction, including energy and power storage/output. While some customers may be satisfied with current technologies and commercial ECs, others are not, which has led researchers to continue seeking new materials, production procedures, and innovative cells to improve their performance. Careful optimization of these parameters is necessary to achieve the best performance for a specific application and the analyte being detected (Zuo *et al.*, 2017; Zhao and Zheng 2015; Iro *et al.*, 2016).

Instruments and measurements used in characterization performance of SCs: In order to obtain optimum result for any instrument or device, the performance of such device must be evaluated using standard instrument to Characterization and validate its workability in practice. Supercapacitors are not an exemption to this very vital procedure of optimization and validation. The entire constituent parts of a supercapacitor add to its performance consequently, the total contribution of its constituent parts should harmoniously account for its overall characterization (Zhao and Zheng 2015; Iro *et al.*, 2016).

Capacitance: To evaluate feat of electrodes with various masses, specific capacitance (Cs), which is simply defined as capacitance stabilized by mass of active material, is used ((Kim *et al.*, 2015; Zakira *et al.*, 2019). Common names for specific capacitance Cs include gravimetric capacitance ($\mathrm{F/g}$), volumetric capacitance ($\mathrm{F/ml}$), normalized capacitance ($^{\mu F}/_{cm^2}$) and areal capacitance ($^{F}/_{cm^2}$) or linear capacitance ($\mathrm{F/cm}$). (Zhang and Pan, 2015). Supercapacitors' capacitance is typically affected by experimental setups such as double electrode or triple electrode setups as well as electrode configurations such as single electrode, symmetric electrode, or asymmetric electrode. While electrode thickness is sum quantity active material on current collector, mass loading can be understood as mass of active material in each electrode. (Zhang and Pan, 2015). These two parameters are very important in determining the manufacturing process of SCs; they also influence capacitance of SCs and consequently energy density of SCs. Research has shown that in order for SCs to function effectively, mass loading should be minimum $55^{mg}/_{cm^2}$ and thickness should be in the range of 50 to 200 μm (George *et al.*, 2016; Zhang and Pan, 2015). The capacitance of materials is determined using galvanostatic charge/discharge (GCD) and cyclic voltammetry (CV) experiments.

Equivalent Series Resistance (ESR): As previously acknowledged ESR, measured in Ohms (Ω), is calculated using GCD and electrochemical impedance spectroscopy (EIS). ESR is indispensable at some stage in determining power yield and energy efficiency of ECs(George *et al.*, 2016; Zhang and Pan, 2015). As (ESR) diminishes, effectiveness of SCs electrochemical performance increases(Melchior *et al.*, 2018; Yan *et al.*, 2014). Presently, two key factors influence the accuracy of ESR, which are dwelling time and size of SC especially when measured using galvanostatic charge/discharge (GCD) test.

Operating Voltage: Operating voltage (V_0) is said to be practical potential to a system or appropriate voltage window where cell can operate without degrading. It is measured using cyclic voltammetry and charge-discharge curve (Lee *et al.*, 2018; Vaquero *et al.*, 2013). Factors influencing V_0, for ECs embrace solvents in electrolytes and cell arrangement. To overcome influence of solvent in electrolytes, ionic liquid electrolytes with values between 3.0 – 6.0 V is recommended; while the challenge of cell configuration can be overcome via usage of varied SCs materials in order to initiate extra electrochemical potential difference (Zhang and Pan, 2015).

Time Constant (τ): The product of ESR and specific capacitance in SCs is known as time constant. Time constant is determined by supercapacitor mechanism, its material and manufacturing process(George *et al.*, 2016; Zhang and Pan, 2015).

$$T = R_{ESR} \times Cs \quad (5)$$

As the time constant increases, the sensitivity of the supercapacitor lessen. Therefore it is necessary for the supercapacitor to possess a small time constant in order to increase its response time. Usually, different companies have different values for time constant (τ) set for SCs made by them (George *et al.*, 2016; Zhang and Pan, 2015).

Energy and power densities: When determining SCs deliverable performance for practical applications, calculating its energy and power densities is crucial. A supercapacitors energy density and power density are calculated gravimetrically or voluminously$\frac{Wh}{Kg}$ and $\frac{Wh}{L}$ to represent sum of electrical energy stored or deliverable and $\frac{W}{Kg}$ or $\frac{W}{L}$ to explain effectiveness in energy uptake/delivery, respectively. As a result, energy and power densities can be calculated using either CV or GCD methodologies.

Energy Density: Energy density is calculated by multiplying the cell potential difference between the negative and positive electrodes (in Volts) by the cell capacity (Demir-cakan *et al*., 2020). Because of this, it is described as amount of energy a supercapacitor is capable of storing per unit of mass or volume (George *et al.*, 2016). Cell design, which comprises entire mass of "sedentary mass" like wrapping foils, pole tabs, separators, or electrolyte, and "active mass" such as electrode materials, is what determines how much energy is stored by SCs. Specific energy density (that is, watt hour per kilogram) can be obtained when active mass of electrode is considered and incorporated into energy density equation, given as equation (3).

$$E = \frac{CV^2}{2} \quad (6)$$

$$E = \frac{CV^2}{2} = \frac{1}{2}C_sV^2 = \frac{1}{2}\frac{C}{m}V^2 \quad (7)$$

Where Cs is specific capacitance, and active mass is given as m. Equation (6) demonstrates that the specific energy density, capacitance, and active electrode mass are all closely related(Kim and Zhang, 2015b). The squared potential range (v^2) demonstrates how energy density is significantly influenced by operating voltage window(Kim *et al.*, 2015b; Pan and Feng, 2014a; Tomiyasu *et al.*, 2017). Electrolytes have a significant impact on the operational potential range of SCs, as was previously noted. In order to increase the potential window that these electrolytes can reach and so address Supercapacitors' lack of high energy density, researchers have centered their efforts on organic electrolytes in addition to ILs. However, there are some drawbacks to this as well because ILs and other electrolytes with large potential windows are known to have poor power density. Therefore researchers need to work out a way of increasing potential windows of electrolytes without reducing their power density.

Power Density: Power density describes how rapidly device sends energy to external loads when the current density is constant (Watt per kilogram). Formula in equation (8) is used to convey this(George et al., 2016; Kim *et al.*, 2015b; Pan and Feng, 2014a).

$$P = \frac{V^2}{4R} \quad (8)$$

Only when there is "matching load condition" can this criterion, represented as equation (4), be considered to have been satisfied (Zhang and Pan, 2015). When the external load equals the ESR resistance, the matched load condition is satisfied, resulting in maximum power density shown in equation (4). Yet, achieving this condition is actually incredibly challenging and rare (Zhang and Pan, 2015). Using a straightforward relationship between average power density and energy density, this results in:

$$P_{avaerage} = \frac{E}{\Delta t} \quad (9)$$

Where Δt is the cell's discharge rate, also referred to as time constant (τ) (Kim *et al.*, 2015b). This equation demonstrates how cell time constant $\tau = T = R_{ESR} \times Cs$. Directly connects energy and maximum power densities. Energy density can be raised by raising capacitance or operating voltage, but rising capacitance alone causes time constant, which will result in less reactive cells even if ESR remains constant (Zhang and Pan, 2015). Although maintaining the same value, and increasing voltage can dramatically broaden both power density and energy density (Zhang and Pan, 2015).

Self-Discharge Leakage: Self-discharge also known as current leakage is the voltage drop that occurs owing to leakage of current between electrodes especially

under open circuit condition, and it does not involve any chemical process (George *et al.*, 2016; Ike and Iyuke, 2015; Jakobczyk and Biegun, 2013; Jakobczyk and Rudnicka, 2015; Menzel *et al.*, 2020; Zhang and Pan, 2015), is used to calculate ability of ECs to sustain potential whilst not in utilize. (Jakobczyk and Biegun, 2013), in their work on electrochemical double layer capacitors self-discharge proposed three mechanisms of self-discharge, which are self-discharge due; to a leakage-current, faradaic reactions and charge redistribution(Ali *et al.*, 2018; Bissett *et al.*, 2015). Self-discharge in SCs is recorded as a compensating current in fully charged capacitors after 72hrs (George *et al.*, 2016; Zhang and Pan, 2015).

Self-discharge, also referred to as current leakage, is the voltage drop that results from current leaking between electrodes, particularly when the circuit is open (George et al., 2016; Ike and Iyuke, 2015; Jakobczyk and Biegun, 2013; Jakobczyk and Rudnicka, 2015; Menzel *et al.*, 2020; Zhang and Pan, 2015). Self-discharge is used to determine how well SCs can maintain their rated potential when not in use.

Main types and structures of carbon based super capacitors

Electric double-layer capacitors (EDLCs): Electrostatic charge in EDLC is generated and discharged by depositing or removing electrons from either the negative or positive electrode. One of the electrodes becomes negative as the EDLC is connected to a power source (charging), and an electric field forms there that attracts negative electrons and repels positive electrons. At the positive electrode, the opposite is true. The electrolyte's ions and electrode's electrons are held together by concentrated electric field; when discharged, the opposite happens. When a charge double layer forms and potential energy is stored, this is referred to as development of charge double layer where energy that is stored, is potential energy(Soheila *et al.*, 2014; Aziz *et al.*, 2019). According to equations (1) and (2), large surface area, great porosity, and good pore dispersion are required to achieve good capacitance in EDLCs (Zhang *et al.*, 2019). Carbons of diverse forms have been used as precursors for production of electrodes in EDLCs, such as activated carbon(Misnon et al., 2015), carbon nanotubes (CNTs) (Chen *et al.*, 2013; Che *et al.*, 2014; Salisu *et al.*, 2018), and carbon aerogel s(Aken and Pérez, 2015; Moreno-Castilla *et al.*, 2012).

Activated Carbons (ACs): An efficient material that has found numerous use in various fields is Activated carbon.(Taer *et al.*, 2018). One of such areas is production of electrodes for EDLCs when large surface area is needed. This procedure was demonstrated by(Taer *et al.*, 2018), they produced seven types of activated carbon electrodes using banana peel waste, the electrode synthesis was made by conditions of carbonization and activation. Results obtained at the end of experiment, for specific surface area (S_{BET}) $581 m^2/g$, specific capacitance (C_{sp}) $68\ ^F/_g$, specific energy$0.75\ ^{Wh}/_{kg}$, and specific power$31\ ^W/_{kg}$. (Salisu *et al.*, 2018), showed the relationship between ACs and activating agents in their work. They employed ACs made from oil palm leaves (OPL) and palm kernel shells (PKS) with H_3PO_4 as activating agent at various concentrations. According to Brunauer-Emmett-Teller (BET) studies they obtained, surface area reduces when H_3PO_4 concentration drops. This shows that activating agents like H_3PO_4 can influence the surface properties of ACs. This according to(Shi *et al.*, 1995; Barbieri *et al.*, 2005), is caused by the reduction in concentration of acidic surface functional groups and oxygen content. When electrode produced was tested with cyclic voltammetry, it gave capacitance value of$434 Fg^{-1}$. The gas absorption volume, and BET specific surface area data obtained showed surface area of activated carbon materials (ACMs) and electrochemical properties relates significantly. Theoretically and experimentally, a number of factors sway electrochemical performance of carbon material especially, pore size, pore distribution, and pore volume as was shown by(Misnon *et al.*, 2015). ECs characteristics of AC made from oil palm kernel shell (PKS) were weighed against those of other biomass-derived ACs. They carbonized PKS by pyrolysis, afterward activated PKS by physical and chemical procedures. Result obtained showed chemically AC had wider pore distribution while physically activated AC has identical pores. This was in line with findings of (Wu *et al.*, 2005), which states that, chemically activated ACs usually has bigger pores and more mesoporous whereas the physically activated AC has identical ink-bottle type of pores. To assess the electrochemical characteristics of two forms of AC, they employed CV, CDC test, and EIS in three electrode configurations. At the end of experiment, they obtained these results; specific capacitance for chemically and physically ACs (C_{sp}) were($210\ F/g\ and\ 123\ F/g$), respectively. The PKS AC EIS, results gave low series resistance, which implies fine power density in EDLC showing relationship between power and ESR as displayed in equation (3). These ability of EDLCs not to lose capacitance can be attributed to two factors according to (Frazier and Burkett,.2011). Firstly, there is a short transfer distance of ions through the EDLCs solution, which gives them good capacitive values as established by equation(1). Secondly, the production style applied in

making their good graphitic characters, making them conducting materials. Yet, as they become more hydrophobic due to their increased graphitic character, amount of water that can enter their pores reduces, lowering their effective surface area and, consequently, the material's capacity when utilized in aqueous electrolytes. These factors explain why most common activated carbon electrodes have poor electrical conductivity and little electrolyte accessibility (Frazier and Burkett, 2011). As a result, carbon nanotubes (CNTs) are proficient candidates to substitute carbon materials.

Carbon Nanotubes (CNTs): The storage mechanism utilized by carbon materials in EDLCs is produced at periphery connecting electrode and electrolyte, this character, can be enhanced to obtain large capacitance when given a large surface. But, surface area alone cannot sufficiently control or influence electrochemical properties of EDLCs. As a result, additional crucial elements such pore shape, structure, pore size distribution, surface functioning, and electrical conductivity cooperate to influence an EDLC's electrochemical capabilities (Iro *et al.*, 2016). Majorly, CNTs exists in two forms, single-walled nanotubes (SWNTs) and multi-walled nanotubes (MWNTs). SWNTs walls are associated with an atom layer thickness while MWNTs walls encompass multiple layers of graphitic carbon. SWNT often appears as single graphene sheet spin on itself to form a tube. Whereas, MWNTs is often metallic, or semiconductor materials depending on sheets that are used in making the rolled tubes, which could be zigzag, armchair and chiral forms (Zhai *et al.*, 2011; Baughman *et al.*, 2002). CNTs either as composites or non-composites are good materials for electrode production owing to their distinctive pore structure, mechanical strength, and ability to withstand heat, advanced electrical properties along with ability to be modified or improved upon. (Chen *et al.*, 2017). These enhanced performances have attracted attention of researchers into looking at CNTs as suitable materials for supercapacitor electrodes, as was established by(Niu *et al.*, 2012), they produced a superior CNTs by disassembling a catalytically grown CNTs made from nitric acid, the disassembled CNTs was annealed to produce a new functionalized electrode that is randomly entangled, thermally cross-linked with uniform thickness. Also, no binder was needed given that functionalized carbon nanotubes are self-binding. Due to this modifications and treatments, it was observed, that surface area improved from 250 to $430 m^2/g$ with 38 wt.% H_2SO_4 in water as electrolyte, and specific capacitance of $102\ ^F/_g$ was attained. This high value is attributed to entangled mesoporous nanotube network granting easy access to ions at electrode/electrolyte interface for double layer formation, and technique applied (Riccardo *et al.*, 2009). As stated, CNTs can be customized to suit different need performance of electrode materials. This modification to serve has been demonstrated by modifying the chirality of nanotubes to produce single walled or multiple walled nanotubes, and has equally given both types of nanotubes some comparative rewards and shortcomings as long as production and usage is considered. SWNTs are made to have high degree of purity and MWNTs can be synthesized to take different forms (by doping with other materials to have multiple functionalities added to their structures) and shapes. These functionalities facilitate improved compatibility to an electrolyte to make best use of electroactive surface area and thus improve performance(Dennis et al., 2013). Surface area to a large extent determines capacitance of an electrode material. The surface area, when fully utilized leads to high capacitance. Again, high surface area does not lead to high capacitance for reason that capacitance also depends on some properties like pore size distribution and electrical conductivity. This implies that to optimize capacitance, a regular pore structure, large pore size, good pore distribution and a suitable electrolyte is desirable. Studies have shown that well aligned CNTs perform better than their random counterparts when applied electrochemically(Wen., 2010). The aligned structures make available superior charge storage/ delivery properties, for the reason that the constituent aligned tubes connects individually to electrode in order to allow them contribute effectively to charging/discharging process (Chen and Dai, 2013). In the report by (Lu *et al.*, 2009) vertically aligned carbon nanotubes was used as electrodes in synergy with ecofriendly ionic liquids (ILs) as electrolytes. CNTs structure were perpendicularly line up and properly spaced in ionic liquid electrolyte. Plasma etching was used to form an aperture at end tips of nanotubes thereby, initiating defects and creating oxygen functionalized nanotubes, this act makes available distinct surface area for every constituent tube to be easily reached by ions, furthermore enriching capacitive performance of carbon nanotubes. Additionally, aperture at end tips of nanotubes given right condition like Plasma etching, permit electrolyte contact to inner cavity of tubes. It has been discovered that altering shape of electrodes has an effect on supercapacitor parameters even though area of electrodes remains constant. The electrode shape allows electrolytes ions to access surface area of electrodes thereby varying capacitance and other parameters. Research connected to this was conducted by (Singh and Karandikar, 2015). They discovered that making and designing electrodes

based on shape has strong impact on it usage in practice. When designing electrochemical materials, it is beneficial, to have patterned structures for CNTs so their property can be easily accessed making it easy for them to be successfully integrated into devices (Azam *et al.* , 2013; Azam *et al.*, 2013). This denotes an increased energy density for capacitor due to collective charge from all individual tubes of aligned electrode. In turn, ability to quickly deliver stored energy through each electrode tube results in outstanding power density for capacitor (Wen, 2010). Surface condition of carbon nanotubes affects its capacitive performance to a large extent. CNTs chemically tailored by strong acid oxidation have established pseudocapacitive behavior ensuing from Faradaic redox reactions of rich surface functionality leading to higher capacitive values(Du and Pan, 2007). Nevertheless, capacitance and stability of pure CNT-based supercapacitor does not solely depend on surface area alone as earlier stated, but it is also affected by factors like specific surface area, pore size, pore distribution, conductivity and shape engineering of electrode etc. these factors, need to be optimized and improve upon using some of these suggested methods like doping, oxidization, functionalization and combination with other materials to form composites. The usage of CNTs as supercapacitor materials is currently constrained by their low inherent capacitance and low energy density. Modifying CNTs to greatly improve their electrochemical characteristics is one way to address these shortcomings. Redox-based materials such as ruthenium oxide (RuO_2), cobalt oxide (Co_3O_4), manganese oxides (MnOx), nickel hydroxide ($Ni(OH)_2$), and others can be combined with CNT to achieve this goal (Ates and Fernandez, 2019; Deshmukh *et al.*, 2013; Fan *et al.*, 2019; Tao *et al.*, 2015). Metal oxides when added to make better specific capacity behavior of CNTs, form composite electrode by bringing in their reversible redox reaction, allowing them to store and release more electric charge; much more than is obtainable in carbon materials alone (Hepel, 2016). Composites conducting polymers and carbon nanotubes also show huge potential when used as electrode materials.

Carbon Gels: By using the right components and formulation, the surface area, porosity, and surface chemistry of carbon gels can be altered. These materials can be made from high-purity components and can be shaped in various ways, such as pellets, microspheres, irregularly shaped powders, and films (Zapata-Benabithe *et al.*, 2016). This has been proven by Zapata-Benabithe *et al.* (2012), who created carbon aerogels by carbonizing organic aerogels from polycondensation reactions of formaldehyde, resorcinol, and pyrocatechol under basic or acid conditions, respectively. The resulting carbon aerogels were then treated with KOH to activate the surface and included surface oxygen and nitrogen functionalities. KOH activation resulted in creation of surface area with sizable gravimetric capacitance, of$1935\,{}^{m^2}/_{g}\ and\ 220\,\frac{F}{g} respectively$. To have better or increased performance, carbon gels can also be used to create composite materials. This was shown through (Dang *et al.*, 2016), in their research, single-walled carbon nanotube (SWCNT) thin films were created using aerosol deposition, then used as EDLC electrode. SWCNT films displayed specific capacitance of $178F/g$ and mass specific capacitance of${}^{482F}/_{g}$. Numerous production techniques give room for manipulation of concentration of carbon aerogels pores independently, making aerogels fitting material for electrode production. However, if a metal precursor is present, it causes a change in pH, pyrolysis, activation, etc., making it challenging to control consistency of pore (Sumaiyah, 2019). CNTs are known to offer both high surface area and good specific-conductance (Zhai. *et al.*, 2011), drawbacks such as high cost of production amid stressful production path, have restricted it scale-up process for larger scale manufacturing (Genc *et al.*, 2017). Other disadvantages were also outlined by (Mensah-Darkwa *et al.*, 2019). It is therefore necessary; to conduct more research on carbon based electrode materials, even as other materials with pseudocapacitive qualities and potential to overcome these shortfalls experienced by carbon materials are looked into.

Pseudocapacitors: EDLCs are distinct from pseudocapacitors or redox Faradaic supercapacitors. Given that swift and reversible redox processes (Faradaic reaction) occurs on/in electrode materials when an electric potential is supplied to a pseudocapacitor, this results in transfer of charge across double layer (Gupta, 2017; Salinas-torres et al., 2019). Surface redox pseudocapacitance and intercalation pseudocapacitance are two categories under which pseudocapacitance materials fall (Zhang *et al.*, 2019). electrode material for pseudocapacitors are MnO_2 which was studied by (Brousse *et al.*, 2006; Yang *et al.*, 2013), $RuO_2{\cdot}nH_2O$ (Ates and Fernandez, 2019; Frazier and Burkett, 2011)and conductive polymers (Shi and Yu, 2016; Vlad *et al.*, 2018) etc. In their research (Wang & Hu, 2005), examined impact of composition on capacitive qualities and textural aspects of binary nanostructured hydrous ruthenium-tin oxides ($(Ru\text{-}Sn)O_x.nH_2O$), produced by modified sol-gel technique, found that $(Ru\text{-}Sn)O_x.nH_2O$'s capacitance and redox reversibility improved after being annealed in air for two hours at temperatures

between 150 and 250°C. They achieved total specific capacitance of $690\ F/g$. The improved electroactive sites of deformed RuO_2 nanocrystals are responsible for high specific capacitance achieved. Despite hydrous ruthenium oxide's advantages as electrodes, their shortcomings—such as a lack of shape support, conductivity, crystallinity, adhesion, etc.—limit their performance as supercapacitors and prevent future advancement (Li *et al.*, 2018). Manganese dioxide(MnO_2) compounds with various structural variations were produced by (Brousse *et al.*, 2006), they achieved capacitance for group of MnO_2 compounds with greater Brunauer-Emmett-Teller (BET) surface areas$150\ F/g$, $125\ m^2/g$ respectively. Their research demonstrated that increasing surface area does not increase capacitance, and that capacitance depends more on structure than surface area. For most part, conducting polymers are viewed as electrode material of future for supercapacitor production due to two factors: (1) they provide high specific capability, which does not end at surface alone but involves bulk materials in mechanism of charge storage; and (2) they have a high conductivity in charged state, especially when combined as composites (Gupta, 2017).

In their analysis of carbon nanotubes and conducting polymers, Peng et al. (2008) and colleagues demonstrated that these materials exhibit enhanced mechanical, electrical, and electrochemical properties better than conducting polymers alone, resulting in range of applications. Express electron transfer is possible between redox species in electrolyte and electrode material, because electrolyte's redox processes may facilitate transport of electrons to redox active electrode. (Akinwolemiwa *et al.*, 2015). This was shown by Hashemi *et al.* (2018), who presented technique for rising energy density through in situ electrocatalytic redox additive-assisted regeneration. They used nanostructured conjugated polyaniline electrode and quinone-based redox electrolyte to accomplish this, they repeatedly regenerated reactants to produce redox supercapacitor with high energy density of $1091\ ^{Wh}/_{kg}$ high power density of$196\ ^{kW}/_{kg}$, and 84% capacity retention after 7000 cycles at$35\ A/g$. Electrolyte cations(Na^+, K^+, Li^+, H^+, etc.) intercalate or deintercalate layers of electrode materials in conjunction with faradaic charge transfer that doesn't involve phase shift (Yu *et al.*, 2018; Zhang *et al.*, 2019). When compared to EDLCs, their faradic process-connected pseudocapacitor enables superior specific capacitance and energy densities (Iro *et al.*, 2016).

Electrolyte Materials for Electrochemical Supercapacitors: Because of the crucial roles that electrolytes play, electrochemical capacitors (ECs) require them to have wide voltage window, good electrochemical stability, low resistivity, low toxicity, and other benefits (Aiping *et al.*, 2015). According to Morallón, (2016), solvent used to create an electrolytes has a significant impact on working voltage of supercapacitors. Additionally, relationship between electrolyte and electrodes and electrolyte's ionic conductivity play a significant role in internal resistance. increase in resistance associated with reduced cycle life is linked to poor electrolyte stability and poor chemical stability(Aiping *et al.*, 2013). As can be seen in equations (3) and (4), the best route to enhancing energy density of ECs, is increasing value of voltage (V) and reducing resistivity as much as possible. Consequently, cautious selection of appropriate electrolyte is essential in assembling high performing ECs (Kim *et al.*, 2015). An ideal electrolyte should have low volatility, cheap cost, low flammability, low degradation potential, wide range of operating temperature, low viscosity broad potential window, and strong electrochemical stability to ensure safe usage and operation of ECs. Yet, as will be seen, no single electrolyte can possess all of these desirable characteristics. The types of electrolytes now are aqueous, organic, and ionic liquids (ILs) (Qin *et al.*, 2020).

Aqueous electrolyte: High ionic conductivity, cost efficiency, non-flammability, non-corrosiveness, intrinsic safety, and ease of assembly in air are all benefits of these types of electrolytes (Kim *et al.*, 2015b; Zhao and Zheng, 2015). The majority of researchers frequently employ aqueous electrolytes including aqueous potassium chloride (KCl), H_2SO_4, KOH, Na_2SO_4, and others. They come in acid, base, and neutral electrolyte forms, giving researchers a variety of alternatives for better functionality and application (Kim *et al.*, 2015b; Ramachandran. and Wang, 2018). According to research, carbon-based capacitors (EDLCs) can have their energy density enhanced by switching from an alkaline-acidic electrolyte to one that is more neutral. This was shown by Li *et al.* (2019), they put together symmetric carbon-based SCs, with electrodes made of activated carbon derived from biomass and an alkaline-acidic electrolyte. With remarkable cycle stability over 10,000 cycles and high stable working voltage, this aqueous symmetric SC demonstrated outstanding energy density of $36.9\ ^{Wh}/_{kg}$ at $248\ ^{W}/_{kg}$which is significantly greater than typical energy and power densities $8.8\ ^{Wh}/_{kg\ and}\ 4083\ ^{W}/_{kg}$ respectively for aqueous symmetric SCs. Most aqueous electrolytes

have two fundamental drawbacks that prevent them from being used in ECs: (1) a narrow voltage window (i.e., narrow ESW) and (2) an incompatible electrode potential range (Gou *et al.*, 2020). According to Demir-cakan *et al.*, (2020); Dushina (2016); Gou *et al.*, (2020), the limited thermodynamic voltage stability of water, which is ranged at 1.23V and is dependent on pH range; over which water decomposes, is what causes narrow voltage window. Numerous researchers have employed a variety of techniques to get around this limitation of the narrow voltage window. Some of these techniques include the use of redox active additives (Akinwolemiwa *et al.*, 2015; Wang *et al.*, 2015), fine-tuning of pH through the use of combined electrolytes (Gorska *et al.*, 2019), employing water-in-salt electrolytes (Qin *et al.*, 2020; Sun *et al.*, 2017). The drawback of incompatible electrode potential range typically results from a different energy storage procedure (Gou *et al.*, 2020). Increasing the ESW of electrolyte, improving characteristics of electrodes, and creating aqueous asymmetric supercapacitors can all help to combat this (Gou *et al.*, 2020; Tomiyasu *et al.*, 2017; Wan *et al.*, 2020).

Organic electrolytes: In order to determine rate capability of SCs, an electrolyte's ionic conductivity must be high and its electronic properties must tolerate wide electrochemical window, (Balbuena and Balbuena, 2016; Zhang *et al.*, 2020). Supercapacitor charge loss is compensated for by electrolytes, which also aid in improving ionic conductivity and, thus, energy density. Equation (2) demonstrates that if electrode materials are within working voltage range, electrochemical window of ECs depends on electrolyte utilized. Typically, operating potential range for organic electrolytes is between 2.5 and 2.7V. Conductive salts and organic solvents make up the natural system of this electrolyte. Acetonitrile and propylene carbonate are two organic solvents that are most frequently used (Aiping *et al.*, 2013; Aza *et al.*, 2007; Kim *et al.*, 2015b; Pan and Feng, 2014b; S. Wang *et al.*, 2018). Tetraethyl ammonium tetrafluoroborate (TEABF4) (Aza *et al.*, 2007; Zhang *et al.*, 2016), and $LiPF_6$ are most often utilized salts with these solvents. Organic electrolytes in manufacturing of ECs are constrained by few drawbacks. These limitations include need for controlled environment (water-free atmosphere) for production and use of organic electrolytes, which drives up cost of overall electrolyte manufacturing. Another limitation is that process known as thermal runway causes an electrolyte's temperature to rise steadily often causes device to vaporize, catch fire, or explode (Sanchez, 2012). They also struggle with high ESR, which results in a substantially higher self-discharge (Aiping *et al.*, 2013), in addition to macroscopic events like gas evolution, an increase in mass of electrodes, local separation of coating layer from metallic collector, etc., supercapacitors (SCs) made from activated carbons and organic electrolytes may start to show capacity fading and resistance increase over time. So, in order to avoid such drawbacks, it is crucial to study SCs built of organic electrolyte and its ageing mechanisms (Aza *et al.*, 2007).

Ionic Liquids (ILs): Salts having cations and anions and melting temperatures lower than 100 °C are referred to as ionic liquids (Ferraz *et al.*, 2011; Rogers and Voth, 2007). When an organic symmetric or asymmetric organic anion combine to form ILs, a combination of these cation and anion type results in ILs having a low melting point (Cheng *et al.*, 2015) according to research, ILs can be customized into a wide range of compositions depending on needs of user. By carefully choosing component ions or mixing component ions, desired physical, chemical, and biological properties is obtained (Rogers and Voth, 2007). ILs are useful in diversity of fields and industries, including engineering, chemistry, and pharmaceuticals, due to unusual physical and chemical properties, which include high thermal and chemical stability, which allows for operation at high voltage windows up to 5V, non-flammability, low volatility, and ability to dissolve in and mix with several organic compounds (Castner and Wishart, 2010; Cheng *et al.*, 2015; Deng *et al.*, 2013; Ferraz *et al.*, 2011). Imidazolium, pyrrolidinium, ammonium, sulfonium, phosphonium, and other elements are frequently used as ILs cations for ECs. The anions are dicyanamide (DCA^-), bis(trifluoromethanesulfonyl)imide ($TFSI^-$), bis(fluorosulfonyl)imide (FSI^-), bis(trifluoromethanesulfonyl)imide ($TFSI^-$), tetrafluoroborate (BF_4^-), hexafluorophosphate (PF^-_6), and bis(fluorosulfonyl (Cheng *et al.*, 2015). Because of high viscosity, poor ionic conductivity, and high cost, ILs is frequently disadvantaged, which limits their use in creation of ECs (Aiping *et al.*, 2013). As was already said, the improved ion mobility (kinetic energy) that results from ILs' strong thermal stability allows them to overcome the problem of low ionic conductivity at high temperatures, which raises conductivity and increases device power and response time (Aiping *et al.*, 2013). By combining ionic liquids with other electrolytes, such as propylene carbonate and acetonitrile, and by making sure that high potential windows of ILs and improved conductivity and power of organic electrolytes are properly balanced, it is possible to overcome the low conductivity of ILs (Aiping *et al.*, 2013; Ferraz *et al.*, 2011; Yu and Chen, 2019). Using such a mixture can increase security,

lessen toxicity, and create a device with a high energy density (Aiping *et al.*, 2013).

Conclusion: Considering the demands for superior energy density, power density and prolonged cycling life, scientists seem to believe that designing top-notch electrode materials is key to bring out full potential of SCs. Studies reviewed in this research paper indicate that choice of electrode material is most critical factor in performance of SCs. Carbon materials served as primary energy-storage mechanisms for electrode materials in EDLC materials, while conducting polymer electrode materials and transition-metal oxides/hydroxides serve as primary energy-storage mechanisms for pseudocapacitors. When created as individual electrodes, these electrodes suffered from flaws such as low energy density and expensive fabrication costs. Two strategies are recommended in order to get over these drawbacks. First, the development of novel materials for electrodes and knowledge of ion transport process in pores of electrode materials needs to be studied and improved upon in order to enhance total capacitance. It is also necessary to employ composite electrodes and electrolytes, adding redox additives to traditional electrolytes, or use IL electrolytes in addition to appropriate or proper selection of electrodes. Second, usage of asymmetric supercapacitor integrating electric double-layer as anode and cathode for redox reaction is recommended to increase cell voltage. However, compared to batteries, research on electrode materials for SCs has advanced significantly, and it has led to many creative innovations on how to advance effectiveness of supercapacitors.

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