



## Challenges and Future Perspectives of Supercapacitor Materials: Mini Review

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### ABSTRACT

Supercapacitors (SCs) formed an integral part of the contemporary energy storage systems thanks to their unique traits of power delivery and charge storage capabilities coupled with long cyclic stability ( $>10^5$ ). In its basic form, a Supercapacitor (SC) consists of two separated electrodes impregnated with an electrolyte. The performance of SC is largely a function of the chosen electrode-electrolyte materials. The choice of the materials depends on the prevailing conditions at hand and is always at a tradeoff between certain merits and de-merits. Herein, we reviewed the different types of SC materials (both electrodes and electrolytes) with emphasis on the challenges associated with each. This will create an avenue in determining the direction and future perspective of SC materials.

**Keywords:** Supercapacitor; Electrodes; Electrolytes; Challenges

### INTRODUCTION

Fossil fuels contribute to about 85.5% of the global energy source, however, with the global rapid population growth, the key issues are environmental degradation, fast rate of depletion and uneven resource distribution (Ediger, 2019; Majumdar et al., 2020). Recently, much attention is given to more abundant, inexhaustible and environmentally friendly renewable energy sources (Seetharaman et al., 2019; Huaizhi Wang et al., 2019). Supercapacitors (SCs) are promising renewable energy storage devices with the potential for diverse applications (Hashim & Sa'adu, 2014; Sa'adu et al., 2014; L. Sa'adu & Hashim, 2020; Lawal Sa'adu et al., 2014). The basic requirements of an efficient energy storage system are extreme specific power, high energy per unit mass, swift charge cum discharge behaviour, considerable life cycle, and cost-effectiveness (Horn et al., 2019; S. Liu et al., 2020). Until

now, no single system matched all these requirements. While Supercapacitors recently received great attention, they cannot be used as independent energy storage systems due to challenges of low energy density and low voltage per cell compared with the Li-ion batteries. The energy density (E) of SCs is a function of both the capacitance (C) and the square of the cell's potential operating window (V) according to the relation (Conway et al., 1997; Tomiyasu et al., 2017):

$$E = \frac{1}{2}CV^2 \quad (1)$$

Supercapacitors consists of two separated electrodes submerged in an electrolyte (Miller et al., 2018; J. Sun et al., 2017). To improve the energy density of SCs, high capacitance (C) is needed. For EDL SCs, the capacitance is a function of the electrode surface area, the shape, size and dispensation of the pore, and also the electrical resistance (Lim & Suk, 2021; J. Zhao et al., 2019). In

Pseudocapacitors, the capacitance is based on the Faradaic redox reactions occurring at the exterior of the electrode material, hence, the orientation and charge storage kinetics of the electrode affects the capacitance (Ye et al., 2021). All these factors are directly related to the SC electrode. Therefore, proper selection of electrode material is a key issue regarding the performance of SCs. Moreover, the energy density can be increased by adopting a remarkably wider potential window as envisaged from equation 1. The operating potential window is directly related to the employed electrolyte whether aqueous, organic or ionic liquids (Godse et al., 2019; Gou et al., 2020). Hence, proper choice of electrolyte is also a key issue in the performance of SCs. So far, it is clear that the right choice of electrode-electrolyte combination is eminent for the optimal performance of SCs. Herein, we reviewed the different types of supercapacitor materials (both electrode and electrolytes) with emphasis on the challenges associated with each. This will create an avenue in determining the direction and future perspective of supercapacitor materials.

### Supercapacitor Electrode Materials

The scientific community is struggling to enhance remarkably the energy and power density of SCs by developing novel electrode materials (Fallah et al., 2020; Meyer et al., 2020; Shi et al., 2021). All the contemporary SCs electrode materials are associated with different challenges. The centre of attention is on carbon-based materials, conducting polymers, transition metal oxides, and the likes of metal-organic/nitrates frameworks, along with covalent organic frameworks which are new materials.

### Carbon-based Electrode Materials

The main factors to be considered when designing a reliable SCs electrode are high

capacitance per unit mass, large charge-discharge cycle lifetime, cost effectiveness, and also the environmental impact of the active material (Q. Meng et al., 2017; Redondo et al., 2020; Shi et al., 2021). Carbon-based materials like carbon nanotubes, aerogels, template carbons, carbide-derived carbons and graphene are characterized by high capacitive performance, good electro-thermal stability, and superb robustness but are associated with high cost and difficult synthesis process which limits their commercial application as SCs electrode material (Samal et al., 2021; Yifan Wang et al., 2021; Zeng et al., 2019). For example, Fekri and Ghoranneviss synthesized symmetric paper SCs with carbon nanotube (CNTs) structure, while the specific capacitance of the CNT's SCs was found to be in the range of  $87 \text{ F g}^{-1}$ – $410 \text{ F g}^{-1}$  using a scan rate of  $(20\text{--}150) \text{ mVs}^{-1}$ , the power density ranged from 12 to  $13.6 \text{ kWKg}^{-1}$  respectively (L et al., 2018). Though the achieved result is appreciable, the synthesis method is difficult and not cost-effective (Baddour & Briens, 2005; Shoukat & Imran, 2021). Moreover, Sui et al. (Sui et al., 2015) synthesized Nitrogen-Doped Graphene Aerogels SCs electrode using a facile hydrothermal reaction method, in which the carbon and nitrogen were derived from graphene oxide (GO) and ammonia. The prepared electrode exhibited a large surface area of  $830 \text{ m}^2 \text{ g}^{-1}$  with a specific capacitance of  $223 \text{ F g}^{-1}$  at  $0.2 \text{ Ag}^{-1}$ , however, capacitors fabricated with these electrodes are polarity-sensitive and must be connected in series due to their low voltage of about 2.75 V (Gurav et al., 2010).

In a separate study in which a porous carbon/silicon carbide (HPC/SiC) SCs electrode was synthesized from bamboo waste biomass through facile method, a specific capacitance of  $234.2 \text{ Fg}^{-1}$  at a current density

of  $1 \text{ A g}^{-1}$  was realized (Tang et al., 2021). However, the setback is that the facial method involves using hot and hypercritical pressure water, usually in the temperature range of  $200\text{--}1000 \text{ }^\circ\text{C}$  and pressure of up to thousands of kPa, moreover, the carbon yield decreases with an increase in pressure (Presser et al., 2011). On the other hand, carbon template techniques (hard, soft and self templates) are among the cutting-edge synthesis method of porous carbons with clear-cut pore structures and pore size distributions (W. Zhang et al., 2021). In a particular research involving the preparation of 3-Dimensional self-supported pillared porous carbon via hard-template method, an average pore size of approximately  $7 \text{ nm}$  and specific surface area of  $883 \text{ m}^2 \text{ g}^{-1}$  with a specific capacitance of  $289 \text{ F g}^{-1}$  at  $2 \text{ mVs}^{-1}$  was achieved (Fan et al., 2012), however, the electrochemical features of SCs electrodes synthesized via this technique mainly depend on the template itself. The electrodes usually consist of macro and mesopores, however, their specific surface areas (SSA) are fairly stunt hence, thermal decomposition via chemical activation method should be employed to achieve high specific surface areas, good micro-porosity and appreciable pore volume. This in turn, complicates the synthesis and removal of the hard template processes and thus making it cost ineffective.

In another research involving soft-template technique, research group of Li et al. (Peng et al., 2019) employed graphene oxide nanosheet as the substrate, 1,3,5-trimethyl benzene as swelling agent with soft template polaxamer (F127) to form associated colloids. When this material is used as SC electrode, an ultra-fast charge-discharge time of  $1.52 \text{ s}$  is achieved with an energy density of  $15.2 \text{ W h kg}^{-1}$  at a power density of  $36 \text{ kW kg}^{-1}$  and a specific capacitance of  $402.5 \text{ F g}^{-1}$  at  $1 \text{ A g}^{-1}$ . However, the downside of this technique is

stunt specific surface area coupled with reduced specific capacitance arising from the mesopore structure of the material. Again, the synthesis method is complicated and thus inhibiting the possibility of vast production (W. Zhang et al., 2021). For graphene-based electrodes, Martinez et al. (Martinez et al., 2021) demonstrated how the specific capacitance of graphene-based SC was increased from  $135$  to  $2585 \text{ F g}^{-1}$  as well as the cycle-life of greater than  $80\%$  after  $1000$  cycles to almost  $100\%$  after  $20\,000$  cycles. Nevertheless, graphene-based SC electrodes are associated with unresolved challenges of agglomeration, thereby making it impossible for graphene to attain the hypothetical specific surface area and conductivity. Moreover, graphene needs to be doped with nitrogen for example to achieve desirable pore size distribution (Yang, 2021).

Primarily, the SSA and pore size of an electrode material determine the corresponding electrochemical performance of SCs, the smaller the porosity the higher the capacitance and consequently the lower the power density due to an increase in the overall pore resistance (Largeot et al., 2008; Rajagopal et al., 2022). Activated carbon-based materials are nowadays the most favoured electrode materials thanks to their tremendous specific surface area with appreciable electrical conductivity, cost-effectiveness, ease of preparation and high electrochemical stability (Jiangqi et al., 2018), however, activated carbon-based materials are linked with reduced energy density and a declining affordability of fossil based-carbon source (Ching-fang Liu et al., 2019; D. Wang et al., 2016; Yan Zhang et al., 2021). The low energy density of ACs is linked to their high porosity of up to greater than  $500 \text{ \AA}$  (Sharma et al., 2019).

To overcome the challenges of solely carbon-based SCs electrode materials, much attention

is given to carbon composites such as carbon-metal oxide (C/MO) and carbon conducting polymer (C/CP) composites. In C/MO composites, Manganese oxide for example is integrated into the electrode material to address the challenges of wholly carbon electrodes. As an example, the synthesis of a ternary (rGO)/MnO<sub>2</sub>/carbon C/MO electrode leads to the realization of a specific capacitance of up to 356.5 F g<sup>-1</sup> using a scan rate of 10 mV s<sup>-1</sup> with 93.6% capacitive retention after 2000 cycles (L. Zhang et al., 2017). Though a remarkable power density of 13.5 kW kg<sup>-1</sup> was achieved, it is clear that the tradeoff between robustness and high performance at lower carbon concentration remains a challenge (Jia et al., 2018). Another way of forming C/MO composite is by the addition of Titanium oxide to graphene for minimizing the graphene's stunted mechanical flexibility (Li et al., 2019). For instance, TiO<sub>2</sub>-rGO C/MO composite was synthesized and specific capacitance of up to 572 F g<sup>-1</sup> using a scan rate of 1 Ag<sup>-1</sup> was achieved (Yue et al., 2019). A stunted capacitance retention of almost 84% after 5000 cycles was also achieved (using a scan rate of 10 Ag<sup>-1</sup>). However, it was demonstrated that the electrochemical performance of the SCs using this electrode is a function of the morphologies of the composite which is determined by the percentage composition by mass of TiO<sub>2</sub> and rGO.

For C/CP composites, conducting polymers are added to carbon for the realization of electrodes with lofty capacitance and appreciable thermal stability (Li et al., 2019). As a representative case, a C/CP composite with PPy as the active compositing agent on a multiwalled carbon nanotube was reported by Xiao and Zhou (Xiao & Zhou, 2003). The capacitance and specific energy of the SCs were reasonably enhanced in comparison with SCs with all carbon nanotubes. For the

composite system, a capacitance and specific energy of up to 87 F g<sup>-1</sup> and 1.82 W h kg<sup>-1</sup> were achieved against 21 F g<sup>-1</sup> and 0.58 W h kg<sup>-1</sup> respectively, of the non-composite system. However, the poor cycling stability cum poor rate performance of PPy remains a drawback (Karim et al., 2019). Moreover, the brittleness and insolubility of PPy makes it difficult to process (Deshmukh et al., 2017).

### Conducting Polymer-Based Electrode Materials

Conducting polymers are conductive organic materials composed of macromolecules crafted from multiple reciprocating molecules (Namsheer & Rout, 2021). Their distinct characteristics include ease of preparation, robust conducting attributes and high mechanical stability (Lo et al., 2020; Namsheer & Rout, 2021; Yaqun Wang et al., 2019). Conducting polymers are characterized by pseudocapacitance active functional groups and hence can store charge through both electrostatic and electrochemical processes (Snook et al., 2011). Consequently, the capacitance per unit mass of conducting polymer-based SCs is higher than that of carbon-based. Common examples of conducting polymers are polyacetylene (PPy), polythiophene (PTh), ethylenedioxythiophene (PEDOT), and polyaniline (PANi) (Snook et al., 2011). Research on SCs electrodes based on conducting polymer is now at an advanced stage. The most well-researched conducting polymer-based electrode material is PANI due to its excellent conducting property concerning others. For example, the research team of Subramania and Devi prepared PANI-based nanofibres electrodes by surface active agent assisted dilute polymerization. The prepared electrodes showed a specific capacitance of almost 300 F g<sup>-1</sup>. Nevertheless, the electrochemical processes result in the general distortion and rupture of the PANI

network, thus, impoverishing the cycle stability.

In a separate study, Zhou et al. (H. Zhou et al., 2005) prepared PANi SC electrode by the pulse galvanostatic technique using a stainless steel based substrate with  $0.5 \text{ mol}^{-1}$  aqueous solution of  $\text{H}_2\text{SO}_4$ . Although a specific capacitance and specific density of  $609 \text{ Fg}^{-1}$  and  $26.8 \text{ Wh kg}^{-1}$  were achieved respectively using a discharge current density of  $1.5 \text{ mA cm}^{-2}$ , degradation of the capacitance occurred after 1000 cycles. Moreover, the resulted structural instability during charge/discharge processes could result in a relatively narrow potential window of the SCs electrode (Huanhuan Wang et al., 2016). These challenges necessitate the emergence of PANi composite configurations that integrates in the PANi matrix, additives such as carbon-rich substances or metal oxides. In this regards, an additive such as *p*-phenylenediamine could enhance the interfacial interaction of PANi with the employed electrolyte resulting in an enormous specific capacitance of  $550 \text{ F g}^{-1}$ . In a study involving polythiophene (Pth), it was reported that: 500 cycles without loss of capacity;  $40 \text{ mAh g}^{-1}$  at  $20 \text{ mV s}^{-1}$  was realized. Nonetheless, the authors acknowledged that still work has to be done to decrease the internal resistance of the synthesized network (binder effect) thereby improving the corresponding cyclability (Laforgue et al., 1999).

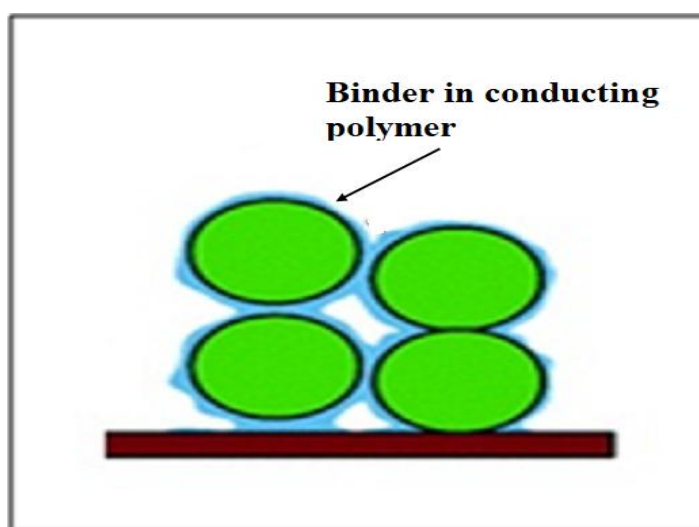
The main problem with CP-based SCs electrodes has to do with the design. Conventionally, the CPs are blended with binders to construct the electrode by using slurry-based wet chemical polymerization process (Bhadra et al., 2020). The ionic

diffusion rate of such electrodes during the Faradaic process is too slow because of the resistance provided by the binder materials, and this leads to substantial swelling and contraction of the CP and thus, poor durability and an insufficient cyclic performance of the electrode (C. Meng et al., 2010; Shown et al., 2015; Snook et al., 2011). Researchers are still striving to find ways of improving the cycle lifetime of CP-based electrode without sacrificing the corresponding benefits. In this regard, conducting polymer binary or ternary composites based electrodes is receiving great attention as a means of annihilating the problems of reduced cycle stability (arising from volume changes during charge-discharge processes) affecting the conducting polymer solely based electrodes.

These composites are usually conducting polymer-carbon (CP/C) or conducting polymer-metal oxides (CP/MO) fabricated by either ex-situ, in-situ, or one-pot fabrication methods (C. Zhao et al., 2020). As an example, transparent, binary  $\text{RuO}_2/\text{PEDOT}$  CCP/MO SC electrodes were fabricated using the aerosol-jet thermal spraying method (C. (John) Zhang et al., 2016). These composite electrodes displayed appreciable conductivity of  $279 \text{ S/cm}$  and high areal capacitance of  $1.2 \text{ mF/cm}^2$ . However, as reported by Bheekhun et al. (Bheekhun et al., 2018), aerogel powders are very difficult to coat by thermal spray guns, a manifestation of their incompatible granulometric and morphological features. Table 1 presents the strengths and weaknesses of different CP electrode materials while Figure 1 illustrates the CP-binder interface where high resistance manifests.

**Table 1:** Strengths and weaknesses of conducting polymer-based electrode materials (Q. Meng et al., 2017)

CP Electrode Material	Strength	Weakness
polyaniline (PANI)	Robust, superior conductivity, high specific capacitance, simple synthesis method, can easily be doped, tunable ionic conductivity	The capacitance per unit area depends on the fabricating conditions, reduced durability, can only work with specific electrolytes
polyacetylene (PPy)	Robust, ease of fabrication, relatively high specific capacitance per unit volume, high durability	Low capacitance per unit mass, can only be used as a cathode electrode, cannot be easily doped
polythiophene (PTh)	Robust, simple synthesis technique, good durability, environmental stability	Low conductivity, low capacitance per unit area



**Figure 1:** Effect of binder on conductive polymer-based SCs electrode

### Transition Metal Oxides (TMO) Electrode Materials

Transition metal oxides are compounds consisting of oxygen atoms bound to central transition metals forming different configurations. They are alluring electrode materials exhibiting bountiful redox chemistry thanks to their numerous oxidation states and coordination geometries. The commonest transition metal oxides (TMOs) used as supercapacitor electrode materials are manganese oxide ( $\text{MnO}_2$ ), vanadium oxide ( $\text{V}_2\text{O}_5$ ), ruthenium oxide ( $\text{RuO}_2$ ),  $\text{Co}_3\text{O}_4$ , cerium oxide ( $\text{CeO}_2$ ), zinc oxide ( $\text{ZnO}$ ), and cobalt oxide ( $\text{Co}_2\text{O}_3$ ) (Kuan Yew et al., 2018; Liang et al., 2021). Transition metal oxides-based SC electrodes are characterized by

tremendous energy storage ability, low cost, environmentally friendly, and high specific capacitance (Cui & Meng, 2020). In their pure form, transition metal oxides display appreciable electrochemical activities; however, their rate capability is low due to poor electronic and ionic conductivities and hence, their practical application is limited (Zhu et al., 2014). For example,  $\text{MnO}_2$  as electrode material for zinc-ion batteries is faced with the challenge of  $\text{Mn}^{2+}$  ions being continuously dissolved from the manganese oxide electrodes into the electrolyte leading to agglomeration and grave structural transformations in the course of insertion and extraction of the hydrated  $\text{H}^+/\text{Zn}^{2+}$  ions. The result of which is low rate capability (Y. Zhao et al., 2020).

For the above reasons and to achieve higher electrochemical performance, composite manganese-based materials such as Lithium Manganese Oxide ( $\text{LiMn}_2\text{O}_4$  or LMO) are being employed. However, challenges are still being faced with the composite compounds notably, low ionic conductivity at room temperature matched with reduced Lithium ions mass diffusivity (Yi Zhang et al., 2020). For example, Ahn et al. (Ahn et al., 2017) reported that  $\text{LiNi}_0.5\text{Co}_0.2\text{Mn}_0.3\text{O}_2$  based electrode displayed good reversibility of up to  $\sim 200 \text{ mAh g}^{-1}$  but with a reduced rapid capacity which is related to active medium deterioration attributed to the phase transition to the spinel and rock-salt at a high operational voltage of about 4.5 V. Similarly, it was demonstrated that while Co-HIPA and Ni-HIPA can retain reversible capacities of 1043 and 532  $\text{mAh g}^{-1}$  at 200  $\text{mA g}^{-1}$  for 200 cycles, it is not the case with Mn/Fe/Zn-HIPA as anode electrode because of the decay in the

electrolyte (Du et al., 2021). On the other hand, vanadium oxide is famous for its multiple electron transition with a theoretical capacity yield up to 294  $\text{mAh g}^{-1}$  with 2  $\text{Li}^+$  extraction, However, the challenges of poor ionic conductivity which triggers slow reaction kinetics, being less electronically reactive, capacity fading due to the amphoteric nature of vanadium, and toxicity issue limits its application as electrode material (S. Zhang et al., 2020)

With these TMO challenges, new strategies aimed at improving the conductivity/redox activity via tuning their nanostructures have been developed. These include inserting the TMOs into a conducting membrane, doping the metals, as well as mixing oxide composites with multiple oxidation states (Cui & Meng, 2020). Table 2 presents some advantages and disadvantages of some selected TMO electrode materials.

**Table 2:** Strength and weaknesses of transition metal oxide SCs electrode materials

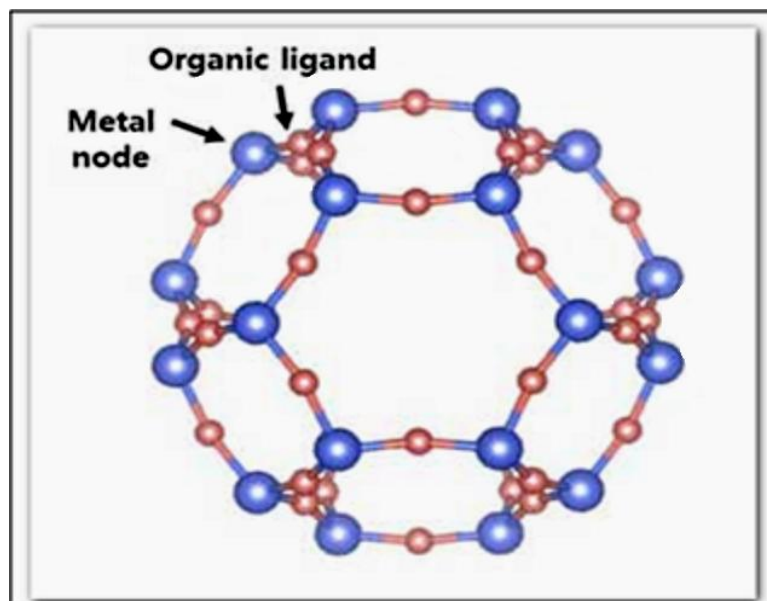
TMO Electrode Material	Strength	Weakness	Reference
Ruthenium oxide ( $\text{RuO}_2$ )	High specific capacitance (1400-2000 $\text{Fg}^{-1}$ ), good electrochemical reversibility, long life cycle, superb electrical conductivity, high chemical/thermal stability, wide voltage window	Too expensive, agglomeration effect	(Dipanwita et al., 2019; F. Yu et al., 2021)
Cobalt oxide ( $\text{Co}_3\text{O}_4$ )	High theoretical capacitance (up to 3560 $\text{Fg}^{-1}$ ), cost effective, environmentally friendly, superb chemical durability.	wide disparity between real and predicted specific capacitance, low electrical conductivity, reduced specific energy, large volume expansion-contraction, agglomeration effect, and low ionic diffusion	(Lu et al., 2020; X. Wang, Fu, et al., 2020; X. Wang, Hu, et al., 2020)
Manganese oxide ( $\text{MnO}_2$ )	Environmentally friendly, high theoretical capacitance (up to 1380 $\text{Fg}^{-1}$ ), wide potential range	Poor conductivity, sluggish ionic diffusion rate, weak structural stability	103-105, 106-108, (Wu et al., 2020)
Zinc oxide ( $\text{ZnO}$ )	Cost effective, environmentally friendly, large surface area superb ionic diffusion, high chemical/thermal stability, easy to dop	low conductivity, high volume shrinking in the cyclic process	(Geetha et al., 2019; Q. Yu et al., 2019)

### Novel Materials: Metal-organic Framework (MOFs) Electrode Materials

MOFs represents a novel class of sophisticated porous crystalline materials fabricated from an assembly of metal ions and organic ligands (Xinyu et al., 2022). Metal-organic frameworks (MOFs) as SCs electrode materials are receiving much attention from the scientific community, due to their tunable pore size distribution, superb porosity, ease of preparation, and structural flexibility (S. Zhou & Sun, 2021). Furthermore, blending with diverse electrode materials, such as metal oxide-based and carbons based decreases the resistance of MOFs and consequently increases the conductivity and chemical stability, an essential pre-requisite of SC electrode materials. Common MOFs materials used as SCs electrode material are the pristine and composites MOFs, MOFs-extracted nanoporous carbons, and MOFs-extracted metal oxides. On the downside, the challenges

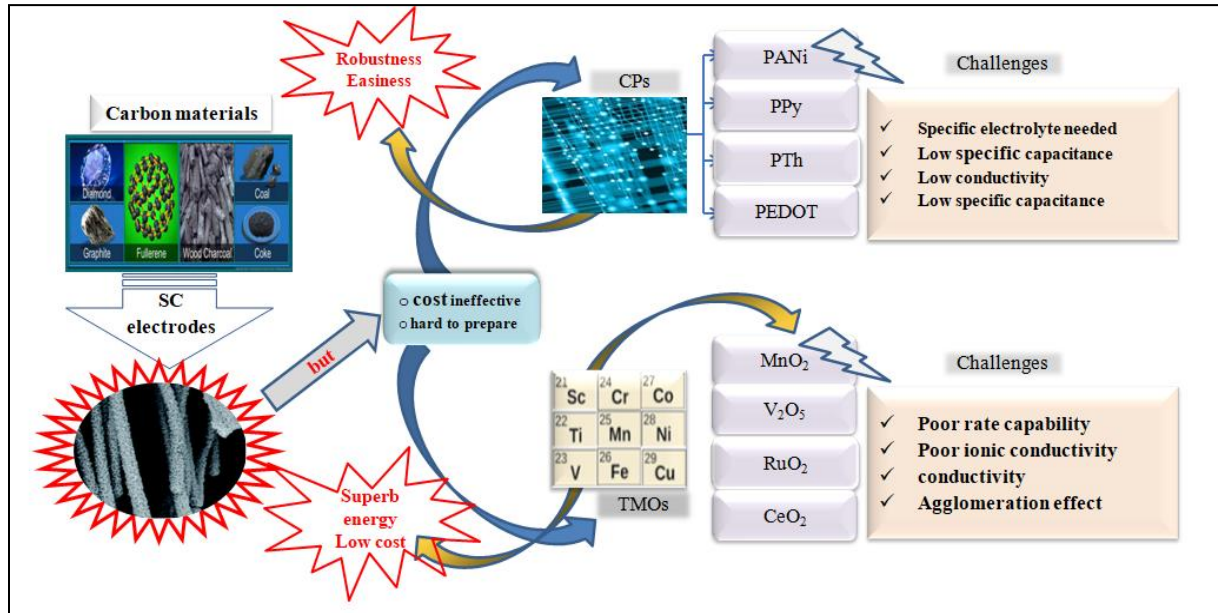
associated with MOFs are: theoretical prediction of the influence of MOFs microstructure on the electrochemical kinetics of MOFs-based SC is still insufficient, flaws (such as high internal resistance) resulting in stunt conductivity and reduced cycle stability manifests.

These drawbacks limit their application as SC electrode material. moreover, the structure-composition-fabrication method of MOFs are not yet advanced for the production of SC electrode materials for ensuing industrial application, the optimum-performance design of electrodes and supercapacitor devices has not been achieved due to lack of study coupling experimental description and theory (Jayesh et al., 2020; Xinyu et al., 2022). Figure 2 demonstrates the schematic illustration of MOFs consisting of metal ions and organic ligands whereas Figure 3 highlights the different types and challenges of SC electrode materials.



**Figure 2:** Schematic illustration of MOFs consisting of metal ions and organic ligands





**Figure 3:** Highlights of the different type of electrode materials with their challenges

### Supacapacitor Electrolytes

Electrolytes as materials medium through which the SCs electrodes are connected, play an integral role in the performance of supercapacitors (Qin et al., 2020; Verma et al., 2021). Conductivity is a key parameter to the performance of electrolytes, and it depends on the concentration of free ions and ionic mobility (Bo et al., 2018; Hao et al., 2013; Z. Zhao et al., 2015). Ions are produced via different chemical reactions and upon application of potential difference, an electric field is generated between the electrodes (Chernysheva et al., 2018; Chunli Liu et al., 2021). The energy density of SCs is highly dependent on the operating and decomposition voltages emanating from the ionic conductivity of the chosen electrolyte (Zhong et al., 2015). Hence, improving the electrolytic cell voltage remains a key challenge (Schutjajew et al., 2019). The basic requirements of an optimal SCs electrolyte are large potential window, extensive range of working temperature, low viscosity, large ionic conductivity, high electrochemical and

thermal stability, environmentally feasible, cost-cutting and low flammability (Patel et al., 2021). As of present, no single electrolyte satisfied all these requirements (Chodankar et al., 2020). Founded on the ion type, ion size, ion concentration, and ion interaction with solvent, electrolytes are categorized into three groups: liquid, solid/quasi-solid and redox-active (Ulihin et al., 2013) and are reviewed hereunder.

### Liquid Electrolytes

Liquid electrolytes are either aqueous, non-aqueous or ionic (Hannan et al., 2017; Roy et al., 2021). All electrolytes serve the same purpose in electrochemical devices (Xu, 2004). They are medium through which ions migrate to their opposite charge electrode with their volume un-depleted (Chunli Liu et al., 2021; Miller et al., 2018). The mobility of ions depends on the nature of the electrochemical kinetics and electrode materials (W. Liu et al., 2022; Tao et al., 2020). For its unsophisticated laboratory examination procedure, liquid electrolyte is the most extensively researched (Pal et al., 2019;

Zhong et al., 2015). However, low potential window, water-decomposition, leakages, low-mobility, corrosions, flammability, chemical degradation, large viscosity, and volatility are the key issues (X. Cheng et al., 2017; Roy et al., 2021). For example, Webber and Ue, study on ionic electrolyte showed that more ions with large size were realized, this give rise to high viscosity and consequently low-mobility (Ue, 1994; Webber, 1991) Moreover, packaging difficulties of SCs with liquid electrolytes results in a surge in the bulkiness, weightiness and cost of materials (Roy et al., 2021). Kim *et al* (Kim et al., n.d.) discussed the fabrication of organic and ionic electrolyte in dry environment. It was observed that introducing water in either organic or ionic electrolyte facilitate gas evolution at large operating voltage and consequently lead to high pressure and bulkiness of the cell.

### 3.1.1 Aqueous electrolyte

Aqueous electrolyte is a mixture of solute and solvent in a proportion that make a desirable concentration (Pal et al., 2019). The conductivity of aqueous electrolyte depends on dielectric constant and viscosity and is antagonistic. Moreover, aqueous electrolyte has the highest literature upon all the electrolytes. Zhong et al. (Zhong et al., 2015) reported that more than 80% of electrolytes literature in 2014 were published on aqueous electrolyte. However aqueous electrolyte has narrow potential voltage which limits its industrial production and commercialization. Based on the nature of the solutions, aqueous electrolytes are divided into three groups: acidic, alkaline, and neutral. Examples are  $\text{Na}_2\text{SO}_4$ ,  $\text{H}_2\text{SO}_4$ ,  $\text{KOH}$ ,  $\text{NaOH}$ ,  $\text{KCl}$ ,  $\text{NaCl}$  and  $\text{NaAc}$  (Zhong et al., 2015). While acidic electrolyte produced large ions, the electrolyte volume increases due to reduction of oxygen to water leading to the rapture and damaging of the cell (Tao et al., 2020). Srinivasan and Weidner (V. Srinivasan, 2000) demonstrated

the large increase in the specific capacitance of nickel oxide electrode from (16 to 155) F/g upon exchanging  $\text{H}_2\text{SO}_4$  with  $\text{KOH}$  electrolyte. However, water decomposition mount pressure within the system that led to cell rapture (Kim et al., n.d.).

Alkaline electrolytes on the other hand are characterized by non-toxicity, low ionic resistance, and are commonly available, the downside is that of narrow potential voltage window which drastically lower the energy density of the corresponding SCs (Huang et al., 2019; Kim et al., n.d.). The cell can easily be damaged on increasing the potential voltage due to water decomposition (Kim et al., n.d.). The report of Xiong et al. (Xiong et al., 2018) confirmed the appreciable potential window of neutral electrolytes relative to that of acid and alkaline electrolytes. In the report a potential window in the range of (1.8–2.2) V of neutral  $\text{Na}_2\text{SO}_4$  aqueous electrolyte was achieved, however, the potential window is still shallow due to water decomposition. They also pointed out that carbon electrode despite its good qualities exhibits low potential activity in a neutral aqueous electrolyte. This emanate from the large hydrogen evolution which limits the potential window at 1V and consequently lead to cell rapture (Kim et al., n.d.; Xiong et al., 2018). Chloride ions from neutral electrolytes such as potassium chloride cause corrosion on the metal electrode. Kim et al. (Kim et al., n.d.) reported that strong acid or strong base electrolyte induced corrosion on nickel and aluminum electrode.

### *Non-aqueous Electrolyte*

This class consists of organic and ionic liquid electrolytes of abundant ions with little or no water. Ionic liquid electrolytes are molten salt at low temperature that consist of ions only (Armand et al., 2009). They have large potential window but with low ionic

conductivity due to large internal resistance. Nonetheless, the high flammability of its unique solvent poses great danger when used in any electrochemical device, again, toxicity and unprofitability limits its application (Qingyun Dou et al., 2018; Suo et al., 2015). Azais et al. (P Azais et, 2007) found large potential window in the range of (2.2 – 2.7)V in acetonitrile organic electrolyte. However, high flammability and toxicity of acetonitrile make it un-safe. Moreover, El-Kady (El-Kady et, 2012) compared efficiency of laser-scribed graphene (LSG) in aqueous, organic and liquid ionic (LI) electrolytes, LI based LGS appeared to be more efficient. However, it suffered from low conductivity and high cost.

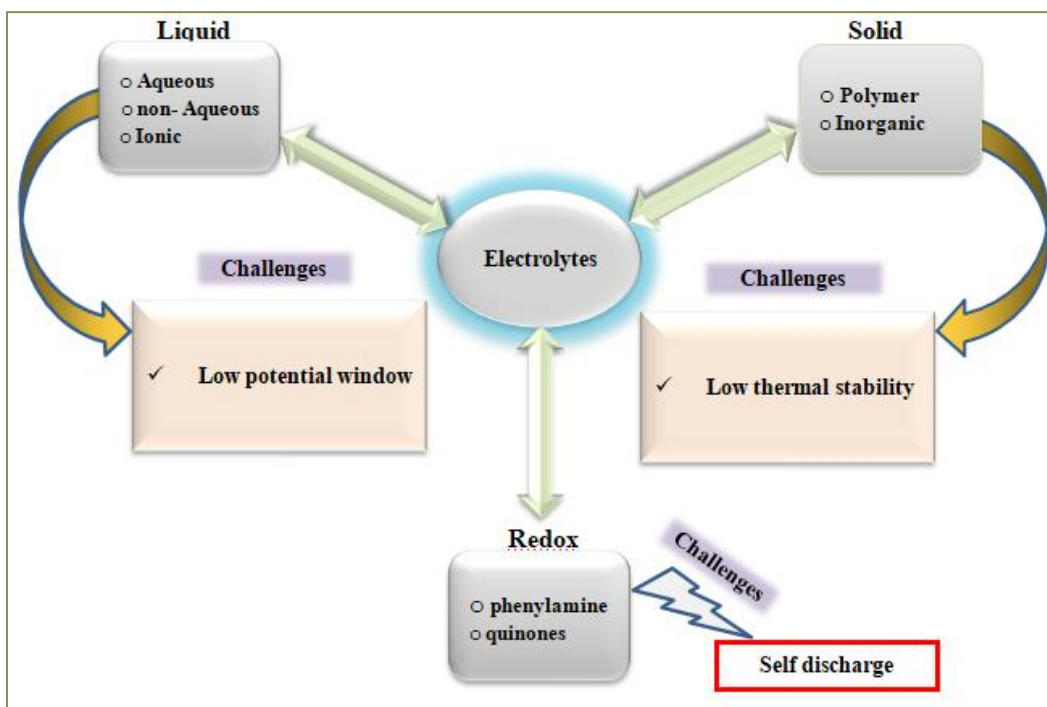
### Solid/Quasi-Solid Electrolytes

Different types of solid electrolytes are widely used in the fabrication of SCs. Reddy et al. (Govinda Reddy et al., 2018) successfully synthesized Alumino based lead nitrate ( $\text{PbNO}_3$ ) solid electrolyte. The characterization of the electrolyte was done based on impedance spectroscopy which revealed an improved ionic conductivity, however, it was well established that results obtained based on this spectroscopy is not reliable and thus the derived conclusions could be dubious and misleading (Drvarič Talian et al., 2022). Solid state electrolytes developed from solid polymer (organic solid electrolyte) have poor working temperature and continuity, however, with robust mechanical strength (Y. Cheng et al., 2020). On the other hand, Quasi-solid-polymer electrolytes are electrolyte with both low mechanical strength and poor working temperature range. To date, all efforts to improve the ionic conductivity and working temperature of polymer gel based electrolytes without compromising the corresponding

benefits proved abortive (X. Cheng et al., 2017). Ulihin et al. (Ulihin et al., 2013) demonstrated the use of inorganic solid electrolyte to provide large working temperature, but a mighty challenge of avoiding discontinuity between the electrolyte and the electrodes manifest.

### Redox Electrolytes

In redox based electrolytes, redox active species such as halide ions, transitional metals ions, phenylamine and quinones, are doped in aqueous or non-aqueous electrolytes. Significant increase in the electrochemical performance of SCs using these hybrid electrolytes is reported, yet the low decomposition voltage of water (1.23V) drastically reduces the energy density (Akinwolemiwa et al., 2015; Chodankar et al., 2020). Redox electrolytes can either be redox additive in which the active species are added to facilitate electron transfer at electrode/electrolyte interface, or redox active which could naturally exhibit redox reaction. SCs consist of two electrodes in between a membrane called separator. If redox additive is added, a redox polymer electrolyte may be formed. However, the active polymer is sensitive to electrons and could not be used as an electrolyte (Akinwolemiwa et al., 2015). To illustrate this, the report of Sun and Yan (Y. Sun & Yan, 2017) is considered. The report demonstrated that a double-electrode DSSCPS designed by Hagfeldt et al. with lithium trifluoromethanesulfonate ( $\text{LiCF}_3\text{SO}_3$ ) and propylene carbonate solution as the electrolyte suffered short circuit and self-discharging, because most of the  $\text{I}^-/\text{I}_3^-$  located on the surface of  $\text{TiO}_2$  dissolved in the electrolyte. Figure 4 highlights the challenges associated with the different types of S electrolyte.



**Fig. 4** Highlights of the different types of electrolytes with their challenges

### CONCLUSION

Proper choice of SCs material is the main issue regarding their performances. Here, we reviewed the different types of SC electrode and electrolyte materials with emphasis on their corresponding setbacks. To date no single electrode material whether carbon based, conducting polymer based, or metal oxide based matches all the desired electrode characteristics. Each is associated with its hiccups. Same applies to electrolyte materials. The scientific community is struggling to enhance remarkably the energy and power densities of SCs by developing new electrode and electrolyte materials. This review will create an avenue in determining the direction and future perspective of SC materials.

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