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Review

Comprehensive Review on Multifaceted Carbon Dot Nanocatalysts: Sources and Energy Applications

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Abstract: In recent decades, several studies have been conducted on sustainability progress with high efficiency of renewable energies by utilizing advanced nano-module catalysts. Some collaborative studies advocate the unique characteristics of unconventional materials, including carbon nanotubes, nanosheets, nanoparticles, conducting polymers, integrated nano polymers, nano enzymes, and zero-dimensional nanomaterials/carbon dots (CDs) at the atomic and molecular level to generate efficient energy from various biomass substrates. Nanotechnology-based catalysts are considered a crucial tool for revolutionizing various energy-related applications. This review article addresses the sustainable and scarce biomass resources to synthesize CDs, properties, mechanisms, and insights with the advancement of research on CDs as nanocatalysts in the field of energy applications. These materials possess exceptional and rapidly expanding features such as being non-toxic, biocompatible, having excellent electrocatalytic activity and photoluminescence, and being highly dispersible in water. Because of these advantages, they are appealing for use in energy conversion and as storage material. Moreover, the emphasis is placed on the function of CDs as nanocatalysts for energy storage devices, and relevant instances are provided to clarify the concepts. These advanced strategies of nanotechnology for energy storage and conversion are expected to play a vital role in promoting sustainability.

Keywords: carbon dots; biomass; precursors; biofuel cell; energy storage; nanocatalyst

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1. Introduction

Researchers are concerned about how the preservation of the ecosystem and human health is threatened by the rise in atmospheric pollution and rising energy consumption. To tackle these issues, several technologies are being actively pursued, such as renewable energy sources, alternative energy storage [1–3], biofuel cells [4], sustainability [5], advanced nanocatalysts such as two dimensional (2D) materials, one dimensional (1D) materials, zero-dimensional materials, or carbon dots (CDs) [6–10].

In the field of electronics, nanomaterials such as ambipolar graphene quantum dots and other 2D materials play a significant role in excellent carrier mobility in phototransistors with excellent light-harvesting properties. The distinctive structure of 3D graphene allows it to efficiently absorb light while maintaining excellent electrical conductivity [11,12]. Additionally, there has been significant interest in the use of CDs, specifically group IV–VI quantum dots, due to their excellent light-harvesting capabilities in the infrared region. These quantum dots can be conveniently integrated with silicon substrates through a solution process. This integration method offers a practical and efficient approach to incorporating quantum dots into silicon-based devices [12]. CDs emit efficiently in the blue-green range, with the peak shifting towards longer wavelengths as excitation increases. A limited understanding of photoluminescence (PL) in CDs hinders researchers due to the complex structure and variability of the PL centers [13]. Nevertheless, theoretical calculations offer insight into the excited states and electronic structures of different CDs in the context of their optical properties [14].

According to several reports [8,15,16], CDs were first introduced in 2004, and Sun's research group later gave the name 'fluorescent CDs' in 2006 [17]. Xu's group found CDs during the downstream of single-wall nanotubes (SWNTs) by the gel phoresies of carbon soot [6,8,15,16]. CDs have quasi-spherical even shapes with ultra nano size (almost less than 10 nm) that are primarily made of sp^2 or sp^3 amorphous carbon along with nanocrystalline graphene layers and some functional groups such as O (almost 5 to 50 wt%), S, $-NH_2$, N, $-OH$, and $-COOH$ [15,18], which mostly depends on the technique involved [15,16,19]. Due to their ubiquitous optical, electrical, thermal, biological, and physicochemical properties [20], CDs are potential replacements for traditional bio-based nanomaterials in several domains [21–24]. Additionally, CDs possess excellent electron-transferring abilities due to the uniform dispersion quality of quantum dots [16]. The increased usage of CDs has been hindered by numerous debates due to the rapid publication of emerging nanomaterials, which creates significant obstacles to comprehending their natural properties, thus substantially impeding their widespread adoption [22].

Among the several nanomaterials like graphene, graphene oxide [10,25], and reduced graphene oxide, CDs are becoming a subject of growing interest in various fields and are being examined as a potential substitute for traditional energy storage materials, specifically semiconductor quantum dots, enzymatic biofuel cells, and electronic devices [26] due to their high tunable band gap, higher surface-to-volume ratio, and quantum confinement effect [27].

Some researchers have reported CDs as carbon quantum dots (CQDs)/multi-layered graphene quantum dots (m-GQDs) due to the uncertainty surrounding the classical quantum confinement effect. This is because there is some uncertainty regarding whether the quantum confinement effect is present in CDs or whether their unique properties are due to other factors, such as surface functionalization or defects. Some researchers may use these alternative names to reflect the particles' properties more accurately and avoid confusion regarding their true nature [28]. The wide range of synthesized processes and materials is essentially the main cause of the diversity of CDs [29]. Currently, the synthesis of CDs is still in its primary stage [16]. Two major techniques are adapted for the synthesis of CDs. Top-down techniques (breaking down large carbon particles into smaller sizes) employ harsh and powerful processes, including electrochemical oxidation [30], arc discharging [31], chemical oxidation [16], and laser ablation [29]. For bottom-up techniques, CDs are formed from small molecules or polymer precursors such as ethylenediaminetetraacetic acid (EDTA) [32], citric acid, ethylene glycol, etc., [33–35] under relatively simple and benign conditions, like microwave-assisted pyrolysis, hydrothermal treatment, and ultrasonic reaction [31,33,34]. The drawbacks of the adapted techniques include low yield, time-consuming, synthetic conditions, treatment processes, high cost, and toxicity [36]. This promotes the requirement of a green synthesis technique for producing superior luminous CDs for practical applications [33,37] and heteroatom doping for promoting catalytic activity [36].

Natural sources for CDs include materials like vegetables (onion, ginger, and cabbage) [33] and fruits (orange juice, banana juice, and winter melon) [33,36]. Several other sources include biomass such as betel leaf [37], sugarcane [38], soy milk, bovine serum albumin, gelatin, pomelo peels, bottles, silkworm, chitosan, grape juice, salicin cortex, and papaya powder [33,39].

Numerous reviews advocate the luminance properties of CDs for applications in luminescent devices [8], solar cells [36], bioimaging, fluorescent inks [6], light-emitting diodes [34], sensors [40], supercapacitors, Li-ion batteries, K-ion batteries, sodium-ion batteries, and metal-air batteries [6,40] with significant performance. Extensive research has been dedicated to the development and modulation of CDs as nanocatalysts due to the drive to minimize carbon footprints and advance clean energy conversion and storage technologies in a smooth and rapid manner. It is important to compare the current state-of-the-art technologies to establish a benchmark for progress. This review provides an in-depth understanding of CDs' potential as nanocatalysts, including their resources,

properties, mechanisms, and recent progress in their applications for energy conversion and storage. These applications include using CDs to convert transferring electrons into electricity in biofuel cells for self-powered biosensors or portable electronic devices, as an electrocatalyst accelerates the hydrogen evolution reaction and stores electrical energy in supercapacitors. The article provides valuable insights to encourage further research on the use of CDs as an energy material. The review also provides valuable insights to encourage green synthesized CDs as catalysts and further research prospects and economic analysis that can be used in the advancement of sustainable energy applications.

2. Synthesis Techniques of Carbon Dots and Precursors

Both natural and synthetic carbon-containing substances have been employed as precursors to produce CDs. CD synthesis approaches are mainly classified into two groups: top-down and bottom-up. Several studies advocate the utilization of both techniques for synthesizing CDs [6,16,41,42]. Top-down techniques involve the chemical or physical breakdown of larger carbon substances into smaller CDs. Processes including laser ablation, acidic oxidation, arc discharge, electrochemical synthesis, and others are employed to degrade carbon-rich materials, namely carbon nanotubes (CNTs), graphite rods, activated carbons (ACs), carbon fibers, carbon ash, etc., to ultimately produce CDs. This method favors high crystallinity and well-preserved nanostructures [41,43]. Conversely, the bottom-up approach involves polymerization and oligomers (intermediate products in polymerization reactions) under ambient temperature and pressure to generate CDs. The mechanisms resulting in the excellent luminescence properties of CDs have been discussed [44]. Carbon-based nanoparticles (CNDs) exhibit 0D nanostructures with 10 nm dimensions and a 'core-shell' structure consisting of an ordered sp^2 core and disordered sp^3 shell. Varied sp^2/sp^3 ratios result from synthesis and environmental factors, affecting optical properties like visible emission, excitation-dependent redshift, and competition with organic dyes and quantum dots. Due to this mechanism, carbon nanodots have varied and controllable emission spectra, making them useful for a variety of applications, including optoelectronics and bioimaging [44].

Amorphous CD structures can be produced with more abundant surface functional groups and multiple doping sites. This is attributed to its excellent performance in chemical sensing and cell imaging applications [36,42]. Glucose, ascorbic acid [45], citric acid [46], ammonium citrate [36], graphite [47], EDTA [32], polyethylene glycol [48], urea, and thiourea [49] are various commonly employed chemical carbon sources as a precursor for preparing CDs. However, these chemicals have resulted in significant environmental, economic, and social challenges due to their reliance on environmentally hazardous solvents and organic compounds with high energy consumption. As a result, significant efforts are being made to adapt the eco-friendly synthesis of CDs employing natural carbon sources, biomass, or less toxic precursors.

2.1. Green Biomass as Carbon Dot Precursors

Over the past decades, due to increased social awareness and a sense of responsibility for sustainability, there has been a noticeable growth in the use of biomass. The precursor biomass materials include a variety of vegetables, leaves, fruit, and organic sources, including proteins, alkaloids, carotenoids, and carbohydrates (Figure 1). Arul et al. (2017) synthesized graphitic-natured nitrogen-doped carbon dots (N-CDs) using dragon fruit (*Hylocereus undatus*) extract and aqueous liquor ammonia (nitrogen source) using the hydrothermal method that exhibits strong blue fluorescence at 400 nm when excited at 320 nm with an ultra-small diameter of 2.5 nm [30].

It was explored that N-CDs emit strong blue luminance and good biocompatibility on both Michigan Cancer Foundation-7 (MCF-7) cells and lymphoblastoid-929 (L-929). They also demonstrated superior catalytic activity for dye reduction, especially methylene blue reduction via sodium borohydride [30,50]. Shen's group studied probes for Fe^{3+} detection. They synthesized highly fluorescent CDs from sweet potatoes using a

hydrothermal technique, which resulted in CDs with a yield of 8.64%. CDs demonstrated great water dispersibility due to the availability of functional groups on their surfaces. Furthermore, cytotoxicity tests demonstrated that CDs were non-toxic at concentrations up to 100 µg/mL. These fluorescent CDs were appropriate for probes in cell imaging within a range of 1–100 µM and a minimum detectable limit of 0.32 µM [50].

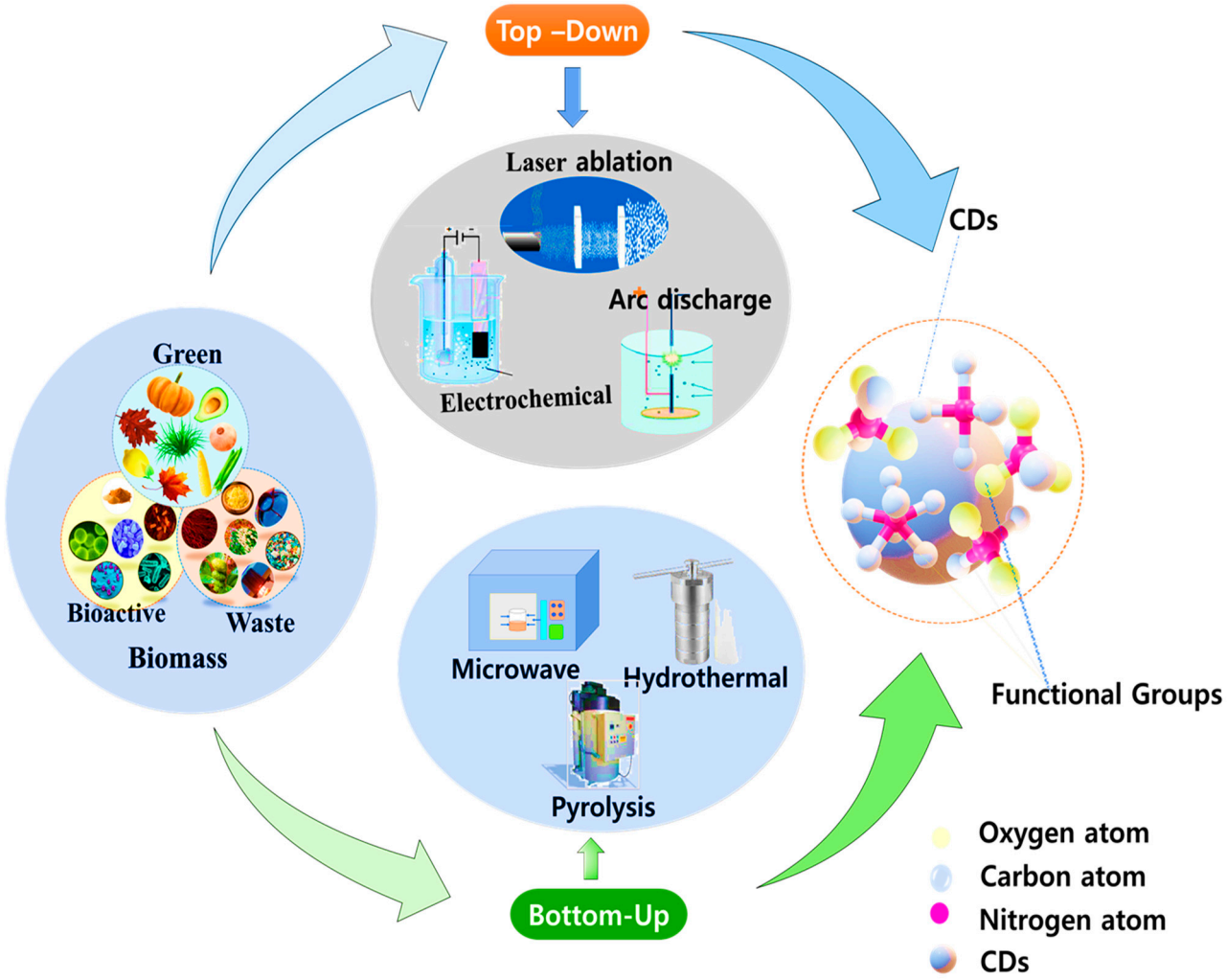


Figure 1. Synthesis techniques and various biomass for CDs.

CDs synthesized from water-soluble citric acid were considered to overcome size control and high energy consumption issues and were synthesized from empty fruit bunches (EFB) biochar employing the autoclave hydrothermal technique with a 4 nm average size. Furthermore, it was noted that the growth rate of bacteria co-cultured with these CDs declined with the increase in the concentration of CDs, resulting in up to 100% antimicrobial activity. For *E. coli* and *S. aureus*, the minimum inhibitory concentration (MIC) was 60 µg/mL [46]. In a similar investigation, biocompatible copper oxide nanoparticles (CuNPs) were synthesized using Morinda fir extract [51]. It showed higher nociceptive responses and anti-inflammatory potential without any harmful effects, which was confirmed with preclinical research, including peritoneal leukocyte infiltration, carrageenan-induced paw edema, and carrageenan-induced air pouch tests [51]. Researchers in the field of energy have been focusing on the utilization of polymer-CD composites for energy storage devices. Oskueyan et al. (2021) synthesized integrated polypyrrole and graphene carbon dots from carrot juice through a hydrothermal process and in situ polymerization to improve the electrochemical efficiency of the supercapacitor. They developed a high-potential window

supercapacitor after integrating a nanocomposite of polyaniline with 10 wt% CDs with polypyrrole-graphene, demonstrating a maximum specific capacitance of 396 F/g under a current density of 5 A g^{-1} [52].

Additionally, unusual bioresources have been used as carbon precursors for CDs. For example, Zhao et al. (2018) developed a nanosensor probe (detection limit of 0.05 μM) for the diagnosis of uric acid in human serum and urine samples using a pig source as a carbon precursor with a high quantum yield (17.3%) and superior chemical stability. However, some other sources, including food waste materials, bioactive molecules, and microorganisms, are better for the synthesis of CDs. Therefore, the recent developments related to synthesizing CDs from these biosources are a focus [53].

2.2. Synthesis of Carbon Dots from Waste Biomass

The increase in population and the growing demand for horticultural products are major obstacles to effectively managing waste biomass. Nonetheless, the focus has shifted towards utilizing waste biomass to create value-added products such as valuable chemicals, biofuels, bio-oil [54], nano-biochar [55], energy storage products [56], etc. Waste biomass can be considered a sustainable and affordable material for the synthesis of CDs. Fe^{3+} ions could be detected with the CDs obtained from onion waste [33]. These water-dispersible CDs were employed as blue, green, and red fluorescent probes for the detection of Fe^{3+} ions via fluorescence quenching under different conditions, including a wide range of molarity (0–20 mM), pH, high ionic strength, and continuous irradiation with a detection limit of 0.31 mM for Fe^{3+} ions. It had good biocompatibility for both cancerous (HeLa) and normal (HEK-293) cells [33].

Likewise, CDs were produced using sugarcane molasses as a carbon source from industrial waste [38]. These CDs had a small size of 1.9 nm and a quantum yield of 5.8%. This study found that the illuminance quenching intensity of the CDs decreased upon the addition of Fe^{3+} or by giving them a sunset yellow color. Furthermore, the researchers assessed the biocompatibility of CDs in vivo and in vitro and discovered their ability to perform bioimaging in MCF-7 cells [38]. To create smaller N-CDs, Pankaj et al. (2018) synthesized EDTA as a nitrogen source and sodium lauryl sulfate from waste-candle soot. The researchers were able to decrease the size of N-CDs to as small as 2−5 nm to selectively detect Fe^{3+} and Hg^{2+} ions. By maintaining a pH level of 4–5, a functional group with a negative net surface charge (−10.4 mV) was successfully produced [57]. Thakur et al. (2019) also utilized pyrolysis to synthesize CDs from the waste pulp of *Citrus limetta* for the detection of Fe^{3+} ions. The CDs had an ultra-small size range of 4–7 nm and achieved the highest quantum yield of 63.3% among other green-synthesized CDs. These CDs showed promising results in various applications, including electrolysis during the photoelectrochemical current density of 6 mA/cm^2 for water splitting, methylene blue reduction, and antibacterial activities against *S. aureus* and *E. coli* [58].

2.3. Microorganisms and Bioactive Molecules as a Carbon Dot Precursor

Microorganisms are regarded as efficient materials for synthesizing various nanomaterials such as ZnO, Cu_2O, Au, CdS, and MgO nanoparticles [59]. This method is remarkable for environmental protection and minimizes waste. Recently, microorganisms like yeast, bacteria, cyanobacteria, algae, actinobacteria, etc., and bioactive molecules such as thiamine pyrophosphate (Vitamin B_1), malonic acid, ascorbic acid, gelatine, dopamine, bovine serum albumin (BSA), etc., have also been utilized as green precursors in synthesizing CDs [9,60–62]. Wu et al. (2018) synthesized CDs by a novel one-step approach from yeast extract powder through the hydrothermal method at 160 °C with a high yield of 65.8% and a size of 3.36 nm. They achieved good dispersibility in water. They also prepared CDs integrated with polyvinyl alcohol (PVA). It showed good water-induced shape memory performance at the optimum temperature (room temperature). Additionally, it can be synthesized economically on an industrial scale without harming the environment [9].

A similar study was conducted by Bakhshi et al. (2016) on the cost-effective, eco-friendly, and replicable production of cadmium sulfate carbon dots (CdS-CDs) with a particle size of 4.4 nm at room temperature using *Bacillus licheniformis* ptcc1320 as a surfactant [63]. Lin et al. (2018) reported the formation of biofilms and their detrimental effects on biomedical and industrial applications. In their investigation, carbon dots from *Lactobacillus plantarum* biomass were prepared through single-step hydrothermal carbonization without using any chemicals. These CDs demonstrated low cytotoxicity and good biocompatibility. Additionally, it has been demonstrated that these CDs effectively inhibit the formation of biofilms by *E. coli* without hindering the growth of *E. coli*, thus overcoming the cytotoxicity issues associated with many current antibiofilm agents and becoming a novel and safe approach for biofilm treatment [60].

Gong et al. (2017) fabricated a fluorescent probe using N, S, and P co-doped CDs derived from *Saccharomyces cerevisiae* via the hydrothermal pyrolysis method. This probe could detect manganese (VII) with a detection limit of 50 nmol/L and L-ascorbic acid (L-AA) with a detection limit of 1.2 µmol/L in medicinal herb, river water, and tap water samples through label-free and 'on-off-on' detection techniques. Additionally, it is worth developing an 'AND' logic gate using the same probe for environmental monitoring, disease diagnosis, and cellular labeling [64]. Investigations are being accomplished on different bioactive molecules to achieve two objectives: first, to minimize the severity of the synthesis conditions, and second, to synthesize particles with ubiquitous properties [62]. Ganjkhanlou et al. (2022) synthesized dual fluorescence carbon dots. These CDs, as nanosensors, have immense promise and enormous potential in the fields of life and materials sciences [65].

CDs were synthesized from bioactive molecules such as glutathione and formamide through the solvothermal method at 180 °C [65]. They observed two fluorescence CDs (red-shedding porphyrin and blue-shedding carbon dots) as synthesis products. They separated a mixture of red-emissive and blue-emissive kaolinite [65]. To enhance the accumulation of astaxanthin in microorganisms, Ghose et al. (2017) prepared N-CDs derived from carbonized BSA. The carbon dots exhibited blue emission with a quantum yield of up to 44%, which surpasses the CDs synthesized using bioactive molecules. In *Haematococcus pluvialis* culture, astaxanthin (dihydroxy-3,3′ dioxo-4,4′ bêta-carotène) production increased by double (66 mg/L) in just one week after adding the N-CDs (1 mg/L), contrary to two weeks during the reddening stage for the control (29 mg/L) [62]. These results proved their photostability and reusability because their fluorescence spectra displayed no crucial variation after a month of continuous irradiation. N-CDs have a promising future in the natural astaxanthin industry and other value-added products produced by microorganisms [62]. Similarly, Amjad et al. (2019) prepared semiconductor quantum dots (QDs) from bovine gelatin through a hydrothermal technique to overcome their toxicity. Surprisingly, the resulting carbon bovine gelatin nanotubes exhibited QDs-like fluorescence characteristics despite their size exceeding (59.45–67 nm) the Bohr exciton radius.

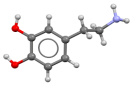
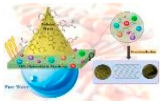
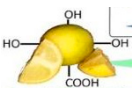
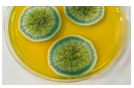
CDs were also prepared from *Pectinodesmus* sp. algal biomass under the same hydrothermal conditions (200 °C and 3 h), resulting in CA-PHM3$_{algae}$ nanodots [66]. A comparison of CG and CA-PHM3$_{algae}$ nanodots revealed that surface chemical composition played a significant role in determining their surface states and PL properties. CGs were found to be non-toxic and suitable for bacterial cells and imaging plants, while both CA-PHM3$_{algae}$ and CGs showed promising anticancer properties against HCC 1954 (breast cancer) and HCT 116 (colorectal cancer) cell lines [66].

The value-added utilization of waste biomass for the synthesis of CDs, an affordable and environmentally friendly resource, not only addresses the pressing need for large-scale synthesis of CDs but also drives the advancement of sustainable management practices. However, it is important to consider certain aspects considering the recent progress in the green synthesis of CDs. First, most of the current synthetic methods have been carried out on a laboratory scale. The production of CDs on a larger scale using these green precursors has not been extensively explored, leaving uncertainty regarding whether the synthesized

samples would exhibit the same unique properties observed in lab-scale preparations. Considering the high demand for CDs in numerous applications, large-scale production from green sources would have significant economic and environmental implications.

Table 1 lists a variety of biomass precursors that have been mentioned in the literature and have been used to synthesize CDs.

Table 1. Synthesis methods, applications, and derivative CD types from various precursors.

No.	Precursor	Image	Size	Quantum Yield	Synthesis Method	Type of CDs	Application	Reference
1	Onion waste		0.21 nm	28%	Autoclave	CDs	Antimicrobial activity against HEK-293 cells, HeLa cells, Fe^{2+} ion probe detection	[67]
2	Aloe peel		<10 nm	NA	Hydrothermal	N-CDs	Electrode	[68]
3	Betel leaf		<10 nm	NA	Hydrothermal	N-CDs	Picric acid and Fe ion detector probe	[37]
4	Sugarcane bagasse		1.9 nm	5.80%	Hydrothermal	GQDs	Bioimaging (MCF-7 cells)	[38]
5	Apple seeds		<10 nm	NA	Pyrolysis	CDs	Detection of 4-nitrophenol, bioimaging	[69]
6	Coriander leaves		<10 nm	NA	Hydrothermal	CDs	Fe^{3+} detection, bioimaging for lung normal (L-132) and cancer (A549) cell lines	[70]
7	*Citrus limetta*		4~7 nm	63.30%	Pyrolysis	G-CDs	Photoelectrochemical water splitting, dye removal (MB), Fe (III) ions sensor, anti-bactericidal activity	[58]
8	Carrot		<10 nm	NA	Hydrothermal	Polymer CDs	Supercapacitor	[52]
9	Animal skin waste (Collagen)		<10 nm	NA	Hydrothermal	CDs, biofilm	Nanophotonics	[71]
10	Dopamine		2–4 nm	NA	Sonochemical	CQDs	Fe^{2+} detection, nanothermometer to sense temperature both in water and in cells	[72]
11	Ultra-pure water impregnated by TiO_2		3–6 nm	NA	Hydrothermal	TiO_2/CDs nanohybrid	Photocatalytic activity, peroxidase mimicking activity	[73]
12	Candle soot		2–5 nm	NA	Hydrothermal	N-CDs	Hg and Fe ions probe detector	[57]
13	Citric acid with Ppy * and PANI *		<10 nm	NA	Hydrothermal	Ppy-CDs	Electrode	[20]
14	Graphite Powders		1 nm	NA	Laser ablation	N-CDs	Catalytic and biosensor	[74]
15	*Penicillium chrysogenum*		10.28 nm	NA	Gamma-ray irradiation	CDs	Antimicrobial agents against *Enterococcus faecalis*, *Candida albicans*, and *Klebsiella pneumoniae*	[59]

* Ppy: polypyrrole, PANI: polyaniline.

3. Properties and Functionalization of Carbon Dots

Jiang et al. (2022) and Yao et al. (2019) have noted that CDs can be produced using various precursors and methods, resulting in a wide range of sizes, morphologies, and chemical structures. This complexity leads to significant variations in the physicochemical properties of CDs [40,75]. Good dispersibility is essential for the processing and application of CD solutions. Most carbon nanodots are hydrophilic due to oxygen-containing functional groups (−OH, −COOH, CO, etc.) that are either derived from precursors or generated during the synthesis procedure. The ability to manipulate the wettability of CDs in response to environmental factors has been achieved, which could broaden the range of applications for carbon dots [15,76]. Kumar et al. (2022). elevated the electron recovery by adding CDs, which were synthesized from peanut shells through pyrolysis at optimum conditions (250 °C, 2 h) into the microbial fuel cell. In the investigation, CDs were employed as an oxidizing agent for organic substrate (acetate) at the anode, and by dispersing CDs, the greater surface area of CDs in the anode chamber enhanced the conductivity of anolyte by reducing the ohmic overpotential [76].

Ramanavicius and Ramanavicius (2020) prepared integrated red-emitting CDs with MXenes and found that DNA has a significant role in dispersing the Ti_3C_2-based nanosheets and in the creation of Pd/Pt and Pd structures. The Ti_3C_2-based MXene provided conductive support. A novel electrochemical biosensor (MXene/DNA/Pd/Pt/GCE) electrode was fabricated for the measurement of dopamine, with a detection range of 0.2–1000 mM and a minimum limit of detection of 30 nM [77]. Despite having a wide range of chemical structures, most CDs display a similar UV-Vis absorbance spectrum, with a pronounced absorption band that decreases from the UV to the visible region. However, some CDs are visibly distinct due to various precursors in different solvents [17]. CDs exhibit their optical characteristics through absorption and fluorescence measurements. Specifically, their absorption properties in the UV range (230–320 nm) are used to describe their optical properties. The peak absorption at 230 nm is primarily due to the π-π* transition of aromatic C-C bonds in the carbon core of the CD. Additionally, a smaller absorption peak (shoulder peak) at 300 nm is attributed to the n-π* transition of C=O bonds [78,79]. This absorbance is attributed to the p-p* transitions of conjugated electrons and n-p* transitions from oxygen or heteroatoms. The shape and intensity of the curve can be adjusted through surface modification or heteroatom doping employing the same precursors [78,80].

Dias et al. (2019) reported three distinct CDs from kiwi, pear, and avocado fruits through the same hydrothermal approach. These showed a high fluorescent intensity range of 470–542 nm [81]. Bioenergy generation through microbial extracellular electron transfer (EET) was reported by Zhang et al. (2022). The PL intensity of CDs significantly decreased after the reaction between CDs and cytochrome due to the accumulation of CDs induced by cytochrome. The UV-Vis absorption spectra were used to confirm the changes in the cytochrome with CDs. The characteristic Soret band (408 nm) and Q-band (529 nm) of native cytochrome remained unchanged even after adding CDs, which indicates that the structure of hemoprotein remained pristine, which could lead to new applications in the coming year [82].

Tripathi et al. (2022) prepared anode CDs coated with Fe (II, III) oxide (Fe_3O_4) for applications in microbial fuel cells. The obtained maximum PL was 1 mg/cm^2, which was higher than the graphite anode [83]. According to Guo et al. (2019), a new way to modify the bacteria's surface was achieved using positively charged N, S-doped carbon dots (m-NSCDs), which can attract negatively charged bacteria through electrostatic attraction. These m-NSCDs not only improve the bacteria's conductivity but also act as efficient photosensitizers that enhance biocatalysts under light, leading to a significant increase in EET during bio-anodic oxidation techniques in microbial fuel cells. Additionally, doping both S and N on carbon dots via carbonization and etching of hair fiber via H_2SO_4 resulted in good luminescence stability [84].

The minimal toxicity of CDs is crucial for their successful biological applications in various biomedical fields. Typically, CDs have low toxicity at low concentrations, which is

mainly due to their bulk form, and at this size, they either decompose or are eliminated by the excretory system [85]. In certain situations, the toxicity of CDs is greatly heightened when exposed to an external stimulus [86]. Various types of anodic materials have been reported to enhance electron transfer rates, wastewater treatment, and energy-generation processes in microbial fuel cell (MFC) systems [87]. The significant surface area of the anodic substances has a crucial role in achieving high efficiency in removing pollutants and generating a current, as it promotes the growth of bacteria at the anode's surface, whereas graphene-based CDs cannot be preferred because of their poisonous effect on bacteria [88].

The formation of a biofilm indicates that microbes can strongly adhere and have good biocompatibility, which helps with extracellular electron transfer and improves energy conversion efficiency. However, if the biofilm becomes too thick, the mass diffusion coefficient may be negatively impacted. Therefore, it is necessary to investigate the optimal thickness of the biofilm to improve the efficiency of MFCs [89]. Sn-based CD alloys have been extensively studied as anodic substances in MFCs because of their electrocatalytic activity and biocompatibility with living organisms [89]. The research was conducted for the synthesis, characterization, properties, and bioimaging capabilities of CDs derived from widely cultivated fruits such as pear (*Pyrus*), avocado (*Persea americana*), and kiwi (*Actinidia*) [81]. Among them, they found that kiwi-derived CDs were highly toxic for zebrafish embryos and the epithelial cell lines of humans [81]. CDs possess a remarkable combination of optical, surface, and biocompatibility properties and are thus expected to have a wide range of potential uses in industries including biomedicine, energy storage, environmental sensing, electrochemical electrocatalysis, and optoelectronics.

4. Carbon Dots as Nanocatalysts in Energy Storage and Conversion

4.1. Biofuel Cells

Biofuel cells (BFCs) have shown great potential as a power source for portable biomedical devices and self-powered biosensor portable electronic devices by transferring electrons between enzymes and electrodes as well as the degradation of organic substrates [18,90–92]. This is due to their superior efficiency, volumetric power density, biocompatibility, low working temperature, and neutral pH [93–95]. Mostly, BFCs' power supply capacity is in the range of 10 µW–450 mW with a voltage range of 0.5–1 V [93,95] and a power density of 3.7 mW/cm^2 [91]. However, the major limitations of the technology include limited mass transportation, the minimal utility of enzymes, low durability and lifetime, and slow electron transfer [91,94,96].

Bioenergy generation through microbial extracellular electron transfer (EET) was reported by Zhang et al. (2022). They reported that by incorporating CDs, the efficiency of electron transfer could be enhanced five times from the pure dissimilatory metal-reducing bacteria (DMRB) due to the enhancement in biofilm immobilization and riboflavin secretion [82]. To create advanced BFCs, direct electron transfer (DET) between electrodes and redox enzymes is the best solution. It provides higher stability and better power supply without a redox mediator at an optimal voltage [79,90]. Since it only relies on the position and direction of the active site within the protein, DET requires an immediate connection between the enzyme and the electrode, which is not always possible [92]. Barelli et al. (2019) reported that during successful DET, the tunneling distance should be about 1.5 nm. The effective DET electron transfer process demonstrated that the highly conductive, porous macro or nanofabricated electrode material is the turning point for enhancing the number of wired enzymes per unit volume [18,90].

Earlier studies have explored important avenues for the efficient utilization of carbon-based materials such as CDs, nanofibers, graphene, nanowires, and carbon black in BFCs for implantable and low-power device applications [90,91,97].

Researchers have developed advanced nanoelectrode materials and immobilization techniques for enhancing the efficiency of BFCs (Figure 2) and biosensors [26]. Recently, nanoscale technologies have addressed the issue of the low electron transfer efficiency between the enzyme and electrode surface, along with permitting the assimilation of

a higher enzyme load to improve the efficiency of kinetic processes in BFCs through modified fabrication. Similarly, Wu et al. (2017) prepared CDs from candle soot to design a laccase-based electrode. It was reported that the laccase activity was 220 U/mg, and the CD immobilizing matrixes facilitated high methanol oxidation through direct electron transfer at the anode and oxygen reduction into H_2O at the laccase-based cathode. They achieved a better power density of 68.7 ± 0.4 μW/cm^2 and an open-circuit voltage (OCV) of 0.71 ± 0.02 V after fabricating the immobilizing matrixes of CDs [18].

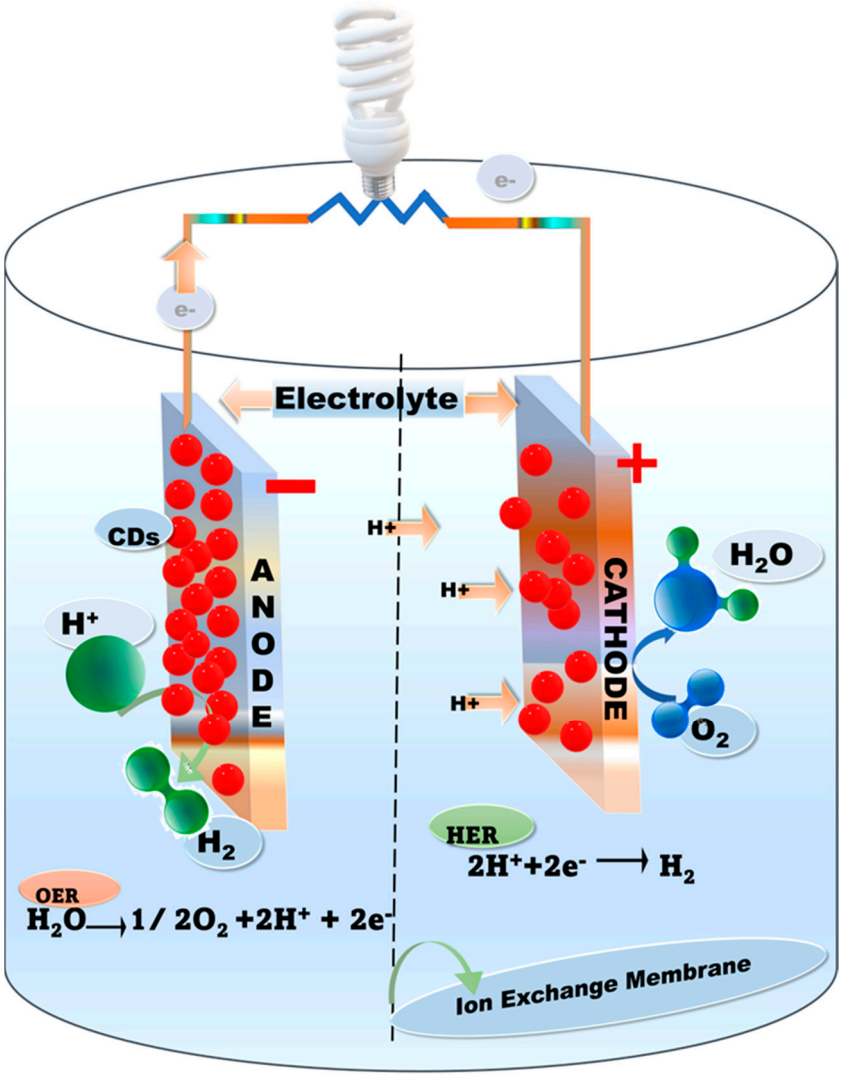


Figure 2. Schematic illustration of the full-cell configuration with CDs as electrocatalysis.

Zhao et al. (2015) developed a biosensor for glucose detection using direct electron transfer reactions of glucose oxidase (GOx) and bilirubin oxidase (BOD) immobilized on CDs. The biosensor has a detection range of 0–0.64 mM and an optimum high sensitivity of 6.1−1 μA mM with an optimal detection limit of 1.07 μM. Additionally, the fast DET high rate constant of the CD electrodes trapped in GOx was 6.28 ± 0.05 s, and the apparent Michaelis–Menten constant for dextrose affinity was as low as 0.85 ± 0.03 mM [92]. Further, due to their excellent direct bioelectrocatalytic performance, CDs were discovered to be effective for the development of bioelectrochemical sensors and BFCs. Using CD electrodes, a DET-type glucose–air enzymatic BFC was successfully modified employing CD electrodes entrapped with GOx that oxidized glucose at the anode. With an OCV maximum voltage of 0.93 V and a maximum power density of 40.8 μW/cm^2, BOD reduced oxygen at the biocathode.

According to these findings, CDs hold potential as materials for immobilizing enzymes and creating effective bioelectrochemical devices [92]. Through in situ coupling with MFCs, Zeng et al. (2019) prepared a novel sustainable self-energy conversion system to produce renewable CDs. It was found that the generation of CDs highly enhanced electricity production [12]. So far, few studies have been conducted on CD-based BFCs. However, the potential of CDs in BFCs is yet to be realized. Due to their ultra-nano size, CDs have the potential to integrate with the biocatalysts and directly contact their active centers. By capitalizing on these exceptional qualities, biomass CD (BCD)-based BFCs can be developed with better performance.

4.2. Electrocatalysts for Energy Conversion

The performance of CDs as carbon nanomaterials for electrocatalysts has been advocated due to their high dispersibility in polar solvents, strong coordination, and distinctive electron transfer ability [98]. Additionally, CDs can be combined with other nanomaterials or metal to create 2D and 3D nanostructures along with $-COOH$, $-NH_2$, $-OH$, or other similar functional groups via self-crosslinking or splicing, hydrophilic terminals, or covalent bonds with rich edge structures. Thus, it accelerates the hydrogen evolution reaction (HER) by significantly expanding the three-phase boundary where reactants, electrolytes, and electrons converge [87].

Moreover, the flexible carbon structure and the surface chemistry of the CDs allow optimization of the electrocatalysts based on CDs while also promoting the development of advanced biofuel cells [98,99]. The CD-based electrocatalysts have used several synthetic techniques, including hydrothermal treatment, calcination, electrodeposition, and reflux. These catalysts can be classified as metallic or non-metallic based on the availability of the metal components [42,98,99].

HER is an extensively researched topic in the electrocatalyst field and involves an uncomplicated proton–electron transfer process without any accompanying reactions. A three-electrode system (working, reference, and counter electrode) is used to measure HER. The reference electrode's potential is measured via the reference electrode [56].

According to Volmer's equation, the intermediate step entails the adsorption of hydrogen onto the electrode's surface. Electronic interactions and thermodynamics with catalyst materials have a significant impact on hydrogen generation.

The binding energies are determined by the kinetics and thermodynamics of the reactions occurring on the electrode surface, as well as other factors such as solvent contribution. In the case of transition metal catalysts, the catalyst performs according to the Sabatier principle [100].

Once absorbed, the HER proceeds through the Heyrovsky or Tafel equation, depending on the pathway. The following Volmer–Heyrovský mechanism involves the reaction of the HER electrode system [56,100].

$$H^+ + e^- \rightarrow H \text{ (Proton-coupled electron transfer step (PCET))} \quad (1)$$

$$H^+ + e^- + H \rightarrow H_2 \text{ (Heyrovsky reaction)} \quad (2)$$

$$H + H \rightarrow H_2 \text{ (Tafel reaction)} \quad (3)$$

In the following studies, to facilitate the HER process, a suitable catalyst is necessary for lowering the overpotential and activation energy. Pt metal is a frequently used catalyst for HER due to its long lifespan and minimal overpotential. However, due to its high cost and limited availability, researchers are working on developing suitable, cost-effective substitute catalysts for HER [101]. Moreover, the use of Pt as a catalyst in an alkaline environment is hindered by hydroxyl ion ($-OH$) poisoning, necessitating the substitution of a highly stable and low overpotential Pt catalyst. In this context, CDs, along with their metallic and non-metallic counterparts, could serve as an efficient electrocatalyst [56,101,102].

Developing an effective electrocatalyst to facilitate HER from water is a significant but difficult task that must be addressed to facilitate the ongoing energy crisis. Yang et al. (2015) prepared CDs (TiO_2/CD nanohybrids) for the development of an alkaline electrocatalyst for HER (1 M KOH). Hybrid facile fabrication of a nickel nanoparticle/carbon quantum dot (Ni/CQD) displayed outstanding catalytic activities for HER, with an initial potential the same as the Pt wire and a low Tafel slope of 98 mV/dec, due to the Ni-O-C interface between Ni nanoparticles and CDs. The stability of the Ni/CDs hybrid was also found to be high, as is observed by the negligible current loss after 1000 cyclic voltammetry. Additionally, the Ni/CDs hybrid exhibited improved catalytic performance and UV-light illumination, with a lower Tafel slope of 77 mV/dec [101].

Moreover, it is also crucial to establish techniques for the precise characterization and quantification of active sites in reactions 1, 2, and 3. However, the number and type of active sites are often not reported, and determining turnover frequency can be a challenging task. It is crucial to conduct meticulous investigations of catalytic activity using flat electrodes with controlled catalyst density, develop electrodes with well-defined active sites, and explore techniques for quantifying active sites that are critical for determining the true activity of catalytic materials [100]. Overall, hybrid CDs are potentially efficient electrocatalysts for HER, which could contribute to the development of sustainable energy solutions.

Studies focusing on the preparation methods and applications of various CDs in different energy storage and conversion fields are summarized in Table 2.

Table 2. Reported data on the utilization of various CD composites and their application in energy conversion and storage.

Catalyst Name	Synthesis Method	Size (nm)	Current Density (mA/cm^2)	Applications	References
Graphitic N	Hydrogel	3–5	1.62	ORR, HER, photocatalysis	[47]
Nitrogen-deficient g-C_3N_4	Hydrothermal	<10	10	HER	[102]
C_3N_4/$(Co(OH)_2$/$Cu(OH)_2$	Microwave-assisted	0.27	8.9	CO_2 reduction	[103]
Ni-CDs	Electrochemical	2–5	0.23	HER	[101]
Polyaniline Carrot derived-CDs	Hydrothermal	6–8	5 A/g	Window supercapacitor	[52]

4.3. Supercapacitor

Supercapacitors or ultracapacitors are based on redox reactions to store energy (Figure 3). There are three types of supercapacitors based on the type of electrochemical reactions occurring: pseudo-capacitors (PCs)/Faraday capacitors, electrostatic double-layer capacitors (EDLCs), and hybrid ion capacitors (HICs) that combine the two capacitors [40]. When conducting polymers, metal oxides, or metal nitrides are used as electrode materials in a pseudo-capacitor, a reduction–oxidation reaction takes place, resulting in a higher transfer of electron charges between the electrode and the electrolyte, resulting in a higher electrochemical pseudo-capacitance. Therefore, compared to electric double-layer capacitances (EDLCs), PCs and HICs perform better [40,104].

Despite the advancements in renewable energy storage devices, high-capacity HICs like potassium-ion capacitors, sodium-ion capacitors, and lithium-ion capacitors face challenges in achieving high energy density and power density due to an imbalance in capacity and kinetics between the anode and cathode materials. [104–106]. In the pursuit of supercapacitors that can charge and discharge faster and have higher energy densities, it has become crucial to explore various high-performance electrode materials. To achieve this, different approaches have been adopted, such as utilizing nanomaterials and modifying existing electrode materials, creating materials based on the compatibility of electrolytes

and electrodes, and exploring novel materials [43,106]. Previous reports advocate that the addition of CDs to composites can improve their conductivity and reduce the electrolyte diffusion length during the charge–discharge process. Some composites that combine CDs and metal chalcogenides were successfully synthesized with promising results.

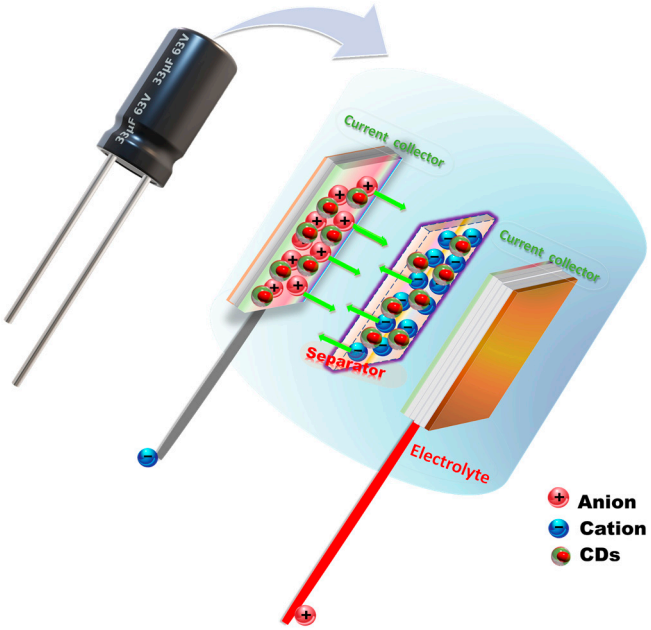


Figure 3. The schematic mechanism illustration of fabricated asymmetric supercapacitor devices based on CDs.

Wang et al. (2020) investigated the effect of CDs on a rechargeable supercapacitor with light assistance. They created CDs by utilizing pro-anthocyanidin precursors for light-assisted supercapacitors (OPC-CDs-700), resulting in the generation of CDs that act as a photoactive medium to stabilize charge under light illumination. This allows for greater charge accumulation on the material surface and the storage of more energy under light illumination. The researchers were able to achieve a 54.4% increase in specific capacitance (312 F/g at 0.1 A/g) in light conditions as compared to dark conditions. This was due to CDs providing a larger specific surface area for OPC-CDs-700, which promotes mass transport and charge transfer [105].

Utilizing CDs with integrated graphene to create various three-dimensional porous structures is a proven effective approach for preventing graphene agglomeration, improving electrical conductivity and mechanical strength, and increasing wettability. Jin et al. (2018) synthesized nitrogen and oxygen-co-doped carbon nanodots (N-O-CDs) from discarded fiberboards using a sequence of carbonization, acid treatment, centrifugation, and dialysis purification procedures. These CDs could be integrated with graphene oxide to form a graphene hydrogel for supercapacitor electrodes. The composite hydrogel was created at an ideal mixing ratio with the desired porosity and optimum loading weight of 200 g. The composite hydrogel electrode displayed a specific capacitance of 335.1 F/g at 1 A/g and excellent mechanical strength, retaining approximately 90.6% of its capacitance after 500 bending and unbending cycles. Furthermore, the flexible symmetric supercapacitor demonstrated cycling stability of 83.4% after 10,000 charge/discharge cycles at 5 A/g with a high specific capacitance of 121.0 F/g [107].

Numerous studies have concentrated on exfoliating layered materials and restacking the 2D exfoliated nanosheets such as MoS_2 to form electrodes, as well as the enhanced electrochemical response, to promote higher conductivity with the formation of metallic octahedral structures during the intermediate state (1T-phase) and account for the enhancement of the electrochemical performance of the electrode [108]. Gao et al. (2016)

investigated the utilization of thiourea as a precursor for N-doped CDs, confirming the effect of hindering the agglomeration of MoS_2 and increasing the interlayer spacing of MoS_2 through the generation of NH_3 during the hydrothermal process. The integration of MoS_2/reduced graphene oxide at polyaniline (MoS_2/RGO @ PANI) resulted in a synergistic effect that led to exceptional energy storage performance, with remarkable capacitive value (1224 F/g at 1 A/g), excellent rate capability (721 F/g at 20 A/g), and a high level of cyclic stability of 82.5% after 3000 cycles [92]. Additionally, the symmetric cell that utilized MoS_2/RGO @ PANI demonstrated favorable capacitive properties (160 F/g at 1 A/g) along with impressive energy (22.3 W h/kg) and power density (5.08 kW/kg) [109].

As aforementioned, CD-based supercapacitors and graphene at metal–organic frameworks (graphene @ MOF) integration with CDs can improve the properties of electrodes by merging physical and charge storage mechanisms into one. This combination can provide an ideal mixture of energy and power density due to EDLC from graphene-contenting materials and the pseudo-capacitance from metal-based MOFs [94].

4.4. Photocatalysts

In photocatalysis, CDs have proven to be adaptable materials with a variety of uses. They are extremely useful for capturing solar energy in a variety of catalytic processes due to their special qualities. The wide absorption spectrum extending into the visible range is one of the main advantages of being able to use sunlight effectively. This characteristic distinguishes CDs from traditional semiconductor photocatalysts such as titanium dioxide (TiO_2) and enables them to effectively catalyze environmentally important reactions. CDs are used in environmental remediation to degrade colors and medicines and to break down organic contaminants in wastewater [110]. CDs are also essential for hydrogen generation via water splitting, which helps produce hydrogen in a greener manner. In the process of reducing carbon dioxide, CDs help to convert CO_2 into organic molecules or valuable hydrocarbons, facilitating the capture and usage of CO_2, and are used to increase solar cell efficiency, which helps convert sunlight into power. Furthermore, their antimicrobial qualities make them useful for sterilizing and purifying water. When exposed to light, they help break down organic pollutants and are integrated into self-cleaning surfaces [111]. Additionally, CDs participate in selective photoredox reactions, providing excellent selectivity in the production of different compounds and medications. Researchers continue to find ways to modify CDs to further improve photocatalytic efficiency, which would increase the number of sustainable and energy-efficient processes in which they can be used [110,111].

5. Economic Analysis

Due to their potential to transform chemical energy stored in biofuels into electrical energy, biofuel cells have emerged as a promising renewable energy technology. Carbon-based catalysts improve biofuel cell performance by facilitating efficient electrochemical processes. BCDs made from sustainable biomass materials offer enormous potential as a cost-effective and environmentally friendly alternative to traditional catalytic materials. However, there is a lack of detailed economic analysis of BCDs in biofuel cell applications. It is very important to address the economic feasibility of employing BCDs in commercial biofuel cells.

The economic study is divided into three primary steps: production cost estimation, scalability assessment, and prospective economic benefit analysis. First, the cost of BCD manufacturing is determined by considering the costs of raw biomass sources, synthesis processes, purification techniques, and post-treatment operations. Energy consumption, labor costs, and equipment investments are all considered. Second, scalability is evaluated by examining BCD production capacity and associated costs at various sizes, from laboratory to industrial. Finally, the potential economic benefits of using BCDs in biofuel cells are assessed by considering parameters such as increased energy conversion efficiency, longer lifespan, lower maintenance costs, and overall system performance. The economic study

demonstrates that the choice of biomass feedstock, synthesis process, and purification techniques has a substantial impact on the cost of producing BCDs. However, because of the abundance and low cost of biomass feedstocks, BCDs have numerous cost advantages over standard catalytic materials. Furthermore, the scalability analysis suggests that BCD production may be easily scaled up without significant cost increases. Increased power output, lower fuel use, and extended lifespan of biofuel cells are all potential economic benefits of BCD adoption, resulting in cost savings and improved overall system efficiency. BCD implementation can result in enhanced energy conversion efficiency, lower operational costs, and increased sustainability. To realize their full economic potential in commercial biofuel cell applications, further research and development activities should focus on optimizing synthesis techniques, discovering novel biomass feedstocks, and improving BCD performance.

6. Conclusions and Prospects

Recent green synthesis of CDs was emphasized using biomass materials, particularly novel, contemporary, and more sustainable precursors. To promote sustainability, these methods rely on the use of renewable resources, including biomass, biowaste, and microorganisms, which are both inexpensive and environmentally friendly. Meanwhile, the current synthesis processes have only been conducted on a small scale. Therefore, it is still unclear whether the unique properties exhibited by the lab-scale preparation of CDs can be replicated in large-scale production using green sources. If large-scale production can be achieved, it would have significant economic and environmental benefits, as CDs are highly sought-after materials for many applications. Various factors affecting CDs, including their size, functional groups, the crystallinity of the carbon core structure, and other factors, decisively influence their properties. To deal with these, further deep studies of the structure-activity relationship of CDs are needed.

CDs offer distinct advantages and enormous potential for clean energy conversion and energy storage. However, the complex mechanism of CDs as an electrocatalysis material in the energy storage field makes it challenging. To address this challenge, new in situ analysis and kinetic characterization methods should be introduced for utilizing CDs as an excellent model. Although CDs might be the most efficient material for energy applications, they have provided valuable insights into catalytic mechanisms and catalyst design principles. With the continued development of advancement, BCDs can display distinctive optical and electrical features with electrocatalytic properties. These properties make BCDs a fitting choice for various applications in the sustainable energy domain, such as enhancing energy conversion efficiency by serving as nanocatalysts for electrodes, mediators, and electrocatalysts for water splitting in hydrogen fuel production. This review presents an idiosyncratic opportunity in the field of energy to expand the possibility of green synthesized CD nanocatalysts with multifunctional properties. Based on current global research, CDs as modulated nanocatalysts with potential applications in clean and green energy are an exciting and quickly evolving field that holds great promise for promoting sustainability.

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