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Review Article

Recent Advances on Use of Graphene, Metal Oxides and Metal Organic Framework as Electrode Materials for High-Performance Supercapacitors: A Review

Sa'adu Lawal, Yusuf Abdulmalik, Ajibade I. Isa

Department of Physics, Federal University Gusau, P.M.B. 1001, Gusau Zamfara.

Abstract:

The increasing strain on finite fossil fuel reserves, coupled with the inherent limitations of renewable energy conversion technologies, has intensified the search for efficient energy storage solutions. Supercapacitors, renowned for their high-power density, extended cycle life, and rapid charge-discharge capabilities, are pivotal components of modern energy storage systems. This review synthesizes findings from previous studies that investigated the electrochemical performance of graphene, metal oxides, and metal-organic frameworks (MOFs) as electrode materials for supercapacitors. The analysis focuses on key performance indicators, including specific surface area, capacitance, energy density, power density, and cyclic voltammetry. Comparative analysis of the literature indicates that graphene consistently demonstrates superior performance, followed by MOFs, highlighting their potential for advancing high-performance supercapacitor technology. Further study into the optimization of these materials is necessary.

Keywords: Energy storage, electrochemical performance, specific surface area, capacitance, energy density, power density.

1. Introduction

Due to the continuous degradation of fossil fuels and other non-renewable energy sources, it is necessary for innovative ways to harvest, convert and store energy (Aliyu and Yusuf, 2019; Algarni *et al*, 2023). Numerous studies have been carried out on energy conversion and harvesting, but storage has been a serious concern, which necessitate researchers to come up with new storage technologies and constantly optimize existing technologies (Hoseini *et al*., 2023; Liu *et al*., 2023).

Amongst other storage devices, Supercapacitors, also referred to as Electric Double Layer Capacitors (EDLC) or Ultracapacitors form an integral part of the contemporary energy storage systems, thanks to their unique properties of power delivery and charge storage capabilities coupled with long cyclic stability (Yadlapalli *et al*., 2022; Satpathy *et al*., 2023; Sa'adu *et al*., 2024). A liquid/solid electrolyte divides two carbon-based electrodes in an EDLC arrangement, and a thin, porous separator sits between the electrodes and the electrolyte (Khan, *et al*., 2020; Otgonbayar, *et al*., 2023), as opposed to batteries, which store charge on either the cathode or anode electrode in response to faradaic redox reductions (Kumar *et al*., 2023). Supercapacitors have emerged a popular choice amongst other energy storage technology due to its high-power density, extended longevity, quick charge-discharge rate, cheap maintenance costs, and environmental (Kamila *et al*., 2021; Nguyen, *et al*., 2023). Supercapacitors are a popular and affordable battery substitute because of their higher performance (Thomas *et al*., 2019).

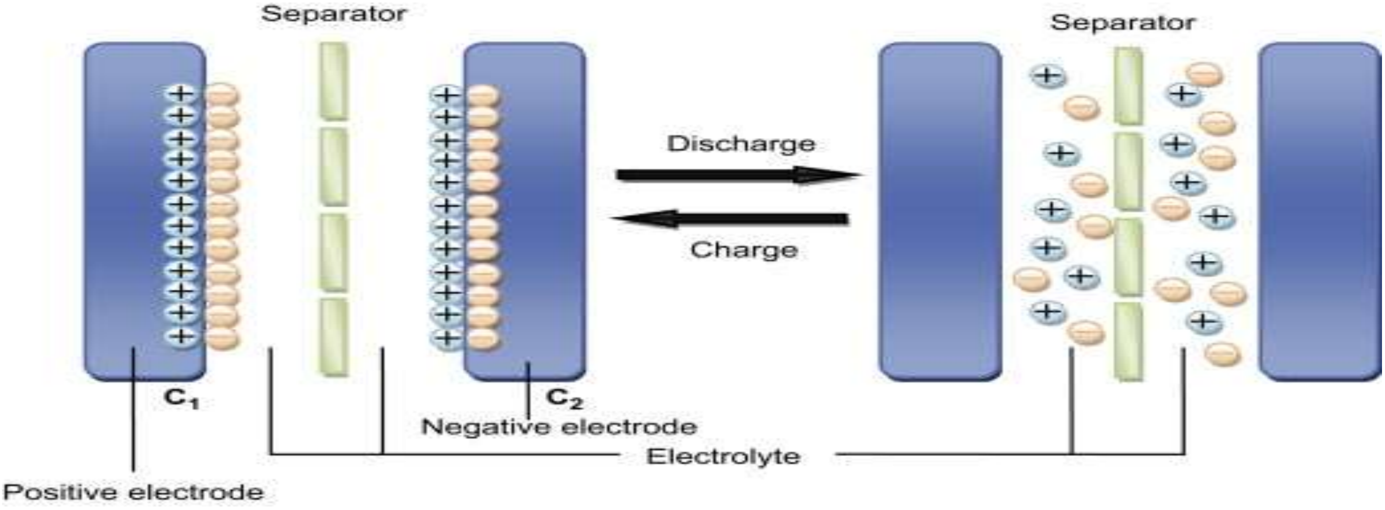

Figure 1: Schematic Diagram of a Supercapacitor (Li and Wei, 2013).

To achieve high capacitances, Supercapacitors use electrode materials with high specific surface area and thinner dielectrics (Jalal *et al.*, 2021; Babu *et al.*, 2024). The dependance of capacitance on the surface area of a material is shown in the Equation (Yusuf *et al.*, 2019; Chung, 2023; Zarzycki, 2023):

$$C = \varepsilon \frac{A}{d} \qquad (1)$$

The electrode materials utilized in Supercapacitors are electrochemically inert, such as carbon materials with the added benefits of high conductivity, bigger surface area, high capacitance and high temperature stability, they store energy by accumulation of electric charge (Gaikwad *et al.*, 2023Sriram *et al.*, 2024). Due to the highly developed pore structures and vast internal surface area of activated carbon, it is widely used in many industrial applications (Tian *et al.*, 2020; Ajibade and Maduka, 2024). Energy stored in a supercapacitor is a dependent variable of capacitance and its potential difference, where increase in either the capacitance or potential difference, results to an increase in the energy, as found in Equation 2 (Yusuf *et al.*, 2019; Pholauyphon *et al.*, 2024):

$$E = Cv^2 \qquad (2)$$

Activated carbon is a solid, porous, and carbonaceous adsorbent that is created by carbonizing and activating carbon-based materials at temperatures as high as 800 °C in inert environments (Zhu *et al.*, 2022). The performance of activated carbon as electrode material for supercapacitors will be optimized when doped with materials with good electrochemical properties such as Metal oxides, Metal organic frameworks and/or Graphene (Yan *et al.*, 2022; Yang *et al.*, 2023; Li *et al.*, 2024). Graphene, Metal-oxides and Metal-organic frameworks have displayed unique abilities to perform very well as dopant for supercapacitor electrodes (Ahmad *et al.*, 2024; Saleem *et al.*, 2024), especially Metal-organic frameworks which can be tuned to a desired structural architecture (Chaouiki, *et al.*, 2024).

This study is aimed at reviewing numerous works on Graphene, Metal-oxides and Metal-organic frameworks, in order to identify which among them would be a suitable candidate for incorporating to activated carbon to produce novel electrodes for supercapacitor applications.

2. Graphene

Graphene is layer of carbon atoms arranged in a two-dimensional hexagonal lattice structure, often described as honeycomb lattice (Sa'adu and Hashim, 2020; Bhattacharya and Jana, 2024). It is famous for its exceptional electrical conductivity, strength and thermal properties (Bhattacharya and Jana, 2024). Graphene has gained significant attention from scientists, researchers and engineers due to its unique properties (Santra *et al.*, 2024). It possesses immense potential applications in electronics, energy storage, and composite materials (Devi *et al.*, 2024). This has caused for its popular nickname "a marvel of modern materials" (kausar and Ahmad, 2024). Among its numerous properties are its mechanical strength, large surface area, flexibility, electrical conductivity and transparency (Esteghamat and Akhavan, 2023; Jia *et al.*, 2024).

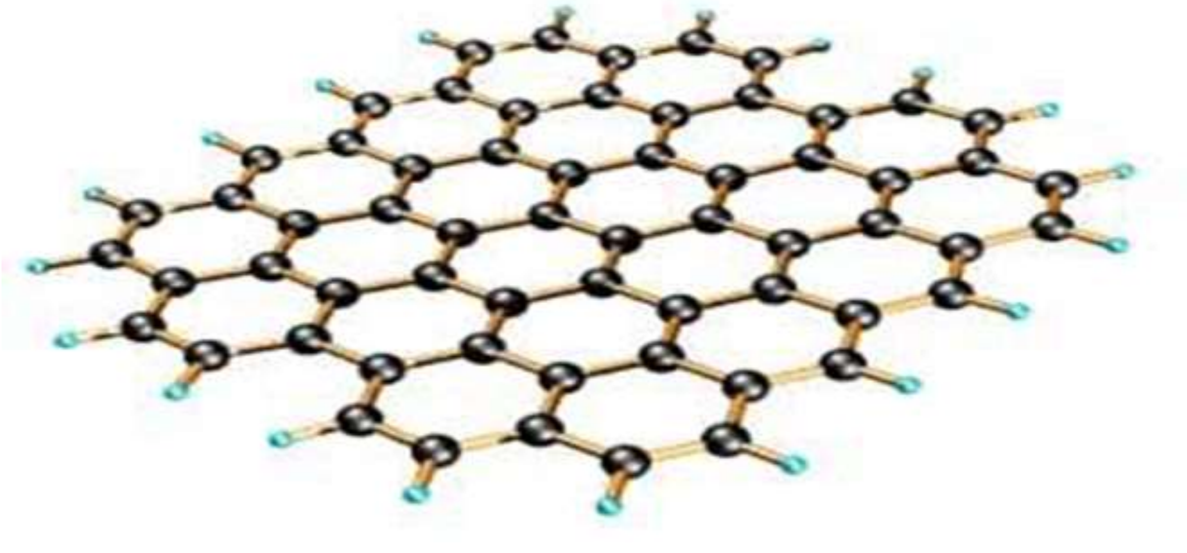

Figure 2: Structure of Graphene sheet (Radadiya, 2015).

Graphene has numerous applications such as Energy storage, Biomedical engineering, Material science, etc. (Ahmad *et al.*, 2024; Priyadarshi *et al.*, 2024).

2.1 Production of Graphene

Graphene is produced via two broad methods called Top-down and Bottom-up (Borane *et al.*, 2024; Nowduru, *et al.*, 2024).

2.1.1 Top-down method of producing graphene

In this method, a bulk of Graphite (which is an allotrope of Carbon) is subjected to a continuous exfoliation, until desired graphene layers are obtained (Satra *et al.*, 2024).

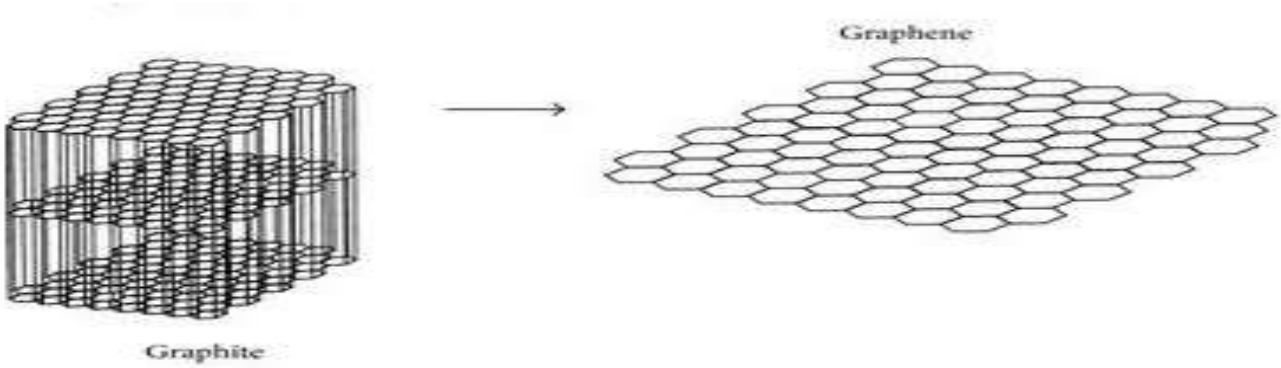

Figure 3: Schematic Diagram of Graphene Production from Graphite. (Skoda et al., 2014).

The exfoliation of graphite to obtain graphene of two types, i.e., mechanical exfoliation and liquid-phase exfoliation (Sumdani *et al*., 2021; Kaur *et al*., 2024).

Scalability, cost and ability to produce larger graphene has made Top-down method a more desirable method, especially for commercial purpose (Nair *et al.*, 2024).

2.1.2 Bottom-up method of producing graphene

In this process, a substrate material such as copper or nickel is subjected to chemical vapor deposition (CVD) at high temperature, where hydrocarbon gas is decomposed on the metal, forming graphene (Anisur *et al*., 2024). The process offers a great control over the properties and nature of the graphene produced (Olatomiwa *et al.*, 2022).

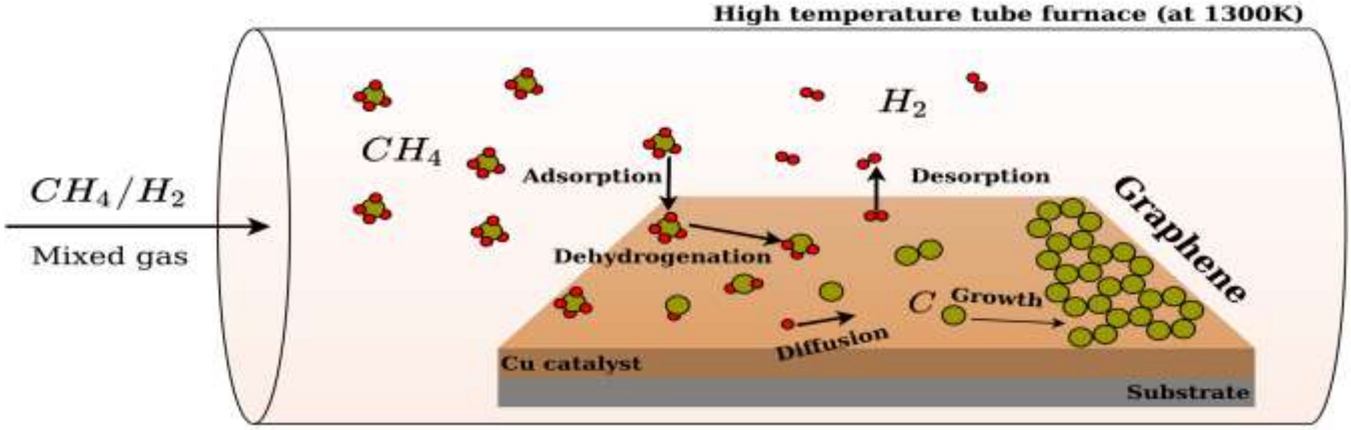

Figure 4: Production of Graphene using CVD (Esmaeilpour et al., 2023).

Graphene produced in this method are of high quality, with no defects, and their thicknesses and layer numbers can be controlled, as against those produced with top-down method (Abid *et al.,* 2022; Goethem *et al.*, 2024).

2.2 Applications of graphene

Graphene has numerous applications such as: Electronics, energy storage, material science, Biomedical Engineering etc. (Karaci and Acarali, 2024).

Graphene and Graphene-Oxides were used as precursor materials, mostly doped with different materials, depending on the choice of the researcher.

Edison *et al.* (2021) electrochemically exfoliated graphite to obtain graphene sheet, which yielded a specific capacitance of $40.83Fg^{-1}$, relatively small energy density of $3.03Whkg^{-1}$ and a power density of $562.5Wkg^{-1}$ was obtained at a current density of $0.1Ag^{-1}$. Qiu *et al.* (2023) used pyrogallic oligomers to dope Graphene, and a specific capacitance of $835Fg^{-1}$ was obtained, with an energy density of $4Whkg^{-1}$, power density of $90{,}000Wkg^{-1}$ at a current density of $0.5Ag^{-1}$. 90% retention was achieved after $20{,}000$ cycles. The process of freeze-drying and carbonization was used to fabricate a 3D carbon aerogel using pristine graphene, doped with 2,2,6,6-tetramethylpiperidine-1-oxyl, specific capacitance of $134.09Fg^{-1}$ was obtained at a current density of $0.5Ag^{-1}$, an impressive 99.3% of its capacitance was retained after 5,000 cycles (Wang *et al.*, 2023). Duisenbek *et al.* (2024) used potassium hydroxide to dope graphene. The material possesses a surface area of $1924m^2g^{-1}$, a specific capacitance of $131Fg^{-1}$, but the energy density was $4.54Whkg^{-1}$ and power density was $280Wkg^{-1}$ at a current density of $1Ag^{-1}$. Mohamed *et al.* (2024) incorporated Activated carbon to graphene to obtain an electrode with a specific capacitance of $490Fg^{-1}$, energy density of $58.9WhKg^{-1}$, power density of $9210Wkg^{-1}$ at $1Ag^{-1}$. After $5{,}000$ cycles, it retained up to 88.5% of its capacitance.

Kiafiroozkoohi *et al.* (2024) came up with a tungsten oxynitride nitrogen-doped graphene (WON-NG) electrode for supercapacitor, the specific capacitance of $1079.4Fg^{-1}$ was obtained, the energy density and power density of $81.6Whkg^{-1}$ and $5005.4Wkg^{-1}$ respectively at $1Ag^{-1}$. 88.7% of its capacitance was retained. Jiang *et al.* (2024) used a boron-doped graphene aerogel as an electrode where $3056.4Fg^{-1}$ was obtained as the specific capacitance, its energy density was $50.2Whkg^{-1}$ and power density was $800Wkg^{-1}$. And 95.6% of its capacitance was retained after $10{,}000$. Zhang *et al.* (2024) doped graphene with Zinc and obtained an energy density of $129.9Whkg^{-1}$, its capacitive retention was 99.8% after $10{,}000$ cycles. Imtiaz *et al.* (2024) was able to get a specific capacitance of $1403.35Fg^{-1}$ and a reasonable energy density of $50.2Whkg^{-1}$, a power density of $254.1Wkg^{-1}$ at a current density of $1Ag^{-1}$ after doping graphene with Cerium vanadate ($CeVO_3$). Mathan *et al.* (2024) used graphene alone as a precursor material to fabricate an electrode, and its specific capacitance of $106.5Fg^{-1}$, the energy density was $44.4Whg^{-1}$ and the power density was $1{,}000Wkg^{-1}$ at $1Ag^{-1}$ current density.

Graphene-oxide has also been used as precursor material alone or doped with some other compounds to fabricate high efficiency electrode for supercapacitor applications. Dong *et al.* (2023) and Kumar *et al.* (2024) used graphene-oxide alone as a precursor material without a dopant. A large surface area of $159.53m^2g^{-1}$ was observed within the graphene and a specific capacitance of $104.3Fg^{-1}$ by Dong *et al.* (2023), though the values of the energy and power densities were not mentioned by the author, but 93% of its capacitance was retained after $5{,}000$ galvanic cycles at a current density of $2Ag^{-1}$. As for Kumar *et al.* (2024), the specific capacitance of the graphene was $75Fg^{-1}$, its energy density was $15Whkg^{-1}$ and its power density was $600Wkg^{-1}$ at $1Ag^{-1}$.

Quite a number of dopant materials were used for graphene-oxide by various researchers yielding good performances, with the earliest being Yuan *et al.* (2023) and Choi *et al.* (2023). Yuan *et al.* (2023) incorporated carbon nanotubes (CNT) to graphene and found the energy and power densities to be $60.6Whkg^{-1}$ and $850.2Wkg^{-1}$ respectively. Whereas, Choi *et al.* (2023) used carboxymethylated cellulose nanofibrils (c-CNF) to dope graphene-oxide, this resulted to a specific capacitance of $96.5Fg^{-1}$, an energy density of $2.14Whkg^{-1}$ and a power density of $49.9Wkg^{-1}$ at a current density of $0.5Ag^{-1}$ being the least value recorded in the literatures reviewed.

Other compounds like $NiCo_2S_4$, ZnMgAl, $NiFe_2O_4$, Ni_3S_2, $Ni/CoMgSO_4$ and Polythiophene/N were used to dope graphene-oxide as in *Table 1*, but those that have shown excellent results are $NiCo_2S_4$ doped on graphene-oxide, which yielded a specific capacitance of $1505Fg^{-1}$, an energy density of $25Whg^{-1}$ and a power density of $7227wkg^{-1}$ at $1Ag^{-1}$ (Ahamed and Kanagambal, 2024) and $Ni/CoMgSO_4$ doped on graphene-oxide by Salami and Sanjabi (2024). The specific capacitance was $667Fg^{-1}$, the energy density was $63.3Whkg^{-1}$ and an impressive power density of $11214.5wkg^{-1}$ at a current density of $1Ag^{-1}$. 94.5% of its capacitance was retained after $10{,}000$ galvanic cycles.

Sasikumar *et al.* (2024) also obtained an impressive result from the core-shell synthesis of nano-composite, consisting of graphene-oxide and nickel sulphide (Ni_3S_2) which produced a specific capacitance of $1052.5Fg^{-1}$, energy and power densities of $61.9Whkg^{-1}$ and $585.7Wkg^{-1}$. Abaft *et al.* (2024) used $NiFe_2O_4$ to dope graphene-oxide, the surface area was $16.60m^2g^{-1}$, the specific capacitance was $1393Fg^{-1}$, the energy and power densities are $14.39Whkg^{-1}$ and $168.157Wkg^{-1}$ respectively. ZnMgAl has proven to also be a good candidate for doping graphene-oxide, Siwach *et al.* (2024) fabricated an electrode with a specific capacitance of $656.7Fg^{-1}$, energy density of $31.73Whkg^{-1}$ and power density of $784Wkg^{-1}$ at a current density of $1Ag^{-1}$. The supercapacitor was able to retain 89% of its capacitance after $15{,}000$ galvanic cycles.

Table 1: Recent Reviews of Performances of Graphene and Graphene-Oxides.

Precursor Material	**Dopant Material**	**S.A** $(m^2g^{-1}$	**S.C** (Fg^{-1})	**E.D** $(Whkg^{-1})$	**P.D** (Wkg^{-1})	**C.D** (Ag^{-1})	**Cycle**	**Retention** (%)	**Reference**
Graphene	Nickel	-	40.83	3.03	562.5	0.1	-	-	Edison *et al.*, 2021
Graphene	Pyrogallic oligomers	-	835	4	90000	0.5	20000	90	Qiu *et al.*, 2023
Graphene	$C_9H_{18}NO$	-	134.09	-	-	0.5	5000	99.3	Wang *et al.*, 2023
Graphene	KOH	1924	131	4.54	280	1	-	-	Duisenbek *et al.*, 2024
Graphene	Activated Carbon	-	490	58.9	9210	1	5000	88.5	Mohamed *et al.*, 2024
Graphene	WON-N	-	1079.4	81.6	5005.4	1	10000	87.7	Kiafiroozkoohi *et al.*, 2024

Graphene	Boron	-	3056.4	50.2	800	-	10000	95.6	Jiang *et al.*, 2024
Graphene	Zn	-	-	129.9	-	-	10000	99.8	Zhang *et al.*, 2024
Graphene	$CeVO_3$	-	1403.35	25.05	254.1	1	5000	-	Imtiaz *et al.*, 2024
Graphene	-	-	106.5	44.4	1000	1	-	-	Mathan *et al., 2024*
Graphene-Oxide	CNT	-	-	60.6	850.2	-	-	-	Yuan *et al.*, 2021
Graphene-Oxide	-	159.53	104.3	-	-	2	5000	93	Dong *et al.*, 2023
Graphene-Oxide	c-CNF	-	96.5	2.14	49.9	0.5	-	-	Choi *et al.*, 2023
Graphene-Oxide	-	-	75	15	600	1	2000	-	Kumar *et al.*, 2024
Graphene-Oxide	$NiCo_2S_4$	-	1505	25	7227	1	5000	85.6	Ahamed and Kanagambal, 2024
Graphene-Oxide	ZnMgAl	-	656.7	31.73	784	1	15000	89	Siwach *et al.*, 2024
Graphene-Oxide	$NiFe_2O_4$	16.60	1393	14.39	168.157	8	1000	82	Abaft *et al.*, 2024
Graphene-Oxide	Ni_3S_2	-	1052.5	61.9	585.7	1	10000	94.5	Sasikumar *et al.*, 2024
Graphene-Oxide	$Ni/CoMgSO_4$	-	667	63.3	11214.5	5	3000	83.2	Salami and Sanjabi, 2024
Graphene-Oxide	Polythiophene/N	-	455	13.1	279	-	1500	94	Khanary *et al.*, 2024

S.A: Surface area, S.C: Specific capacitance, E.D: Energy density, P.D: Power density, C.D: Current density.

3 Metal-Oxides

Metal-oxides are a class of inorganic compounds that consist of a metal cation bonded to an oxide anion (O^{2-}) (Talaat *et al.*, 2021; Gupta *et al.*, 2024). They are mostly solid at room temperature, although, they can be crystalline or amorphous (Lemieszek *et al.*, 2024; Peng *et al.*, *2024*).

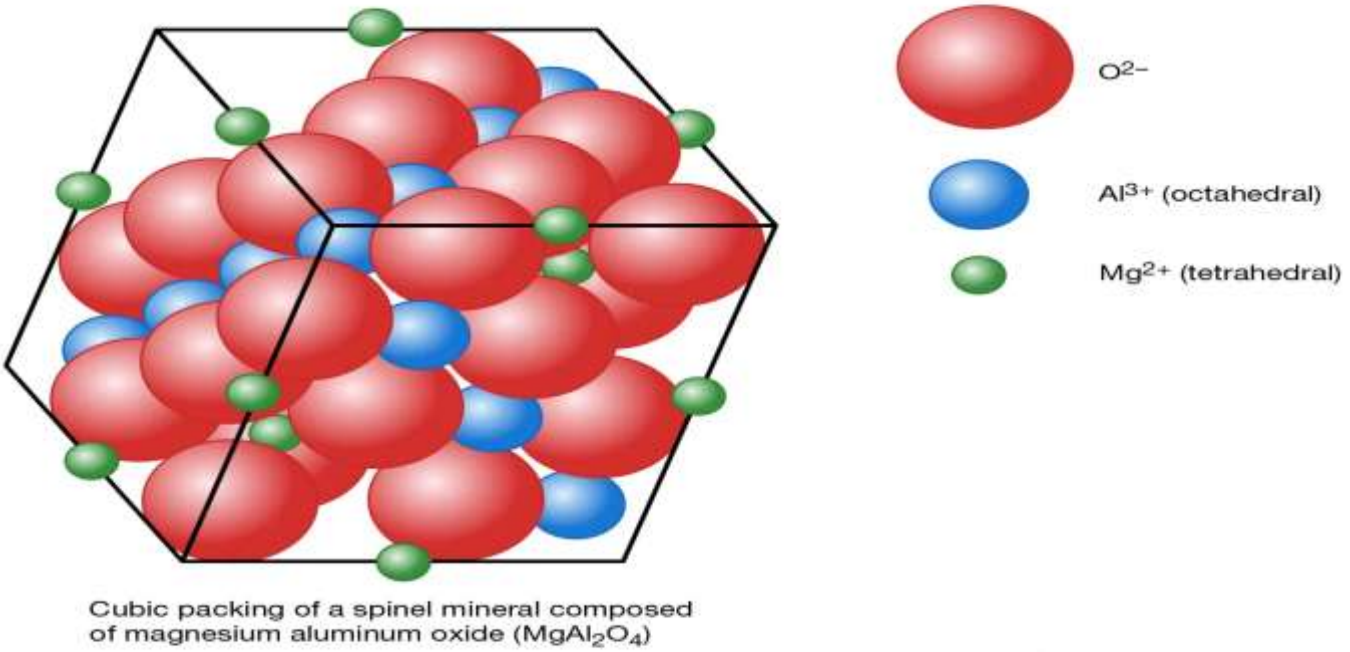

Figure 5: Atomic Structure of Metal-Oxide (Encyclopedia Britannica: Klein, 2024).

Supercapacitor electrode precursors have been doped with metal-oxides, due to the improved capacitance, enhanced conductivity and structural stability of metal-oxides (Qi *et al.*, 2024; Tatrari *et al.*, 2024).

3.1 Production of Metal-oxides

Being metal-oxides compounds formed from the combination of a metal and oxygen (Zhou *et al.*, 2024). Here are some of the methods of producing metal-oxides:

3.1.1 Thermal decomposition

Thermal decomposition is an endothermic process in which a larger compound breaks down into two or more smaller products when heated (Gong *et al.*, 2024). In this process, metal salts, hydroxides or carbonates are heated in air or oxygen (Wu *et al.*, 2024; Pawar and Kim, 2024). Example is heating of copper carbonate to give copper oxide and carbon dioxide:
$CuCO_3 \xrightarrow{yields} CuO + CO_2$ (Ali *et al.*, 2022).

3.1.2 Combustion

Combustion is a chemical reaction that occurs between fuel and an oxidizing agent, usually oxygen, accompanied by the release of energy (thermal energy) (Gulec, 2024).
In this process metals or compounds containing metals are burnt in the presence of air or oxygen (Siegmund *et al.*, 2024). Such as burning of Magnesium to get Magnesium oxide (Dai *et al.*, 2024):
$2Mg + O_2 \rightarrow 2MgO$ (Ilango *et al.*, 2024).

3.1.3 Hydrolysis

Hydrolysis is the process in which a compound is broken down after reacting with water (Azizi *et al.*, 2024). In this case, metal salt reacts with water or base to form hydroxides which can further be decomposed to metal oxides (Tichit *et al.*, 2024):
$AlCl_3 + 3\,NaOH \rightarrow Al(OH)_3 + 3\,NaCl$ (Ndubueze *et al.*, 2024).

3.1.4 Electroplating

Electroplating is the use of electrolysis to deposit metal oxides on a substrate (Rajoria *et al.*, 2024).

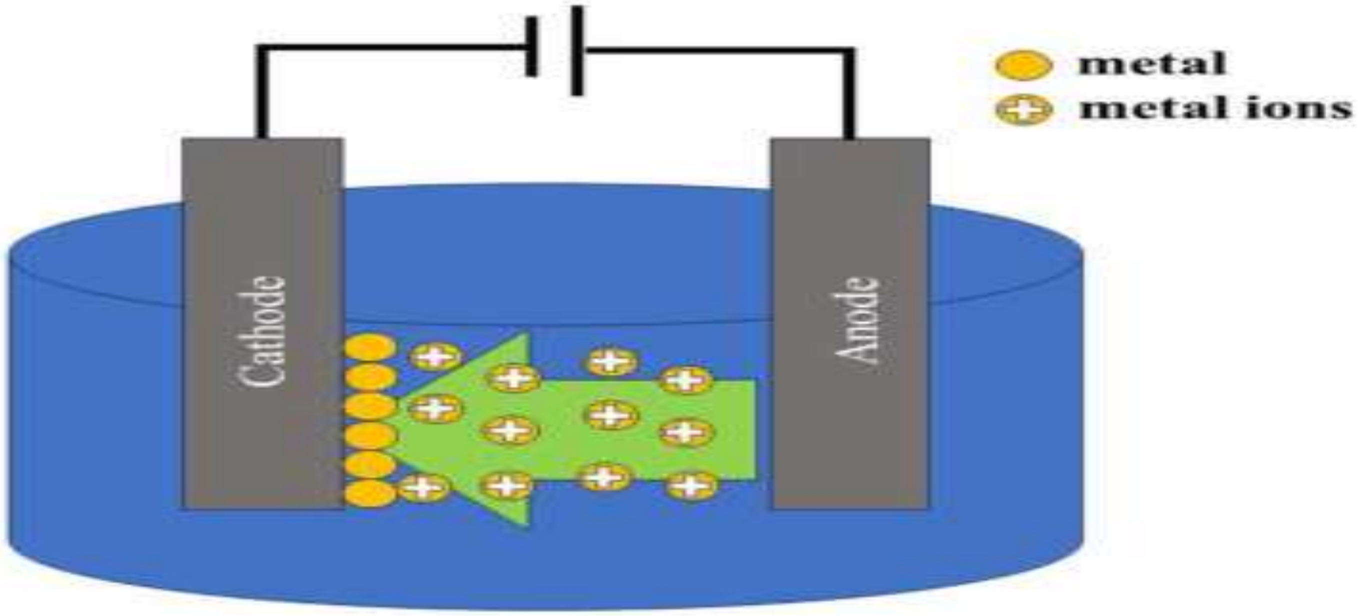

Figure 6: Electroplating process of a metal (Zhu et al., 2024).

3.1.5 Chemical vapor deposition

Chemical vapor deposition is technique used to deposit thin-films of materials onto a substrate (Weston *et al.*, 2024).

3.2 Applications of Metal-oxides

Metal-oxides have numerous applications which include: catalysis, electronics, pigments and colors, materials etc. (Marouzi *et al.*, 2024).
Latif and Maqsood (2024) synthesized a mixed metal-oxide of Lanthanum and Zinc, using a sol-gel method. The specific capacitance was $\mathbf{188.54Fg^{-1}}$ at a current density of $\mathbf{1Ag^{-1}}$, with a capacitive retention of $\mathbf{82\%}$ after $\mathbf{5,000}$ galvanic cycle. Surulinathan *et al.* (2024) developed a novel metal-oxide mixture of Cobalt oxide and Cerium oxide through a one-step hydrothermal method, and the performance of the supercapacitor was impressive, with energy and power densities of $\mathbf{45.3Whkg^{-1}}$ and

$\mathbf{4320Wkg^{-1}}$ respectively. The capacitive retention was up to $\mathbf{93.1\%}$ after $\mathbf{5,000}$ cycles. Ramadoss *et al.* (2024) noticed the relatively low intrinsic conductivity of CuO, hence the need to enhance the performance by doping it with Ti_3C_2Tx, which is an appropriate transition metals ion. The surface area was $\mathbf{20.5m^2g^{-1}}$, energy and power densities were $\mathbf{93.34Whkg^{-1}}$ and $\mathbf{3679.25Wkg^{-1}}$ at a current density of $\mathbf{1Ag^{-1}}$. $\mathbf{95.4\%}$ of its capacitance was retained after $\mathbf{10,000}$ cycles. Sivakumar *et al.* (2023) used a two-step synthesis strategy comprising hydrothermal and heat treatment to prepare $NiWO_4/WO_3$ electrode, with a specific capacitance of $\mathbf{108Fg^{-1}}$, energy density of $\mathbf{33.77Whkg^{-1}}$ and a power density of $\mathbf{896.39Wkg^{-1}}$ at a current density of $\mathbf{1Ag^{-1}}$. After $\mathbf{10,000}$ cycles, it was able to retain more than $\mathbf{88\%}$ of its capacitance. Wang *et al.* (2021) developed interactive ternary metal oxide nano-ribbon electrode comprising of Nickel oxide, vanadium oxide and Manganese oxide, that possessed a specific capacitance of $\mathbf{788Fg^{-1}}$, energy density of $\mathbf{Whkg^{-1}}$ and a power density of $\mathbf{450Wkg^{-1}}$, and was able to retain $\mathbf{83.6\%}$ of its capacitance after $\mathbf{10,000}$ cycles. Talluri *et al.* (2021) developed a high entropy spinel oxide (HEO) as a nanoparticle-based supercapacitor electrode material. The HEO has a chemical composition of $(CrMnFeCoNi)_3O_4$, with a specific capacitance of $\mathbf{239Fg^{-1}}$, energy density of $\mathbf{24.1Whg^{-1}}$, and a capacitance retention of $\mathbf{76\%}$ after $\mathbf{1,000}$ cycles at $\mathbf{0.5Ag^{-1}}$. Ma *et al.* (2021) used metal hydroxide to dope nickel oxide. This resulted to significant specific capacitance of $\mathbf{2774.18Fg^{-1}}$, an energy and power densities of $\mathbf{65.33Whkg^{-1}}$ and $\mathbf{849.97Wkg^{-1}}$ respectively, at a current density of $\mathbf{1Ag^{-1}}$. Wang *et al.* (2024) prepared metal oxide electrode using Granular Activated Carbon (GAC) as a substrate to study the degradation of phenol by three-dimensional electrocatalytic oxidation for water treatment. The electrode was able to yield an impressive energy density of $\mathbf{750Whkg^{-1}}$. Hsu *et al.* (2024) explored the use of metal ratio and bimetal nanoarchitectonics of ammonia-based fluoride complex induced nickel hydroxide and manganese oxide composites as active materials of energy storage device. The material yielded a specific capacitance of $\mathbf{1385.2Fg^{-1}}$, energy density of $\mathbf{11.45Whkg^{-1}}$ and a power density of $\mathbf{350Wkg^{-1}}$. It was able to retain up to $\mathbf{87.5\%}$ of its capacitance after $\mathbf{10,000}$ galvanic cycles. Zhu *et al.* (2020) developed a ternary phase Aluminum-nickel-cobalt-Oxide (AlNiCo-O) electrode, prepared using hot-air oven method. Testing at a current density of $\mathbf{1Ag^{-1}}$, the energy and power densities were $\mathbf{63.3Whkg^{-1}}$ and $\mathbf{881.4Wkg^{-1}}$ respectively. With an impressive specific capacitance of $\mathbf{1008.5Fg^{-1}}$.

Table 2: Recent Reviews of Performances of Metal-Oxides.

Metal-oxide Material	**Dopant Material**	**S.A** (m^2g^{-1})	**S.C** (Fg^{-1})	**E.D** ($Whkg^{-1}$)	**P.D** (Wkg^{-1})	**C.D** (Ag^{-1})	**Cycle**	**Retention** (%)	**Reference**
La & Zn	-	-	188.54	-	-	1	5,000	82	Latif and Maqsood, 2024
Co_3O_4/CeO_2	-	-	-	45.3	4320	-	5,000	93.1	Surulinathan *et al.* (2024)
Ti3C2Tx/ Mo-CuO	-	20.5	-	93.34	3679.25	1	10,000	95.4	Ramadoss *et al.*, 2024
$NiWO_4/WO_3$	-	-	108	33.77	896.39	1	10,000	>88	Sivakumar *et al.*, 2023
$NiO/V_2O_5/MnO_2$	-	-	788	138	450	-	10,000	83.6	Wang *et al.*, 2021
$(CrMnFeCoNi)_3O_4$	-	-	239	24.1	-	0.5	1,000	76	Talluri *et al.*, 2021
NiO	Metal (OH)	-	2774.18	65.33	849.97	1	-	-	Ma *et al.*, 2021
Fe_2O_3/CeO_2	GAC	-	-	750	-	-	-	-	Wang *et al.*, 2024
$Ni(OH)_2/MnO$	NH_4BF_4/NH_4HF_2		1385.2	11.45	350		10,000	87.5	Hsu *et al.*, 2024
AlNiCo-O	-	-	1008.5	63.3	881.4	1	5,000	84.2	Zhu *et al.*, 2020

S.A: Surface area, S.C: Specific capacitance, E.D: Energy density, P.D: Power density, C.D: Current density.

4 Metal-Organic Frameworks

Metal-organic frameworks (MOFs) are a specific class of porous materials made-up of metal ions precisely coordinated to organic ligands, forming high surface area three-dimensional structure (Manzoor *et al.*, 2024). Amongst all the materials that have previously been studied, MOFs have shown good potentials, due to its unique electrochemical properties (Chettiannan *et al.* 2024; Huang *et al.*, 2024). These unique properties include its tunability, compatibility with other dopant materials like metal oxides, chemical and thermal stability, low weight, etc. (Li *et al.*, 2024; Sajid *et al.*, 2024).

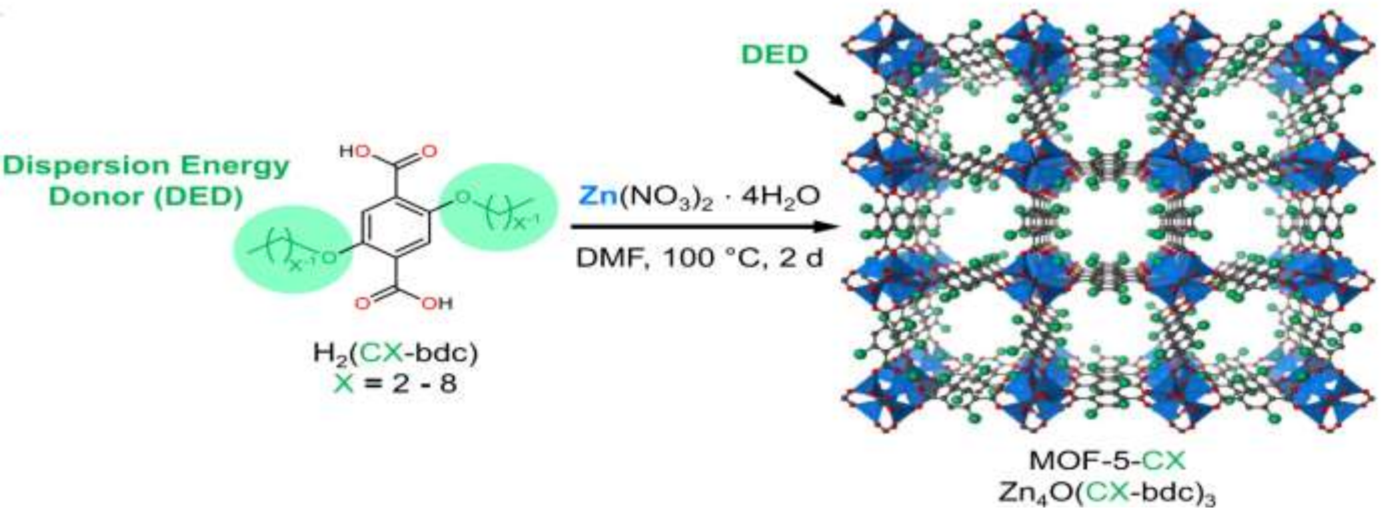

Figure 6: Structure of Metal-Organic Framework (Pallach et al., 2021).

MOFs have displayed potential for speeding reactions such as Oxygen reduction reaction (ORR), Hydrogen evolution reaction (HER) and Carbon dioxide reduction, which are key players in energy conversion technologies (Guo et al., 2024). This implies that, there are vast opportunities for researchers to delve into extensive and continuous studies on harnessing the abundant potentials of this magical material.

4.1 Production of Metai organic framework (MOF)

MOFs are typically produced through the synthesis of metal ions or clusters with organic linkers in a suitable solvent (Hayat *et al.*, 2024). Here are some of the methods of producing MOFs:

4.1.1 Solvothermal synthesis

Solvothermal synthesis is the process to produce Metal-organic frameworks in which metal salts and organic linkers are heated in a sealed container at high temperature and pressure (Muslim *et al.*, 2024).

4.1.2 Hydrothermal synthesis

Hydrothermal synthesis is similar to solvothermal synthesis, but in this case, it involves the reaction of the metal salts and organic ligands in aqueous solution at elevated temperature and pressure (Huo *et al.*, 2023).

4.1.3 Microwave-assisted synthesis

Microwave-assisted synthesis of metal-organic frameworks is an innovative technique that utilizes microwave radiation to speed the rapid formation of MOFs (Wei *et al.*, 2024; Xu *et al.*, 2024). This method is much embraced because of its fast reaction time, improved yield and potential for the control of the sample morphology (Soni *et al.*, 2024).

4.1.4 Mechanochemical synthesis

Mechanochemical synthesis of MOFs is an innovative approach that uses mechanical energy to facilitate their formation (Wang *et al.*, 2024). This method involves mechanical grinding or milling of solid samples typically metal salts and organic ligands, to induce chemical reaction, without the use of solvents or high temperature (Penczner *et al.*, 2024).

4.1.5 Electrochemical synthesis

Electrochemical synthesis is the method of using electrical energy to facilitate the synthesis of MOFs (Shi *et al.*, 2022; Zhang *et al.*, 2023). Here, electrochemical process is used to deposit MOFs onto a conductive substrate (Babu and Varghese, 2023). Its advantage over other methods is because thin film of MOFs can be produced (Sabzehmeidani *et al.*, 2024).

4.1.6 Template-based synthesis

Template-based synthesis is simply the use of a template, usually a polymer or a surfactant to modify the growth and/or morphology of MOFs (Jose *et al.*, 2024). Due to the fact that multiple MOFs can be reproduced with the same shape and size, this method is favorite for manufacturers of large-scale MOFs (Chen *et al.*, 2024).

4.2 Applications of MOFs

MOFs have demonstrated tremendous potential due to their endless applications (Zheng *et al.*, 2024). MOFs are used for gas storage and separation, catalysis, sensors, drug delivery, water purification, energy storage, electronics, optics, etc. (Haruna *et al.*, 2024). Hassan *et al.* (2024) developed a MOF, which is a combination of Zinc cobaltite ($ZnCo_2O_4$) and Nickel cobalt (NiCo). Hydrothermal and sonication technique was employed for the synthesis. The MOF was found to possess a specific capacitance of $984Fg^{-1}$, the energy and power densities were $92.4Whkg^{-1}$ and $930Wkg^{-1}$ respectively, at a current density of $1.5Ag^{-1}$. It was able to retain 92.5% of its capacitance after $12{,}000$ galvanic cycles. Liu *et al.* (2024) used an in-situ approach to synthesize Nickel-cobalt MOF, based on a thiophene ligand. The specific capacitance of the MOF was up to $1243Fg^{-1}$, with an energy density of $54.6Whkg^{-1}$ and a power density of $800Wkg^{-1}$ at a current density of $1Ag^{-1}$. It was able to retain 84.7% of its capacitance after $5{,}000$ cycles. Nechiyil *et al.* (2024) unveils a new free-standing carbon nanotube aerogel-metal organic framework electrode, which have shown exceptional dual functionality, both as an electrode and a current collector. It is a combination of functionalized carbon nanotubes (FCNT) and the metal-organic framework. The electrode exhibited an impressive specific capacitance of $465Fg^{-1}$ at $2Ag^{-1}$. The electrode had a cyclic stability of 92% after $6{,}000$ cycles. The energy and power densities were $40.5Whkg^{-1}$ and $1{,}600Wkg^{-1}$ respectively. Iqbal *et al.* (2024) fabricated a metal-organic framework made of pyridine 3,5 dicarboxylate (PYDC) ligands in combination with Cu and Co. The energy density recorded was $17Whkg^{-1}$ and power density of $2550Wkg^{-1}$. Kuo *et al.* (2024) utilized a simple and time-effective precipitation. The cobalt, nickel and manganese were used to design MOF-derived LDH with different metal species and numbers. The MOF presents a high specific capacitance of $1113.7Fg^{-1}$, the energy and power densities are $61.5Whkg^{-1}$ and $750Wkg^{-1}$. 92.5% of its capacitance was retained after $10{,}000$ galvanic cycles. Kim *et al.* (2024) prepared a porous nanocarbon by hybridizing MOF ZIF-8 and fluorinated polyimide, with a surface area of $1096m^2g^{-1}$. Its energy density is $10.7Whkg^{-1}$ and power density of $5{,}000Wkg^{-1}$. Cheng *et al.* (2023) fabricated a bimetallic MOF of cobalt and manganese, with a specific capacitance of $670.1Fg^{-1}$, an energy density of $17.9Whkg^{-1}$ and a power density of $785.7Wkg^{-1}$. It was able to retain 78% of its capacitance after $10{,}000$ galvanic cycles. Rozveh *et al.* (2022) synthesized a series of isoreticular Cu-MOFs with various metal ions (Co, Zn, Ni). The best performing mixed metal was Cu-Co with an energy density of $3.39Whkg^{-1}$ and a power density of $750Wkg^{-1}$. 86% of its capacitance was retained after $10{,}000$ galvanic cycles. Abid *et al.* (2023) used wet-chemical method to fabricate an Eu-MOF. The MOF has a specific capacitance of $1543Fg^{-1}$, the energy and power densities are $97Whkg^{-1}$ and $658.8Wkg^{-1}$, with an impressive capacitance retention of 93.58% after $10{,}000$ galvanic cycles. Pamei *et al.* (2022) synthesized two amino functionalized MOF, Co-BDC-NH_2 and Cu-BDC-NH_2, considering cobalt and copper as the metal sources. Its specific capacitance was $291Fg^{-1}$, the energy density was $16Whkg^{-1}$ and a power density of $749Wkg^{-1}$ at a current density of $1Ag^{-1}$. After $1{,}000$ cycle, the electrode was able to retain 86% of its capacitance.

Table 3: Recent Reviews of Performances of Metal-Organic Framework.

Metal Organic Framework	**Dopant Material**	**S.A** (m^2g^{-1}	**S.C** (Fg^{-1})	**E.D** ($Whkg^{-1}$)	**P.D** (Wkg^{-1})	**C.D** (Ag^{-1})	**Cycle**	**Retention** (%)	**Reference**
$ZnCo_2O_4$@NiCo	-	-	984	92.4	930	1.5	12,000	92.5	Hassan *et al.*, 2024
NiCo	-	-	1243	54.6	800	1	5,000	84.7	Liu *et al.*, 2024
MOF	FCNT	-	465	40.5	1,600	2	6,000	92	Nechiyil *et al.*, 2024
Cu-PYDC-MOF	-	-	-	17	2550	-	-	-	Iqbal *et al.*, 2024
CoNiMn	-	-	1113.7	61.5	750	-	10,000	92.5	Kuo *et al.*, 2024
ZIF-8	fPI	1096	-	10.7	5,000	-	-	-	Kim *et al.*, 2024
CoMn	-	-	670.1	17.9	785.7	-	10,000	78	Cheng *et al.*, 2023
Cu-MOF	Co/Zn/Ni	-	-	3.39	750	-	10,000	86	Rozveh *et al.*, 2022
$EuZrSe_3$	-	-	1543	97	658.8	3	10,000	93.58	Abid *et a.*, 2023
Co-BDC-NH_2/ Cu-BDC-NH_2		-	291	16	749	1	1,000	86	Pamei *et al.*, 2023

S.A: Surface area, S.C: Specific capacitance, E.D: Energy density, P.D: Power density, C.D: Current density.

5 Conclusion

The choice of materials for the fabrication of high-performance electrodes of supercapacitor plays a vital role in energy storage technology (Girirajan *et al.,* 2024; Phor *et al.,* 2024; Shah *et al.,* 2024).

From the performances of all the reviewed studies, graphene has proven to be the best candidate to use as a precursor material for the fabrication of supercapacitor electrodes, as it has the highest of all the individual electrochemical performance. This can evident in Table 1, where the largest surface area was recorded for the graphene at $1924m^2g^{-1}$ (Duisenbek *et al.*, 2024). This surface area is the largest surface area recorded for the three materials. Similarly, Jiang *et al.* (2024) obtained the highest specific capacitance of $3056.4Fg^{-1}$ from doping graphene with Boron. As for energy density and capacitive retention, Zhang *et al.* (2024) doped graphene with Zinc, where they recorded the highest energy of $129.9Whkg^{-1}$ and an impressive capacitive retention of 99.8% after 10,000 galvanic cycles.

Comparing the performance of metal-oxides and metal-organic frameworks in Table 2 and Table 3, metal-organic frameworks show noteworthy capabilities, considering its significant power density of up to $5{,}000Wkg^{-1}$.

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