

## Review Article

# Capacity Imbalance and Diffusion Kinetic Between the Electrodes of Hybrid Supercapacitor: A Review

S.E. Umoru<sup>1\*</sup> 

<sup>1</sup>Dept. of Natural Science, National Institute of Nigerian Languages, Aba, Nigeria

\*Corresponding Author: [efemenamoru@gmail.com](mailto:efemenamoru@gmail.com)

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**Abstract**—The need for energy storage devices that have significant power and energy density, outstanding cycle stability and charging and discharging cycles, affordability, and excellent environmental protection has propelled the creation and advancement of hybrid supercapacitors. Because hybrid supercapacitor utilizes two types of electrodes made of different materials, the mechanisms of charge storage are also different and hence there are gaps in terms of capacity imbalance and diffusion kinetics. This work reviews the different promising methods or techniques that can be used to tackle these problems militating against the performance and commercialized applications of hybrid supercapacitors. The control of electrodes open circuit voltage (OCV), utilization of redox couple materials, utilization of water in salt (WIS) electrolytes, the use of core-shell hybrid nanostructures, mass and Charge Balancing approach, Electrodes' structural design and control, incorporating of two-dimensional carbon materials, utilization of Materials with Good conductivity, choosing an appropriate electrolyte are some of the different techniques reviewed in this work. This article also examines the difficulties associated with several approaches for future study aimed at enhancing hybrid supercapacitor functionality.

**Keywords**—Hybrid supercapacitor, Energy storage device, Electrode, Electrolyte, capacity imbalance, diffusion kinetics,

## 1. Introduction

The amount of energy output alone won't be enough to meet the challenges brought on by the present energy crisis but also the time of production. Consequently, a long-term sustainable strategic plan is needed to accommodate the rising demand for commercial energy. Even when advances are made to alternative power producing techniques and sources that can offer renewable energy, the inclusion of devices that can store energy will be demanded to sustainably store and distribute the energy produced over time [1]. Electronics that may store electric energy include capacitors and batteries.

Batteries are electrochemical devices that allow the storage and release of electrical energy. But because of the electrochemical procedure that is going on inside the device, the energy content is high but possesses low power content. Electricity is stored in capacitors via an electrostatic charge storage process. It charges quickly and immediately releases the stored charge. Capacitors consequently produce more electrical power and have a longer lifespan than batteries with less degradation due to their charging process. Comparatively speaking, ordinary capacitors possess lesser energy density than batteries, which leads to a lower usage rate than batteries.

On the other hand, supercapacitors are the advanced forms of the capacitor with cutting-edge designs. Interest in supercapacitors have grown during the last few years owing to its great power weight, long longevity, and environmental safety [2,3]. The use of supercapacitors in commercial level have been limited because of their low energy density [4,5]. But in the other way, Rechargeable batteries have a wider range of applications. Hybrid supercapacitors were developed to close the low difference between the energy densities of batteries that can be recharged and supercapacitors.

### 1.1 Causes of capacity and diffusion kinetic imbalance in hybrid supercapacitors

To bridge the minor disparity in energy density between rechargeable batteries and supercapacitors, hybrid supercapacitors were created. This made it possible to combine mechanisms that are faradaic and those that aren't in a single storage device [6]. The materials made of transition metal, which store electrical charge through the processes of faradaic and redox reaction, were used to create the battery-type electrode, whereas the materials made of activated carbon with which electrical charge storage is possible through electrostatic double layer mechanism are the primary materials for the capacitive-type electrode. This has led to imbalance in terms of capacity and diffusion kinetics the

between the types of electrodes [7,8]. Better stability as well as faster diffusion kinetic is typically provided by capacitor-type electrodes, while battery-type electrodes typically give greater capacity [9, 10]. The charge-storage process in battery-type electrodes is controlled by the kinetics of metallic ion diffusion and the cyclical intercalation/de-intercalation of metal ions to/from the electrode [11, 12]. It is well known that the adsorption or desorption of ions to the electric double layer of the capacitive electrode, for example, is a capacitive energy storage process that happens much faster than intercalation (redox reaction). [13]. Because different electrode types (battery and capacitive type) are used in one hybrid supercapacitor device, Bailmare *et al.* (2022)[14] found that the electrical charge storage mechanisms between the two different electrodes are also different [15]. Removing these limitations is one requirement for enabling hybrid supercapacitors to operate effectively. According to Syed *et al.* (2023) [16], attention must be given to the limitations in order for the two electrodes in the hybrid supercapacitor to work together and enhance performance. . This review study is divided into sections. Section 1 gives the introduction of the whole study, section 2 highlighted some related strategies that have been employed to enhance the performance of hybrid supercapacitors, Section 3 contains the strategies needed to balance the capacity and diffusion kinetics between the electrodes of hybrid supercapacitors. section 4 is the concluding part of the work.

## 2. Related Strategies that Have Been Employed

Numerous initiatives have been made to solve these problems associated with the electrodes of hybrid supercapacitors by modifying the composition, shape, and porosity of performance-enhancing electrode materials [17]. Modifying nanostructures, ionic fixing, and surface effect are some of the ground-breaking technologies [18]. Many hybrid supercapacitor devices with exceptional performance have been made possible by the thoughtful application of these technologies [19]. Table 1 shows a few excellent examples along with the energy density of the most recent devices that have been suggested. It can be seen from table 1 that different materials have been combined to form the hybrid structures. The performance of the device depends on the material composition and the electrolyte used as depicted in table 1.

Table 1: hybrid supercapacitors that were reported.

Device	Electrolyte	Cell Voltage/V	Energy Density/Wh kg <sup>-1</sup>	Power Density/kW kg <sup>-1</sup>	Ref.
LDH-NPs/CH-NWs //AC	6 M KOH aqueous solution	0 ~ 1.6	58.9	51.5	[20]
PSNC-3800//PSOC-A	1M NaClO <sub>4</sub> in EC/DEC	1.5 ~ 4.2	201	16.5	[21]
CTAB-Sn(IV)@Ti3C2 //AC	1 M LiPF <sub>6</sub> in EC/EDC	1.0 ~ 4.0	105.6	10.8	[22]
GNSs//HN-PPs	1 M LiPF <sub>6</sub> in EC/EDC	1.0 ~ 4.8	265	5	[23]
Sn@NRT//NRT	1 M LiPF <sub>6</sub> in EC/EDC	1.75 ~ 4.35	274	22.8	[24]
CoxNi1-x(OH) <sub>2</sub> @rGO//	2 M KOH aqueous	0 ~ 1.6	72	16.7	[25]

PPD/rGO	solution				
3DFC//3DFAC	1M NaClO <sub>4</sub> in EC/DEC	0 ~ 4.0	111	20	[26]
V-FG//MnO <sub>2</sub>	1 M Na <sub>2</sub> SO <sub>3</sub> and 1 M Na <sub>2</sub> SO <sub>4</sub> aqueous solution	0 ~ 2.0	75	25	[27]

The study of hybrid supercapacitors is now going through a thrilling phase of development with impressive advancements made. Several outstanding review articles provided an overview of recent developments with a particular emphasis on electrode materials [28]. According to (Zoller *et al.* 2019)[29] “Florian *et al.* created flexible composite paper based on molybdenum disulfide using straightforward vacuum filtration and high-energy shear force grinding, demonstrating its benefits for electrochemical energy storage.” Similarly, According to (Aravindan *et al.* 2014) [30], Madhavi *et al.* specifically analyzed insert form of electrode materials for hybrid supercapacitors. As of now, there are challenges to overcome in the technique of enhancing the efficiency of hybrid supercapacitors by the modification of electrode materials (Xia *et al.* 2020) [31]. Other approaches to improving the hybrid supercapacitors' practical performance have been put forth. These techniques, which go beyond conventional approaches on electrode materials, have focused particularly on other significant but not well-known factors, like the open-circuit voltage (OCV) of electrode as well as electrolyte composition [32, 33, 27]. Achieving a great deal of energy and power performance in hybrid supercapacitors has been made possible by rational manipulation of these factors [34, 27]. This hot topic has already generated interest across the globe in recent years. Therefore, there is an urgent need for an update on the state of research in this exciting field. Table 2 highlighted some studies carried out in this field.

This review study will primarily pay attention on these cutting-edge approaches that go beyond material engineering methods in order to enhance the efficiency of hybrid supercapacitors.

Table 2: Study on enhancing the efficiency of hybrid supercapacitors

Reference	Study method	Energy density/Wh kg <sup>-1</sup>
[35]	Cobalt-nickel double hydroxide supercapacitor design for flexible asymmetric capacity	42.5
[36]	Li-ion capacitor made of porous graphene macroform and hybrid Li <sub>4</sub> Ti <sub>5</sub> O <sub>12</sub> .	72
[37]	Highly-Efficiency Anode: Mesoporous Niobium Pentoxide/Carbon.	74
[38]	Composite of 3D Porous VN Nanowires and Graphene	64
[39]	Interconnected Chain Anode of Titanium Carbide Nanoparticles	67.5
[40]	Electrochemical capacitors with improved redox state	100
[41]	active electrolyte	113
[33]	Controlled Electrochemical Charge Injection	175
[42]	Nano-sized pyroproteins Redox-active heteroatoms found in pyroproteins.	217

[43]	“Water-in-Salt” Electrolyte	133
[44]	Using electrolytes made of water-in-salt as electrode	233

### 3. Strategies to Balance the Capacity and Diffusion Kinetics Difference In hybrid supercapacitors' Electrodes

Capacity imbalance refers to the situation where each electrode of a hybrid supercapacitor has different level of charge storage capacity. This can occur due to variations in electrode material properties, manufacturing processes, or other factors. A capacity imbalance may cause the hybrid supercapacitor to operate poorly overall and inefficiently. Diffusion kinetics on the other hand refers to the process of ions diffusion within the hybrid supercapacitor's electrode materials. It has a significant impact on how well the device performs. Several elements including the electrode material's conductivity, surface area, and pore size dispersion. Hybrid supercapacitors typically utilize various materials for their cathode and anode electrodes to optimize their energy storage capacity. However, these materials can possess various charge storage mechanisms that can bring about imbalance of the specific capacity and diffusion kinetic between the electrodes. Therefore, kind of materials used to make electrodes for hybrid supercapacitors is a significant factor in determining how well they can work. According to Sahil & Kushal (2023) [45] and Nimra *et al* (2023) [46], a lot of work has been put into this field of increasing hybrid supercapacitors' effectiveness over the last 10 years, and significant advancements have been made.

By controlling OCV, the charge distribution between the electrodes of hybrid supercapacitors can be balanced and address the issue of capacity imbalance. Developing advanced control and monitoring systems that can accurately measure the OCV of each electrode and make the required modifications is essential. A feedback control system can also be used to minimize capacity imbalances by ensuring that each electrode receives an equal amount of charge.

#### 3.1 Control of the open circuit voltage (OCV) of the electrode

When there is no external load attached to the hybrid supercapacitor, the voltage differential between the electrodes' positive and negative terminals is referred to as the open circuit voltage (OCV) of that electrode. It is the measure of the electrochemical potential difference between the two terminals. The open circuit voltage can play a role in addressing the problems of capacity imbalance and diffusion kinetics between the electrodes of hybrid supercapacitors [47, 48,49]. The use of hybrid architecture is the most advantageous method for expanding the voltage range of supercapacitors [50, 51]. However, as the real devices' practical voltage range rarely aligns well with the maximum voltage that can be reached, there is frequently a waste of profitable potential range [52]. Theoretically, the electrode degradation voltage and electrolyte dissolution voltage at their respective opposite ends should be the only limits on the device's available voltage window [33]. In reality, several

variables limit this range, including the OCV of the electrode. The electrode's OCV is a factor that can affect the voltage range in actual operation [52, 53, 54, 55]. For instance, in lithium-ion capacitors, the negative electrode (capacitive electrode) typically operates in the capacitive mechanism and provides a capacity that is less significant than that of the anode electrode (battery electrode). To be able to allow for the equilibrium of electrical charge, the battery-type electrode is limited to function within extremely little voltage variation [56]. It is clear that a realistic lithium ion capacitor using the electrolyte of a lithium ion battery has a maximum theoretical voltage of 3.0 V [57]. With this kind of asymmetric bulk loading of these electrodes, the voltage range on ground can almost be utilized completely. Overall, the voltage range as previously indicated [58, 33] is the only way to raise the specific capacity of hybrid supercapacitors. Additionally, this technology may result in a progressive decline in power capacity. To specifically control the voltage of the hybrid device when the circuit is open, the operating voltage needs to be adjusted since the operating voltage is a function of the OCV [59]. This can be achieved by first raising the cell's initial voltage to a certain value using a reduced current volume. Before recharging to boost the voltage, it is first allowed to rest for three hours at an open circuit voltage. A hybrid device with outstanding capacitance performance can be created by gradually raising the voltage to 4.5 V and then relaxing, with the cathode operating between 1.5 and 4.5 V and the anode operating between 0 and 1.0 V [59]. At last, a hybrid supercapacitor that possesses an exceptional capacitance functionality and the positive electrode operating in 1.5 ~ 4.5 V may be achieved by slowly increasing the voltage to 4.5 V and slowly relaxing. The possibility of OCP modulation may result from variations in the self-discharge rates of the two electrodes, as suggested by (Du Pasquier *et al.* 2002) [60]. Koji Yamada *et al.* first proposed this idea in the manufacture of LICs [61]. They operated the LIC's AC cathode in the range of 1.5 to 4.5 V vs. Li<sup>+</sup>/Li, as opposed to 3.0 to 4.5 V, as they did in their research. The energy and power densities as a result were much higher. That being said, the authors did not reveal how the cathode stored charge in the extended potential range or how the OCP was adjusted. In a similar development, Lingfang Chen *et al.*(2019) [62] reported an aqueous asymmetric supercapacitor (Fe<sub>2</sub>O<sub>3</sub>/MnO<sub>2</sub>//rGO/Fe<sub>2</sub>O<sub>3</sub>) which operate with a voltage window of 0 ~ 1.2V was made to operate at a voltage window of 0 ~ 2.4. The authors stated that “the as constructed ASC show energy density of 57.0 Whkg<sup>-1</sup> and outstanding cycling stability (with 88.9% retention at 10 A g<sup>-1</sup> after 10,000 cycles) and good rate performance”. Furthermore, good capacitance, ratibility, and voltage window (2.3 V) are displayed by the Fe<sub>2</sub>O<sub>3</sub>/MnO<sub>2</sub>//rGO/Fe<sub>2</sub>O<sub>3</sub> all-solid-state Asymmetry supercapacitor. By injecting electrons, the operating voltage range might also be modified. H. M. Cheng *et al.* established that charge injection is a practical method to adjust the open circuit voltage of electrodes by demonstrating that the hybrid supercapacitor electrode's materials function with the surface charge density [63].

Problems with capacity imbalance and diffusion kinetics between the electrodes of hybrid supercapacitors can be

solved by employing the Control of Electrode's OCV method. It does, however, have some drawbacks. These drawbacks are discussed as follows

Effectiveness in addressing capacity imbalance is limited. While the control of OCV approach might somewhat reduce the issue, it might not be sufficient. Hybrid supercapacitors' capacity imbalance can be caused by a variety of factors, such as variances in electrode aging and manufacturing processes and different electrode materials. Perfect balance between the various electrodes might not be possible with only the control of the OCV approach.

Challenges in controlling the diffusion kinetics is another limitation in the use of OCV. The use of various electrode materials with unique diffusion kinetics is common in hybrid supercapacitors. It's possible that regulating this various diffusion kinetics presents difficulties that the management of OCV technique does not directly address. Diffusion-related problems in hybrid supercapacitors may not be completely resolved by it, despite the fact that it might indirectly affect diffusion kinetics by affecting charge distribution and electrode potential.

Trade-off between capacity and performance is a limitation in the use of OCV. The control of OCV method may involve adjustments to the charging and discharging processes of the supercapacitor to achieve balance between electrodes. However, these adjustments can potentially impact the overall capacity and performance of the supercapacitor. To Achieve optimal balance while maintaining high capacity and performance can be a delicate trade-off.

By controlling OCV, the charge distribution between the electrodes of hybrid supercapacitors can be balanced and address the issue of capacity imbalance. Developing advanced control and monitoring systems that can accurately measure the OCV of each electrode and make the required modifications is essential. A feedback control system can also be used to minimize capacity imbalances by ensuring that each electrode receives an equal amount of charge.

### 3.2 Utilization of Redox couples

Electrolytes, which take up a sizable portion of the device, are used to ionically link the electrodes of electrochemical power housing devices [64, 65]. The electrochemical storage device's performance is significantly influenced by the materials employed in its electrode construction. Redox couples are a class of electrode materials that comprise ions from transition metals, halides, hydroquinone (HQ), phenylamine, and other compounds. These materials are soluble and have the ability to perform a quick Faradaic reaction that occurs at the electrode/electrolyte contact. [64]. When these redox couples are introduced into the electrolyte, the supercapacitor is compelled to work in a hybrid mechanism, which results in a noticeably higher efficiency [66, 67]. Si Wen Zhang *et al* increased the voltage range up to 2.4V and the energy density by including  $K_3[Fe(CN)_6]$  to 1M  $Na_2SO_4$  electrolyte [68]. In a related development, H. Bai *et al*, discovered that adding an extra Hydroquinone (HQ)

at 0.4 M to the electrolyte can increase the specific capacitance of supercapacitors made with electrodes containing carbon-based materials in an electrolyte of 1 M  $H_2SO_4$  by two times the initial amount [69; 67]. The researchers inserted a self-reliance source electrode to track changes in voltage between the electrodes. One issue with supercapacitors is that they have intrinsic electrodes made of redox couple carbon yet have a narrow electrochemical stability range of aqueous solutions. For maximum efficiency, the voltage range should be restricted to voltages below 1 V [66; 67]. This further restricts the improvement of the energy density. The problem has been solved thanks to the carefully chosen electrode materials that have replaced the intrinsic carbons. Reduced grapheme/ $Fe_3O_4$  (rG/ $Fe_3O_4$ ) nanocomposites were used by R. Kaner *et al.* to demonstrate a Hybrid supercapacitor with  $K_3Fe(CN)_6$  redox couplings working at 1.8 V in an electrolyte of aqueous  $Na_2SO_4$  [70]. The energy density has increased 10 times in comparison to other devices using electrodes made of reduced grapheme (rG). The balanced voltage range of the subsequent electrolyte has more than doubled to 2.0 V thanks to the design of an asymmetric electrode. Specifically, the operating voltage window has increased by 10% as a result of replacing one of the electrodes made of iron oxide that utilize a composite electrode made of  $MnO_2$  and CNT [71]. Additionally, 2.2 V can be added to this voltage range by carefully choosing electrodes containing vastly different individual work functions, such as cathode (negative electrode) containing  $ZrO_2$  /CNTs and  $WO_3$  /CNTs as the anode (positive electrode). Because of significant energy required for both the hydration of the lithium and sulfate ions, the balanced voltage range of the aqueous electrolyte of  $Li_2SO_4$  is as high as 2.2 V. A  $MnO_2$  /CNT composite electrode has been substituted for one of the electrodes based on iron oxide, specifically expanding the operating voltage window by 10% [38]. The voltage space can also be boosted to 2.2 V by carefully deciding on electrodes containing significantly different individual work functions, such as cathode (negative electrode) containing  $ZrO_2$  and CNTs and anode (positive electrode) of  $WO_3$  and CNTs. The balanced voltage range of electrolyte containing aqueous  $Li_2SO_4$  is up to 2.2 V and this is because, both water absorption of the lithium and sulfate ions demand a large amount of energy.

In HSCs, Yu Zhang *et al.* (2020)[72] offer an entirely new concept for asymmetric electrolyte design: the NiCo layered double hydroxide (LDH) battery-type electrode works in the KOH electrolyte, while the carbon capacitive electrode is purposefully exposed to electrolyte-soluble redox couples. The capacity imbalance issue in the two electrodes with equal mass loading is resolved by the redox couplings' additional faradic capacity contribution to the capacitive carbon electrode. The pristine imbalanced device's specific energy of 20.3 Wh/kg is roughly 4 times lower than the exceptionally high specific energy of 79.6 Wh/kg that the optimized HSC offers. Yuan Wang *et al.*(2022)[73] proposed an innovative aqueous Zn-ion hybrid supercapacitors where Zn foils, porous sheet carbons, and dual redox-active ions were employed as the electrolytes, positive electrodes, and negative electrodes, respectively. The involvement of double redox-active ions in

the electrolytes in electrochemical energy storage can greatly boost the specific capacity of such devices. For both redox-active ions ( $\text{Mn}^{2+}$  and  $\text{Br}^-$ ), a high specific capacity of  $1.25 \text{ mAh/cm}^2$  ( $312.2 \text{ mAh/g}$ ) is obtained at  $0.8 \text{ mA/cm}^2$ . This capacity is 2.66 times greater than Zn-ion hybrid supercapacitors without redox-active ions and exceeds most reported values in the literature.

Drawback associated with the use of Redox couple in addressing capacity imbalance and diffusion kinetic in hybrid supercapacitors is electrode-electrolyte compatibility, Redox Potential, and Stability and Cyclability

The electrode materials and electrolyte selected for hybrid supercapacitors should be compatible with the redox couples being used. The electrode-electrolyte interface needs to be a productive site for redox reactions in order to operate at its best. But attaining adequate compatibility between the redox pair, electrode material, and electrolyte can be difficult. Secondly, it is also suggested that the hybrid supercapacitor's operating voltage range ought to match with the redox potential of the selected redox couples. This maximizes the use of the redox couples by ensuring that the redox reactions take place within the desired voltage structure. And lastly, for long-term performance, the redox couples should show strong stability and cyclability. Redox reactions should be able to be performed on them repeatedly without causing them to significantly degrade or lose capacity.

### 3.3 The utilization of water in salt (WIS) electrolytes

The application of devices that contain water electrolyte is largely constrained since they produce little energy due to purely small voltage space, despite the fact that these systems are safer, less expensive, and more toxic than organic electrolytes [74]. Been encouraged by favorable effects of solid electrolyte inter-phase (SEI) on the expansion of actual ranges of voltage in organic electrolytes, solid electrolyte inter-phase (SEI) in water electrolytes was found by C. Wang *et al.* in a contemporary method [75]. The application voltage window for water in salt electrolytes was increased (1.23 to 3.0 V) by successfully constructing solid electrolyte inter-phase on electrode that operates in water electrolytes. A significant number of batteries based on aqueous electrolyte with appreciable upward voltage window have been designed by C. Wang *et al.* thanks to the clever use of solid electrolyte inter-phase (SEI) [76, 77]. The intriguing discovery that falls under the SEI (solid electrolyte inter-phase) is micro-pores that we had all believed would be prevented by solid breakdown can still exist and increase capacitance. As a means to increase the potential window of operational possibility for supercapacitors, this modernized concept of electrolyte made of water in salt (WIS) was immediately embraced. An electrolyte made of water in salt (WIS) is a type of electrolytes that use water as the solvent and a salt with very high concentration ( $> 3\text{M}$ ). The high salt concentrations in WIS electrolytes result to a significant reduction in the amount of water molecules that surround each ion. This has resulted to a reduction in the activity of ions and a clampdown of the ion diffusion coefficient. Consequently, the WIS electrolyte has been able to regulate

the ion transfer properties in a manner that address the problems of capacity imbalance and diffusion kinetics between the electrodes.

With this sort of electrolyte, the utilization of substances with the like of carbon, transition metal sulfides and transition metal oxides in the role of electrodes can create a significant voltage window [78, 79, 80]. In unison, the working voltage range of 3.0 V and exceptional pseudo-capacitance performance, Lin *et al* created electrodes utilizing  $\text{TiS}_2$ -coated CNTs [80]. Designing a new electrode for this WIS electrolyte will also boost performance. Using WIS electrolyte, Hasegawa created a variety of porous carbon electrode devices that were powered by electrodes of two carbons and had a voltage of 2.4 V output voltages [78 81]. Hybrid supercapacitor devices with various electrodes have also been built as a result of the potential for several kinds of electrodes to work in electrolytes containing WIS [82, 83, 84, 85]. An amalgamated (hybrid) supercapacitor with a cathode of  $\text{MnO}_2$  and a carbon anode that operates in a salt-water electrolyte and an upgraded running potential range of 2.2 V was proposed by Belanger and his colleagues [86, 87]. The electrolyte that is utilized ultimately determines conductivity. Reber *et al* study the conductive properties of different WIS electrolyte and discovered that quantity and cationic radius had an effect [85, 82].

Precisely, the employment of WIS electrolyte increases the kinetics of both electrodes and reduce capacity imbalance. It also brings about an improved energy density of the device which is necessary for commercial. There are still restrictions to take into account, even if WIS electrolytes appear to be a promising solution to the capacity imbalance and diffusion kinetics problems in hybrid supercapacitors. The operating voltage window of hybrid supercapacitors may be constrained by the usage of WIS electrolytes. Compared to organic electrolytes, WIS electrolyte often has a narrower electrochemical stability window. This is because there is reduction in the activity of ions and a clampdown of the ion diffusion coefficient This restriction lowers the hybrid supercapacitor's maximum operating voltage, which has an impact on both its energy density and general performance.

It is therefore suggested that there should be right balance of water content in the salt electrolyte. High water content can lead to excessive swelling of the electrode materials, affecting their stability and performance. On the other hand, low water content can hinder proper ion transport and decrease the overall capacity of the system.

### 3.4 The use of core-shell hybrid nanostructures

Core-shell hybrid nanostructures can help to alleviate the capacity imbalance by increasing the surface area that both electrodes have to interact with and thereby increasing the system's overall capacity [88]. The core of the nanostructure can act as a base to support the shell of the nanostructure, which may be made of a different substance that is more appropriate for electrochemical operations [89]. For instance, carbon-based electrodes' electrochemical performance can be enhanced by metal oxide shells. As a result, hybrid

supercapacitors may have a larger overall energy storage capacity and longer-lasting stability. Additionally, the shell material can improve the overall nanostructure's structural integrity, halting deterioration over time. Additionally, the utilization of core-shell nanostructures may enable better control over the size and shape of the active material particles, improving the application's electrochemical performance and efficiency [90]. Figure 1 shows a typical core-shell hybrid nanostructure with multifunctionality that is applied in hybrid supercapacitors. It shows that carbon cloth and rGO were utilized for this core-shell hybrid nanostructures.

With NiO-decorated CNT/ZnO core-shell hybrid nanocomposites (CNT/ZnO/NiO HNCs), Nagabandi et al. (2020)[91] constructed an electrode with an exceptional capacity of  $67\text{Ahcm}^2$  at a current density of  $3\text{mAcm}^2$ , and excellent cycling stability of 112% even after 3000 cycles of continuous operation, the CNT/ZnO/NiO HNCs-based electrode has demonstrated better performance. Zhang et al.(2015) [92] used twin coaxial electrospinning to create core-shell Si/C fibers with an interior carbon architecture resembling a honeycomb. The findings showed an exceptional rate capability, a high specific capacity ( $3612\text{mAh g}^{-1}$ ), steady cycling performance (71% after 150 cycles), and exceptional Si's accessibility. The distinct hierarchical core-shell architectures of Si/C fibers were credited with the improved electrochemical performance. In addition to improving the conductivity within these fibers, the porous carbon structure in the core region may be able to support silicon's volume growth. In order to create a stable solid-electrolyte interphase on the fiber surface, the compact carbon was able to prevent electrolyte from seeping into the cores. According to Narasimharao Kitchamset al. (2023)[93], an effective supercapacitor electrode with a substantial surface area and numerous channels for  $\text{OH}^-$  ion diffusion was created by using the core-shell structure of hierarchical 2D Manganese Dioxide ( $\text{MnO}_2$ ) nanoflakes and 1D Nickel Titanate ( $\text{NiTiO}_3$ ) (NTO) mesoporous rods. With a specific capacitance of  $1054.7\text{ F/g}$ , specific power of  $11879.87\text{ W/kg}$ , and specific energy of  $36.23\text{ Wh/kg}$ ,  $\text{MnO}_2@\text{NTO}$ 's two-step chemically processed hybrid porous core-shell hetero-architecture fares well. Additionally, 5000 cycles show 85.3% retention in capacitance without a decline in the surface morphological characteristics.

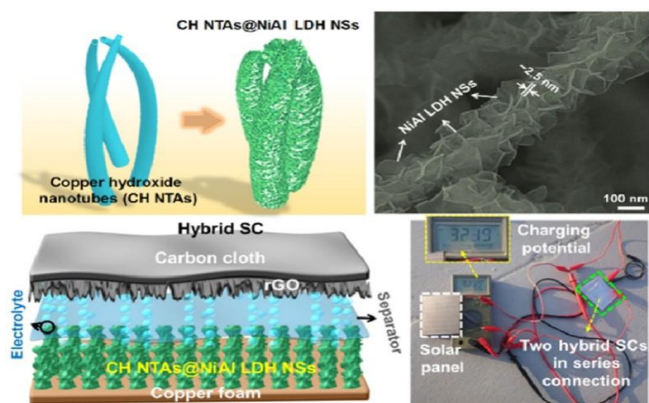


Fig 1; Multifunctional core-shell-like nanoarchitectures for hybrid supercapacitors [94]

By combining various materials in a core-shell arrangement, these nanostructures can offer improved energy storage and performance.

One limitation in the application of this technique is the structural stability of the core-shell nanostructures. The electrodes' frequent expansion and contraction during the charge/discharge cycles can put mechanical strain on the nanostructures. The core-shell structure may deteriorate as a result, reducing performance and shortening the supercapacitor's lifespan. To improve structural stability, for instance, self-assembled monolayer integration or the use of polymer coatings can be used. The size and connection of the pores inside the nanostructures can still limit the rate of ion diffusion even though the core-shell structure can create channels for ion transport. This may lower the supercapacitor's total charge and discharge rates. The property of nanostructured membranes having pores the size of subnanometers is called ion selectivity, which allows certain ions to be transported preferentially while blocking the passage of others. This ions selectivity has the potential to boost the total ion diffusion rate in the intended direction. Accelerated ion diffusion can also be achieved by improving the electrical conductivity of hybrid nanostructures, for example, by adding conductive materials or using carbon nanotubes.

### 3.5 Mass and Charge Balancing approach.

One method for dealing with the imbalance between the electrodes in hybrid supercapacitors is to make sure that the mass and charges of the cathode and the anode are equal [95]. The mass and charge balance between the positive and negative electrodes in hybrid supercapacitors is critical because it considerably affects the device's performance. [96]. Mass balance between the two electrodes can be adjusted using an electrochemical method that employs conducting polymers, metal oxides, or carbon-based materials [97]. Charge balancing between the electrodes of hybrid supercapacitors can be accomplished in a variety of ways, including as mass balancing, changing the electrode potential, and using redox electrolytes. Charge balancing is essential to address the issue of capacity imbalance and create a superior and efficient hybrid supercapacitor because excessive charge accumulation on one electrode would lower voltage and diminish the supercapacitor's capacity to produce energy [97, 98]. In hybrid supercapacitors, the act of altering the mass or coating ratio between the two different types of material for the electrodes is known as "mass balancing." A better and more efficient hybrid supercapacitor can be created by properly balancing the mass of the two different types of electrodes and adjusting the charge distribution [99, 100].

This is because mass balancing permits an equivalent amount of charge to be stored in electrodes with high capacitance and high energy density, resulting in a greater energy output and a longer device lifespan [101, 102]. Another approach to achieve charge balancing is to change the electrode potential from the point of zero charge (PZC) to obtain equal charge on both electrodes [103, 104]. At PZC, the electrode material doesn't show any excess charge (a state of being uncharged).

As the electrode potential is adjusted from its PZC, the charge density of electrode material will change. Doha M. Sayed et al.(2020)[105] presented innovative and straightforward electrochemical principle that serve as a guide for selecting a safe and appropriate operating potential window profile for carbon-based hybrid supercapacitors. They use PZC as the initial vertex potential of cyclic voltammograms (CVs). Nevertheless, the storage mechanism as reported is clarified using the CVs recorded at different potential scan speeds throughout the entire studied potential window, and then the ultimate potential limit is selected. Furthermore, after assessing the potential window of the two independent electrodes according to the authors, the mass and charge balance of the created hybrid electrodes are logically designed. By employing those tactics, a hybrid device consisting of carbon derived from biomass and carbon derived EDTA (Ethylenediaminetetraacetic acid) salt achieves record performance abilities. In addition, they reported that the assembled device demonstrates a high capability rate, specific capacitance of 265 F/g at 5 mV/s and 221 F/g at 1 A/g. Attaining a battery-like energy level of 99.2 W h/kg as presented by the authors is a proof of idea that validates the proposed electrochemical fundamental methods for monitoring the mass ratio balancing of the hybrid cell electrodes. Finally, the device demonstrates exceptional stability over 10,000 cycles with 100% capacitive retention and nearly 100% columbic efficiency.

Redox electrolytes are used in hybrid supercapacitors to help in establishing charge balance by supplying an equivalent number of ions that can store charge on the two electrodes. [106, 107, 108].

A hybrid supercapacitor that combines the capabilities of a high-energy battery and a high-power supercapacitor in a single unit was reported by Jun Feng *et al.*(2018) [109]. This device uses asymmetric LiFePO<sub>4</sub> (LFP) lithium intercalation and electric double layer activated carbon (AC) electrodes. In the activated/lithium half cell, the active electrode has a capacity of 44.5mAhg<sup>-1</sup> and an operating voltage more than 2V (Li/Li<sup>+</sup>), which allows it to display an ion adsorption process like that of a supercapacitor. The hybrid cells with AC/LiFePO<sub>4</sub> mass ratios of 0.33, 1.20, 1.93, and 3.19 demonstrate the transition from a redox reaction-dominated system, like a battery, to a system in which both faradaic redox and the physical electrostatic adsorption process forming an electric double layer are fully balanced in terms of stored charges on both electrodes.

Electrode-electrolyte interaction is the major drawback associated with the mass and charge balancing method of solving the problems of capacity imbalance and diffusion kinetics between the electrodes of hybrid supercapacitors [110]. The mass and charge technique does not take into account the complex interactions between the electrodes and the electrolyte. Poor ion diffusion, reduced charge transfer kinetics, and elevated internal impedance are some of the problems that may result from this interaction [111]. To solve the problem of electrode-electrolyte interaction in hybrid supercapacitors, it is possible to improve the interaction

between the electrode and electrolyte by using interface engineering approaches. This may entail applying coatings or interfacial layers that enhance charge transfer kinetics and ion diffusion [112].

### 3.6 Electrodes' structural design and control

The problems of capacity imbalance and diffusion kinetics in hybrid supercapacitors can be resolved by designing and controlling of the electrode configuration [113].

The availability of the same quantity of active material in each electrode is crucial in order to address the issue of capacity imbalance. One strategy is to design the electrode structure so that its properties are homogeneous and reliable [114]. To make sure that the same quantity of active material is present in each electrode and to achieve balance capacity, for example, it is important to ensure that the porous structure of the electrode is the same across all electrodes. The active material can also be made to have a high surface area and good pore interconnectivity [115, 116]. This can be accomplished by utilizing a range of materials, such as porous carbon, conductive polymers, metal hydroxide, and metal oxide, which can offer a large surface area and enhance reactants' accessibility and product transport to and from the active material [117].

Guilin *et al* loaded  $\alpha$ -MnSe nanoparticles into a hollow cubic CuSe through ingenious design which maintain high conductivity. The CuSe@MnSe composite possesses a high capacity of 635.32Cg<sup>-1</sup> over 7000 cycles of stability (91.62% capacity retention) [113]. Wang *et al* developed an efficient sulfurization tactic to advance the electrochemical assets of metal phosphates via creating porous nanotube structure and improving the overall conductivity and the as-prepared sulfur-doped CoP nanotube arrays disclosed about 1.8 times Sophisticated capacity than that of CoP [118].

In hybrid supercapacitors, the design of the electrode structure can also affect how quickly ions diffuse. The performance of hybrid supercapacitors can be increased by structuring the electrode to have a large surface area, strong pore interconnectivity, and the right pore size [119, 120].

The problem of diffusion kinetics can also be addressed by controlling the manufacturing process. For instance, it is possible to regulate and optimize the rate of ion diffusion by precisely adjusting the size and distribution of the pores in each electrode structure [121]. Additionally, it is possible to modify the electrode structure and control the diffusion kinetics by using additives and binding materials [122].

Utilizing fork-shaped electrodes is one structural design method of achieving equilibrium between the diffusion kinetics and capacity between the electrodes of hybrid supercapacitors. Hybrid supercapacitors operate better thanks to the special structure of fork-shaped electrodes, which has multiple benefits. In comparison to conventional flat electrodes, fork-shaped electrodes have a greater surface area. Because of the greater surface area, there can be more electrochemical processes, which increase capacity and energy storage capacity. Pengfei Hao *et al.*(2023)[123]

successfully synthesized single crystalline fork-like potassium vanadate ( $K_2V_8O_{21}$ ) using the electrospinning method followed by an annealing step. The authors state that the resulting  $K_2V_8O_{21}$  forks have a distinct layered structure layer by layer. High specific discharge capacity and good cyclic stability are demonstrated by the as-prepared fork-like materials when utilized as cathode materials for lithium-ion hybrid supercapacitors. With current densities of 50 and 500  $mA\ g^{-1}$ , respectively, high specific discharge capacities of 200.2 and 131.5  $mA\ h\ g^{-1}$  can be produced. Furthermore, after 300 cycles at 500  $mA\ g^{-1}$  with a fading rate of only 0.043% each cycle, the  $K_2V_8O_{21}$  electrode maintains a remarkable long-term cycling stability, maintaining a capacity of 108.3  $mA\ h\ g^{-1}$ . The findings show how they might be used in high-performance, next-generation lithium-ion batteries

Lack of precise control over the electrode structure and composition is the main barrier to using electrode structural design and control approaches to address the issues of capacity imbalance and diffusion kinetics between the electrodes of hybrid supercapacitors. Ion diffusion rates can be affected by the electrode composition and structure, which results in slower charge and discharge rates. Achieving a balanced capacity requires precise control over the electrode structure and composition.

### 3.7 Incorporating of two-dimensional carbon materials

Due to their unique physical and chemical properties like large specific surface areas, abundance of ionic active sites (surface and functional groups), and wide layer-to-layer gaps, two-dimensional carbon materials like carbon nanosheets, graphene, and graphite are potential candidates for hybrid supercapacitors. The many vital component elements of supercapacitors are constructed from carbon-based materials. The cathode of a capacitive electrode is made of materials like activated carbon, porous carbon, and grapheme [123, 124]. The carbon materials used in the battery-type electrodes (anode) are graphite, graphene, unstructured carbon, and carbon N-doped nanotubes [125, 126]. The carbon materials used to make the negative electrode must contain an adequate number of ionic active sites for reversible anion adsorption and desorption to take place. Large size cations like  $Na^+$ ,  $K^+$ , or  $Zn^{2+}$  etc. must be added to or removed from the battery-type anode reversibly, which necessitates a large gap between layers. It was also found that functional groups on carbon with oxygen in them increase capacitance as well as the distance between the layers of carbon, which facilitates ions diffusion. [127]. Specific requirements need to be satisfied for two-dimensional carbon materials to function well as hybrid supercapacitor electrode material; there must be sufficient ionic active sites for reversible anion adsorption and desorption. Graphene must be kept from rebuilding in order to enhance the density of ionic active sites. This can be done by using structural supports like carbon nanotubes or polymer strands. [128, 129]. According to Zhao *et al*, by modifying the surface chemistry and linkages between the polyimide and graphene, the capacity of the hybrid to store sodium ions was dramatically boosted, going from 50  $mAh\ g^{-1}$  for pure polyimide to 225  $mAh\ g^{-1}$  for the hybridized polyimidegraphene. [130].

Two-dimensional carbon materials for battery-type electrodes (anodes) must have enough of a space between layers to permit the reversible entry or removal of large-size cations. It has been demonstrated that the spacing between the layers of carbon can be extended by heavily modifying the carbon structure with nitrogen and phosphorus heteroatoms [131]. The heteroatom doped carbon material may also increase the density of cations adsorption sites. [132]. Modified carbon materials might also improve the layer-to-layer gap and cations adsorption sites, which would increase the functionality of capacitors [90]. Figure 2 represent several types of common ultrathin 2D nanomaterials.

By the process  $-C \rightarrow \bar{O} + e + M^+ \rightarrow -C - O - M^+$  for instance, oxygen-modified carbon nanofibers may be able to store more  $M^+$  cations. Oxygen-containing functional groups might also increase the distance between the layers of carbons, which would improve cations diffusion. Unlike capacitive-type electrodes (cathodes), battery-type electrodes (anodes) go through sluggish cation diffusion. In order to tackle the issue of the different diffusion kinetics between the two electrodes, two-dimensional carbon materials are commonly used as conducting frameworks to build graphene hybrid electrodes [133, 134]. Huang *et al*. described a hybridized  $MoS_2/rGO$ -based hybrid sodium supercapacitor. The constructed hybrid sodium supercapacitor exhibited remarkable performance when utilized as an anode material, yielding a 580  $mAh\ g^{-1}$  capacitance at a 100  $mAh\ g^{-1}$ . This was made achievable by the ability of the graphene skeleton frame to transmit enough charges and the capability of the greatly layer-to-layer enhanced  $MoS_2$  to permit rapid ion diffusion. [135]. Somnath *et al*. (2023)[135] have achieved three-dimensional (3D) architecture by growing NiSe nanoparticles on two-dimensional (2D) graphitic carbon nitride (g-C<sub>3</sub>N<sub>4</sub>) nanosheets. The NiSe/g-C<sub>3</sub>N<sub>4</sub> nanocomposite material's specific capacity was increased by the 2D support, high nitrogen concentration, and g-C<sub>3</sub>N<sub>4</sub> characteristics. The resultant nanocomposite exhibits a specific capacity that is significantly higher than virgin NiSe, measuring 320  $mA\ h\ g^{-1}$  at a current density of 1  $A\ g^{-1}$ . Figure 2 Illustrate many kinds of typical ultrathin 2D nanomaterials, such as metals, oxides, graphene, h-BN, BP, graphene, TMDs, MOFs, COFs, MXenes, and LDHs

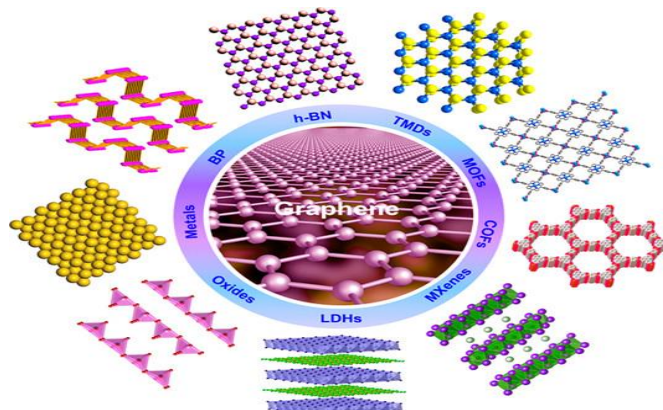


Fig 2 : Schematic representation of several types of common ultrathin 2D nanomaterials, including oxides, metals, BP, graphene, h-BN, TMDs, MOFs, COFs, MXenes, and LDHs [136].



Two-dimensional carbon materials have demonstrated significant promise in improving the balance of capacity and diffusion kinetics between the electrodes in hybrid supercapacitors. This finding is supported by investigations and research done in the area of energy storage by various researchers. Researchers were able to overcome the challenge by introducing two-dimensional carbon compounds. These materials enable effective charge and discharge operations because to their huge surface area and strong electrical conductivity, which speed up ion diffusion. This ultimately results in increased energy storage capacity and quicker rates of charging and discharging. Moreover, the hybrid supercapacitors' overall stability and durability are enhanced by the mechanical strength of two-dimensional carbon materials. For long-term performance and dependability, this is essential. Two-dimensional carbon materials provide further potential for flexible energy storage systems and device miniaturization. These materials' thin and light properties enable the building of compact and light supercapacitors, which makes them perfect for use in wearable technology, electric cars, and portable electronics.

### 3.8 Utilization of Materials with Good conductivity

Hybrid supercapacitors, as was previously said, combine the high energy density of batteries with the high power density of supercapacitors, making them desirable for energy storage applications. They do, however, have difficulties with regard to diffusion kinetics and capacity imbalance. These issues can be solved and hybrid supercapacitors' performance enhanced by using materials with good conductivity [137].

One issue in hybrid supercapacitors is capacity imbalance, where the positive and negative electrodes have different charge storage capacities [138]. This imbalance can cause uneven charge distribution and limit the overall energy storage capacity of the device [139].

By using materials with good conductivity, the charge transfer between the electrodes can be facilitated, reducing the potential for capacity imbalance [140]. When the charge transfer is efficient, both positive and negative electrodes can contribute equally to the overall energy storage capacity of the supercapacitor [141].

Rapid and more effective charge transfer is made possible by electrode materials with high conductivities, resulting in balanced charge storage and increased device performance. [142].

Ion and electron diffusion kinetics within the electrodes' materials present a further challenge in hybrid supercapacitors. Reduced performance and power density can be the result of sluggish diffusion kinetics, which can restrict the rate at which charges can be stored or released [143].

Materials with good conductivity provide pathways for rapid ion and electron transport [144]. High electrical conductivity makes it possible for charges to travel more quickly between electrodes, resulting in effective charge storage and release processes [145]. This not only improves the power density but

also contributes to faster charging and discharging rates of the supercapacitor.

Additionally, by lowering resistance and eliminating energy losses related to charge transfer, materials with strong conductivity can improve the kinetics of diffusion [146]. As a result, the supercapacitor can achieve better overall performance, with higher power density and improved charge storage capabilities.

Guilin *et al* loaded  $\alpha$ -MnSe nanoparticles into a hollow cubic CuSe through ingenious design which maintain high conductivity. The CuSe@MnSe composite possesses a high capacity of  $635.32\text{Cg}^{-1}$  over 7000 cycles of stability (91.62% capacity retention) [109]. Wang *et al* [147] developed an efficient sulfurization tactic to advance the electrochemical assets of metal phosphates via creating porous nanotube structure and improving the overall conductivity and the as-prepared sulfur-doped CoP nanotube arrays disclosed about 1.8 times Sophisticated capacity than that of CoP.

The issues with capacity imbalance and diffusion kinetics can be resolved by using materials with strong conductivity in hybrid supercapacitors. Overall improvement of the device's ability to store energy can be achieved by having good conductivity, which makes it easier to transport charges across electrodes in a balanced manner. Additionally, it improves the diffusion kinetics, permitting quicker charging and discharging rates and raising the supercapacitor's power density. The imbalance in kinetics between the two electrodes of hybrid supercapacitors is one of the drawbacks of adopting materials with good conductivity to address the issues of capacity imbalance and diffusion kinetics between the electrodes. This mismatch may prevent the hybrid supercapacitor from using its full potential energy. Multiple ions migration, a large ionic radius, and a high interfacial resistance can all contribute to the slow kinetics of the multi-electron transfer process in the electrodes.

### 3.9. Choosing an appropriate electrolyte

Ions are transported between the electrodes of supercapacitors via an electrolyte, which is made up of a solution that is conductive and additives. According to Fig. 1, there are various categories of electrolytes used in supercapacitors. The electrolyte is a crucial component of supercapacitors and is essential for the transfer and balancing of charges between electrodes in hybrid supercapacitors.

In a hybrid supercapacitor, the term "diffusion kinetic" describes the flow of ions in and out of the electrodes. The size and shape of the electrode, the composition of the electrolyte, and the concentration of ions on each side of the electrode are only a few examples of the variables that affect the kinetics of diffusion [148]. The interaction between the electrode and electrolyte in all electrochemical processes can affect the inner structure of active materials and the state of the surface between the electrode and electrolyte [149]. Consequently, by influencing the ion diffusion kinetics, selecting an appropriate electrolyte for a hybrid supercapacitor can help to resolve this problem. Ions' ability

to pass through an electrolyte swiftly can be affected by properties such as viscosity, ions' conductivity and diffusivity, and resistance, which different electrolytes have [150, 151]. Hybrid supercapacitor designers can make sure that the diffusion kinetic is balanced over all of the electrodes by choosing an electrolyte with the right properties. This helps to avoid capacity imbalances while also preserving the device's overall performance. By selecting an electrolyte with high diffusivity, low viscosity, and low resistance, the rate of diffusion as well as the charge/discharge rate of the device can be increased [152]. Some examples of electrolytes with such properties are; aqueous electrolytes (H<sub>2</sub>SO<sub>4</sub>, KOH etc), acetonitrile (CH<sub>3</sub>CN) electrolyte, ionic liquids electrolytes (1-ethyl-3-methylimidazolium bis (trifluoromethylsulfonyl) imide ([EMM][TFSI]), 1-butyl-3-methylimidazolium tetrafluoroborate ([BMM][BF<sub>4</sub>)]), gel polymers electrolytes (which consist of polymer matrix such as polyethylene oxide (PEO) or polyvinylidene fluoride (PVDF), combined with a liquid electrolyte or ionic liquid), water in salt electrolyte [153], hydrogel electrolyte. See figure 3 for the classification of electrolytes. Table 3 shows the comparison of electrolytes in supercapacitors' applications. With table 3, the selection of electrolyte will help to booster the capacity of hybrid supercapacitor. Figure 4 also shows some electrolytes and the factors affecting their properties.

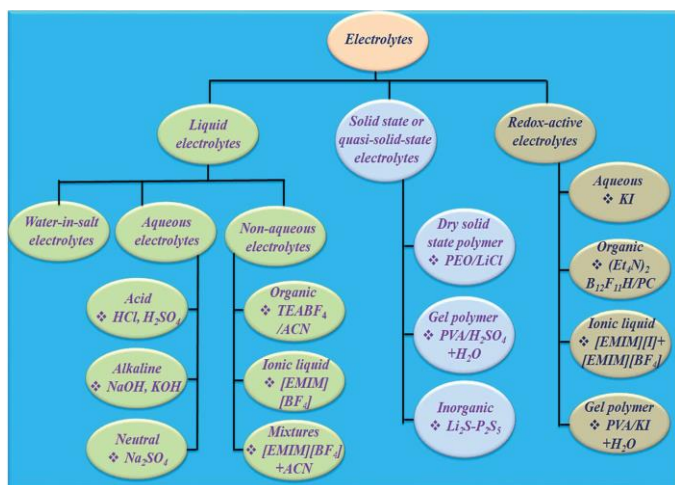


Fig 3: Classification of electrolytes for electrochemical supercapacitors [154]

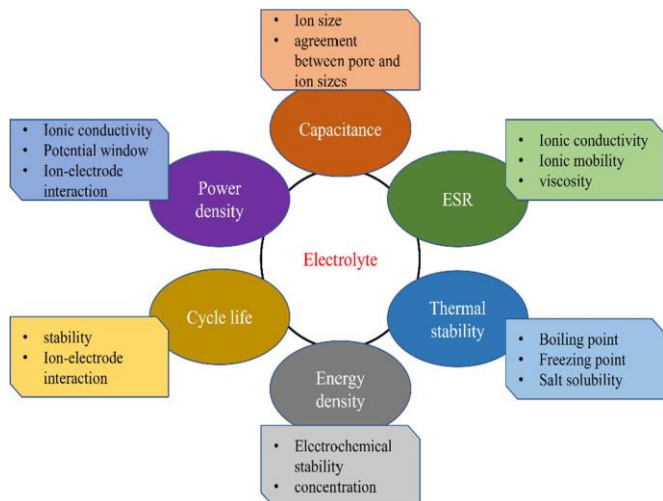


Fig. 4 Factors affecting electrolyte properties [155].

Table 3: Comparison of Electrolytes in supercapacitors

Electrolyte Type	Operating Potential Range	Ionic Conductivity	Energy Density	Power Density
Aqueous Electrolytes	Low (1.0–1.3 V)	High	Low	Relatively high
Organic Electrolytes	Wide (2.5V–2.7 V)	Moderate	High	Moderate
Ion-Liquid Electrolytes	Wide (3.5–4.0 V),	High	High	High
Hydrogel Polymer Electrolytes	Low to Moderate	Low to Moderate	Moderate	Moderate
Solid Polymer Electrolytes	Low to Moderate	Low to Moderate	Moderate	Moderate
Polyelectrolytes	Wide	Moderate	Moderate	Moderate
Redox Electrolytes	Wide	Moderate to High	Moderate	Moderate

Note: The values in the table are based on general trends and may vary depending on specific

The primary chemical reaction in supercapacitor electrolytes is the dissociation of the electrolyte into positive cations and negative anions, which results in electrical conductivity as shown in in equation 1 and 2. Table 4 display some reported electrolytes and their performance in supercapacitors.

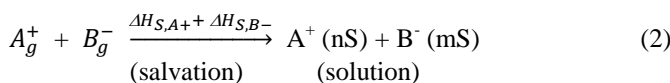
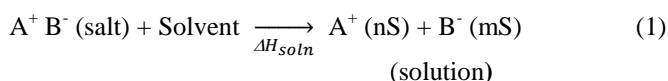


Table 4: Some reported electrolyte and their performance in supercapacitors

Electrode material	Electrolyte type	Electrolytes	Cell voltage (V)	Power density (W kg <sup>-1</sup> )	Energy density (Wh kg <sup>-1</sup> )	Specific capacitance (Fg <sup>-1</sup> )	reference
Microporous carbon	Aqueous electrolyte	1M H <sub>2</sub> SO <sub>4</sub>	1.0	100	3.8	100	156
p-CNT <sub>n</sub> /C GBs	Aqueous electrolyte	6M KOH	0.9	150	4.9	202	157
Microporous carbon	Aqueous electrolyte	0.5M Na <sub>2</sub> SO <sub>4</sub>	1.8	40	7.0	60	156
Graphene-CNT composite	Organic electrolyte	1M TEABF <sub>4</sub> ADN	3.0	400	34.3	110	158
Heteroatom-doped porous carbon flakes	Organic electrolyte	LiPF <sub>6</sub> /EC-DE	3.0	243	29.0	126	159
Ac/poly(3-	Ionic Liqui	[BMIM][PF <sub>6</sub> ][PYR <sub>14</sub> ][TFSI]	4.0	140	177	11.06	160

methyl- thiophen e)	d electr olye							
Fe <sub>2</sub> O <sub>3</sub> /G /Zn-Ni- Co- oxide@	Gel polym er electr olyte	Poly(vinyl alcohol) PVS/KOH	1.3	15.	70	87.5	161	
NiMoO <sub>4</sub> NaI@C/ /I <sub>2</sub> @C	Gel polym er electr olyte	Poly(vinylde ne fluoride) PVDF/LiCF <sub>3</sub> S O <sub>3</sub>	2.0	400	49	209	162	

### 3.10 Modeling of Electrode

Numerous investigations have been carried out on the modeling of electrodes, whole batteries, or supercapacitor cells. For instance, Popov et al. have developed a one-dimensional model to investigate the behavior of a hybrid system under pulse discharge currents, based on a Sony 18650 battery and a Maxwell's 10F supercapacitor. According to the authors, the proposed model has shown that the hybrid system can operate at a high power density and still provide a higher energy density than the battery-alone system. Furthermore, when compared to actual experimental settings, it has accurately predicted the power-energy relationship. In addition, many models have been presented by researchers, including statistical circuit modeling and numerical modeling, to replicate the changes in electrochemical behavior that occur when charge storage systems transition from battery-type to capacitor-type. In order to solve the capacity imbalance and diffusion kinetics issues between the electrodes in hybrid supercapacitor electrodes, numerical modeling is essential. Researchers can attain a balanced capacity by optimizing the electrode composition and design using numerical simulation. Through the simulation of electrode charge-discharge behavior, researchers are able to pinpoint the causes of capacity imbalance and suggest mitigation measures. These strategies may include adjusting the electrode materials, modifying the electrode structure, or optimizing the operating conditions.

Numerical modeling also allows researchers to study and optimize the diffusion kinetics within the electrodes. By simulating the ion transport processes and analyzing the concentration profiles within the electrode material, researchers can identify factors that affect diffusion kinetics and propose strategies to enhance it. These strategies may involve optimizing the pore structure of the electrode material, adjusting the electrolyte composition, or modifying the electrode-electrolyte interface.

Fenghua Guo et al (2018)[163] propose a numerical modeling that studies ionic transport and electro-kinetics within a spherical solid particle Pseudocapacitive Process for Energy Storage Devices. The authors discovered that the OCV of the solid materials at various concentration of the charge carrier strongly affect the entire charge storage mechanism.

Applying statistical circuit basis modeling to a hybrid supercapacitor can also help balance the diffusion kinetics and capacity between the electrodes. An analysis of the

behavior of the hybrid supercapacitor and modifications to its design can be done with a statical circuit model. With the use of this modeling technique, it is possible to optimize a number of parameters, including the electrolyte composition, electrode thickness, and material composition, in order to balance the diffusion kinetics and capacity. The statical circuit model evaluates the hybrid supercapacitor's electrical properties, including its capacitance, diffusion resistance, and equivalent series resistance (ESR). By adjusting these parameters, the capacity and diffusion kinetics of the hybrid supercapacitor can be balanced and improved.

**Fabio Corti et al.(2021)** [164] Reported a paper which provided two precise methods for modeling the dynamic and non-linear behavior of a hybrid supercapacitor. The authors stated that the novel techniques that have been suggested can be applied to time-domain simulation of an electrical network, including hybrid supercapacitors. The methods based on the Non-Linear Dynamic Model (NLDM) equivalent circuit can be used to evaluate power losses and provide an intriguing look into the inner workings of the hybrid supercapacitor.

## 4. Conclusion and Future scope

This review has identified capacity imbalance and diffusion kinetics as the major challenges to the performance of hybrid supercapacitors for commercial applications as a result of the different charge storage mechanisms from its different electrode types. Addressing the issues of capacity imbalance and diffusion kinetics requires a thorough strategy that takes into account the control of open circuit voltage (OCV), redox couple utilization, application of water in salt electrolyte, use of core-shell hybrid nanostructures, mass and charge balancing approach, electrodes' structural design and control, incorporation of two-dimensional carbon materials, utilization of materials with good conductivity, and selection of suitable electrolyte techniques.

Controlling the open circuit voltage (OCV) of the electrodes is essential in maintaining the stability and functionality of hybrid supercapacitors. The charge storage capacity can be optimized and the device's overall energy density can be increased by carefully managing the OCV. Additionally, OCV control contributes to the supercapacitor's longevity, cycling stability, and reduction of self-discharge rate.

Utilizing redox couples is a crucial part of dealing with diffusion kinetics and capacity imbalance. The charge transfer kinetics and overall electrochemical performance of the hybrid supercapacitor can be improved by choosing the right redox partners. This results in enhanced power density and better energy storage capacities.

It has been demonstrated that using water in salt electrolyte can improve the performance of hybrid supercapacitors. Water functions as a solvent, promoting ion transport and enhancing the electrolyte's ionic conductivity. Better charge transfer between the electrodes as a result improves the supercapacitor's overall functionality and efficiency.

Core-shell hybrid nanostructures have produced encouraging outcomes when used to address capacity imbalance and diffusion kinetics. The surface area, ion diffusion efficiency, and overall electrochemical performance can all be improved by adding core-shell nanostructures to the electrode materials. The problems with hybrid supercapacitors can be effectively solved by this method.

Approaches for mass and charge balancing are crucial for hybrid supercapacitors to work at their best. Reduce capacity imbalance and encourage uniform charge distribution by carefully balancing the mass and charge between the electrodes. As a result, the device's ability to store energy is boosted, as is the device's general performance.

In order to solve capacity imbalance and diffusion kinetics, electrode structural design and control are required. The ion diffusion paths, active surface area, and overall charge storage capacity can all be improved by fine-tuning the electrode shape. With this method, the electrode materials may be used more effectively, which improves the supercapacitor's performance and efficiency.

Two-dimensional carbon materials have a lot of potential for solving the problems with capacity imbalance and diffusion kinetics. Graphene and carbon nanotubes are two-dimensional carbon materials that have a high electrical conductivity, a sizable surface area, and superior mechanical qualities. The charge storage capacity, ion diffusion kinetics, and overall performance of hybrid supercapacitors can all be improved by adding these materials to the electrode structure.

Utilizing materials with good conductivity is crucial in addressing capacity imbalance and diffusion kinetics. By selecting electrode materials with high electrical conductivity, it is possible to improve the charge transfer kinetics, reduce the resistance, and enhance the overall performance of the supercapacitor. This approach leads to improved energy storage capabilities and increased power density.

Finally, in order to solve capacity imbalance and diffusion kinetics, it is critical to choose the right electrolyte approaches. The ion transport characteristics, ionic conductivity, and overall electrochemical performance of the supercapacitor can all be improved by carefully selecting the electrolyte. As a result, the device's energy storage capacity and efficiency both improve.

With the help of these methods and strategies, hybrid supercapacitors' performance, energy storage capacity, and efficiency may all be improved, opening the door to their broad use in a variety of energy storage systems.

Research on the capacity imbalance and diffusion kinetic imbalance between the electrodes of hybrid supercapacitors has a wide and exciting future ahead of it. Scholars can make noteworthy progress in tackling these issues in a practical manner by concentrating on the strategies reviewed in this work. These developments will help create hybrid

supercapacitors that are more balanced, efficient, and capable of storing and delivering power

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