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Recent development on nanocomposites based on quantum dots and metal oxides for supercapacitor applications: A mini-review

Ahmad Ismael Saber[1], Hasan Khalid Dabis[2], Nahed Mahmood Ahmed Alsultany[3], Heba Muhammad Hussein Abdulwahab[4], Aseel Salah Mansoor[5], Nasr Saadoun Abd[6], Fariba Alimola[7], *

[1]*Faculty of Dentisry, University of Al-Noor, Iraq*
[2]*Faculty of Chemistry, University of Ahl Al Bayt, Iraq*
[3]*Faculty of Dentisry, University of Science and Technology, Dhi Qar, P.O. Box 64001, Iraq*
[4]*Faculty of Chemistry, University of Al- Zahrawi, Iraq*
[5]*Faculty of Chemistry, University of Gilgamesh Ahliya, Iraq*
[6]*Faculty of Medical Technical, University of Al Farahidi, Iraq*
[7]*Research Laboratory of Polymer, Department of Organic and Biochemistry, Faculty of Chemistry, University of Tabriz, P.O. Box 51666, Tabriz, Iran*

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ABSTRACT

In this study, the supercapacitor performance of metal oxides and nanocomposites based on metal oxides and quantum dots to overcome the limitations of metal oxide electrode materials. By investigating the supercapacitor performance of nanocomposites based on metal oxide and quantum dots in three categories of carbon quantum dot, graphene quantum dot, and polymer dot, the performance of each quantum dot in improving the supercapacitor behavior of metal oxides was discussed. The results confirmed that the synergistic effect of quantum dots and metal oxides is improving the supercapacitor performance of nanocomposites. Also, the functional groups on the surface of the quantum dots accelerate the rate of penetration of ions. The results of this study confirmed that polymer dots have better performance among quantum dots with polymer properties and quantum dots properties at the same time which provides research opportunities for researchers in the field of supercapacitors.

1. Introduction

Due to the environmental pollution that exists in the use of fossil fuels, the need for an alternative energy source has become an important strategy in the industry. [1-7]. Environmental pollution caused by industry not only increased environmental problem but also threatened human health. Therefore, an alternative energy source was considered as a fundamental challenge [8-13]. With the advancement of technology, fuel cells, and supercapacitors were considered alternative energy sources. The limitation of sodium and lithium resources and the use of batteries in the industry has faced problems [14-16]. Also, the short life cycle and low power density of batteries have limited the use of these energy sources in industry. Supercapacitors as energy storage systems with cheap and available electrode materials and long life cycles have become a promising energy storage system in the industry [17, 18]. Renewable energies (energy of wind and solar) are considered suitable alternatives to fossil fuels, but the many needs of industry for energy limit the use of these sources [19-21]. Recently, batteries, fuel cells, and supercapacitors were considered as an appropriate technology for fossil fuels [22-24]. High energy density batteries as energy storage devices have limitations such as production cost due to lack of lithium resources, low safety, and high weight. [7, 25-33]. According to the studies, supercapacitors are a suitable technology in energy storage systems. In other words, supercapacitors have a higher power density than batteries,

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* Corresponding author; e-mail: fariba.alimola@gmail.com
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and a higher energy density than ordinary dielectric capacitors [33-37]. However, supercapacitors have limitations, such as the low energy density. According to formula 1, the energy density was improved with the increase of the potential window and specific capacitance [38-41].

$E=1/2\ CV^2$ (1)

Therefore, targeted electrode materials synthesis is essential to achieve optimal properties (supercapacitors with high energy density) [37, 42, 43]. Supercapacitors are divided into three categories based on the electrode materials: I-electrical double layer supercapacitors (EDLC): supercapacitors distribute charge by absorption and desorption of electrolyte ion on the electrode/electrolyte interfaces. The graphene, graphene oxides, and carbon nanotubes are EDLC electrode materials. The advantages of EDLC supercapacitors are high charge/discharge rates, but these electrode materials have low energy density [44-46]. II-Pseudocapacitor: the charge storage process in this category of supercapacitors is done through the faradic process, so the energy density and specific capacity in these supercapacitors are higher than EDLC. Conductive polymers are quasi-capacitive electrode materials, and these polymers have little cyclic stability due to the swelling and shrinking of polymer chains during the doping/doping process [47-49]. III-The hybrid system is a combination of EDLC and pseudocapacitor, which has favorable supercapacitor properties due to the good cyclic stability of EDLC and the high specific capacity of the pseudocapacitor [50-54]. The defect of the hybrid supercapacitor is to avoid converting a normal supercapacitor into a battery [55-58]. Metal oxides are pseudocapacitor electrode materials that have high specific capacitance, but these materials have low power density [59-61]. To improve the power density of metal oxides [62], composites of carbon materials [23], conductive polymers [24, 63], and metal oxides were prepared [64]. To improve the electrochemical performance of metal oxide electrode materials, the composite based on metal oxide and (g-C3N4) Graphitic carbon nitrides were synthesized. The g-C3N4 nanoplate has metal electrodes including high nitrogen content, which is suitable for matching the electrode with electron accumulation, as well as nitrogen atoms in the form of pyridine, which improves the supercapacitance behavior, and quaternary nitrogen with a positive charge accelerates the speed of ion transfer. The high density of nitrogen in the g-C3N4 structure provides a suitable substrate for trapping ions, so the limitations of using metal oxides in supercapacitors can be overcome with the composite of g-C3N4 with metal oxides [65]. Beyond their role in energy storage, metal oxide-based nanomaterials and their nanocomposites have also gained significant attention in various fields of chemistry, such as organic synthesis, due to their high surface area, stability, and tunable surface properties [66, 67]. These nanocatalysts efficiently facilitate various organic transformations, offering advantages such as improved reaction rates, selectivity, and easy recovery, thereby supporting greener and more efficient synthetic processes [68].

In addition to their electrochemical and catalytic applications, nanostructured materials—including those based on metal oxides and quantum dots—are widely explored for environmental purposes. Computational studies play a crucial role in understanding the interactions between nanostructured adsorbents and pollutants, offering insights into adsorption mechanisms and guiding the design of advanced materials [69, 70]. These computational approaches can also be applied to nanocomposites based on metal oxides and quantum dots, deepening our understanding of their behavior in both energy storage and environmental remediation.

In recent years, various types of nanocomposites based on quantum dots and metal oxides were synthesized, and the synergistic effect of quantum dots and metal oxides improved the final supercapacitor performance [71]. Quantum dots alone as a microconductor in composites based on quantum dots and metal oxides have supercapacitor properties [72-75]. In other words, the functional groups on the surface of quantum dots as conductive components in composites accelerate the speed of ion penetration and increase the specific capacitance [76-78]. In the current review, our team reviewed the advances in recent years in electrode materials based on metal oxide composites of quantum dots. The effect of improving the performance of metal oxide supercapacitors by being composited with quantum dots (CQD, GQD, and PD) was investigated separately. The effect of each of the quantum dots in improving the performance of metal oxides and the synthesis methods of various types of quantum dots as well as the synthesis methods of nanocomposites based on the type of quantum dots were discussed and investigated. In the last part, it was mentioned the one-step methods of nanocomposite synthesis based on polymer dots and metal oxide, that in these methods, metal oxide and polymer dots are formed in one step and composited at the same time. Finally, recommendations and suggestions for electrode materials development based on metal oxide composites with quantum dots were discussed.

2. Structure of the supercapacitor

Supercapacitors are a group of energy storage devices whose structure has three main parts: current collector, separator، and electrolyte. The general structure of these energy storage devices is that the cathode and anode electrodes dipped in electrolytes are separated by a separator [79, 80].

2.1. Separator

The separator causes ions to move by creating insulation between the electrodes without an electrical connection. By reducing the thickness of the separator, the specific capacity increases. In other words, the performance of separators is such that they affect power and energy density by preventing short circuits. It is important to use a flexible separator for the flexible supercapacitor. The surface porosity of the separators must be sufficient to provide the electrolyte ion to the electrode [37, 81].

2.1.2. Natural Separator

To reduce environmental problems, using natural materials is a challenge. Separators should have properties such as macropores and thin membranes, which was confirmed in natural separators. In the studies conducted egg skin, duck egg skin, and goose egg skin were used as natural separators. Examining the physical and electrochemical properties of duck shell, goose shell, and eggshell confirmed the membrane of the eggshell has less resistance and better supercapacitor properties. In the field of using separators derived from natural materials are limited studies, which can be the field of new research for researchers [82].

2.2. Electrolyte

Choosing the appropriate electrolyte in supercapacitors is very important. In supercapacitors, the cell potential depends on the breakdown voltage of the electrolytes and also on the energy density. The power density of supercapacitors is also dependent on the equivalent series resistance, which subsequently depends on the electrical conductivity of the electrolyte [83, 84]. Electrolytes used in supercapacitors are divided into two categories, aqueous and organic. Aqueous electrolytes usually have a breakdown voltage of about 1 V, significantly lower than organic electrolytes (about 3 V). However, the conductivity of aqueous electrolytes is higher than that of organic electrolytes, which is desirable for high-power devices. Aqueous electrolytes also have important advantages such as low cost and ease of handling [85, 86]. The resistance of aqueous electrolytes is high compared to organic electrolytes however the small radius between ions and high conductivity of aqueous electrolytes offers a large specific capacitance compared to organic electrolytes. The high conductivity associated with aqueous electrolytes reduces the equivalent series resistance (ESR) that supercapacitors offer with high power density. Phosphoric acid, sulfuric acid, and potassium hydroxide are the main aqueous electrolytes used. Propylene carbonate and acetonitrile (ACN) are used in organic electrolytes [4]. Acetonitrile can dissolve large amounts of salt compared to other solvents, but it causes environmental problems due to its toxicity. Propylene carbonate-based electrolytes are biocompatible, have a wide electrochemical window, and have relatively adequate temperature, voltage, and conductivity. However, the important point is that in order not to reduce the voltage of supercapacitors, the amount of water in organic electrolytes should be at most 3-5 ppm [87, 88].

2.3. Current collector

The type of current collector is important according to the physical conditions of making supercapacitors. Current collectors are selected according to physical properties such as resistance to corrosion and heat. Due to the manufacturing conditions of supercapacitors, which require pressing and welding, thus thermal resistance is important [89, 90]. In addition, other criteria are important for choosing the current collector, such as wettability, transparency in some cases, and cost. For the flexible supercapacitor’s fabrication, flexible current collectors such as carbon fabrics, paper, polyamide, etc. were used. Current collectors are divided into two categories: metal foil and plastic. Metal foil collectors such as copper, nickel, and aluminum have good conductivity, compared to plastic current collectors, which have lower conductivity. But plastic current collectors have advantages such as low cost and high flexibility [60, 91].

2.4. Electrode material

Electrode materials play a major role in determining the electrochemical properties of supercapacitors. The electrode materials' characteristics significantly impact determining the supercapacitor's capacity, so choosing the right electrode materials for making a supercapacitor is important. The materials used as electrode materials in constructing supercapacitors include metal oxides, conductive polymers, carbon materials, and quantum dots. Metal oxides are the most common Faradaic active materials that are used as electrode materials for supercapacitors. The transition metal oxides such as RuO_2, Fe_3O_4, MnO_2, and NiO are used as electrode materials in Faradaic supercapacitors. Transition metal oxides are the most common redox pseudocapacitive materials, which have a higher specific capacitance compared to electric EDLC supercapacitors, and the increase in specific capacitance is related to the number of oxide states. However, metal oxides have low electrical conductivity and Faraday reactions in these materials only occur on the surface of the electrode. Quantum dots improve the speed of ion transfer due to having many functional groups on their surface.

3. The performance of metal oxides as electroactive materials of supercapacitors.

Because transition metal oxides have different oxidation states as pseudocapacitors, they can behave as supercapacitors.

3.1. Titanium dioxide

Titanium dioxide is considered an electrode material with low cost, biocompatibility, and high specific surface. Titanium dioxide is widely used as an electroactive material in supercapacitors due to having the highest charge density on the surface [92-94]. Various methods have been used for the synthesis of titanium dioxide [95]. Many researchers used electrochemical doping techniques to synthesize titanium oxide, but the synthesis time is long. Lai. His colleagues synthesized titanium dioxide using the peroxo sol gel method at a certain temperature (200), and hydrogen peroxide was used to neutralize the titanium oxide because the acidic or alkaline environment causes corrosion of the titanium oxide. In other words, by synthesizing titanium dioxide at different temperatures, the optimal temperature of titanium dioxide synthesis for supercapacitor application was determined to be 200 degrees. Finally, the asymmetric device was synthesized using activated carbon and titanium dioxide. Scheme 1a shows the overall scheme of the asymmetric device based on titanium dioxide and activated carbon. Scheme 1b shows the assembly of asymmetric device for activated carbon and titanium dioxide. Scheme 1c and d show the cyclic voltammetry of the asymmetric device at different potentials and the cyclic voltammetry of the asymmetric device at different scan rates, respectively [96]. The asymmetric device had good stability, which maintained 84% of its capacity up to 3000 cycles.

3.2. Cobalt oxide

Cobalt oxide is a group of transition metal oxides whose use in supercapacitor systems was investigated in different synthesis conditions and different morphologies, which can achieve optimal conditions by controlling the morphology.[97]. Rao et al. synthesized Ultra layered Co_3O_4 electrode materials by hydrothermal method. The results showed a specific capacitance of 548 F g^{-1} at 1 A g^{-1} [98]. In another study, Khosla et al synthesized Co_3O_4 nanoparticles with controlled morphology. In this study, by controlling the ratio of ascorbic acid to nickel nitrate (oxidizing agent), the Co3O4 nanoparticle structure was achieved with the same particle distribution, and the results of electrochemical analyses showed a specific capacitance of 166 F g^{-1} at 0.5 A g^{-1} [99].

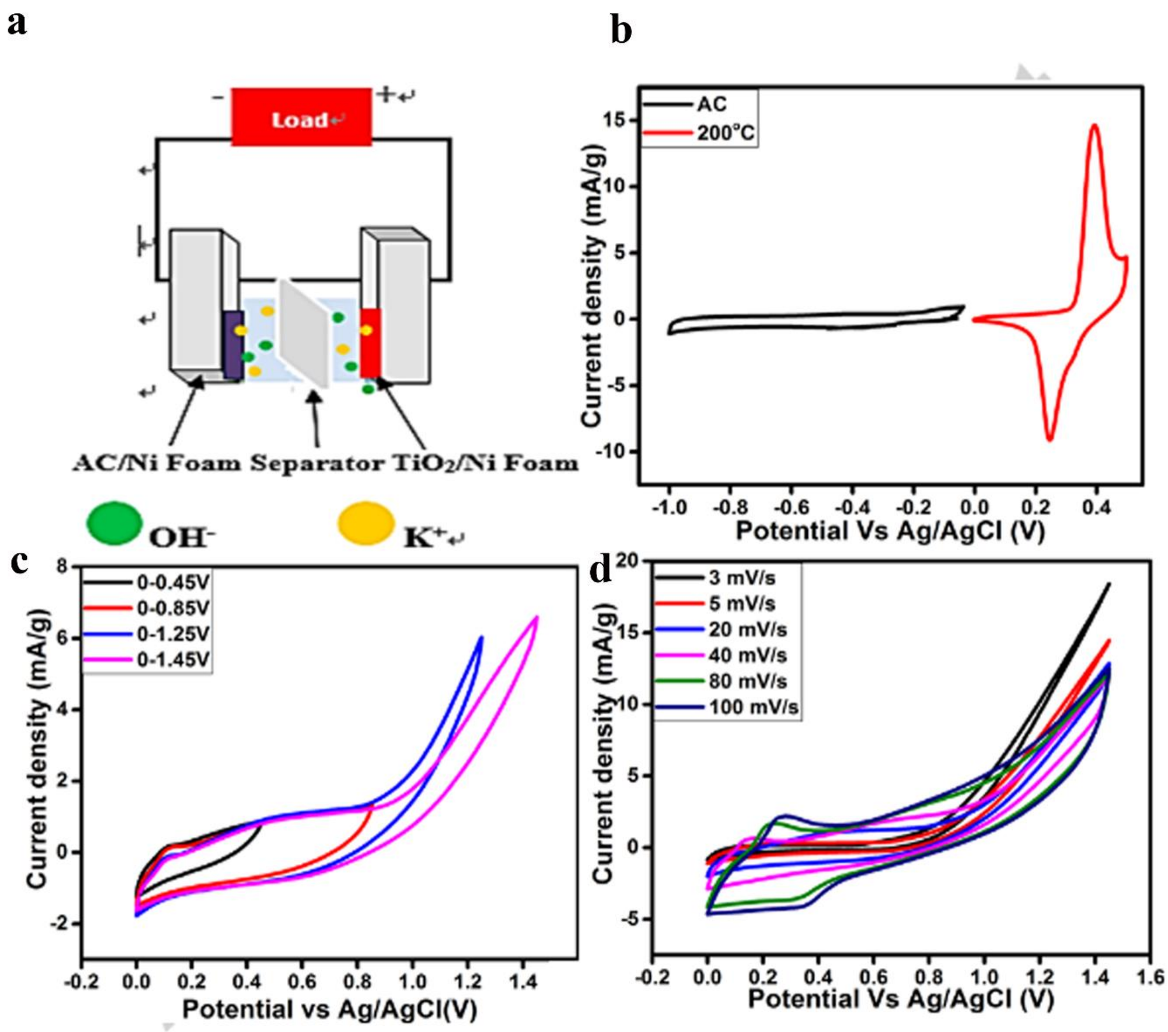
Cobalt oxide has advantages such as low price, and biocompatibility but has disadvantages of poor cyclic stability due to volumetric expansion. Therefore, limited reports are available on the performance of cobalt oxide in supercapacitors. To improve the performance of this material in supercapacitor systems, the composite of this material with carbon compounds such as graphene oxide or conductive polymers has been used. In recent years, cobalt oxide composites and quantum dots have been investigated, and the results have shown an increase in electrical conductivity and an improvement in the final electrochemical behavior of the nanocomposite compared to each of its components, such as a synergistic effect.

3.3. Molybdenum oxide

Molybdenum oxide with nanoplate, nanorod and, nanowire (α-MoO_3) and spherical (β-MoO_3) morphologies are synthesized. Liu et al synthesized α-MoO_3 nanobelt with dichloromethane and sodium molybdate by the hydrothermal method. Electrochemical analysis showed that the maximum specific capacitance of α-MoO_3 nanobelt was 280 F g^{-1} at 30 A g^{-1}, which is more than the nanowire was about 110 F g^{-1} [100]. Hussain et al synthesized molybdenum oxide using the solution combustion method. With increasing temperature, the structural phase reaction changed from beta to alpha. Investigating the electrochemical analyses of molybdenum oxides at different temperatures confirmed that with increasing temperature and changing from beta to alpha phase, the properties of the supercapacitor were improved. An increase in temperature from 300 to 500 degrees caused a phase change from alpha to beta, but with an increase in temperature up to 600 according to Scheme 2, the structure of molybdenum oxide was destroyed and the specific capacitance decreased [101]. In another study, Qi et al. synthesized α-MoO_3 microspheres with $CrCl_3.6H_2O$ and ammonium heptamolybdate as precursors by the hydrothermal method. The type of precursor played a significant role in increasing the specific surface area and improving the electrochemical performance. To improve the movement of electrons to increase the specific capacitance, an annealing operation was performed, which caused the morphology to change from an unstable state (hexagonal) to a stable state (orthotropic), which caused an increase in the specific capacitance (from 62 F g^{-1} to 186 F g^{-1}) [102].

3.4. Vanadium oxide

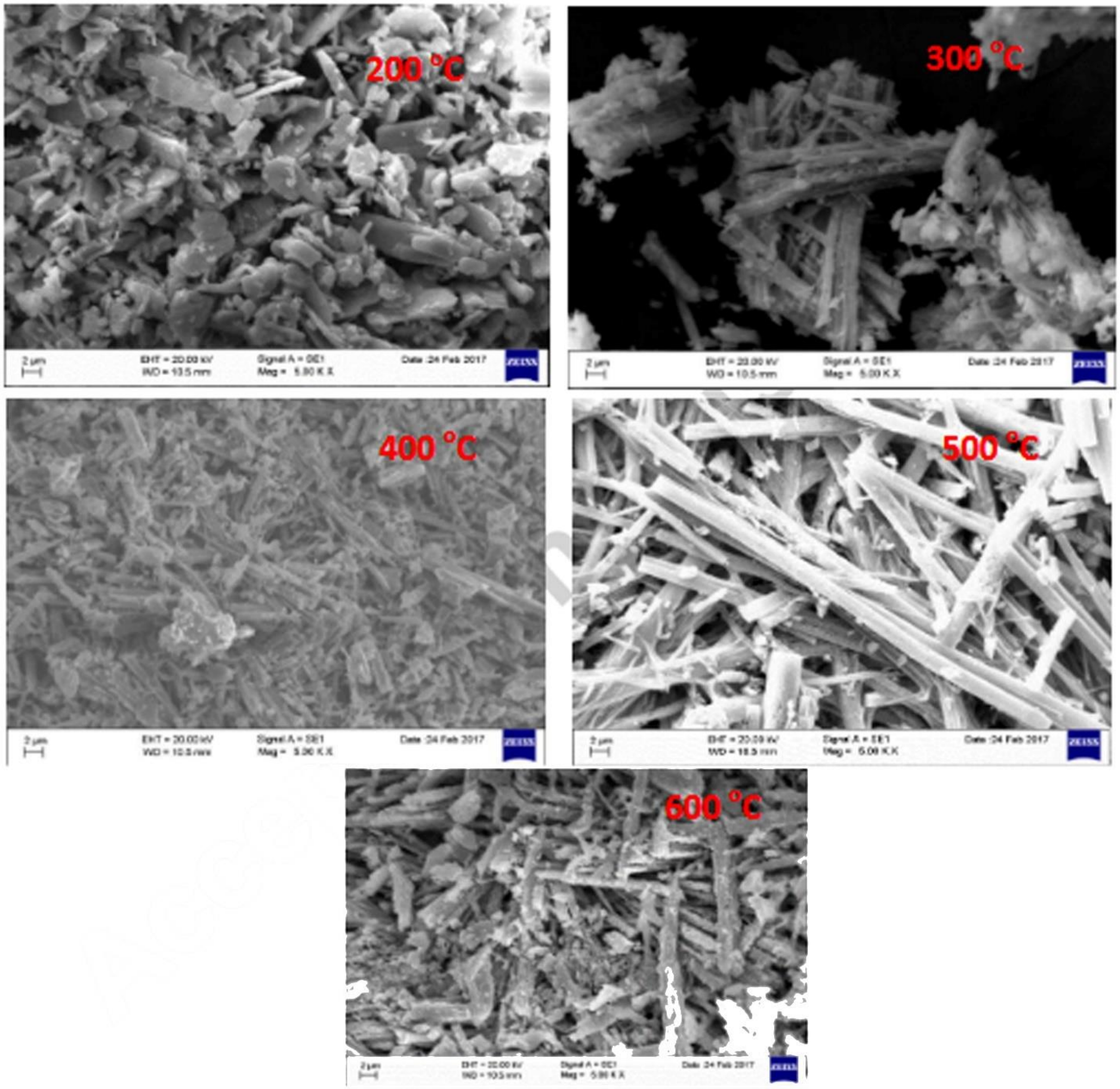
Vanadium oxide is a common electrode material in supercapacitor manufacturing. Vanadium oxide with a layered structure is widely used in organic electrolytes. Various methods such as hydrothermal, sol-gel, etc. were used for the design and synthesis of vanadium oxides. To improve the conductivity, in the study conducted by Wei, vanadium oxide was synthesized with a strip (nanoribbon) morphology with a small width, which increased the specific capacitance. Vanadium oxide with nanoribbon morphology has higher conductivity than vanadium oxide with nanotube morphology, which shows that vanadium oxide nanoribbons respond quickly to the applied potentials on the electrode. Therefore, the specific capacitance of vanadium oxide nanoribbon increased up to 453 F g^{-1} at 2 mV s^{-1} [103]. Therefore, the type of vanadium oxide morphology is important in improving ion diffusion speed and improving conductivity. In another study, Li et al synthesized vanadium oxide nanosheets using the hydrothermal method to obtain asymmetric supercapacitors with high energy density. In this study, using defect engineering, the oxygen content, which was emptied by 25%, was made possible. The space created by oxygen increases the rate of ion transfer.



Scheme 1. (a) Assembly of asymmetric supercapacitor where AC as cathode and TiO_2 nanocrystals calcined at 200 °C as anode (b) CV curves of asymmetric supercapacitor at 20 mVs^{-1} (c) CV curves of asymmetric supercapacitor measured at different potential windows with a scan rate of 20 mVs^{-1} (d) CV curves of asymmetric supercapacitor collected at different scan rates.

3.5. Nickel oxide

Micro/nanomaterials such as nickel oxide have attracted attention as the electrode material of electrochemical capacitors due to the shortening of diffusion paths and fast redox reactions. NiO is one of the other promising pseudocapacitor materials for supercapacitor electrodes, which have attracted attention due to properties such as high theoretical specific capacity, thermal stability, chemical stability, easy synthesis, low cost, abundance, and environmental friendliness. NiO materials can exhibit pseudocapacitor behavior. For example, NiO nanoparticles synthesized via the microwave method show high pseudocapacitor performance as an electrode material for supercapacitors. As well as, a high conductivity of 33.87 Scm^{-1} at room temperature and they show a surface area of 206 m^2g^{-1} [104]. The supercapacitor behavior of nickel oxide has been dependent on the morphology of this metal oxide. To obtain electrode materials based on nickel oxide, efforts have been made to synthesize optimal morphology by Es-Souni et al. The structure of nickel oxide nanotubes was investigated during annealing at 450 degrees Celsius for 25 minutes and 300 minutes, which showed the performance of nickel oxide nanotube electrode materials with a specific capacitance of 2093 F g^{-1} [105].



Scheme 2. SEM images of the molybdenum oxide prepared at various temperatures

3.6. Manganese oxide (MnO_2)

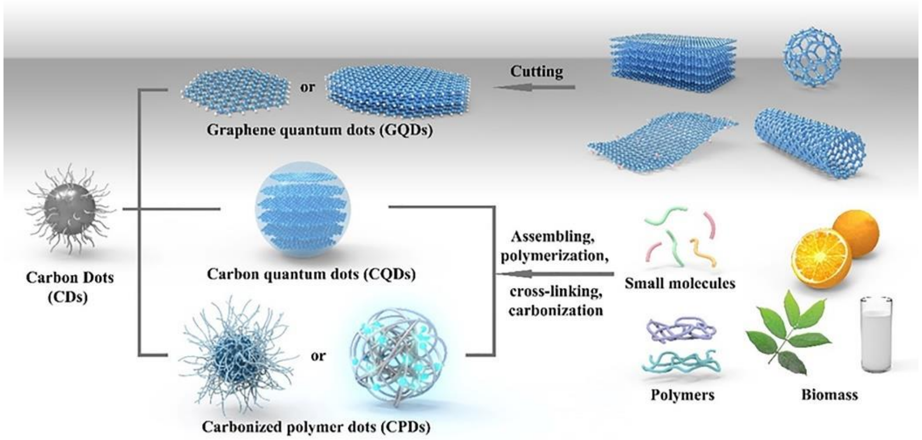
Manganese oxide, due to its superior properties such as supercapacitive performance in aqueous electrolytes, low cost, and low environmental effects, has led to the widespread efficiency of these materials in applications such as biosensors, energy storage and production devices, and catalysis. The lower cost and greater availability of MnO_2 compared to RuO_2-based materials, as well as other superior properties such as wide electrochemical potential window, low toxicity, and high theoretical specific capacitance (1370 $F \cdot g^{-1}$) have made these materials an alternative promising, for electrode material conversion. The MnO_2 charge storage mechanism is according to the following equation [106].

$MnO_2 + C^+ + e^- \rightarrow MnOOC$

The optimal performance of metal oxides as electrode materials requires specific temperature conditions to achieve optimal morphology to increase the specific surface area of metal oxide electrode materials, which limits the industrial performance of these materials. For this purpose, this limitation was overcome by the synthesis of nanocomposites based on metal oxide and quantum dots.

4. Classification of quantum dots

Quantum dots were synthesized in 2004 by Xu et al. through the electrophoresis process during the purification of carbon nanotubes. [107]. Quantum dots have attracted the attention of many researchers due to their unique properties that improve their performance in various applications. [74, 108-110]. Quantum dots have a high surface-to-volume ratio, meaning that a large percentage of the atoms are on the surface of the particles. The structure of quantum dots mainly consists of sp^2 graphite core (aromatic atoms) surrounded by sp3 carbon matrix, which has various functional groups such as C-O, C=O, C-O-C, O=C-OH, etc [111-113]. Quantum dots are divided into inorganic quantum dots and carbon-based quantum dots. Carbon-based quantum dots are divided into three categories: carbon quantum dots, graphene quantum dots, and polymer dots (Scheme 3) [114].



Scheme 3. Classification of quantum dots.

4-1- Inorganic quantum dots

The zinc sulfide (ZnS), cadmium sulfide (CdS), cadmium selenide (CdSe), zinc selenide (ZnSe), and several other nanomaterials are inorganic quantum dots [115-118]. The change in particle size of inorganic quantum dots leads to changes in absorption wavelength and emission wavelength. [119, 120]. Syntheses of inorganic quantum dots from cadmium (Cd), lead (Pb), and mercury (Hg) have disadvantages such as high toxicity. In other words, heavy metals are very toxic even in low concentrations, and production on a large scale is impossible [121, 122].

4-2- Carbon-based quantum dots

The structure of carbon-based quantum dots consists of sp^2/sp^3 carbon groups and is based on oxygen/nitrogen or polymer aggregates. Carbon-based quantum dots are divided into three main categories: carbon quantum dots (CQD), graphene quantum dots (GQD), and polymer dots (PDs) [123-126].

4-2-1- CQD

CQD are spherical and divided into with a crystal lattice and without a crystal lattice. Therefore, photoluminescence properties are different for types of carbon quantum dots. Carbon quantum dots have attracted the attention of researchers due to their unique properties, which include excellent optical properties, photoluminescence (PL), easy surface functionalization, etc. [127, 128]. Carbon quantum dots were investigated in diverse applications such as energy storage systems, medicine, chemistry, food, and the environment due to the ease of manufacturing through the green synthesis method and the availability of raw materials [129, 130]. The modification of carbon quantum dots allows the detection and binding of different analytes on the surface of carbon quantum dots through electrostatic interactions. Carbon quantum dots were synthesized using bottom-up and top-down techniques by carbon-based materials [131, 132]. The top-down synthesis method includes techniques of laser ablation, plasma reactor, ultrasound, exfoliation, and combustion. The bottom-up method includes microwave techniques, hydrothermal, and thermal pyrolysis [133-135].

4-2-2. GQD

Graphene quantum dots are obtained from several graphene sheets cum a size of 1 to 10 nanometers, which distinguishes them from graphene [136-138]. The mechanism of the edge location effect and atomic doping effect (especially nitrogen) for graphene quantum dots is similar to the structure of graphene. Some may think of GQD as a giant polyaromatic molecule, but GQD is distinct from polycyclic aromatic molecules (e.g., pyrene) [139-141]. Graphene quantum dots were enriched by various surface functional groups. The synthesis method of graphene quantum dots is based on the cut of graphitic carbon materials (Top-Down) or the integration of precursor molecules (Bottom-Up) [142, 143]. The top-down method produces monodisperse GQDs with defined structure and molecular size. However, these methods may be difficult, tedious, expensive, and with low production efficiency [144, 145]. The GQDs are prone to π-π stacking due to the absence of functional groups. Therefore, the synthesis of GQDs by bottom-up methods is more suitable. In top-down methods, cheap bulk carbon materials were used as precursors, but low production yields and longer reaction times [146, 147]. Graphene quantum dots were used in catalysts, photocatalysts, biomedicine, and energy storage devices such as supercapacitors [148-150].

4-2-3. PD

Polymer dots are nanomaterials based on zero-dimensional polymer and have the properties of polymeric materials and the fluorescence properties of quantum dots simultaneously. Polymer dots (PDs) were synthesized using conjugated polymers and non-conjugated polymers, which conjugated polymers are more suitable for forming polymer dots due to their π-conjugate structure and excellent photoelectric properties [151-153]. The polythiophene, polyaniline, polypyrrole, and their derivatives were used as conjugated polymers to synthesize polymer dots [154, 155]. Polymer dots (PDs) have several beneficial properties, including water solubility, processability, solvent miscibility, and advanced semiconductor quiddity. The bottom-up is a suitable method for the polymer dots synthesis, which includes mini-emulsion, nano-deposition, and hydrothermal. Polymer dots can be chemically modified and doped to achieve functional properties [156-158]. To improve the properties of polymer dots, they are composited with quantum dots and inorganic nanoparticles, etc. Since polymer dots as a conductive component in nanocomposites have excellent performance in energy storage and transfer systems including supercapacitors. Therefore, their electrochemical properties can be improved through composites polymer dot with metals or metal oxides [159-161].

5. The performance of nanocomposites based on metal oxide and quantum dots as electroactive materials of supercapacitors.

5-1. Nanocomposite of CQD /TMO in supercapacitors

The performance of CQD/TMO-based nanocomposites as electrode materials in supercapacitors has particular advantages, including more space and path for ion transport. Due to the small diameter of carbon quantum dots with a high specific surface, the functional groups of the surface of the quantum dots increase, and the contact between the electrode and electrolyte interface increases.
Finally, the uniform distribution of carbon quantum dots in nanocomposites stabilizes the cyclic performance.
Elumalai et al. synthesized CQD/Bi_2O_3 nanocomposite at 170°C using the hydrothermal method. According to the Scheme 4a and b, the carbon quantum dot used in the nanocomposite structure has caused the formation of an interwoven carbon network throughout the Bi_2O_3 matrix. In other words, the intergranular diffusion path of Bi_2O_3 is filled by conductive carbon quantum dots, which accelerates the speed of ion transfer and reduces surface resistance. The specific capacitance of CQD/Bi_2O_3 nanocomposite electrode to be about 343 F g^{-1} at 0.5A g^{-1}. The asymmetric CQD/Bi_2O_3 nanocomposite device was designed using rGO (negative electrode) and CQD/Bi_2O_3 nanocomposite (positive electrode). According to Scheme 4c, the cyclic voltammetry spectrum of the asymmetric device shows a quasi-oblong shape with redox peaks. By increasing the scanning speed, the current of the anodic and cathodic peaks of the cyclic voltammetry of the nanocomposite increases, which confirms the appropriate supercapacitor behavior. The Ragon diagram of the asymmetric device showed the maximum current density of 88 Wh kg^{-1} in the power density of 2793 W kg^{-1} in Scheme 4d. The asymmetric device showed a maximum

power density of 8400 W kg^{-1}, which is higher than the power densities reported for bismuth oxide.

Yue Wu et al. synthesized $CuCo_2O_4$ /CQDs and Fe_2O_3/CQDs nanocomposites with hierarchical porous structures by hydrothermal method. The results of the electrochemical analyses of $CuCo_2O_4$/CQDs and $CuCo_2O_4$ nanocomposite show an increase in specific capacity and a decrease in the resistance of $CuCo_2O_4$/CQDs nanocomposite compared to $CuCo_2O_4$ due to the improvement of electrical conductivity and charge transfer. The specific capacitance of $CuCo_2O_4$/CQDs electrode material was calculated 298.7 F g^{-1} at 1A g^{-1}. The redox peaks were observed in the $CuCo_2O_4$/CQDs nanocomposite CV, which confirmed the behavior of the battery type.

$CuCo_2O_4+H_2O+e^-\leftrightarrow 2CoOOH+CuOH$ (2)

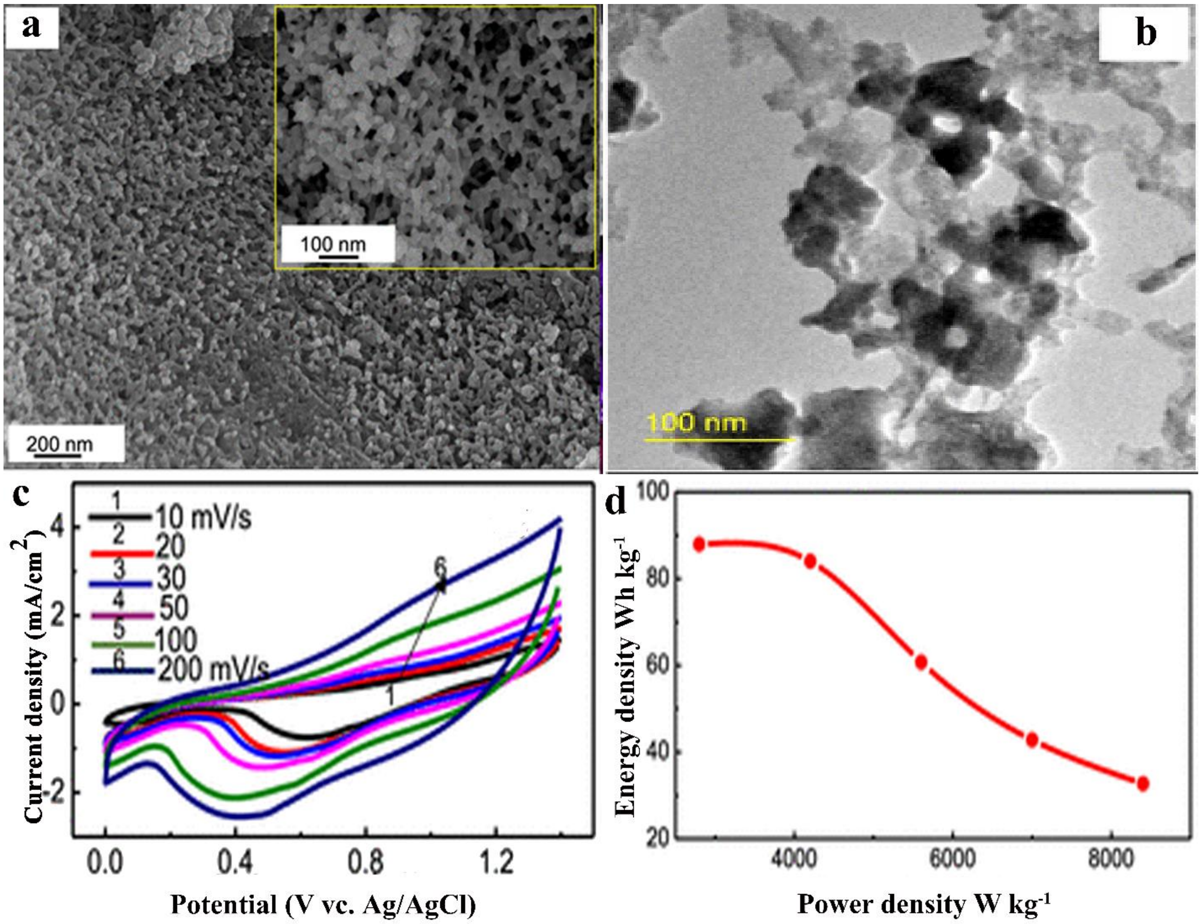
$CoOOH+OH^-\leftrightarrow CoO_2+H_2O+e^-$ (3)

$CuOH+OH^-\leftrightarrow Cu(OH)_2+e^-$ (4)

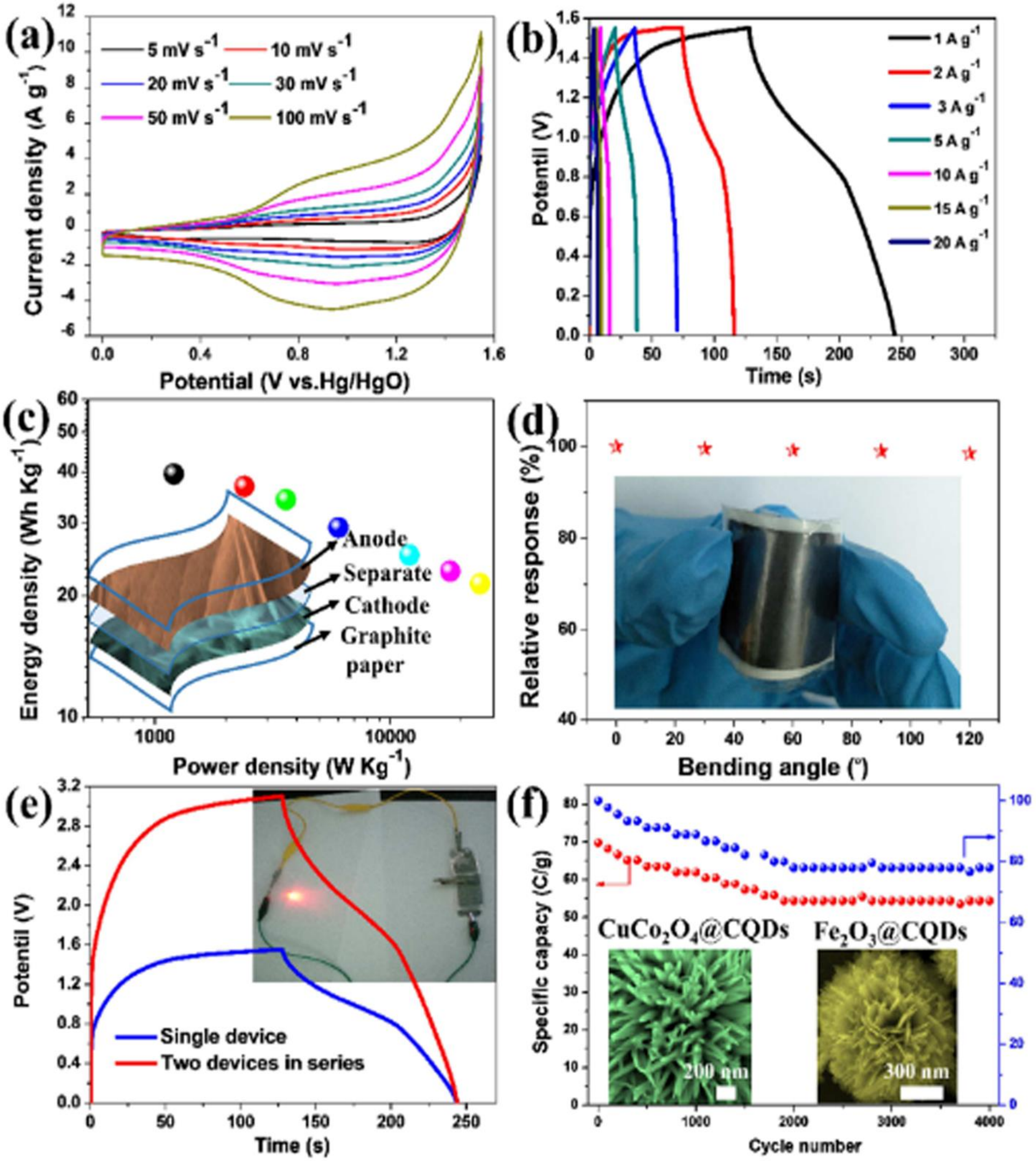
Porous Fe_2O_3/CQDs nanocomposite was synthesized by the same method, and the electrochemical results confirmed the improved performance of Fe_2O_3/CQDs nanocomposite electrode materials compared to Fe_2O_3 electrode materials.

The specific capacitance of Fe_2O_3/CQDs electrode material was calculated to be about 298.7 Fg^{-1}at 1 A g^{-1}. The device (Fe_2O_3/CQDs//$CuCo_2O_4$/CQDs) was made by nickel current collector and $CuCo_2O_4$/CQDs nanocomposite and Fe_2O_3/CQDs nanocomposite as positive and negative electrodes, respectively. The Fe_2O_3/CQDs//$CuCo_2O_4$/CQDs device showed energy density of 96.3 Wh kg^{-1} in 1201.9 W kg^{-1}.

Next, to investigate the performance of these nanocomposites in flexible energy storage devices, an asymmetric device was made using PVA/KOH electrolyte, $CuCo_2O_4$/CQDs nanocomposite, and Fe_2O_3/CQDs nanocomposite on graphite sheet according to Scheme 5c. The CV plot with different scan rates in the potential window of 0–1.55 V is shown in Scheme 5a. The results confirm the pseudocapacitance behavior of the asymmetric device. Scheme 5e shows the LED lamp lit by the flexible asymmetric device, and Scheme 5f confirms the stability of 84.6% of Fe_2O_3/CQDs//$CuCo_2O_4$/CQDs device after 5000 cycles [162].



Scheme 4. (a) and (b) The FE-SEM and TEM analyses of the CQD/Bi_2O_3. (c) yclic voltammograms of the asymmetric device at different scan rates. d) Ragone plot for the CQD−Bi_2O_3 ‖ KOH ‖ rGO asymmetric device.



Scheme 5. (a) CV curves of the FASC measured at different scan rates. (b) GCD curves of the as-fabricated FASC measured at different current densities. c) Ragone plot of the FASC, the inset is the schematic cartoon of the assembled FASC adopted by the $CuCo_2O_4$/CQDs electorde and Fe_2O_3/CQDs electrode. (d) Specific capacitance retention ratio of the FASC device after bending inward to the different angles. (e) GCD curves measured at 1 A g^{-1} for one FASC and two FASCs in series, the inset is the photograph of a red LED powered by two devices in series. (f) Cyclingstability of the FASC over 5000 cycles at 20 A g^{-1}, the inset is the SEM images of the $CuCo_2O_4$/CQDs and Fe_2O_3/CQDs after cycling.

Elumalai et al. synthesized CQD from sour milk as a precursor using the hydrothermal method. Then, the CQD-MnO_2 nanocomposite was prepared by adding potassium permanganate. The CQD-MnO_2 nanocomposite was a suitable supercapacitor behavior due to the high specific surface area of CQD. The supercapacitor behavior of CQD-MnO_2 nanocomposite was investigated in a three-electrode system using 1 M Na_2SO_4 electrolyte. The CQD-MnO_2 electrode recorded the 189F g^{-1} specific capacitance at 0.14 A g^{-1}. The symmetric CQD-MnO_2 nanocomposite device was prepared, which recorded the energy density of 40 Wh kg^{-1} at 3385 W kg^{-1} (power density) [163].

Paoprasert et al. investigated the supercapacitor performance of nanocomposite derived from Titanium dioxide nanoparticles (TiO_2) and carbon quantum dots. The CQD/Tio_2 electrode-specific capacitance was higher than the TiO2 electrode material due to the synergistic effect and increased surface wettability. Carbon quantum dots were considered an enhancing factor due to the proper performance of the supercapacitor [164].

Selvaraj et al. synthesized nanocomposite based on cobalt oxide and carbon quantum dot (Co_3O_4/CQD) by hydrothermal method to investigate its performance in supercapacitor system. The results showed that adding CQD to the cobalt oxide matrix increased the charge transfer process.

The Co_3O_4/CQD nanocomposite electrode-specific capacitance was calculated 193.8 F $g\text{-}^{1}$ at 4 A g^{-1}, which increased compared to the specific capacitance of Co_3O_4 electrode material. Then, an asymmetric device was made with graphene oxide (negative electrode) and Co_3O_4/CQD nanocomposite. The asymmetric Co_3O_4/CQD recorded an energy density of 2.49 Wh kg^{-1} at 426 W kg^{-1} [165].

5.2. Nanocomposite of GQD/TMO in supercapacitors

Graphene quantum dots have better supercapacitor performance than other quantum dots. Many reports have been presented for graphene quantum dots as a conductive component in TMO/GQD nanocomposites. Doong et al. synthesized two-dimensional vanadium oxide nanosheets, and then by the solvothermal method, the graphene quantum dot was placed between the two-dimensional V_2O_5 nanosheets. According to the morphology analysis and the histogram prepared, the particle size distribution of graphene quantum dots was reported to be uniform, which increases the energy storage. To investigate the electrochemical behavior of VNS-GQD nanocomposite, this nanocomposite was synthesized with different weight ratios of graphene quantum dots. The electrochemical behavior was investigated in the three-electrode system in the H_2SO_4 electrolyte. The charge transfer resistance of V_2O_5 is higher than VNS, and among the nanocomposites, VNS-GQD-10 has the lowest charge transfer resistance and suitable supercapacitor behavior. The asymmetric device using VNS-GQD-10 (positive electrode) and MCS (negative electrode) showed a maximum energy density of 20.62 Wh kg^{-1} [166].

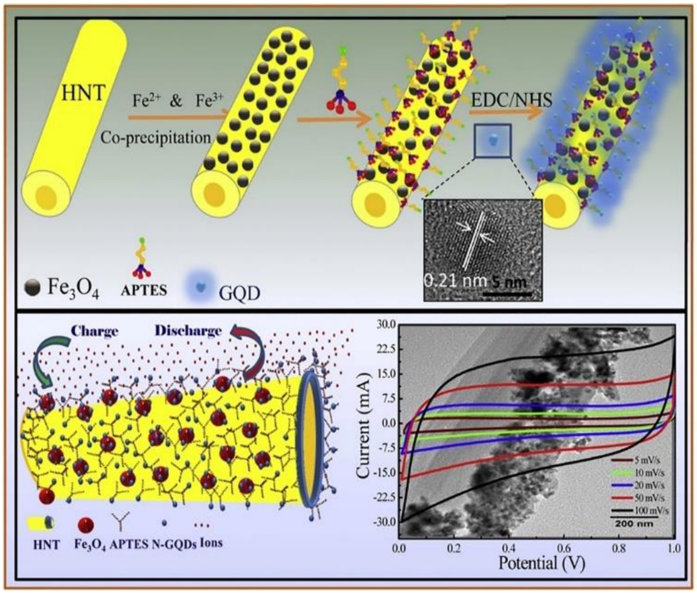
Zaijun et al. investigated the electrochemical behavior of nanocomposite(AD-RuO_2-Trp-GQD-G) derived from tryptophan functionalized GQD/ atomic diffuse of ruthenium oxide (RuO_2) / graphene. The results of investigating the performance of three AD-RuO_2-Trp-GQD-G nanocomposite electrodes in 1 M sulfuric acid electrolyte solution confirmed that the ion transfer process is fast due to the 3-dimensional structure of the AD-RuO_2-Trp-GQD-G nanocomposite. The AD-RuO_2-Trp-GQD-G nanocomposite electrode showed 503 F g^{-1} specific capacitance at 1 A g^{-1}. Then, the flexible AD-RuO_2-Trp-GQD-G nanocomposite supercapacitor was made using a 3D printer with PVA/H_2SO_4 solid electrolyte. The investigation of electrochemical analyses of the flexible supercapacitor confirmed good reversibility due to the oxide reduction of the active groups of Trp-GQD and oxide and reduction of Ru_2O on the electrode surface. The flexible supercapacitor reported a maximum energy density of 332 Wh kg^{-1} [167].

Semnani et al. synthesized GQD using citric acid as a precursor, and then it was composited with $CuMnO_2$ by hydrothermal method. The $CuMnO_2$/GQD nanocomposite-specific capacitance was reported to be twice that of $CuMnO_2$, which confirmed the synergistic effect of graphene quantum dots and $CuMnO_2$. According to the morphological identification analysis, it was confirmed that graphene oxide reduced the particle size of the nanocomposite. The particle size of nanocomposites was lower than $CuMnO_2$, which increased the speed of ion transfer and improved supercapacitor performance. An asymmetric nanocomposite cell was prepared with active carbon (negative electrode) and nanocomposite (positive electrode). The results of the electrochemical analyses confirmed the battery-type behavior of an asymmetric device, and the maximum specific capacitance of the symmetric device was calculated at 47.9 Wh kg^{-1} [168].

In some studies, metal oxides are prepared by annealing metal-organic frameworks. For example, Hong et al. prepared metal oxide CuO by annealing to the metal-organic framework, then composited it with graphene quantum dot by hydrothermal method. In other words, graphene quantum dots with a negative charge are absorbed on the RuO surface, which increases the surface area and improves the supercapacitor performance. According to the morphology of the nanocomposite, the speed of ion transfer and conductivity improves. The results of electrochemical analyses in the three-electrode system for the GQDs/CuO nanocomposite confirmed the specific capacitance of GQDs/CuO to be about 729F g^{-1} at 1 A g^{-1}. The asymmetric GQDs/CuO device was made with active carbon (negative electrode) and GQDs/CuO nanocomposite (positive electrode). The electrochemical analysis of the asymmetric GQDs/CuO device in the window of 1.5 V with 3 M KOH electrolyte confirmed the pesudocapacitive behavior and recorded the maximum energy density of 32.2 Wh kg^{-1} [169].

In other work, Zhi et al. the Co-Ni-BTC metal-organic framework with nickel nitrate, cobalt chloride, and 1, 3, 5-benzene tricarboxylic acid synthesized the using the microwave method. In the next step, a porous multilayer NiO/Co_3O_4 structure was synthesized by hydrothermal method at 500 °C. In the last step, NiO/Co_3O_4/GQDs composite was synthesized with graphene quantum dot and NiO/Co_3O_4 by hydrothermal method. The bimetallic oxide (NiO/Co_3O_4) has a battery-type behavior, which was improved by adding the graphene quantum dot and forming a nanocomposite. The NiO/Co_3O_4/GQDs nanocomposite electrode material showed 1361 F g^{-1} specific capacitance at 1 A g^{-1}. The asymmetric NiO/Co_3O_4/GQDs nanocomposite device (NiO/Co_3O_4/GQDs//AC) was designed and manufactured, which showed a maximum energy density of 38.44 W h kg^{-1} [170].

Kharangarh et al. synthesized Cobaltite and graphene quantum dot composite ($NiCo_2O_4$/GQDs) by hydrothermal method. Investigating the electrochemical behavior of this nanocomposite in a three-electrode system confirmed the increase in conductivity of $NiCo_2O_4$/GQDs nanocomposite compared to $NiCo_2O_4$ due to the C-O bond interaction of graphene quantum dot with $NiCo_2O_4$.The specific capacitance of the nanocomposite was higher than that of $NiCo_2O_4$, which was attributed to the edge effects of graphene quantum dots. The specific capacitance of the nanocomposite was reported to be 481.4 F g^{-1} at 0.35 m A g^{-1} [69].



Scheme 6. Schematic of the synthesis of $NGQD/Fe_3O_4$-HNTs.

Doong and his colleagues synthesized N-doped GQD. In the second step, Fe_3O_4 nanoparticles were synthesized by deposition on HNTs. Then Fe_3O_4 -HNTs were coated with (3-aminopropyl)-triexthoxysilane to create a site for connecting N-GQDs through an amide bond (Scheme 6). The N-GQD-Fe_3O_4-HNTs electrode material showed a specific capacitance of 418 F g^{-1} at 1A g^{-1}. The favorable electrochemical results can be attributed to the synergistic effect of the constituent components. The one-dimensional Fe_3O_4-HNTs matrix shortens the path of electrons and electrolyte ions, and N-GQDs create accessible electroactive sites for electrons [171].

5-3-Nanocomposite of PDs /TMO in supercapacitors

In the study, common electrode materials of quantum dots were discussed. Polymer dots are a new class of carbon-based quantum dots that have been investigated in recent years. Polymer dots were studied in biological imaging, drug delivery, and biosensing due to their unique fluorescence properties (dual fluorescence). However, there are limited studies of polymer dots in energy storage systems such as supercapacitors. Arslan et al. synthesized the nanocomposite derived from cobalt oxide and polymer dots (PD_{JA}-Co_3O_4) by the solvothermal method. The results of electrochemical analyses confirmed good supercapacitor performance of nanocomposite due to the synergistic effect of cobalt oxide with polymer dot. Also, functional groups on the surface of polymer dots (COOH, OH, NH_2) with free chains of polymer led to good supercapacitor properties.

Table 1. Recent investigations of GQD/TMO and CQD/TMO in supercapacitor application.

Materials	Electrolyte	Specific capacitance	Reference
GQDs/MnO_2	Utilizing ionic liquid (IL)	208.2 F g^{-1}	[172]
NGQD/ WO_3 NFs	0.5M H_3PO4	198.15 F g^{-1}	[173]
GQDs /$NiCo_2O_4$	KOH	481.4 F g^{-1}	[174]
GQDs/MnO_2	Na_2SO_4	246 F g^{-1}	[162]
CQDs/$NiCo_2O_4$	2M KOH	856 F g^{-1}	[175]
CQDs/RuO	1 M H_2SO_4	594 F g^{-1}	[176]
GQDs//MnO_2	0.5 M Na_2SO_4	1107.4 µF cm^{-2}	[149]

Table 2. Recent investigations of QD/TMO and TMO in supercapacitor application.

QD/TMO composite	Specific capacitance	reference	TMO	Specific capacitance	reference
Co_3O_4/graphene quantum dots	2435 F g^{-1} at 1 A g^{-1}	[177]	Co_3O_4 nanowire	57.4 at 1 A g^{-1}	[178]
$NiCo_2O_4$/graphene quantum dots	1242 F g^{-1} at 30 A g^{-1}	[174]	$NiCo_2O_4$	49.3 at 1 A g^{-1}	[179]
$NiCo_2O_4$/graphene quantum dots	3940 Fg^{-1} at 0.5 Ag^{-1}	[180]	$NiCo_2O_4$	60 at 1 A g^{-1}	[181]
N-doped graphene quantum dots anchored Fe_3O_4/halloysite nanotubes	418 F g^{-1} at 1 A g^{-1}	[171]	TiO_2	23.24 mF cm^{-2} at 2 mV s^{-1}	[182]
V_2O_5 / graphene quantum dots	572 F g^{-1} at 1 A g^{-1}	[166]	V_2O_5	141.8 F g^{-1}at 1 A g^{-1}	[183]

According to the morphology identification analysis, the nanocomposite derived from polymer dots and cobalt oxide (PD_{JA}-Co_3O_4) increased the rate of ion transport due to its intertwined morphology. The specific capacitance of the symmetrical nanocomposite device was calculated to be about 92 F g^{-1}at 1 A g^{-1}.

6. Conclusion

Supercapacitor energy storage devices are a promising technology to overcome the limitations of using fossil fuels and the environmental pollution caused by the use of fossil fuels. The carbon materials, conductive polymers, and metal oxides have been used to make supercapacitor devices. Metal oxides alone do not have high performance in supercapacitor systems because achieving optimal supercapacitor behavior for metal oxides depends on the morphology and temperature conditions that limit the performance of this class of materials in the industry. To overcome this limitation, composites based on metal oxide and quantum dots have been studied and investigated in recent years. Carbon-based quantum dots are a new class of electrode materials and have been investigated in three main categories: CQD, GQD, and PD in supercapacitor systems. Quantum dots alone have poor conductivity and a small specific surface, so as a conductive component in composites, they lead to improved final performance in supercapacitor devices. The existence of quantum dots as a conductive component in nanocomposites based on metal oxide and quantum dots leads to improved electrical conductivity. The performance of the carbon-based quantum dots (CQD, GQD, and PD) to improve the final behavior of the nanocomposite based on metal oxide and quantum dots confirms the synergistic effect. Due to the very favorable performance of polymer dots, including the polymer properties and quantum dots properties, simultaneously, there are limited reports for polymer dots in supercapacitor systems. According to the results, the easy synthesis method appropriate performance of nanocomposite based on metal oxide and polymer dot allows their industrial production. Therefore, more research on these materials as electroactive materials in supercapacitors is suggested for researchers.

Conflicts of interest

The authors have no conflict of interest.

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