



## Review article

# Applications of carbon quantum dots in electrochemical energy storage devices

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## ABSTRACT

Supercapacitors (SCs), including electric double-layer capacitors (EDLCs), pseudocapacitors, and hybrid capacitors, are esteemed for their high power density and attractive features such as robust safety, fast charging, low maintenance, and prolonged cycling lifespan, sparking significant interest. Carbon quantum dots (CQDs) are fluorescent nanomaterials with small size, broad excitation spectrum, stable fluorescence, and adjustable emission wavelengths. They are widely used in optoelectronics, medical diagnostics, and energy storage due to their biocompatibility, low toxicity, rich surface functional groups, abundant electron-hole pairs, large specific surface area, and tunable heteroatom doping. In this short review, we briefly discussed the advantages and disadvantages of bottom-up and top-down of CQD synthesis methods. The arc-discharge technique, laser ablation technique, plasma treatment, ultrasound synthesis technique, electrochemical technique, chemical exfoliation, and combustion are among the initial top-down approaches. The subsequent section delineates waste-derived and bottom-up methods, encompassing microwave synthesis, hydrothermal synthesis, thermal pyrolysis, and the metal-organic framework template-assisted technique. In addition, this short review focuses on the operational mechanism of supercapacitors, their properties, and the utilization of CQDs in supercapacitors.

## 1. Introduction

The increasing need for energy storage and growing environmental consciousness have emphasized the significance of sustainable energy sources globally. Supercapacitors have garnered significant interest within the scientific community as a promising energy storage device, owing to their impressive attributes such as cost-effectiveness, elevated power density, outstanding cycling stability,

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and prolonged cycle life [1–3]. Nevertheless, the extensive utilization of supercapacitor devices has faced challenges due to their significantly low energy density ( $E$ ). Numerous research initiatives are currently underway to develop supercapacitors with higher energy density. Expanding the voltage window ( $V$ ) or specific capacitance ( $C_s$ ) offers a means to enhance the energy density of supercapacitors, as demonstrated by the  $E$  equation,  $E = 1/2 CV^2$ . However, the electrochemical characteristics of supercapacitors are markedly shaped by the type and composition of the electrode materials [4].

Supercapacitors are categorized into three groups based on their energy storage mechanisms: electric double-layer capacitors (EDLCs), pseudocapacitors, and hybrid capacitors [5,6]. The EDLC acquires its capacitance exclusively through the electrostatic charge accumulation among ions at the electrode [7]. In terms of longevity and stability, this system remains robust as it allows for unlimited charge and discharge over time. Contrastingly, a pseudocapacitor relies on reversible faradaic redox reactions occurring at the interface between the electrode and electrolyte. Similar to the EDLC, this system also allows unlimited charge and discharge over time. However, pseudocapacitor has higher energy density than EDLC due to their compatible electrode/electrolyte and restricted specific surface area [8]. A distinct variety demonstrates an intermediate supercapacitor performance by integrating both EDLC electrode and pseudocapacitive electrode into a unified device. This configuration is commonly known as a hybrid supercapacitor. Considering the energy storage mechanisms outlined earlier, an extensive array of approaches has been explored to evaluate a diverse range of materials suitable for supercapacitor (SC) components. These approaches encompass the exploration of emerging materials, the design of materials considering the energy mismatch between electrolytes and electrodes, and the nanostructuring and functionalization of established electrode materials [9].

In 2004, Xu and others discovered carbon quantum dots (CQDs) while attempting to synthesize single-walled carbon nanotubes (SWCNTs), also referred to as carbon nanoparticles (CNPs) [10]. CQDs exhibit a high concentration of C–C bonds in graphene-type  $sp^2$  hybridization and diamond-type  $sp^3$  hybridization, leading to structural disorder. They are also decorated with abundant functional groups such as aldehyde, hydroxyl, and carboxyl [11]. These particles exhibit a quasi-spherical shape and are typically smaller than 10 nm. Carbon quantum dots exhibit exceptional compactness compared to other carbon materials, leading to uniform dispersion, improved electron transfer/reservoir capabilities, and a unique structure. Since this serendipitous finding, researchers have been captivated by the remarkable optoelectronic properties, photoluminescence (PL), straightforward synthesis methods, surface functionalization potential, excellent biocompatibility, expansive specific surface area, and minimal toxicity exhibited by CQDs [4,12]. Furthermore, in the context of next-generation energy storage applications, CQDs play a promising role as electrodes. The pivotal roles of CQDs in electrodes have been delineated, as depicted in Fig. 1. These encompass enhancing the columbic efficiency of lithium (Li), sodium (Na), and potassium (K) ion batteries; prolonging their cycling durability, providing ample surficial functional configurations for supercapacitors and faradaic reactions, improving interfacial wettability, and fostering the oxygen reduction reaction and oxygen evolution reaction (ORR/OER) in metal–air batteries. As a result, these unique characteristics have garnered significant interest and attention from the scientific community [13–17].

In broad terms, Carbon Dot (CD) synthesis techniques can be categorized into two main types: the “bottom-up” organic approach and the “top-down” nano-cutting technique. The top-down nano-cutting methods typically involve the cutting or modification of various carbon sources such as graphene oxide, carbon fibre (CF), carbon nanotubes, and graphite. Carbonization of carbohydrates, the self-assembly of polycyclic aromatic hydrocarbons, and the organic synthesis of small molecules represent examples of “bottom-up” organic processes [18,19]. Fig. 2 summarizes various synthesis processes for Carbon Dots (CDs) [20], offering a comprehensive overview along with concise descriptions of the chemical and physical properties of CDs, encompassing the diverse types of CDs.

Numerous studies have explored the fluorescence, synthesis, and diverse applications of CQDs in areas such as photocatalysis, sensing, bioimaging, biomedicine, and optoelectronics (including LEDs, photodetectors, and solar cells). However, there is a limited number of studies that specifically investigate the impact of 0-D carbon materials on the electrochemical characteristics of composites containing CQDs. Beyond their captivating and distinctive characteristics, this review offers an overview of diverse synthesis techniques, the adsorption properties of CQDs, their application in energy conversion, and insights into their role in energy systems. The investigated characteristics of CQDs and their controllability have been demonstrated to be intriguing for various research applications.

This review provides an overview of the characteristics, synthesis techniques, and energy-related applications of CQDs, with a

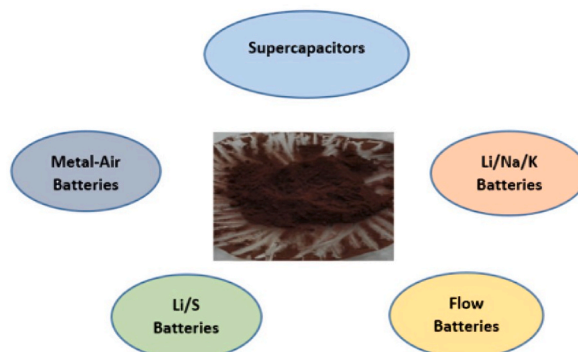


Fig. 1. Different energy storage systems utilize carbon dots (CDs) as electrode materials.

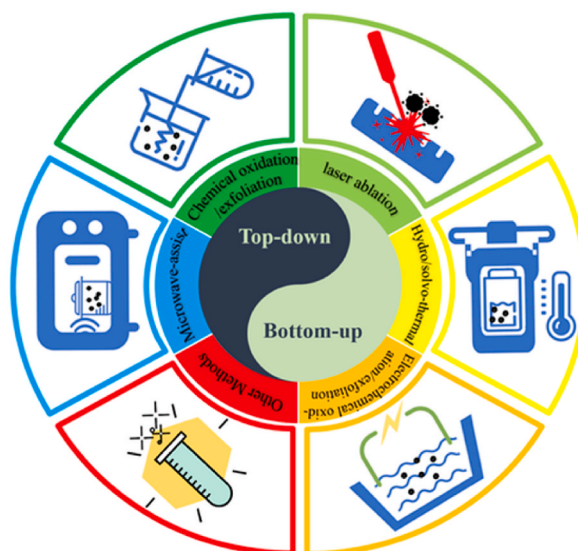


Fig. 2. Illustration depicting various synthesis pathways of carbon dots. Reproduced from Ref. [21] under Creative Commons license.

specific focus on their use in EDLC supercapacitors.

## 2. Working mechanisms of supercapacitors

### 2.1. Electric double layer capacitors (EDLCs)

The EDLC is developed by coating two carbonaceous materials with high porosity, such as graphite sheets, nickel foam, aluminium or titanium foil, metal-organic framework, onto a metal current collector. A porous electrolyte separator is placed between the 2 electrodes (Fig. 3). Ion adsorption forms the foundation for the charge storage process in this coating type due to the non-metallic and inert nature of the coated material. When a voltage is applied, positive charges align at one electrode terminal, and negative charges align at the other electrode terminal. Due to electrostatic attraction, the accumulation of charges on the electrodes facilitates the movement of oppositely charged electrolyte ions towards them. Afterward, ion adsorption occurs at the electrode's surface, resulting in the creation of a double layer at the boundary between the electrode and electrolyte [22]. Materials with high porosity, such as activated carbon (AC) [23,24], carbon nanotubes (CNTs) [25,26], graphene [27,28], carbon aerogels [29,30], carbon foam [31,32], and carbon derived from carbide [33,34], have been utilized to form the electric double layer in supercapacitors. The carbonaceous material used in the EDLC exhibits exceptional stability, enduring an impressive 28,900 charge-discharge cycles [35] and exhibiting low capacity retention (maintaining over 90 % even after 3000 cycles) [36,37]. Additionally, EDLC offers exceptional power density and specific capacitance. Nevertheless, its energy density is limited by non-faradaic reactions inherent in the charge storage mechanism.

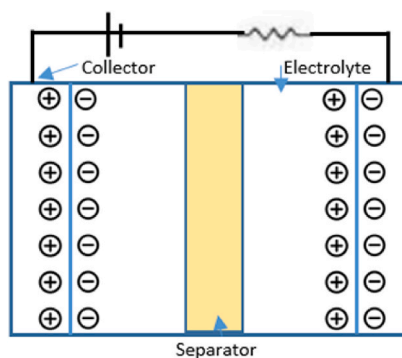


Fig. 3. The diagram illustrates an EDLC type supercapacitor.

## 2.2. Pseudocapacitor

Pseudocapacitors employ a faradaic mechanism for charge storage, distinguishing them from EDLCs by necessitating the electrostatic transfer of charge between the electrolyte and electrode. When a voltage is applied, oxidation takes place on the electrode material of a pseudocapacitor, leading to reduction. Through this process, a charge can traverse the double layer, facilitating the flow of faradaic current within the supercapacitor cell. Pseudocapacitors, generated through this faradaic technique, exhibit superior energy densities compared to EDLCs [38]. In pseudocapacitive materials, ions with a charge migrate towards terminals with the opposite charge when a voltage is applied, accumulating electrons and holes in their respective terminals (Fig. 4).

The cyclic voltammogram (CV) and galvanostatic charge-discharge (GCD) profiles of pseudocapacitors bear resemblance to those of specific batteries employing nanoscale electrode materials. This similarity arises due to their shared operation via redox reactions [39,40]. In contrast to EDLC, pseudocapacitors exhibit lower cycle stability and reduced power density.

## 2.3. Hybrid capacitor

A hybrid supercapacitor setup incorporates both a battery-type electrode for storing energy and a capacitor-type electrode for delivering power, all within a single cell. By selecting the appropriate electrode combination, it becomes feasible to elevate the cell voltage, thereby enhancing both energy and power densities [41,42]. Effective energy storage can be achieved by employing both battery-type and capacitor-type electrodes, utilizing both faradaic and non-faradaic techniques. In comparison to EDLCs, these systems demonstrate superior cycle stability and are more cost-effective [39,43–45].

While pseudocapacitors may not match the power density and cyclic stability of EDLCs, they do address certain drawbacks inherent to EDLCs. To overcome these challenges, scientists have devised a hybrid supercapacitor that integrates both an EDLC and a pseudocapacitor (Fig. 5). Hybrid supercapacitors can be categorized into a) symmetric, b) battery-type, or c) composite hybrid based on the combination of electrode materials employed. While the electrode arrangement is similar for battery types and asymmetric hybrids, the materials used for the electrodes differ in these types. This category exhibits superior power and energy density as it combines the material characteristics of a battery for enhanced energy density and a supercapacitor for heightened power density. Conversely, composite hybrid electrodes operate in a comparable manner and consist of a combination of carbonaceous material and pseudocapacitive substances like metal oxide or conducting polymers. This enhancement improves specific capacitance, corrosion resistance, and the potential window of the supercapacitor. Additionally, the incorporation of carbon enhances the structural stability of conducting polymers [46].

It is also valuable to investigate the hybrid supercapacitor's reduced self-discharge when compared to conventional supercapacitors. This is particularly relevant because self-discharge in supercapacitors often results from electrode materials undergoing a straightforward absorption mechanism. In such cases, self-discharge occurs as ions from the electrode material diffuse back to the electrolyte, driven by a disparity in the concentration gradient [47].

## 3. Carbon quantum dots

A recently developed group of carbon nanomaterials, characterized by diameters less than 10 nm, is referred to as carbon-based quantum dots. This family encompasses graphene quantum dots (GQDs) and carbon quantum dots (CQDs), also known as C-dots or CDs. The initial discovery took place in 2004 during the purification of SWCNT using preparative electrophoresis [48]. Subsequently, in 2006, they were obtained through laser ablation of cement and graphite powder [13]. The captivating characteristics of carbon-based quantum dots have propelled them into the spotlight among the emerging nanocarbon family, given their affordability, abundance, and safety. In broad terms, carbon is a dark material exhibiting faint fluorescence and limited water solubility. However, carbon-based quantum dots, often referred to as carbon nanolights, have attracted significant interest because of their pronounced brightness and excellent solubility [49,50].

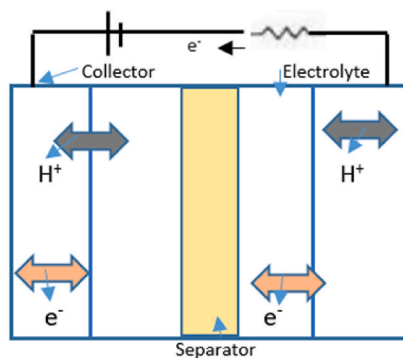


Fig. 4. The diagram illustrates pseudocapacitor supercapacitor.

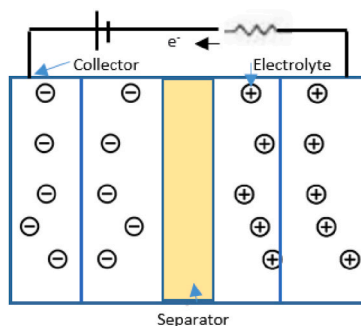


Fig. 5. The diagram illustrates a hybrid supercapacitor.

### 3.1. Properties of CQDs

#### 3.1.1. Absorbance

CQDs commonly exhibit optical absorption primarily in the ultraviolet range, which extends into the visible spectrum. The transitions involving  $sp^2$  carbon to  $\pi-\pi^*$  and the hybridization change of  $n-\pi^*$  with a heteroatom are identified as the primary optical absorption peaks of CQDs in the ultraviolet–visible region. Surface passivation or modification can affect the absorption properties [51, 52]. CQDs possess the ability to absorb a broad spectrum of photons. When incorporated into supercapacitor electrodes or electrolytes, the absorbed light energy triggers charge separation within the material. Furthermore, light absorption by CQDs in the electrolyte can induce changes in its pH, viscosity, and ion mobility, thereby optimizing the electrochemical performance of the electrolyte across different operational conditions. These alterations have the potential to enhance the overall performance of supercapacitors. Jiang et al. devised a simple hydrothermal approach to create red, green, and blue (RGB) luminescent CQDs by utilizing three isomers of phenylenediamines [53]. The UV–visible absorption spectra of the CQDs exhibited a consistent pattern as observed (refer to Fig. 6 (a–c)). The red-shifted absorption transitions in these three CQDs imply that their electronic band gaps are smaller than those of their predecessors. The study’s results demonstrated that, upon a single UV light excitation, the CQDs emit vibrant and stable RGB luminescence both in a polymer matrix and solution. Finally, the three generated CDs exhibit effective multicolor cellular imaging capability under single-wavelength stimulation, and they display minimal cytotoxicity, following a similar trend [53,54].

#### 3.1.2. Photoluminescence

The occurrence of photoluminescence (PL) in CQDs results from the absorption of photons, which are then converted into electromagnetic radiation. In the case of CQDs, the peak luminosity is typically distinctive and is contingent upon the wavelength and intensity of emission. This singular occurrence could be ascribed to the optical selection of nanoparticles or CQDs of different sizes, each possessing distinct emissive traps on their surface [55]. Changes in the intensity or wavelength of photoluminescence can provide valuable insights into the state of charge of the device, enabling real-time monitoring of its performance. Additionally, photoluminescence in CQDs can facilitate energy harvesting in supercapacitors. By converting excess light energy from sources into electrical energy through photoluminescence, CQDs enhance the charging process of the supercapacitor, thereby increasing its energy storage efficiency. The emission spectrum of PL, which is broad and dependent on excitation, mirrors the variability in both particle size and PL emission. A comprehensive analysis of the structural characteristics of the underlying materials and the spectroscopic features of the emissions unveiled that the majority of the observed PL emissions could be classified into two distinct categories. The first category is associated with conjugated  $\pi$ -domains and band-gap transitions, while the second one arises from more complex factors loosely linked to structural anomalies in graphene. Often, these two characteristics may be intertwined, as the formation or

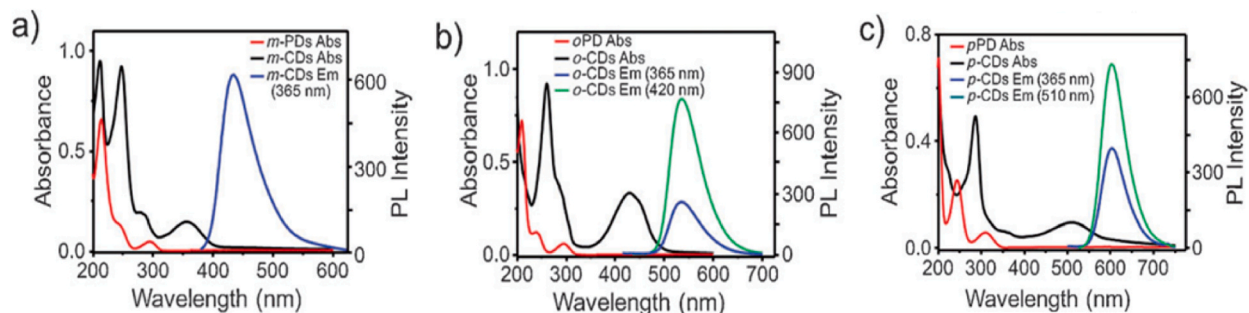


Fig. 6. (a–c) The emission spectra of *m*-CDs, *o*-CDs, and *p*-CDs are represented by blue and green lines, while the UV/Vis absorption spectra of *m*PD, *o*PD, and *p*PD are depicted by the red line. Additionally, the emission spectra of *m*-CDs, *o*-CDs, and *p*-CDs are illustrated by the black line. Reproduced from Ref. [53] with permission from John Wiley and Sons.

induction of  $\pi$ -domains is frequently reliant on leveraging defects in graphene sheets [56].

### 3.1.3. Chemiluminescence

The characteristic of CQDs exhibiting luminescence interruption or excitation following an oxidation event is notable. The chemiluminescence (CL) features of CQDs were observed in the presence of specific oxidants [57]. Variations in the intensity or wavelength of chemiluminescence enable real-time monitoring of the device's performance, providing insights into the state of electrochemical reactions. Chemiluminescence in CQDs facilitates the conversion of chemical energy from electrochemical reactions into light energy, thereby enhancing the efficiency and energy storage capacity of the supercapacitor. The CL characteristics of CQDs were initially uncovered when potassium permanganate ( $\text{KMnO}_4$ ) and cerium (IV) were present alongside them. Electron paramagnetic resonance (EPR) analysis suggests that oxidants like  $\text{KMnO}_4$  and cerium (IV) can induce the introduction of holes into CQDs [58]. This mechanism enhances electron-hole annihilation, augmenting the quantity of holes within CQDs, thereby leading to CL emission and energy discharge. Moreover, the intensity of CL was influenced by the concentration of CQDs within a certain range. Elevating the temperature revealed that the optimal temperature for electron dispersion in CQDs positively influenced CL. An intriguing aspect of this system is that altering the surface groups of CQDs can induce CL characteristics [59].

### 3.1.4. Electroluminescence

Given the established capability of semiconductor nanomaterials to exhibit electroluminescence (EL), it is unsurprising that CQDs have attracted attention in EL research, with potential applications in electrochemical fields and energy storage devices [60]. Electroluminescence in CQDs enables the direct conversion of electrical energy into light energy within the supercapacitor, enhancing both its energy storage capacity and energy conversion efficiency. The emission color of LEDs based on CQDs can be modified by adjusting the driving current, allowing for color-switchable EL from the identical CQDs. This effect, spanning from blue to white emission, was noted at various operating voltages. To delve further into the luminescence mechanism of CQDs, scholars have put forward two theories: one revolving around band-gap emitters within a continuous p domain, and the other highlighting the edge effect arising from diverse surface irregularities [61]. The PL characteristics of CQD fluorescence emission, originating from the conjugated p domain, are regulated by the quantum confinement effect of p-conjugated electrons within the  $\text{sp}^2$  atomic structure. This influence can be altered by the scale, edge orientation, and shape of the conjugated p domain. Surface defects such as  $\text{sp}^2$  and  $\text{sp}^3$  hybridized carbon, along with other surface irregularities of CQDs, contribute to fluorescence emission. Indeed, the fluorescence emission and peak position are correlated with these defects [62].

## 3.2. Synthesis of CQDs

Since their initial identification, numerous techniques have been developed for synthesizing CQDs. Methods for synthesizing CQDs can broadly be classified into top-down and bottom-up approaches [51,63–65]. In the bottom-up approach, small molecules undergo polymerization and carbonization through chemical reactions to form CQDs. Conversely, the top-down approach involves breaking down or dispersing macromolecules into smaller CQDs using physical or chemical methods, as illustrated in Fig. 7.

### 3.2.1. Top-down route

The top-down approach involves employing physical or chemical methods, such as chemical oxidation, laser ablation [66], arc discharge, and electrochemical synthesis [67], to reduce large  $\text{sp}^2$  carbon domains into smaller CQDs. While the precursor is only suitable for materials such as graphene, carbon fibres, and carbon black, which possess large  $\text{sp}^2$  carbon domains, the method is fast, straightforward, and efficient [68,69]. The approach does not allow for precise control over morphological distribution and product size. However, because of the diverse range of sizes and morphologies, it provides an opportunity to create CQDs with adjustable emission wavelengths.

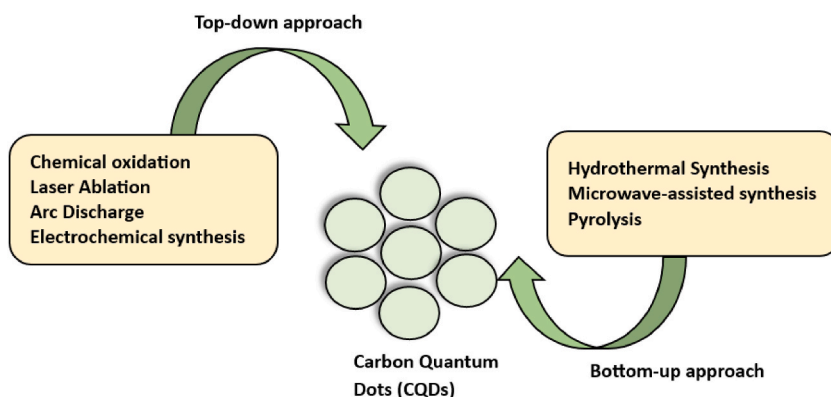
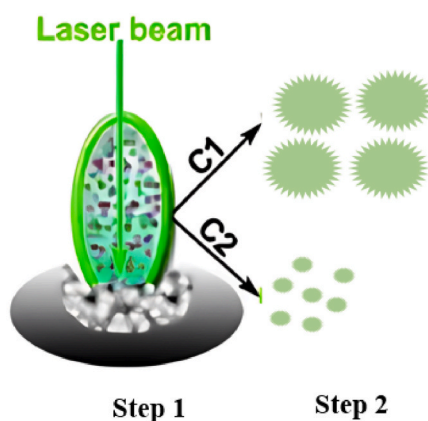


Fig. 7. Common methods used to synthesize CQDs.

**3.2.1.1. Chemical oxidation.** In the process of chemical oxidation to produce CQDs, carbon source precursor materials undergo oxidation using oxidizing agents [70]. Utilizing this approach, carbon raw materials like graphene, CNTs, graphite rods, and reduced graphene oxide (rGO) undergo electrochemical cleavage to generate CQDs. This method involves generating radicals such as  $\text{OH}\bullet$  and  $\text{O}\bullet$  at the anode through the oxidation of water. In CQD production, the  $\text{OH}\bullet$  and  $\text{O}\bullet$  radicals act as electrochemical ‘scissors,’ initiating the exfoliation process typically from the edges and progressing towards the defective areas. In organic solvents, the exfoliation process depends on the electrical tension sustained by the anions intercalating between the layers of graphene [16]. Pan et al. [70] employed an enhanced Hummers technique to oxidize and slice graphite into graphene quantum dots (GQDs). The synthesis conditions for GQDs seem to be more rigorous compared to those for graphene oxide (GO), involving an additional cutting step. The synthesis process entailed oxidizing graphite in an excess of  $\text{H}_2\text{SO}_4$  and  $\text{NaNO}_3$ , followed by high-temperature heating in a controlled autoclave to produce nanoscale GQDs. The resulting GQDs exhibited a quantum yield of approximately 63 % and were uniformly sized, consisting of single or multiple graphite layers. The resulting CQDs from these methods demonstrate high stability, purity, and fluorescence, rendering them appealing materials for applications in drug delivery, biosensors, bioimaging, and solar cells.

**3.2.1.2. Laser ablation.** This technology stands out as a unique and promising approach frequently used to produce CQDs in various geometries. First proposed by Sun et al. [13], this method employs a high-energy laser pulse to illuminate the target’s surface, creating a thermodynamic state characterized by high temperature and pressure. Following irradiation of the carbon raw material (the target material) with a Nd:YAG laser source (1064 nm, 10 Hz), they treated the resulting product with acid. Initially, the produced CQDs did not emit light. However, after surface functionalization with two distinct oligomers, such as poly(ethylene glycol) and poly(propionylethylenimine-coethylenimine), the CQDs exhibited high-intensity PL emission. Following that, numerous research groups effectively showcased the synthesis of CQDs with the desired size by manipulating the laser pulse width to regulate nucleation [13]. Initiating a rapidly expanding bubble involves focusing a high-energy laser plasma pulse at the juncture where the solid target meets the surrounding liquid medium (**step 1 of Fig. 8**). As the bubble contracts under pressure from the surrounding liquid, the cooling core initiates the CQD nucleation process gradually (**step 2 of Fig. 8**) [69]. Hu et al. [71] reported the creation of fluorescent CQDs by laser irradiation of a suspension containing carbon compounds in an organic solvent. They modified the surface state of the CQDs using various organic solvents to achieve tunable light emission. By manipulating the laser pulse width, this technology enables the creation of CQDs with varying cluster densities, each possessing unique characteristics. This process rapidly transforms the target material into a plasma state, followed by vapor crystallization, resulting in the formation of CQD nanoparticles. Laser ablation is a highly effective method known for producing CQDs with precise size distribution, rapid synthesis capabilities, versatility, distinct fluorescence properties, and excellent water solubility. These attributes make them particularly valuable in material science, photodetectors, and drug delivery applications [72,73].

**3.2.1.3. Arc discharge.** The arc-discharge technique involves subjecting a bulk carbon raw material to high-energy conditions, causing it to break down on an anode electrode within a sealed reactor. Plasma is formed between two electrodes positioned at an optimal distance apart under high voltage shown in Fig. 9 [74]. The arc discharge method, initially used to fabricate CQDs, was reported by Xu et al., in 2004. While making SWCNTs using this technique, Xu and his colleagues unintentionally created three different types of carbon-based nanomaterials with varying molecular masses and fluorescence characteristics. The produced CQDs fluoresced in orange, yellow, and blue-green hues at 365 nm. Further tests showed that these CQDs had hydrophilic carboxyl groups on their surfaces, making them soluble in water [10]. Chao-Mujica et al. [75] employed a submerged arc-discharge technique to synthesize CQDs in water. The resulting CQDs displayed excitation wavelengths of 275 nm for band A and 285 nm for band B. They exhibited a quantum yield of 16 % and emitted two distinct bands in the spectra: one at 320–340 nm (band A) and the other at 400–410 nm (band B). These CQDs were utilized as a fluorescent biomarker for L929 murine fibroblasts in cell culture experiments conducted in vitro. The main drawback of this approach is the occurrence of intricate contaminants in the CQDs produced, posing challenges for purification.



**Fig. 8.** Illustration depicting top-down synthesis methods for CQDs: CQD generation via laser ablation: short-pulse laser and long laser pulse widths.

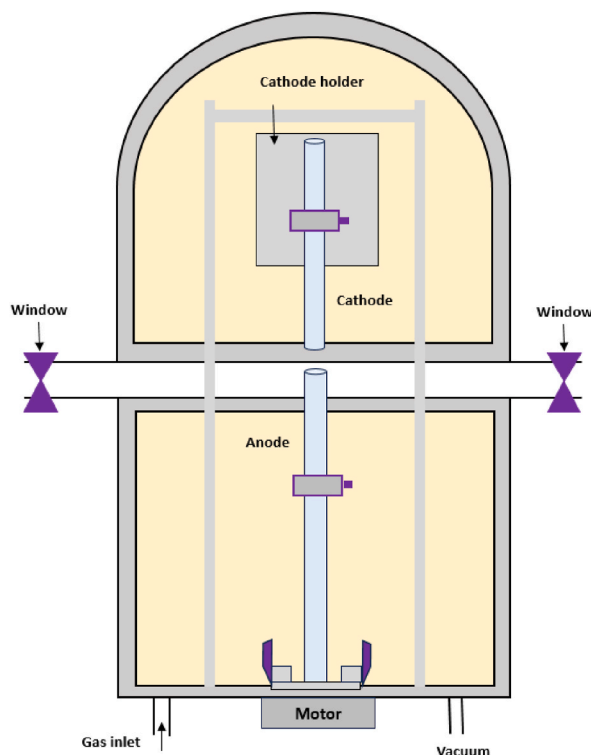


Fig. 9. Illustration depicting the submerged arc-discharge in water (SADW) method for the producing CQDs.

However, most of the time, their particle size distribution tends to be high, resulting in reduced CQD surface area and potentially fewer active reactions. As the particle size increases, the specific surface area of CQDs decreases, leading to a reduction in the number of active reaction sites in the electrocatalytic process [76]. This category of CQDs exhibited limited scalability in terms of particle size and a low quantum yield (QY). However, it demonstrated commendable PL characteristics, and high crystallinity use in applications such as catalysis, electronics, nanocomposites [77,78].

**3.2.1.4. Electrochemical synthesis.** This method is the most utilized, commercially viable, and highly efficient approach for producing CQDs [79–81]. Zhou and colleagues provided the initial demonstration of this method [82]. The authors utilized this method to produce deep-blue luminescent CQDs using multiwalled carbon nanotubes (MWCNTs). In this technique, the synthesis of CQDs involved the addition of 0.1M tetrabutylammonium perchlorate to a three-electrode system. This system comprised a working electrode made of carbon paper with chemical vapor-deposited MWCNTs, a platinum wire counter electrode (CE), and a Ag/AgClO<sub>4</sub>

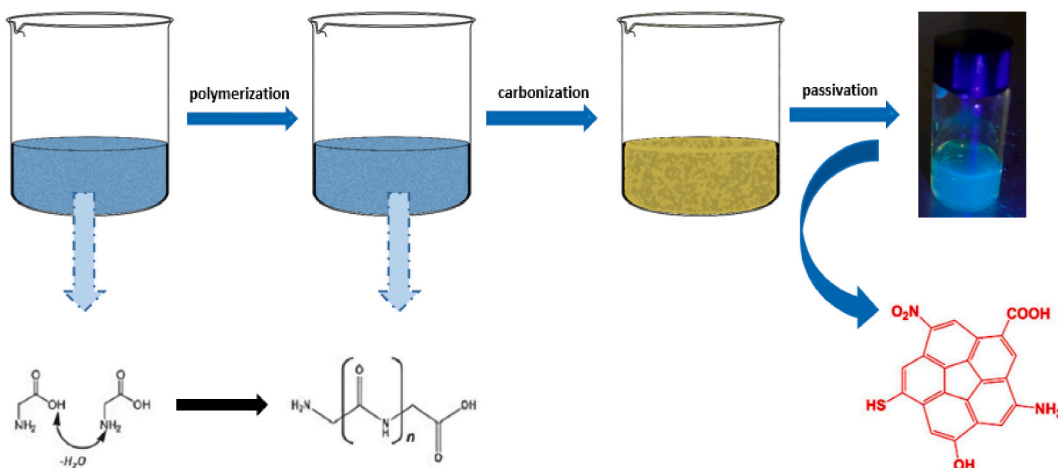


Fig. 10. Diagram illustrating the model for CQD growth following the described steps for glycine.



reference electrode, all submerged in a solution of degassed acetonitrile. This technique yields CQDs characterized by uniform size distribution and excellent crystallinity, potentially finding applications in chemical analysis and biological research and offers several advantages: it is cost-effective, user-friendly, facilitates the production of CQDs with adjustable emission wavelengths across various synthesis conditions, and provides ample opportunity for the preparation of multicolor and long-wavelength emission CQDs for various applications such as optoelectronics, drug delivery, chemical sensors [68,83].

### 3.2.2. Bottom-up route

This method produces CQDs with customizable size and structure by utilizing small molecules as carbon source precursors and combining them with organic molecules to form  $sp^2$  carbon domains. Techniques such as hydrothermal synthesis, microwave-assisted synthesis, and pyrolysis are employed for this purpose. The benefits of this approach include achieving relatively consistent size, the ability to adjust shapes, the use of easily accessible and eco-friendly raw materials, cost-effectiveness, and flexibility to customize the process to suit particular requirements, despite its laborious and intricate nature [68].

**3.2.2.1. Hydrothermal synthesis.** The hydrothermal method, a bottom-up technique, is known for generating uniformly sized particles with a high quantum yield and necessitates minimal system setup. Among the various methods for synthesizing CQDs, the hydrothermal approach stands out as one of the most popular [84]. Using this method, organic precursors undergo carbonization in a sealed hydrothermal reactor lined with Teflon, typically at temperatures ranging between 180 and 200 °C, to synthesize CQDs. Hsu and Chang were the first to discover the hydrothermal approach for synthesizing CQDs. They proposed a growth model for CQDs, outlined in Fig. 10, which comprises four stages: (i) dehydration, (ii) polymerization, (iii) carbonization, and (iv) passivation [85].

Komalavalli et al. [86] utilized Hibiscus sabdariffa leaves as the source material to produce hydroxyl-functionalized luminescent CDs through hydrothermal treatment at 160 °C for 8 h. Meanwhile, Wang et al. [87] employed a hydrothermal method to synthesize various functionalized GQDs, including hydroxyl and amine-functionalized GQDs, by reacting sodium hydroxide and hydrazine hydrate. The hydrothermal process offers precise control and environmental friendliness, making it suitable for applications including energy storage, optoelectronics, and biomedical applications. However, close monitoring of the reaction process is necessary, especially considering the high temperature and pressure conditions involved, which require strict safety measures. Additionally, achieving precise size control for CQDs remains challenging, as the preparation of the CQDs' surfaces and their fabrication occur simultaneously [68].

**3.2.2.2. Microwave-assisted synthesis.** The microwave-assisted method provides high CD yields and is user-friendly, cost-effective, rapid, environmentally friendly, and versatile use in energy storage, bio imaging, and drug delivery [87]. In the synthesis process, molecular-level heating can be induced by the dipolar moments of polar molecules interacting with the alternating electric and magnetic fields present in the solvent [88]. Zhu et al. [89] invented the process of microwave pyrolysis to produce CQDs. They utilized polyethylene glycol-200 (PEG 200) and saccharides (such as fructose or glucose) in a water-based solution, which became transparent and was then heated in a microwave oven. The use of microwave assistance introduces a fresh approach to CQD preparation, offering advantages such as convenient operation, rapid and uniform heating of the reaction, shortened reaction time, and improved yield and quantum yield of CQDs [90]. Shin et al. [91] developed a "top-down" one-pot synthesis technique utilizing high-power microwave processing to produce GQDs in significant quantities. In this process, graphite was dissolved in a strong oxidizing agent consisting of  $H_2SO_4$  and  $KMnO_4$ , with the resulting GQDs generated using low-power microwaves. The resulting GQDs were found to be uniformly dispersed and exhibited a diameter range of 2–8 nm, with an approximate quantum yield of 70 %. Moreover, due to the inability to precisely control the size of the synthesized CQDs with hydrothermal microwave treatment, the microwave-assisted technique applies hydrothermal microwave treatment directly to the reaction solution during the synthesis process [68].

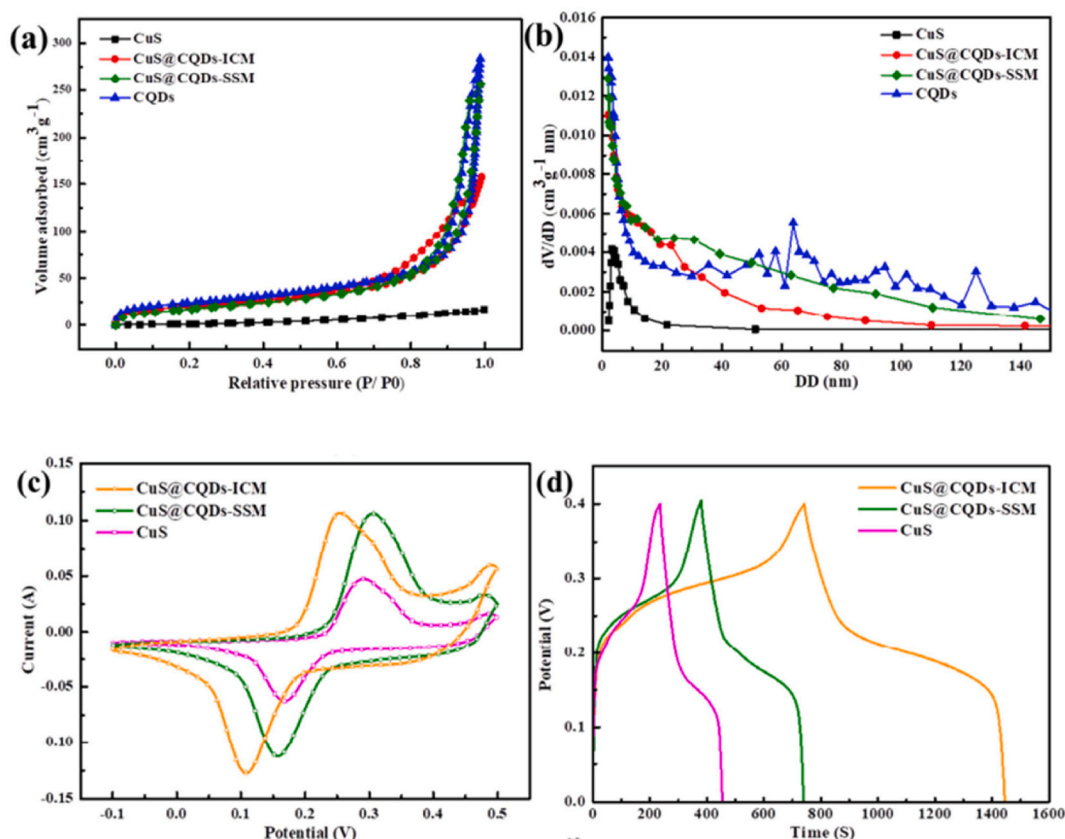
**3.2.2.3. Pyrolysis.** In order to generate luminescent carbon dots, organic small molecules need to undergo carbonization and pyrolysis at a specific temperature. Giannelis and colleagues introduced this technique for the initial synthesis of CQDs. They thermally carbonized various ammonium citrate salts to produce surface-functionalized CNPs for their study [16]. Research indicates that various ammonium citrate salts are employed in the synthesis of carbon dots, where citrate serves as the carbon source and organic ammonium acts as a surface modifier. By selecting appropriate surface modifiers, it becomes feasible to produce hydrophilic and organic carbon dots. The study revealed that bacterial surface proteins contribute to the amide function of CQDs, influencing the generation of reactive oxygen species. Citrate is not the sole small molecule used in pyrolysis for carbon dot creation; polymers synthesized from EDTA, epoxy-enriched polystyrene, and natural substances such as hair, tea leaves, konjac flour, red pepper, among others, have also been employed [92]. The synthesis of CQDs from easily accessible and cost-effective materials offers an economically viable and scalable approach. These CQDs find applications in biomedical fields, such as drug delivery and bimodal imaging, when encapsulated within biocompatible polymers [92,93]. However, purification through dialysis is necessary for the CQDs produced using this method. Nonetheless, by fine-tuning the experimental parameters, this technique can be employed to produce mono disperse CQDs with an average particle size below 10 nm [94].

## 4. CQDs in supercapacitors

Supercapacitors, unlike batteries, are energy storage systems capable of sustaining high output power over prolonged durations [95]. An improved alternative to supercapacitors involves developing electrodes using nanomaterials characterized by higher specific

capacitance and prolonged consistency over time. Due to their exceptional electrical conductivity and customizable size and structure, CQDs find widespread use in supercapacitors. In line with the EDLC mechanism, a double layer of charge forms at the interface between the CQDs and the electrolyte, resulting in charge accumulation. Incorporating CQDs into supercapacitors improves the electrical conductivity of the electrode materials. This enhancement accelerates electron transport from the electrode to the current collector, thereby reducing internal resistance and increasing the overall efficiency of charge/discharge in the device. Consequently, combining metal oxide/sulphide with CQDs to form nano hybrid structures has been identified as a promising approach for fabricating dependable and effective supercapacitors [96]. An electrode made of nanomaterials, exhibiting higher specific capacitance and prolonged stability, holds promise for enhancing supercapacitors. Furthermore, expandable electrodes in supercapacitors prove beneficial for rapid energy storage, enabling significantly high energy densities and swift charging capabilities [97]. Prasath, A et al. [98] developed CQD-MnO<sub>2</sub> nanostructures from an environmentally friendly method utilizing a sustainable waste source, aiming to achieve high power density and specific capacitance. Due to the highly conductive nature of CQDs, structural analysis indicated that CQD- MnO<sub>2</sub> exhibited a larger surface area and improved electrical conductivity compared to pristine MnO<sub>2</sub>. This nano hybrid demonstrated an impressive specific capacitance of 189 F g<sup>-1</sup> along with a long cycle life when employed as the electrode in symmetric supercapacitors. Its swift current-voltage response was corroborated by electrochemical investigations. The remarkable performance of the supercapacitor can be attributed to the existence of a highly conductive CQD nanonetwork and its extensive surface area. The discoveries from this study could establish a sustainable approach for producing innovative CQD MnO<sub>2</sub> electrode materials for energy storage devices.

Transition-metal sulphides are preferred over other materials due to their superior electrochemical activity and high electrical conductivity, making them ideal for electronic functionalities in energy storage systems [99–101]. Incorporating CQDs into sulphide-based materials enhances their stability against degradation during charge/discharge cycles and increases their electrical conductivity. This modification improves ion accessibility, expands the number of active sites available for electrochemical processes, and enhances the overall electrochemical performance of the sulphide material. In 2020, Quan et al. [102] developed a distinctive hierarchical porous nano flower-like nano composite termed CuS@CQDs by integrating CQDs into a CuS nanostructure. To accomplish this, the CuS@CQDs nano composite was synthesized using two different methods: the solvothermal synthesis method (SSM) and the impregnation combination method (ICM). The BET analysis, depicted in Fig. 11a, revealed that these nanocomposites exhibited an improved surface area of 111.2 m<sup>2</sup> g<sup>-1</sup> compared to pristine CuS (22.8 m<sup>2</sup> g<sup>-1</sup>). Additionally, investigations of the pore-size



**Fig. 11.** BET analysis results include (a) N<sub>2</sub> adsorption–desorption isotherms, (b) pore-size distribution curves, and (c) cyclic voltammetry (CV) and (d) galvanostatic charge–discharge curves of an asymmetric device utilizing CuS, CuS@CQDs-ICM, and CuS@CQDs-SSM nanocomposites. Reproduced from Ref. [102] with permission from Elsevier.

distribution indicated abundant mesopore structures in the nanocomposites Fig. 11b. As per the cyclic voltammetry (CV) and charge-discharge profiles depicted in Fig. 11c and d, the electrode fabricated using the CuS@CQDs nano composite exhibited a higher specific capacitance and a power density of  $920.5 \text{ F g}^{-1}$  and  $397.75 \text{ W kg}^{-1}$ , respectively. These outcomes are attributed to the nanocomposites, which likely provide abundant electro active sites and facilitate the transport of electrolytes and electrons. Moreover, even after undergoing 10,000 charge-discharge cycles, the device demonstrated long-term stability, retaining 92.8 % of its initial performance [103]. The synthesized CuS@CQDs-ICM composite exhibits a higher specific capacitance ( $920.5 \text{ F g}^{-1}$  at a current density of  $0.5 \text{ A g}^{-1}$ ) compared to pure CuS ( $369.9 \text{ F g}^{-1}$ ). To assemble an asymmetric supercapacitor, activated carbon (AC) and CuS@CQDs-ICM electrodes are employed as the positive and negative electrodes, respectively. The commendable electrochemical performance of the asymmetric supercapacitor (ASC) device can be attributed to the synergistic impact of CuS and CQDs, which significantly enhanced the material's electron transport properties.

In addition to utilizing nanocomposites, studies have shown that doping carbon quantum dots (CQDs) with different heteroatoms can effectively enhance the specific capacitance and lifespan of energy-storage devices. Several investigations have explored the influence of various dopants, such as nitrogen [103–105], phosphorus [106], iodine [107], oxygen, and boron [108], on the photo-electronic properties of carbon-based materials and the underlying mechanism. Through the chemical oxidative polymerization method, Wang et al. [109] fabricated nanocomposites of nitrogen-doped carbon quantum dots/polyaniline (NCQD/PANI), which were subsequently employed as electrode materials for supercapacitors. The NCQD/PANI nanocomposite exhibited superior electrical conductivity and electrochemical properties compared to PANI or CQD alone, resulting in higher specific capacitance and cycling stability. For the first time, nanocomposites of flake-shaped nitrogen-doped carbon quantum dot/polyaniline (N-CQD/PANI) were synthesized by rapidly combining aniline with nitrogen-doped carbon quantum dots (N-CQDs) abundant in amino groups. The N-CQDs were generated through a microwave pyrolysis process involving 1,2-ethylenediamine (EDA) as a surface passivation agent and citric acid (CA) as the carbon source. PANI chains were either adsorbed through hydrogen bonds or electrostatic contact, or covalently grafted onto the N-CQDs due to their rich surface functional groups such as amino and carboxyl groups. This resulted in enhanced electrical conductivity and overall electrochemical performance, including cycling stability and specific capacitance. Among these, the flake-shaped N-CQD/PANI-3 nanocomposite demonstrated the most outstanding electrochemical performance due to its larger size and reduced aggregation, making it a promising candidate material for high-performance supercapacitors.

Wang et al. [110] employed a binder-free solution immersion and heat treatment method in 2019 to fabricate a novel electrode composite of  $\text{Co}_3\text{O}_4\text{-NiO/graphene}$  foam (GF) derived from a cyanometallic framework. The resultant electrode, benefiting from the synergistic effect of a metal oxide and graphene nanocomposite, exhibited an enhanced specific capacitance of  $766 \text{ F g}^{-1}$  at  $1 \text{ A g}^{-1}$ . This improvement was attributed to the superior electrical conductivity of graphene and the high pseudo capacitance of  $\text{Co}_3\text{O}_4\text{-NiO}$ , respectively. Remarkably, the device incorporating the electrode composite of  $\text{Co}_3\text{O}_4\text{-NiO/GF}$  demonstrated outstanding stability after approximately 5000 cycles of charge and discharge. To enhance the porosity, surface area, electrical conductivity, and other electrochemical properties of nanocomposites, CQDs are often combined with another electrocatalyst. Demonstrating remarkable electrochemical performance, the composite powder exhibits a high capacitance. This study underscores the significant advantages offered by the 9 NiO- $\text{Co}_3\text{O}_4$  composites for supercapacitors. Yu, L. et al. [107] created composite materials wherein carbon dots (CDs) were

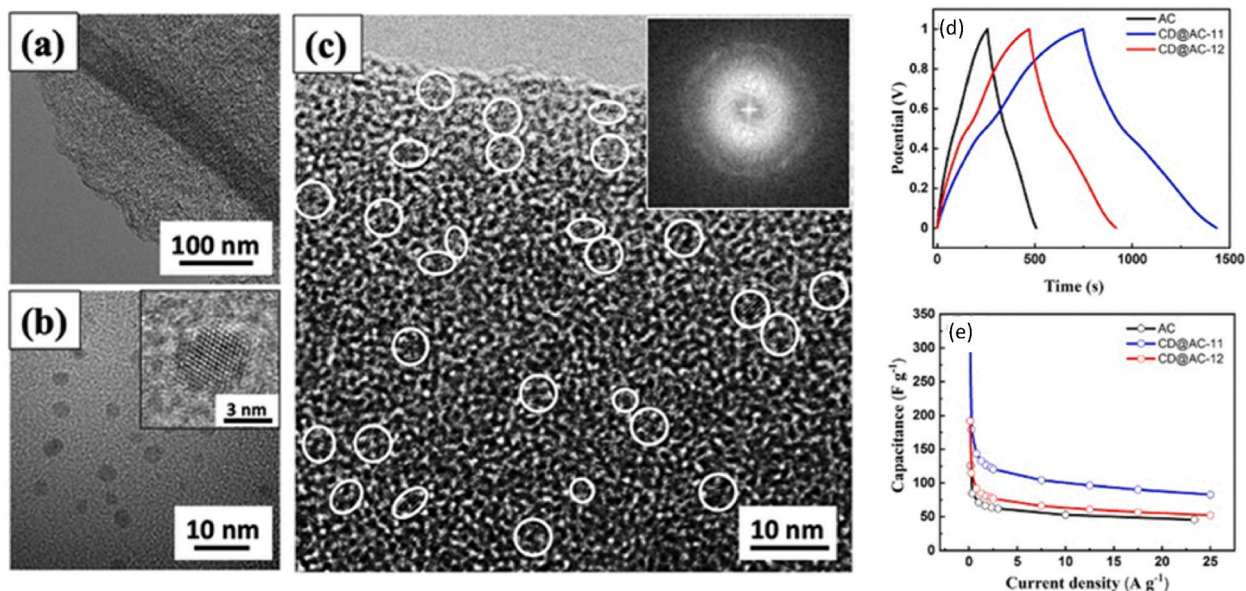


Fig. 12. Transmission electron microscopy (TEM) and high-resolution transmission electron microscopy (HRTEM) images depict (a)ACs, (b) CDs, and (c) CD@AC-11 materials. (d) Galvanostatic charge–discharge (GCD) curves are presented under a current density of  $0.25 \text{ A g}^{-1}$ . (e) The specific capacitance of the obtained samples is measured at various current densities. Reproduced from Ref. [107] under Creative Commons license.

uniformly coated onto lignin-based activated carbons, revealing an interior surface adorned with CD binding sites. The amorphous morphology of the produced activated carbons (ACs) is depicted in the HRTEM image in Fig. 12a. Additionally, the inset of Fig. 12b shows the crystal lattice, indicating a uniform size distribution range of 2 nm–5 nm for the obtained CDs. Following the modification of activated carbons' surface by carbon dots (CDs), numerous CD domains with a crystalline structure are evenly distributed on the AC matrix, as depicted in Fig. 12c. Moreover, the inset of Fig. 12c displays a selected-area electron diffraction pattern indicating bright diffraction rings and spots, signifying the successful transformation of CDs. The hydrophilicity and ultrafine size of CDs significantly enhance electrode penetration in aqueous electrolytes, thereby reducing ion diffusion routes. It is observed that with increasing quantities of CDs, the charging and discharging times progressively lengthen, as shown in Fig. 12d. At 0.15 A g<sup>-1</sup>, the CD@AC-11 electrode exhibits a high capacitance of 301.7 F g<sup>-1</sup>, which is twice as high as that of the AC electrode (125.8 F g<sup>-1</sup>, Fig. 12e). Notably, the CD@AC-12 demonstrates a specific capacitance of 191.8 F g<sup>-1</sup>, indicating a 52 % increase with half the quantity of CDs added, underscoring the impact of CD quantity on capacitance. Enhanced SC performance was achieved by adorning the surface of the activated carbon matrix with uniformly spaced carbon dots (CDs). The release of hydrophilic CDs not only improved the wettability of the electrodes but also created electro active sites. This resulted in a significant increase in capacitance from 125.8 to 301.7 F g<sup>-1</sup>, without compromising high cyclability, owing to the increased effective surface area and improved electrochemical activity. The abundance of polar sites on CDs attracts ions, promoting their clustering and thereby shortening ionic diffusion routes and reducing internal resistance. Research suggests that evenly distributed CDs throughout the carbon matrix can greatly enhance the effective surface area by generating numerous functionalized CD sites, thereby boosting both electric double-layer capacitance and pseudo-capacitance. These findings highlight the efficacy of carbon dot modification in enhancing AC performance, although the understanding in this field is still evolving [111]. By incorporating CQDs with various heteroatoms or transition-metal oxide/sulphides, nanocomposites are produced that serve as both electron donors and acceptors, thereby increasing the number of surface-active sites and enhancing overall device performance. Given the straightforward nature of this method, its provision of additional active sites for improved electrochemical performance, and the synergistic properties it exhibits, further research is expected to focus on synthesizing stable and efficient electrodes through the creation of novel nanocomposites using the heteroatom doping strategy [16].

## 5. Conclusion

This work extensively addresses the working mechanism of supercapacitors, properties of CQDs, synthesis method, and application of CQDs in supercapacitors. Supercapacitors have advanced from their initial role as backup devices for electronic memory to powering grid and energy harvesting systems, as well as being utilized in automobile regenerative braking systems. Currently, there is a growing demand for energy storage devices as part of efforts toward achieving a more sustainable planet. To address their limitations and meet the increasing demand across various applications, the electrochemical performance of supercapacitors is being enhanced. Key factors influencing supercapacitor performance include the electrode and electrolyte, as they respectively facilitate ion transport and intercalation for energy and charge storage. Scientists have devised several methods aimed at enhancing the energy density, power density, and capacitance of electrode materials. These methods typically involve improving the material's ionic conductivity or leveraging synergistic effects. By implementing these techniques, the overall electrochemical performance of supercapacitors can be significantly enhanced. Nevertheless, we suggest that researchers prioritize the electrochemical performance stability and flexibility under bending and stretching when developing flexible supercapacitors. This aspect is crucial for the next generation of electrode materials, especially considering the technological progress in portable and wearable electronic devices by CQDs. Flexible supercapacitors hold significant promise for future applications across various fields, particularly in modern electronics. Their fabrication process requires careful consideration of factors such as fabrication methods, electrolyte composition, electrode materials, and device architecture to achieve optimal electrochemical performance and mechanical stability. However, flexible supercapacitors offer several advantages over conventional counterparts, including improved mechanical durability, uniformity, and cyclic stability [112]. Furthermore, considering that bio waste is cost-effective and environmentally friendly, researchers are encouraged to prioritize the exploration of green raw materials, especially for producing carbonaceous materials from them. The latest paper focuses on synthesizing EDLC devices from CQDs derived from green materials. Therefore, enhancing the electrochemical performance of supercapacitors by integrating recyclable pseudocapacitive materials with green carbonaceous materials also contributes to environmental sustainability. Due to the unique quantum size, ease of surface functionalization, and abundance of functional groups, electrode materials based on CQDs have exhibited outstanding electrochemical performance in supercapacitors, indicating significant potential in the design of electrode materials for this application [113,114]. CQDs have demonstrated significant potential for application in advanced supercapacitors. However, several challenges need to be addressed before CQDs can be effectively utilized in energy storage systems, including the development of scalable fabrication methods and systematic regulation mechanisms [3]. Additionally, the integration of CQDs into supercapacitors is still in its early stages, but future applications may involve modifications to the electrolyte, separator, and current collector.

## Research data policy and data availability statements

No data was used for the research described in the article.

## CRedit authorship contribution statement

**Grishika Arora:** Writing – review & editing, Writing – original draft, Validation, Conceptualization. **Nuur Syahidah Sabran:** Writing – review & editing. **Chai Yan Ng:** Supervision. **Foo Wah Low:** Supervision. **H.K. Jun:** Writing – review & editing, Supervision, Project administration, Funding acquisition, Conceptualization.

## Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: HK Jun reports financial support was provided by Malaysia Ministry of Higher Education.

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