

## Review

## A review of carbon dots in synthesis strategy



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## ABSTRACT

Among the carbon nanomaterials family, the fluorescent carbon dot has emerged as a significant member among the novel types of nanomaterials, and many researchers have introduced studies on its properties and potential applications. Carbon dots (CDs) have numerous outstanding characteristics, such as remarkable photostability, negligible cytotoxicity, exceptional biocompatibility, effortless surface modification, and remarkable chemical inertness. As a result, they have found extensive applications in various domains, including but not limited to cell imaging, *in vivo* imaging, drug delivery, fluorescence sensing, photocatalysis, production of polychromatic light emitting diodes (LEDs), energy conversion and storage, etc. Recently, many synthesis strategies of carbon dots have been developed, but how to synthesize in a controlled way is still challenging. The focus of this article is to provide an overview of the advancements made in the production of carbon dots, with particular attention given to their synthesis techniques along with some discussion and provide a reference for the controllable synthesis and method selection of carbon dots.

## 1. Introduction

Carbon dot belongs to the family of carbon nanomaterials and is regarded as a significant member due to its unique fluorescent properties. It is worth mentioning that CDs were first discovered by researchers accidentally when purifying single-walled carbon nanotubes manufactured by arc discharge method in 2004 [1]. Subsequently, these type of new materials were named by Sun et al. in 2006 [2]. In general, CDs refer to carbon particles that are finely dispersed and have a size of less than 10 nm, which are coated with functional groups like hydroxyl, amino, carboxyl or aromatic ring. CDs have gained significant attention in recent years due to their numerous exceptional properties. Apart from possessing a high quantum yield and adjustable emission wavelength like traditional semiconductor quantum dots, CDs exhibit remarkable photostability, negligible cytotoxicity, exceptional biocompatibility, effortless surface modification, and remarkable chemical inertness [3]. Based on their fundamental composition, CDs can be classified into four categories: graphene quantum dots (GQDs) that have a 2D layered graphene core, carbon quantum dots (CQDs) with a spherical crystal core, carbon nanodots (CNDs) that have an amorphous core [4], and carbonized polymer dots (CPDs) which possess a highly dehydrated crosslinking polymer frame or a slightly graphitized core [5,6]. In addition to the most basic constituent elements C, H, O, doping elements

such as N, S, P are usually introduced into CDs. Fig. 1 illustrates the extensive utilization of CDs in various areas, including cell imaging [7–10], *in vivo* imaging [5,11], drug delivery [12–15], fluorescence sensing [16–24], photocatalysis [25–29], production of polychromatic LEDs [30–33], energy conversion and storage [34–44], etc. As a novel type of fluorescent nanomaterial, CDs have exhibited remarkable promise. However, the controllable synthesis of CDs still faces many challenges. Since the discovery of CDs, various functional CDs have been synthesized with diversified strategies. This review describes various synthetic routes for the production of CDs. Finally, the characteristics of each method are summarized. We aim to create a thorough summary of the various synthesis strategies applied for CDs and offer a useful reference for those seeking to achieve controllable synthesis and select appropriate methods for CDs. Table 1.

## 2. Synthesis methods

Since the discovery of CDs, how to synthesize CDs in a controlled way has been the key for scientists to research. According to different carbon sources, the synthesis strategies of CDs can be roughly divided into “top-down” and “bottom-up” [45,46]. The “top-down” synthesis method involves the stripping process from larger carbon sources, either physically or chemically, to create small CDs. Generally, carbon nanotubes

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[1,47]. carbon fibres, and graphite rods [2] are used as carbon sources for “top-down” method. On the contrary, the “bottom-up” synthesis method employs small carbon sources under molecular or ionic states to synthesize CDs. Most organic small molecules or oligomers include citric acid, glucose, polyethylene glycol and urea are taken into consideration. Common “top-down” and “bottom-up” methods will be discussed in the following sections.

## 2.1. “top-down” strategies

### 2.1.1. Arc discharge

The arc discharge process takes place in a low-pressure arc chamber filled with inert gas. Powdered carbon precursors are usually made into anode and the cathode is usually made of pure graphite. When the system is working, the precursors are evaporated and consumed. As a result, the carbon precursors sublime and travel towards the cathode, where they deposit as nanostructures. CDs were firstly discovered in this way. Xu et al. discovered two novel nanomaterials during the purification of single-walled carbon nanotubes (SWNT) extracted from arc discharge soot [1]. These materials were separated from the coarse soot, and included short tubular carbon as well as a mixture of fluorescent nanoparticles originating from SWNT. Carboxyl functional groups were introduced to the arc-treated soot by oxidizing it with 3.3 mol/L HNO<sub>3</sub> to enhance the hydrophilicity of the carbon material, and then the

precipitate was extracted using a NaOH solution and subsequently separated through gel electrophoresis. It was suggested that the separated fluorescent carbon and short tubular carbon species were a novel carbon material, which was later named CDs. It should be noted that while CDs produced through this method exhibit good fluorescence, the particle size distribution is inconsistent and the yield is exceptionally low, making it unsuitable for large-scale manufacturing. Further research suggested that the spacing of the electrodes played a crucial role in determining the diameter and quality of the carbon nanotubes synthesized [48]. By arranging the magnetic field lines appropriately, the motion of the arc and the production process can be enhanced. The magnetic field also boosted the growth rate of the deposit and improves the purity of the resultant product. This may be a way to improve the purity of CDs obtained by arc discharge method.

### 2.1.2. Laser ablation method

Laser ablation involves utilizing a high-energy laser to irradiate the material, leading to rapid heating. Laser pulses can generate small pits in the sample, consuming the sample and creating plasma flowing through the sample surface, thereby ionizing the sample. Early years, a carbon target was created by Sun and colleagues through the hot-pressing of a combination of graphite powder and cement, followed by a gradual process of baking, curing, and annealing in a flow of argon gas [2]. In the presence of water, argon was used as carrier gas to ablate carbon target

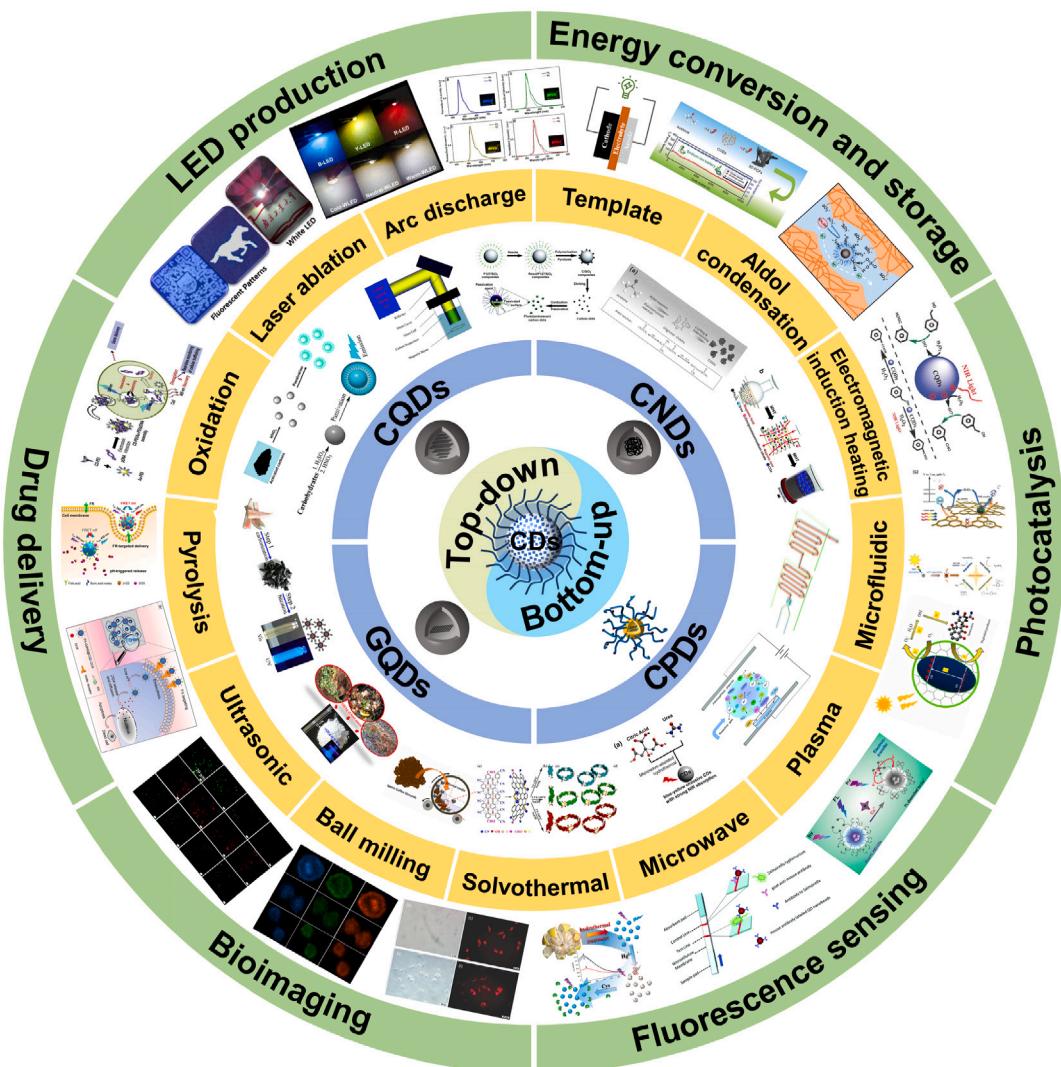


Fig. 1. A summary of the synthesis process and potential application of CDs.

**Table 1**  
Summary of synthetic strategies for CDs.

| Precursor   | Synthetic method   | Application   | Ref.     | Quantum yield  | Advantages   | Disadvantages   |
|---|--|---|----------|----------------|--|---|
| soot  | arc-discharge  | –   | 1        | –              | good fluorescence  | uneven particle size; low yield                                     |
| graphite  | arc-discharge  | –   | 48       | –              |  | uniform particle size; strict equipment requirements                |
| a mixture of graphite powder and cement   | laser ablation   | –   | 2        | 4%-10%         | structure and composition reservation; acceptable yield  |   |
| amorphous carbon  | laser ablation   | –   | 49       | –              |  |   |
| nano-carbon material  | laser ablation   | –   | 50       | –              |  |   |
| graphite powders  | laser ablation   | –   | 52       | 1.2%-12.2%     |  |   |
| carbon cloth  | laser ablation   | cell bioimaging   | 51       | 35.4%          |  |   |
| carbohydrates   | concentrated sulfuric acid and nitric acid oxidation   | –   | 53       | 13%            | effective and convenient; rich raw materials   | consumes a large amount of oxidant and not environmentally friendly |
| activated carbon starch   | nitric acid oxidation<br>300 °C carbonization, HAc and H <sub>2</sub> O <sub>2</sub> oxidation | cell bioimaging<br>cell bioimaging                              | 54<br>55 | 12.6%<br>11.4% |  |   |
| single-walled carbon nanotubes  | electrochemical oxidation  | –   | 47       | 6.4%           | convenient operation; low cost; economic; simple and green; high yield and uniform particle size | requires special equipment; few reactions suitable                  |
| graphite  | electrochemical oxidation  | sensitive detection of Fe <sup>3+</sup> and cell bioimaging     | 56       | 4.6%-11.2%     |  |   |
| L-ascorbic acid   | electrochemical oxidation  | antibacterial   | 57       | –              |  |   |
| screen-printed carbon electrodes  | electrochemical oxidation  | cell bioimaging and solid-state electrochemiluminescence sensor | 58       | 6.6%-10.5%     |  |   |
| watermelon peel   | pyrolysis  | cell bioimaging   | 60       | 7.1%           |  |   |
| thioureapenetrated ZIF  | pyrolysis  | fluorescence sensing  | 63       | –              |  |   |
| boric acid, pyromellitic acid, trimesic acid, and terephthalic acid                                 | pyrolysis  | –   | 64       | 47%-65%        | simple operation; controllable conditions and the raw materials are easy to obtain               | low yield   |
| cellulose acetate butyrate  | pyrolysis  | potassium ion batteries electrode material                      | 65       | –              |  |   |
| ascorbic acid and Cu(Ac) <sub>2</sub> H <sub>2</sub> O  | one-pot synthesis at 90 °C, 5 h  | –   | 73       | –              |  |   |
| strawberry juice  | hydrothermal treatment at 180 °C, 12 h   | detection of Hg <sup>2+</sup>                                   | 22       | 6.3%           | simple; safe and efficient; little damage to the environment; widespread raw materials           | uneven heating; time-consuming and low product yield                |
| citric acid and ethylenediamine   | hydrothermal treatment at 150 °C-300 °C, 5 h   | cell bioimaging   | 19       | 80%            |  |   |
| ascorbic acid   | hydrothermal treatment at 160 °C, 1 h  | detection of F <sup>-</sup>                                     | 74       | –              |  |   |
| glutathione in formamide  | solvothermal treatment at 160 °C, 4 h  | tumor treatment   | 75       | –              |  |   |
| citric acid with hexamine, dodecylamine, octadecylamine, aniline, benzylamine, and phenylethylamine | solvothermal treatment at 180 °C, 12 h   | light emitting diode (LED) production                           | 72       | 20%-39%        |  |   |
| p-phenyldinitrile and p-phenylenedicarboxylic acid  | solvothermal treatment at 120 °C-160 °C, 1 h-2 h   | light emitting diode (LED) production                           | 76       | 30%-46%        |  |   |
| acrylamide, N, N'-methylenebisacrylamide and KPS  | hydrothermal treatment at 200 °C, 8 h  | methanol fuel cell  | 37       | –              |  |   |
| pomelo peel   | hydrothermal treatment at 200 °C, 3 h  | detection of Hg <sup>2+</sup>                                   | 20       | 6.9%           |  |   |
| bamboo leaves   | hydrothermal treatment at 200 °C, 6 h  | detection of Cu <sup>2+</sup>                                   | 21       | 7.1%           |  |   |
| pyrene  | hydrothermal treatment at 200 °C, 10 h   | cell bioimaging   | 77       | 7%-45%         |  |   |
| Polyethylene glycol and saccharide  | microwave oven at 500 W for 2-10 min   | –   | 78       | 3.1%-6.3%      | efficiency; simple operation and equipment demand  | difficult to synthesize on a large scale                            |
| citric acid, L-cysteine and NaOH  | microwave oven at 750 W for 3 min  | light emitting diode (LED) production                           | 32       | 2.88%-54.68%   |  |   |
| neutral red and levofloxacin  | microwave oven at 800 W for 7 min  | cell bioimaging   | 10       | 22.83%         |  |   |

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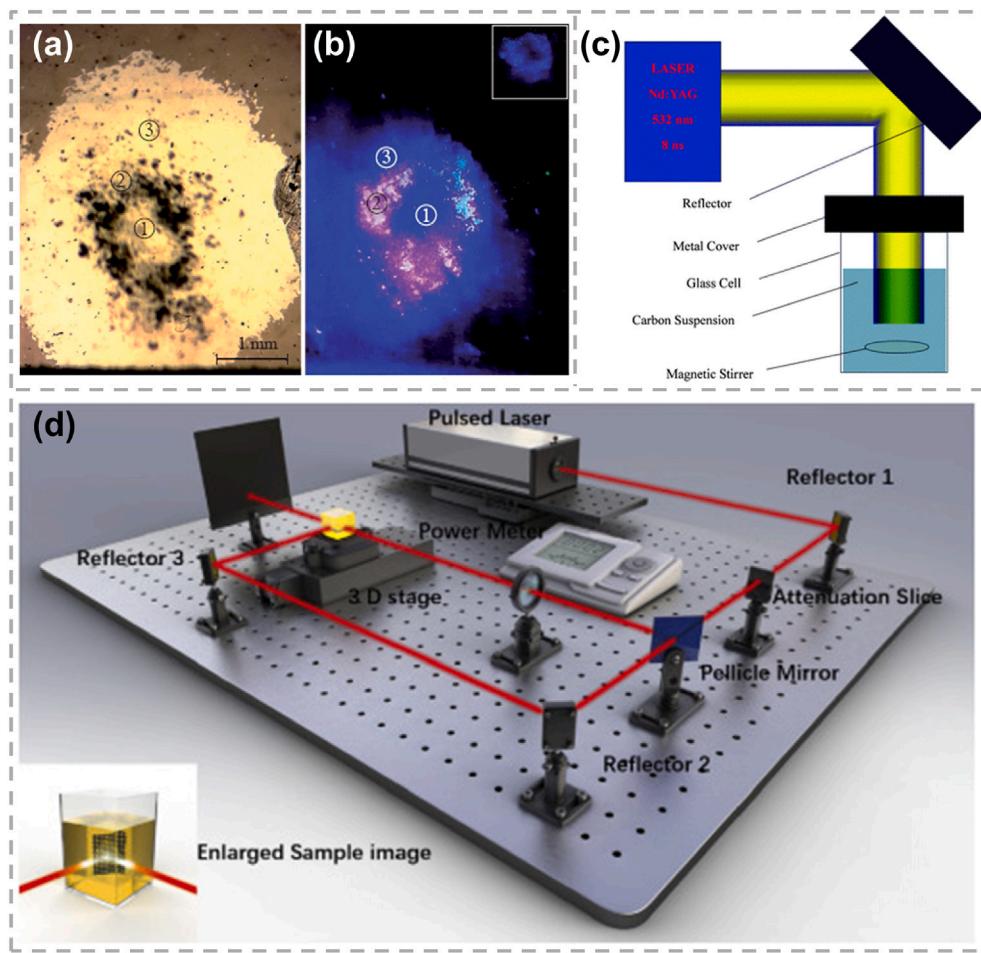
with laser at 900 °C and 75 kPa. The product was refluxed in HNO<sub>3</sub> for 12 h. Through the process of attaching simple organic substances such as PEG1500N (diamine-terminated oligomeric poly(ethylene glycol)) and poly(propionylethylene-imine-co-ethyleneimine) (PPEI-EI) to acid-treated carbon particles for surface passivation, CDs emitting bright light were obtained. This work firstly named this new type of fluorescent carbon nanoparticles CDs, it was also a further exploration of benign nanomaterials with similar optical properties to semiconductor quantum dots but lower toxicity. After this, nanosecond near-infrared pulse laser was utilized to ablate thin amorphous carbon film [49], it was observed that the formation of CDs exhibiting a graphite structure and nanodiamonds, both on the ablation zone (region 1) and the substrate surface around it (region 2). The study indicated that nanodiamonds were predominantly formed at the periphery of the composite plasma region adjacent to the laser torch (region 3), while CDs were formed directly within the ablation zone and its expansion region in the composite plasma, as shown in Fig. 2a-b. It can be seen that different areas of laser ablation will react to different degrees, resulting in different reaction products, which is not conducive to the synthesis of products with single composition. Nanocarbon materials were used as starting materials and simple solvents were used as liquid media to synthesize CDs [50]. Specifically, 0.02 g of carbon nanomaterials were dispersed in 50 mL of solvent. After ultrasonic treatment, 4 mL of suspension dropped into a glass chamber and was excited by a 523 nm wavelength laser unfocused irradiation, as shown in Fig. 2c. CDs can be obtained from the

supernatant. The synthesis method employed in this work is less complex, more convenient, and highly efficient compared to the early manufacturing methods, because the laser beam was not focused. However, the particle size of CDs synthesized by laser ablation is still not uniform. The laser ablation method has fast heating efficiency and can work continuously, while such concentrated high energy brings certain risks and uncontrollability to the operation process, which limits the large-scale application of this method.

The photoluminescence efficiency of CDs depends on their size distribution [52]. Using laser irradiation on a graphite sheet in a polymer solution, the team led by Hu successfully synthesized CDs with an average size of approximately 3, 8, and 13 nm. By adjusting the laser pulse width, it was discovered that the laser-induced condition in the bubble can be altered, resulting in variations in the nucleation and growth process and ultimately leading to different size distributions. This finding implies that a longer pulse width laser may be more effective for the production and size regulation of various nanostructures. CDs can also be produced by ablating low-cost carbon cloth with ultra-high speed and high efficiency dual-beam pulsed laser [51]. In this process, by using a beam splitter, a single laser beam was split into two beams, which reduced the laser ablation time and improved the ablation yield, as shown in Fig. 2d. The resulting CDs exhibited a higher level of uniformity compared to those generated through single pulse laser ablation, with an associated emission quantum yield of up to 35.4%. Further research on laser ablation technology reveals that

**Table 1 (continued)**

| Precursor  | Synthetic method  | Application  | Ref.      | Quantum yield | Advantages  | Disadvantages  |
|--|---|--|-----------|---------------|---|--|
| citric acid and urea   | microwave-assisted hydrothermal treatment                   | –  | 80,<br>81 | –             |   |  |
| urea and D-glucose   | microwave irradiation for 15 min                            | photocatalysis   | 28        | 14.9%         |   |  |
| resols   | template pyrolysis by functionalized silica colloid spheres | cell bioimaging  | 82        | 14.7%         | high yield; uniform particle size and excellent water dispersibility                                    | too complex preparation process                                |
| hexane or acetonitrile   | template pyrolysis by MgO                                   | –  | 83        | –             |   |  |
| acetone and NaOH   | aldol condensation polymerization                           | sodium ion batteries electrode material                  | 38        | –             | easy to large-scale synthesis; composition controllable   | difficult to control reaction rate                             |
| acetaldehyde and NaOH  | aldol condensation polymerization                           | sodium ion batteries electrode material                  | 39        | –             |   |  |
| carbamide or cysteine with acetaldehyde and NaOH                         | aldol condensation polymerization                           | potassium ion batteries electrode material               | 40        | –             |   |  |
| bis(trifluoromethane)sulfonimide lithium salt with acetaldehyde and NaOH | aldol condensation polymerization                           | polymer electrolytes                                     | 41        | –             |   |  |
| polyamide resin  | ultrasonic  | light emitting diode (LED) production                    | 67        | 28.3%         | uniform reaction and low pollution  | long reaction time and purification complexity                 |
| food waste   | ultrasonic  | biomedical imaging and plant seed germination and growth | 68        | 2.85%         |   |  |
| carbon black   | ball milling  | cell bioimaging  | 69        | 2.23%         |   |  |
| spent coffee grounds   | ball milling  | sensitive detection of Fe <sup>3+</sup>                  | 70        | 3.71%-7.82%   | simple and environmentally friendly   | low output and limited raw materials                           |
| citrate and urea   | electromagnetic induction heating method                    | nano-filler in nanofiber for wound healing               | 85        | 50%           | no obvious thermal inertia and high heating efficiency  | consume magnetic nanoparticles and reaction substrate limiting |
| citric acid and carbamide  | electromagnetic induction heating method                    | filler in polymer nanofiber membranes                    | 87        | 67%           |   |  |
| acrylamide   | plasma  | light emitting diode (LED) production                    | 90        | 6%            | in-situ functionalization   | strict equipment requirements                                  |
| ethylene   | plasma  | –  | 92        | 9.6%-13.5%    |   |  |
| carbon film  | plasma  | memory devices   | 93        | –             |   |  |
| egg  | plasma  | printing inks  | 45        | 6%-8%         |   |  |
| glucose and formamide  | microfluidic  | –  | 98        | 9%-37%        |   |  |
| ascorbic acid and dimethyl sulfoxide                                     | microfluidic  | pH detection and bioimaging                              | 99        | 2.6%          | fast heat and mass transfer; shortened characterization time and higher kinetic control reproducibility | complex equipment  |
| acid and different nitrogen sources                                      | microfluidic  | metal nanoprobes and bioimaging                          | 100       | 17%-77%       |   |  |



**Fig. 2.** Photographs of a) The area of the carbon film that has been exposed to radiation and b) luminescence produced by this area. Reproduced with permission [49], Copyright 2018, Institute of Physics Publishing; c) Schematic illustration of experimental setup for unfocused irradiation synthesis of CDs. Reproduced with permission [50], Copyright 2011, Royal Society of Chemistry; d) Schematic of the production process of CDs synthesized by double-beam pulsed laser ablation. Reproduced with permission [51], Copyright 2020, Elsevier.

reducing energy concentration can significantly improve the heterogeneity of products during laser ablation, as well as improve the synthesis efficiency of the target products.

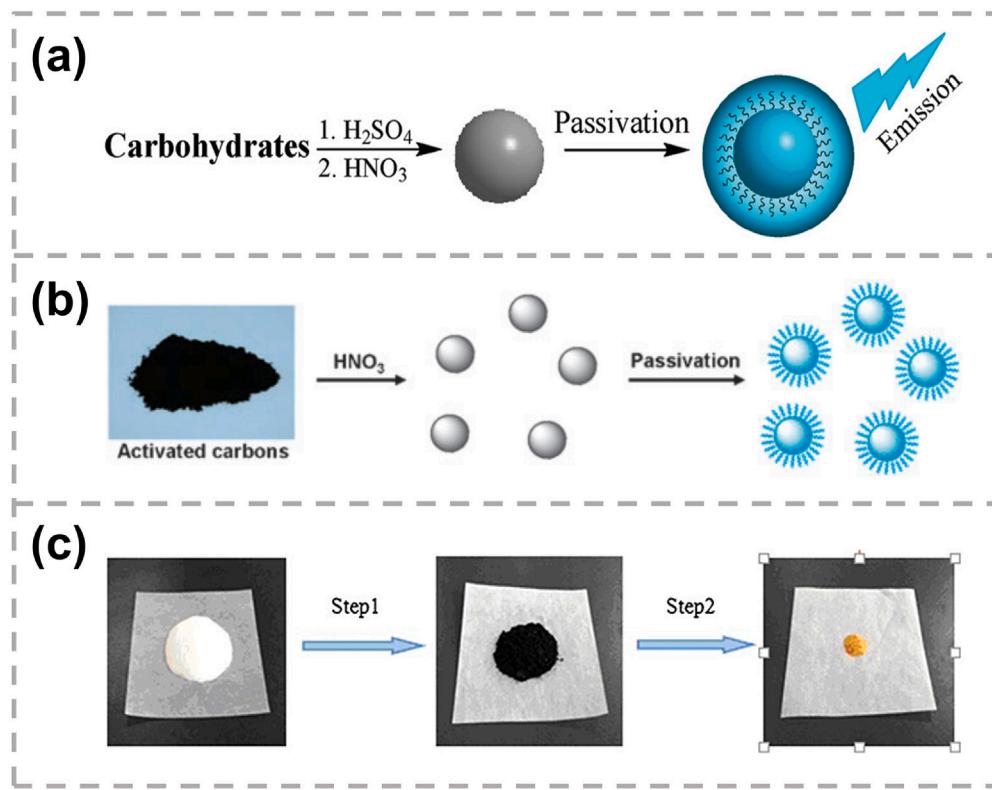
#### 2.1.3. Oxidation method

Oxidation method is a CDs synthesis strategy that oxidizes and decomposes large-sized carbon sources into small carbon nanomaterials. From the perspective of oxidant, it can be divided into chemical oxidation method and electrochemical oxidation method.

In chemical oxidation process, the use of strong oxidants can break the covalent bond in the carbon source to gradually split it into small molecules. The progress of the reaction is largely related to the oxidizability of the oxidant. The commonly used oxidants include sulfuric acid and nitric acid. Peng et al. reported a chemical oxidation method to synthesize CDs [53]. Sulfuric acid was used to eliminate the water of carbohydrate in the aqueous solution, following which the carbon material was decomposed with nitric acid to produce separate carbon nanoparticles. After treating the carbon nanoparticles with amine-terminated compounds, a vivid photoluminescence was detected, as shown in Fig. 3a. Their research results clearly showed that the application of nitric acid treatment not only disintegrated carbonaceous materials into separate carbon nanoparticles, but also resulted in distinct carbon nanoparticle compositions. Analysis of the elements indicated a rise in the oxygen content. Similarly, three commonly used activated carbon sources had been employed to easily prepare photoluminescent CDs [54]. After nitric acid treatment, the amorphous structure of

activated carbon was susceptible to etching into a single form of CDs, which were subsequently treated with an amine terminal compound for passivation, as illustrated in Fig. 3b. The resulting CDs obtained from three distinct sources of activated carbon exhibited excellent dispersity and comparable size. Their results further confirm that the size, structure, and properties of CDs synthesized by chemical oxidation are largely related to the oxidant and oxidation process, while the source of the reaction substrate has little effect on the properties of the final product. Starch is a cheap carbon source, which can also be used to synthesize water-dispersible CDs with excellent fluorescence characteristics by chemical oxidation [55]. Firstly, starch was carbonized under high pressure to produce carbon source. Afterwards, the product was distributed in a mixed oxidant solution consisting of distilled water, HAc, and H<sub>2</sub>O<sub>2</sub>, subjected to ultrasonic treatment and reflux, and subsequently filtered. After neutralizing the filtrate with NaOH and concentrating it, the solution was placed in a dialysis bag and dialyzed. The resulting solution was evaporated and the final product was obtained as a light yellow powder of CDs after vacuum drying, as shown in Fig. 3c. The oxidant not only plays a role in stripping the carbon source, but also endows CDs with oxygen-containing functional groups. This method is applicable to a wide range of carbon sources, and the operation process does not require complex and expensive equipment. However, excessive use of oxidants means the need for partial neutralizing agents, which increases the consumption of reaction media. The use of strong acids and bases is not environmentally friendly either.

Electrochemical oxidation is the process of using a strong oxidation

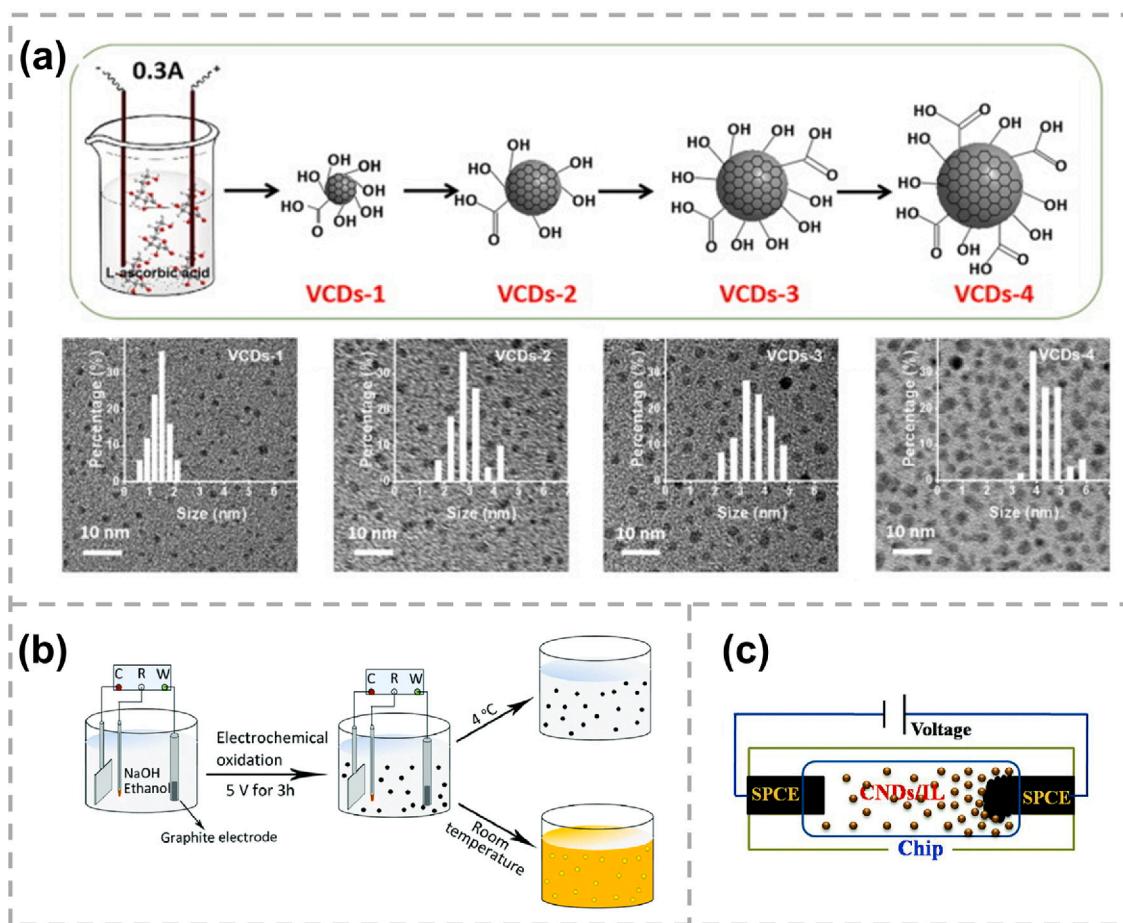


**Fig. 3.** A) Preparation procedure of luminescent carbogenic dots. Reproduced with permission [53], Copyright 2009, American Chemical Society; b) Preparation procedure of multicolor photoluminescent CDs. Reproduced with permission [54], Copyright 2010, Royal Society of Chemistry; c) Processing routing of water-dispersible CDs derived from starch. Reproduced with permission [55], Copyright 2015, Wiley-VCH.

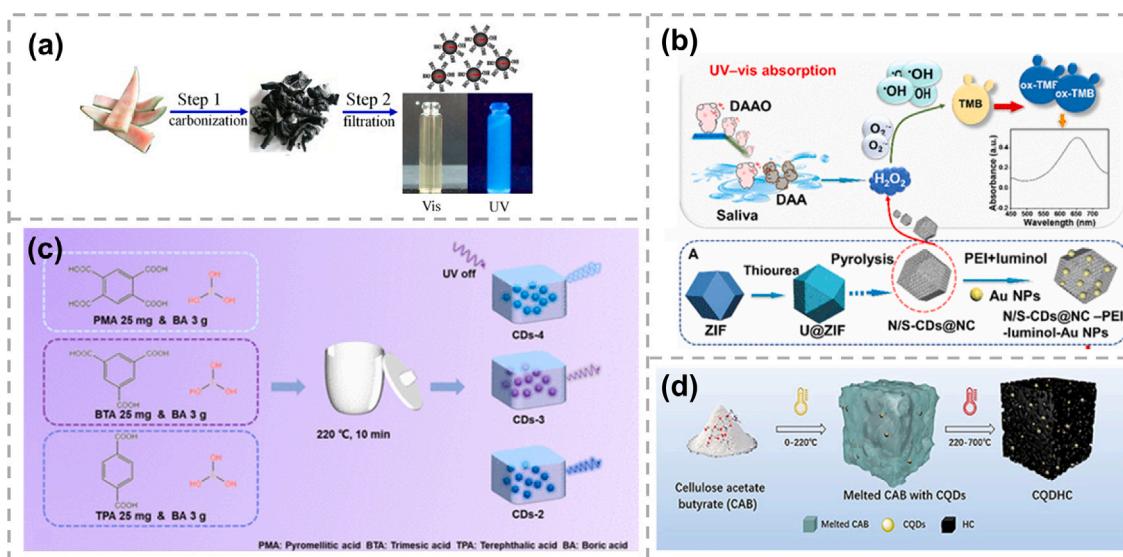
potential to cause the carbon source to undergo oxidative stripping on the electrode to form CDs, which is similar to the principle of arc discharge. Electrochemical synthesis of CDs was first reported in 2007 [47]. The working electrode utilized in this study was composed of multi-walled carbon nanotubes (MWCNTs) grown via chemical vapor deposition on carbon paper. To form the electrochemical cell, the working electrode was paired with a Pt wire counter electrode and an Ag/AgClO<sub>4</sub> reference electrode. A solution of degassed acetonitrile was utilized with the addition of 0.1 M tetrabutylammonium perchlorate (TBAP) as the supporting electrolyte to synthesize CDs via electrochemical synthesis. Applying a potential with a sweep rate of 0.5 V/s in the range of -2.0 V to +2.0 V resulted in a noticeable change in the solution's appearance, transitioning from colourless to yellow and eventually reaching a dark brown. The author predicted that TBA cation is expected to create gaps during the electrochemical cycle and cause damage to the tube structure in the vicinity of these defects. This would lead to the detachment of carbon nanoparticles, which would then enter the electrolyte solution. This study points out the role of solute molecules in electrochemical oxidation, which can strip carbon sources. Graphite electrode can also serve as working electrode, paired with basic alcohol as electrolyte to prepare CDs [56]. The whole process was realized by electrochemical oxidation stripping graphite under 5 V voltage. During the experiment, it was observed that the dispersion of CDs underwent a colour change from colourless to bright yellow upon exposure to air at room temperature, as shown in Fig. 4b. Spectral analysis showed that the colour change resulted from the oxidation of CDs. Finally, the obtained CDs were applied for the sensitive detection of iron ions (Fe<sup>3+</sup>) and effective cell imaging. Small molecule substances can also be converted into CDs through electrochemical synthesis. Du's team used L-ascorbic acid as precursor to prepare CDs by one-step electrochemical oxidation [57]. The working system consists of platinum wire electrodes and L-ascorbic acid solution. Samples were collected on different days, and the product was obtained by dialysis.

Finally, four different sizes of CDs dispersions were obtained, as shown in Fig. 4a. Their results suggested that -OH was continuously oxidized to -COOH in the process of CDs electrolysis, and the average size of CDs increased with the reaction time and the oxidation degree gradually increased with the increase of particle size. In the electrochemical oxidation synthesis of small molecules, functional groups gradually transform into oxygen-containing functional groups with higher oxidation states. Contrary to the stripping process of large molecules, small molecules exhibit aggregation behaviour during the formation of CDs, gradually increasing in size. Therefore, the size and degree of oxidation of CDs can be adjusted by controlling reaction time and electrical parameters.

The technology known as "Lab on chip" is a novel approach that enables conventional laboratory tasks to be carried out using small chip-based devices [58]. Xu and co-workers developed an electrochemical strategy based on a one-step simple chip to directly generate CDs from screen-printed carbon electrodes (SPCEs). Ionic liquids (IL) aqueous solution was applied as electrolyte, as shown in Fig. 4c. It was discovered that the obtained CDs possessed favourable water dispersion and exceptional luminescent property. In approximately 30 min, the synthesis process can be completed until there were no further changes observed in the colour of the solution or the electric current on the DC power supply. The results showed that the duration of the synthesis and the initial current were reliant on the IL concentration in the electrolyte solution. A reduced reaction system is conducive to rapid mass transfer as well as reducing the consume of raw materials, making it easy to explore the effects of various reaction conditions on the synthesis process. However, this synthesis process is limited to the research stage. Considering the uncertainty of the amplification process, it is inappropriate for large-scale synthesis. The process of electrochemical oxidation synthesis has the advantages of ease operation, high yield, and controllable conditions. However, this method requires specialized equipment and has limitations in terms of suitable precursors.



**Fig. 4.** A) Scheme of one-step electrochemical oxidation used to create VCDs-1, VCDs-2, VCDs-3, and VCDs-4 and the particle size distributions. Reproduced with permission [57], Copyright 2023, Elsevier; b) Diagram for the generation of CDs through electrochemical oxidation as well as the colour change of the dispersion upon storage at room temperature. Reproduced with permission [56], Copyright 2016, Royal Society of Chemistry; c) Schematic diagram for the direct generation of CDs on the SPCE-based chip through electrochemical oxidation. Reproduced with permission [58], Copyright 2015, Royal Society of Chemistry.



**Fig. 5.** A) The process of creating water-dispersible fluorescent CDs using watermelon peel. Reproduced with permission [60], Copyright 2012, Elsevier; b) Illustration of the preparation of N/S-CDs@NC. Reproduced with permission [63], Copyright 2022, American Chemical Society; c) Schematic diagram of the manufacturing process of blue-to-violet RTP CDs composites. Reproduced with permission [64], Copyright 2023, American Chemical Society; d) Schematic of the synthesis of CQDHC. Reproduced with permission [65], Copyright 2022, Elsevier.

#### 2.1.4. Pyrolysis

Pyrolysis is a commonly employed technique for the synthesis of CDs. During this process, the organic matter present in the carbon source is subjected to high temperatures in a vacuum or inert atmosphere, leading to gradual conversion into CDs through heating, dehydration, degradation, and carbonization. To decompose the carbon precursors into carbon nanoparticles, high-concentration acids or bases are often utilized [59]. As a novel carbon resource, watermelon peels were chosen to produce fluorescent CDs of high quality by means of low-temperature carbonization and simple filtration [60], as shown in Fig. 5a. This method facilitated the production of CDs on a large scale, without requiring any subsequent post-processing steps. The prepared CDs had small particle size, strong blue luminescence, satisfactory fluorescence lifetime, excellent stability across a broad pH range and high salt concentration. It had been applied to cell imaging successfully. Biomass and its waste are natural organic carbon sources, which is very cheap and convenient for pyrolysis to synthesize CDs. The process of carbonization is endothermic, with temperature playing a critical role. Lower temperatures during pyrolysis may impede the full carbonization of the carbon source into CDs. However, high temperatures will lead to the over-oxidation of carbon source, and the surface structure of the CDs may become damaged, leading to a decline in their optical properties. The effect of reaction time on pyrolysis is very similar to reaction temperature [61]. Inadequate carbonization of the carbon source can result from a short reaction time, whereas a prolonged reaction time may lead to excessive carbonization and damage to the surface structure of the CDs. It is worth noting that the influence of reaction time on the optical characteristics of CDs is intertwined with the reaction temperature. Achieving optimal reaction time is only relevant if the reaction is performed at an appropriate temperature. If the reaction temperature is inadequate, even an extended reaction time would not yield any useful final products [62].

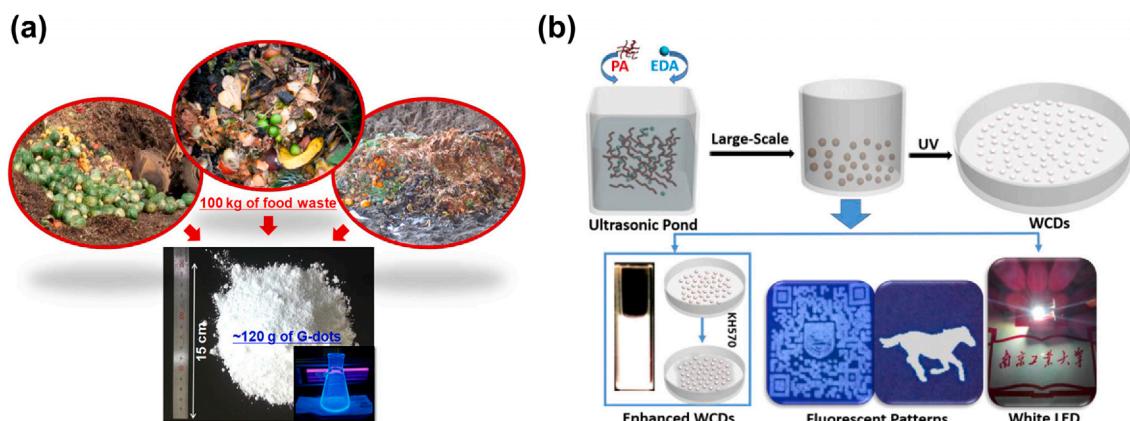
The synthesis of CDs by pyrolysis is not limited to macromolecular bulk carbon sources. In recent years, many researches adopted small molecule pyrolysis to synthesize functionalized CDs composites. Tang's team synthesized a zinc-based zeolite imidazole skeleton (ZIF-MOF) and thiourea was employed to infiltrate the ZIF cavity [63]. N and S-doped N-rich CDs porous carbon nanoenzyme (N/S-CDs@NC) was obtained through simple two-step pyrolysis, as shown in Fig. 5b. In this work, nanomaterial N/S-CDs@NC showed excellent peroxidase-like activity in acid acetate buffer. Combined with magnetic separation and immune recognition, the electrochemiluminescence detection of carcinoembryonic antigen in serum was achieved with high sensitivity and selectivity. The controllability of functional group positions is a major advantage in synthesizing CDs by small molecule precursors. By mixing boric acid with three kinds of aromatic acids and pyrolyzed at a consistent temperature, composites of blue-violet room temperature phosphorescent (RTP) CDs with high colour stability can be produced [64], as shown in Fig. 5c. The phosphorescent colour of the CDs composite can be modified from blue to purple by manipulating the distribution of functional groups in the precursor structure. In addition, CDs can be compounded with hard carbon to obtain CDs/hard carbon composite (CQDHC) by carbonizing cellulose acetate butyrate (CAB) at 700 °C [65]. Without the need for complicated synthesis, purification methods, or physical blending processes, CDs were spontaneously generated and self-dispersed within a hard carbon matrix, resulting in a typical nano-porous structure with an oxygen content of 8.23 at. %, as shown in Fig. 5d. This method employed cheap raw materials and simple process, the structure and properties of CDs were maintained, while also effectively facilitating the distribution of CDs within a hard carbon matrix. The resulting CQDHC demonstrated favourable electrochemical performance when served as an anode for potassium-ion batteries. Pyrolysis method is simple to operate and there are various means of generating heat, such as microwave assisted pyrolysis mentioned in the following chapter, which makes it easy for the pyrolysis method to be widely applied.

#### 2.1.5. Ultrasonic method

It had been indicated in the study of acoustic cavitation that employing sound with sufficient pressure to irradiate the liquid will lead to the formation and oscillation of bubbles, during which extreme conditions (temperature up to 20000 K; pressure of thousands bar) will occur [66]. In this process, sound is a unique source of energy to drive chemical reactions and produce products that are usually unavailable by other standard methods. Furthermore, ultrasound can facilitate the dispersion of reaction products around the substrate, preventing their aggregation. A straightforward technique was created by Park et al. to produce water-dispersible green carbon nanodots (G-dots) on a large scale utilizing various sources of food waste [67]. They treated food waste and ethanol solution with 40 kHz ultrasound, and then centrifuged the solution. G-dots were obtained after filtering and drying at 45 °C, as shown in Fig. 6a. During ultrasonic irradiation, dehydration, polymerization and carbonization occurred in turn, leading to a brief, singular nucleation. Then, the solutes diffused towards the side of carbon nanoparticles and grew along the generated nucleus. It is possible to synthesize approximately 120 g of G-dots with 100 kg of food waste and exhibit promising capabilities for use in biomedical imaging as well as the germination and growth of plant seeds. Similarly, one-step ultrasonic method was a cost-effective and uncomplicated approach to produce white fluorescent CD (WCD) [68]. Polyamide resin and ethylenediamine were put into deionized water along with ultrasonic treatment. Besides, the addition of silane coupling agent KH570 was made to the reaction system mentioned above as a co-passivator to enhance the quantum yield of WCD, as shown in Fig. 6b. WCD exhibited a quantum yield (QY) of approximately 28.3% and was successfully applied to produce white LEDs (WLEDs) without additional phosphors. It was also used as ink to obtain various fluorescent patterns. Unlike traditional thermally driven synthesis, ultrasound can instantly generate the heat required for the reaction locally, which is quickly absorbed by the solvent after completing the reaction. Therefore, there will be no significant temperature changes at the macro level, avoiding the use of high-temperature and high-pressure equipment.

#### 2.1.6. Ball milling

Ball milling is a highly straightforward and environmentally conscious approach. Without the introduction of harsh reagents, it utilizes mechanical energy to fracture chemical bonds and particles. Youh and co-workers developed a dry ball milling method for producing CDs using carbon black and sodium carbonate, which are environmentally friendly and cost-effective [69]. The small particle size and well-formed graphite crystallinity of conductive carbon black make it a favourable precursor to produce CDs. Na<sub>2</sub>CO<sub>3</sub> was used during ball milling to elevate the content of oxygen functional groups present on the surface of CDs, which increased the hydrophilicity of CDs. The CDs present in the mixture was dispersed with deionized water, followed by centrifuging the dispersion and filtering the supernatant. The filtrate was dialyzed to remove ions and freeze-dried to obtain CDs. The prepared CDs showed blue photoluminescence, hydrophilicity and low toxicity, with a quantum yield of 2.23%, and it was a promising material for biological imaging applications. In this method, the utilization of sodium carbonate was pivotal in enhancing both the milling efficacy and functionalization of CDs. In the process of ball milling, a substantial amount of energy was imparted to the target material within the impact area through the collision with the ball, resulting in both the functionalization and decomposition of the carbon black material. While the fine particles within the grinding area experienced frictional forces. Na<sub>2</sub>CO<sub>3</sub> acted as the milling medium and filler, improving the milling efficiency. For the formation of CDs, it is necessary for the supplementary milling medium to be mechanically harder than the precursor and softer than the primary milling ball. Spent coffee grounds (SCGs) are easy to obtain and can be used as carbon source for the synthesis of carbon dots (CFCD) through wet ball milling technology under mild conditions [70]. The addition of small molecules containing oxygen and nitrogen was



**Fig. 6.** a) Diagrammatic depiction of the process involved in the extensive production of G-dots. Reproduced with permission [67], Copyright 2014, American Chemical Society; b) Polyamide resin based WCD synthesis for utilization in WLEDs and fluorescent patterns. Reproduced with permission [68], Copyright 2016, American Chemical Society.

investigated to produce CDs functionalized by carboxylic acid (CA-CFCD) and nitrogen-doping CDs (N-CFCD), as shown in Fig. 7. Compared with unmodified CFCD, CA-CFCD and N-CFCD showed excellent dispersion stability in hydrophilic solvents. Specifically, SCGs and ethanol were introduced into PET plastic containers and subjected to milling at room temperature with a speed of 150 rpm. The resultant dark solution was centrifuged to eliminate any accrued particles, and then dialyzed in deionized water. Finally, pure CFCD was obtained after drying. The ability of micro gel matrix encapsulated by CA-CFCD to detect  $\text{Fe}^{3+}$  ions via fluorescence variation within a matter of seconds confirmed its prospective practical use as a dependable fluorescent probe for metal ion detection. The ball milling method boasts a considerable processing capacity and relatively simple preparation procedures, but the availability of raw materials appropriate for synthesizing CDs using this approach is exceptionally scarce.

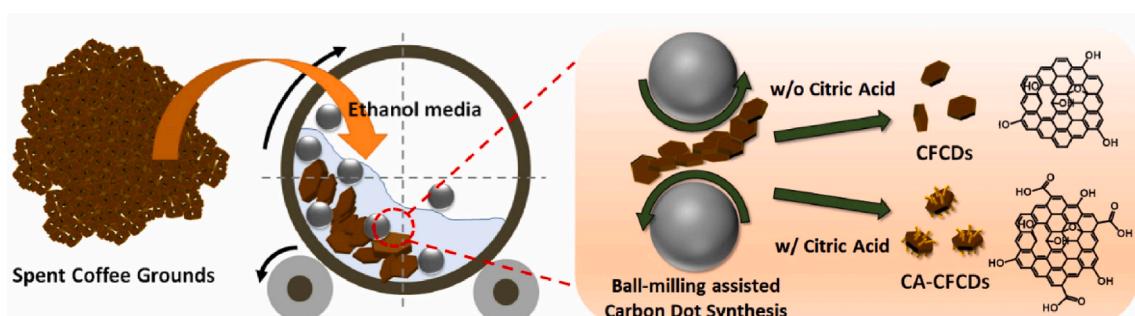
## 2.2. “bottom-up” strategies

### 2.2.1. Hydrothermal/Solvothermal synthesis

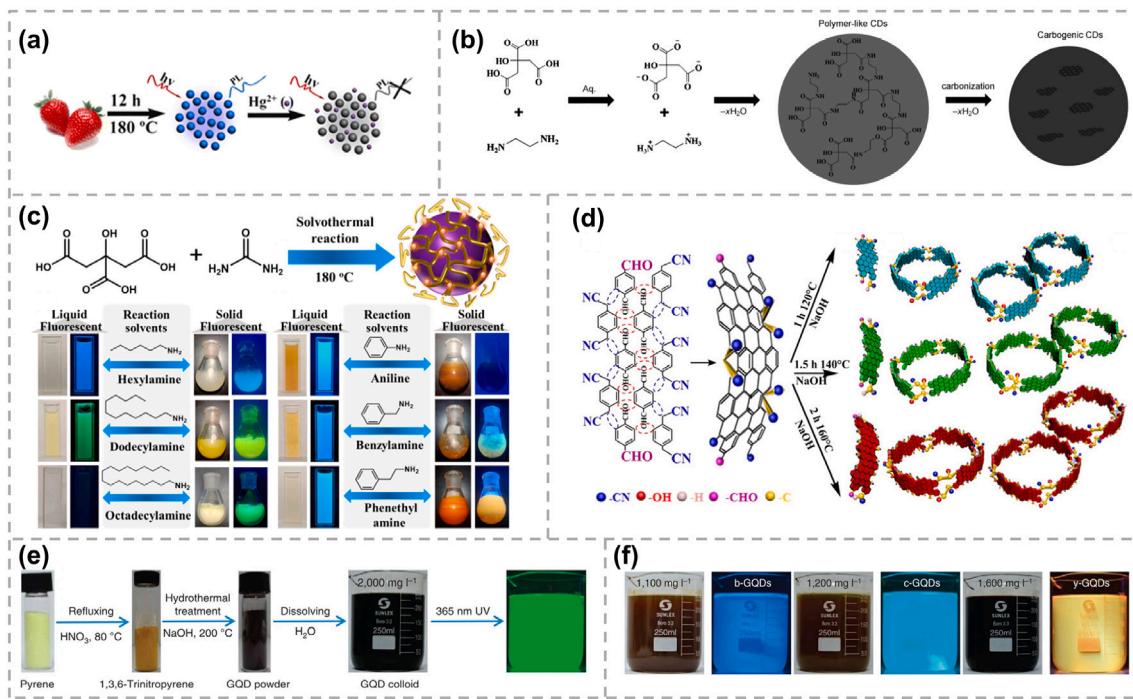
Hydrothermal/solvothermal method refers to a synthesis method in which the reaction mixture reacts with water or organic matter and non-aqueous solvent under a specific temperature and autogenous pressure of the solution in a sealed system, such as an autoclave. They are distinguished by the type of solvent used [71]. Hydrothermal/solvothermal synthesis is a widely used strategy, which is straightforward, secure, and effective. In addition, the raw materials used for hydrothermal reaction are widespread, such as citric acid [19,72], ascorbic acid [73,74], strawberry juice [22], etc. Jia and co-workers proposed a novel approach to easily prepare CDs in one pot by heating with low temperature water [73]. Typically, ascorbic acid solution and  $\text{Cu}(\text{Ac})_2\text{H}_2\text{O}$  was stirred at room temperature and then kept at 90 °C. A

transformation was observed in the reaction mixture as it transitioned from an orange suspension to a clear, golden solution, signifying the generation of CDs. During the synthesis, a large amount of CDs dispersion was obtained after centrifugation. This method is easy to expand. In addition, it is economic and green production. More importantly, it possesses abundant production and effortless post-treatment process. Similarly, fluorescent N-doped carbon nanoparticles (FNCP) containing 6.88% nitrogen can be synthesized through a pot of hydrothermal treatment of strawberry juice [22]. Commonly, the procedure involved transferring strawberry juice to a Teflon-lined autoclave and subjecting it to a temperature of 180 °C for 12 h. The 0.22  $\mu\text{m}$  filter membrane was utilized to filter out the large particles, which were then subjected to centrifugation. Finally, the products were obtained after drying under vacuum for 72 h, as shown in Fig. 8a. The FNCP that was prepared exhibited the highest emission at 427 nm and had a quantum yield of 6.3%. FNCP can be selectively quenched by  $\text{Hg}^{2+}$ . This unique characteristic was utilized to create a fluorescence-based detection method for  $\text{Hg}^{2+}$ .

Most strategies for “bottom-up” synthesis of CDs by solvothermal method have similar steps. The most common method is to load the reaction substrate into a polytetrafluoroethylene lined high-pressure reactor, heat it to high temperature to drive the reaction proceed. Intermediate products containing CDs are obtained after cooling the reactor. Finally, purified CDs can be obtained through subsequent purification and separation. The formation of CDs from small molecular substrates is often accompanied by the interaction between groups. Under high temperature, carbon chain skeleton continuously lengthens with the cross-linking process, and finally forms CDs with different structures. As a widely known method [19], CDs can be synthesized by mixing citric acid and ethylenediamine in deionized water and moving the solution to an autoclave lined with polytetrafluoroethylene for



**Fig. 7.** Schematic illustration of preparation procedure for the spent coffee ground-based CDs. Reproduced with permission [70], Copyright 2023, Elsevier.



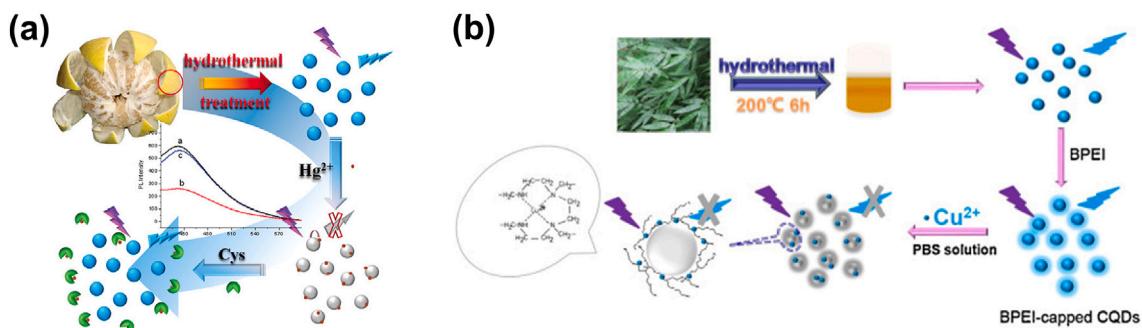
**Fig. 8.** a) Illustration of the synthesis process for FNCP derived from strawberry juice and the application for  $\text{Hg}^{2+}$  detection. Reproduced with permission [22], Copyright 2013, Royal Society of Chemistry; b) The route and formation mechanism of CDs synthesis based on citric acid and ethylenediamine. Reproduced with permission [19], Copyright 2013, Wiley-VCH; c) Schematic diagram of solid fluorescent CDs preparation and images of the obtained CDs both in dispersion (left) and the solid state (right). Reproduced with permission [72], Copyright 2022, American Chemical Society; d) Schematic illustration of the synthesis process for R/G/B-SBF-QRs. Reproduced with permission [76], Copyright 2021, Wiley-VCH; e, f) Synthetic procedure and images of the obtained GQDs dispersed in solution. Reproduced with permission [77], Copyright 2014, Springer Nature.

heating and reaction. After cooling to room temperature, CDs were obtained through dialyzing the brown-black transparent product, as shown in Fig. 8b. When ascorbic acid is used as the precursor [74], water-dispersible green fluorescent CDs can be produced via one-step solvothermal strategy. Similarly, ascorbic acid was dissolved by ultrasonic treatment with water and ethanol, and the hydrothermal process was conducted for 1 h at 160 °C. The final products were obtained by centrifuging the light brown solution to eliminate large particles and subsequently dialyzing it to remove any unreacted precursors. Su and co-workers synthesized CDs through solvothermal method by dispersing glutathione in formamide and treating them at 160 °C for 4 h [75]. They used silica gel column chromatography for purification, which was different from the previous purification process.

The widely used solvothermal synthesis has good controllability. With the variety of reaction substrate, reaction temperature and reaction time, the properties of the products will change greatly. For example, researchers had utilized citric acid as precursor and six alkyl amines as solvent to prepare multicolour solid fluorescent CDs by one-step solvothermal method [72], as shown in Fig. 8c. Their results showed that the interaction between solid CDs was significantly influenced by alkyl chain type of amine solvent, thus affecting the fluorescence properties and quantum efficiency of CDs. Meng and colleagues have announced the successful synthesis of solid-state red/green/blue bandgap fluorescent carbon quantum rings (R/G/B-SBF-QRs) at a gram-scale. The products exhibited impressive quantum yields ranging from 30% to 46% [76]. The reaction substrate was solvothermal treated at 120–160 °C at different reaction time using p-phenyldinitrile containing cyano and p-phenylenediacrylic acid containing aldehyde as precursors, as shown in Fig. 8d. Then it was purified by simple solvent washing procedure. During the reaction, bent carbon quantum bands with different lengths were connected to form carbon quantum rings with different diameters. In another one-step solvothermal addition polymerization and carbonization strategy [37], acrylamide,  $\text{N}_3^-\text{N}^+$ ,

methylenebisacrylamide and potassium persulfate (KPS) were added to deionized water. The substrate was heated at 200 °C for 8 h and cooled to room temperature. The products were filtered through the filter membrane to remove large impurities, and then dialyzed with deionized water in the dialysis bag to eliminate small molecules. Finally, solid CDs were obtained from the solution by freeze-drying. The carboxyl functionalized CDs can be obtained by replacing acrylamide in the reaction substrate with acrylic acid. Pyrene can be employed as a precursor for the large-scale production of high fluorescence water-dispersible GQDs [77]. Firstly, the solid powder was nitrated into 1,3,6-trinitropyrene by refluxing and stirring in hot  $\text{HNO}_3$  at 80 °C for 12 h, followed by ultrasonic dispersion in NaOH solution for 2 h. After a hydrothermal reaction at 200 °C for 10 h and cooling to room temperature, the product was filtered through a 0.22 µm microporous membrane to remove non disperseable carbon products, and further dialyzed in a dialysis bag to remove sodium salts and unfrozen small molecules. GQDs were obtained after drying. Replacing NaOH solution with different concentrations of ammonia or hydrazine hydrate will lead to amino functionalized GQDs, as shown in Fig. 8e-f. The introduction of these alkaline substances enables the exploration of large-scale production of water-dispersible GQDs. The reaction temperature and time have a critical impact on the chain growth, nucleation, and crosslinking of CDs, and the selection of different precursors is an effective means of functionalization of CDs. Accurately grasping the formation mechanism of CDs will help us to use reaction parameters reasonably to synthesize the desired products.

It is worth mentioning that the substrate of solvothermal method is not limited to small molecules, biomass macromolecules can be taken into consideration as well. Pomelo peel, a low-cost waste, can act as precursor to synthesize water-dispersible fluorescent carbon nanoparticles by hydrothermal method [20]. With a quantum yield of about 6.9%, it had been applied for  $\text{Hg}^{2+}$  detection, showing excellent sensitivity and selectivity, as shown in Fig. 9a. Liu and co-workers successfully synthesized CDs with high quantum yield by hydrothermal method

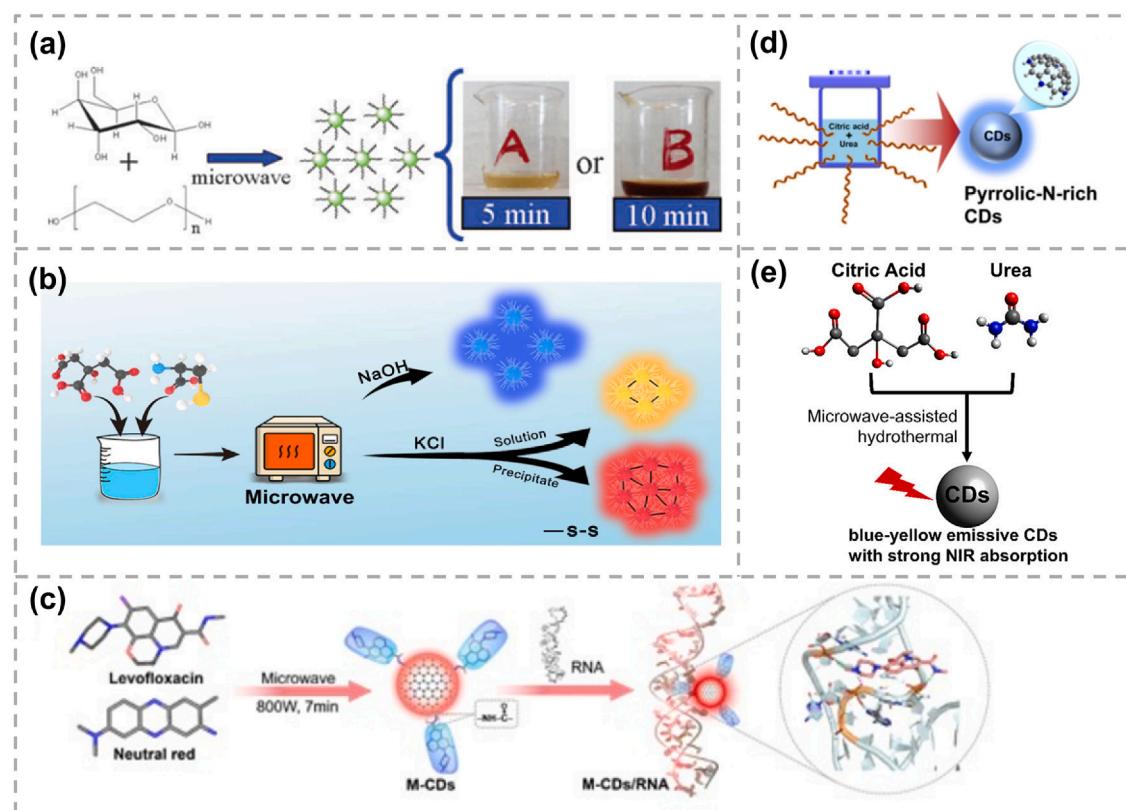


**Fig. 9.** A) The preparation and application of CDs from pomelo peel. Reproduced with permission [20], Copyright 2012, American Chemical Society; b) The synthesis and application of CDs from bamboo leaves. Reproduced with permission [21], Copyright 2014, Elsevier.

utilizing bamboo leaves as carbon source for the first time [21]. Capped CDs were prepared by electrostatic adsorption with branched polyethylene imine (BPEI) coated CDs, which were applied as a fluorescent probe for sensitive and selective Cu<sup>2+</sup> detection, as shown in Fig. 9b. The process of extracting CDs from biomass is similar to cooking. Under high temperature and high pressure, some macromolecules in biomass are broken and dissolved into solution. The substrate solution containing CDs is like a “CDs soup”. Due to the diversity of biomass, the natural CDs extracted by carbon source like this also have abundant properties. Biomass has a wide range of sources and plenty raw materials, which is easy to realize large-scale synthesis. However, synthesis of CDs in this way is somehow blind. It is a process of constant attempts, which is not conducive to the directional synthesis of functional CDs.

### 2.2.2. Microwave synthesis

Microwave method is a commonly employed approach for the direct carbonization of organic matter into CDs under microwave radiation. Owing to its efficiency, simple operation and equipment demand, microwave method is an effective technique with significant potential for the large-scale production of fluorescent CDs. An economical and simple microwave method can synthesize fluorescent CDs in a few minutes [78]. First, the solution containing different amounts of polyethylene glycol and sugar was heated in a 500 W microwave oven for 2–10 min. As time goes by, the solution transferred from colourless to yellow and finally to dark brown, which indicated the CDs had formed, as shown in Fig. 10a. Correspondingly, a synthesis strategy for the rapid preparation of solid-state fluorescent multi-colour CDs with different emission colours by simple microwave method from the same precursors was



**Fig. 10.** A) Microwave approach to CDs derived from polyethylene glycol and sugar. reproduced with permission [78], Copyright 2009, Royal Society of Chemistry; b) Synthesis of the solid-state fluorescent multi-colour CDs. Reproduced with permission [32], Copyright 2023, Wiley-VCH; c) Schematic illustration of the synthesis route for M-CDs and their structure as well as RNA binding behaviors. Reproduced with permission [10], Copyright 2023, Wiley-VCH; d, e) Illustration of the preparation process for pyrrolic-N-rich CDs by microwave-assisted hydrothermal method. Reproduced with permission [80,81], Copyright 2018, American Chemical Society. Copyright 2023, Springer Nature.

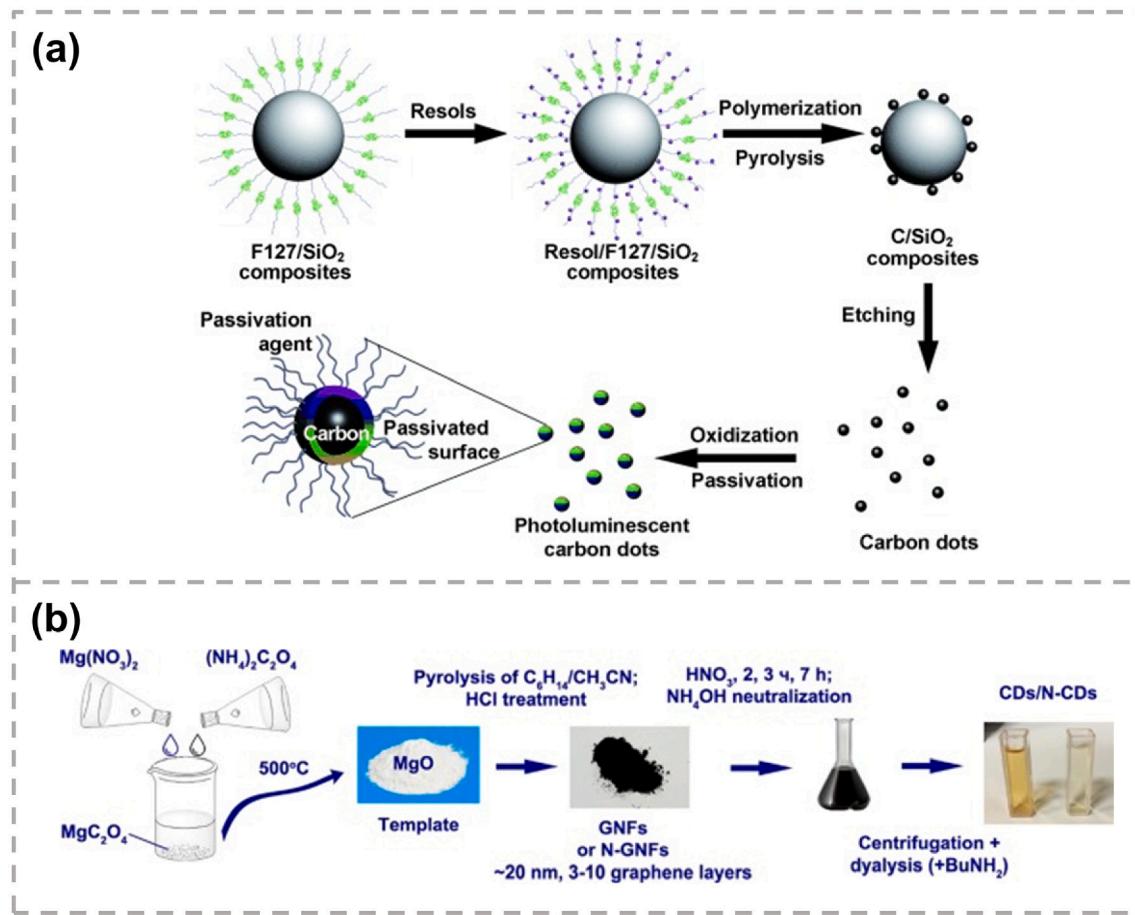
reported [32]. The mixture solution including citric acid, L-cysteine and NaOH was heated in a domestic microwave oven (750 W) at medium and high temperature for about 3 min until a light yellow dispersion was obtained. After cooling to room temperature, the product was dispersed in deionized water and dialyzed for 8 h. Finally, CDs in powder form were obtained by freeze-drying, as shown in Fig. 10b. In the field of biological applications, a new RNA targeted red emission CDs (M-CDs) was designed and synthesized by microwave method [10]. Neutral red and levofloxacin were selected as precursors. In a typical synthesis process, the precursors were heated in a microwave oven at 800 W power for 7 min, and then the purification procedure involving both filtration and dialysis was executed, as shown in Fig. 10c. The M-CDs were obtained with a yield of  $22 \pm 0.6\%$ . It is noteworthy that such one-step microwave synthesis technology is remarkable in its ability to address several issues commonly encountered in traditional hydrothermal synthesis of nanomaterials, such as uneven heating, prolonged processing times, and low product yields [79].

Microwave method is often employed in combination with other synthesis strategies, which is classified as microwave assisted method. Iskandar's team developed a pyrrole-N-rich CDs with absorption peak in the first near-infrared (NIR) window area by one-step microwave-assisted hydrothermal synthesis [80,81]. They dissolved citric acid and urea in ultra-pure water. The precursor solution was moved to a glass bottle and subjected to microwave-assisted hydrothermal treatment at different reaction time and temperature. The product was obtained after purifying the synthesized solution through the regenerated cellulose filter, as shown in Fig. 10d-e. N-doped CDs can be prepared by microwave assisted pyrolysis as well [28]. In typical synthesis, double distilled water was used to uniformly mix urea and D-glucose. The resulting

solution was then subjected to microwave irradiation for 15 min in a domestic microwave oven after clarification. The furnace treatment resulted in the evaporation of all water, leaving behind white precipitates that were subsequently carbonized to produce the final product of N-doped CDs. Different from the methods described before, in the microwave-assisted method, the microwave generates energy to make the system reach the required temperature instead of directly acting on the reaction substrate to initiate the reaction. Compared with traditional heating equipment, the reaction system heated by microwave is faster and more uniform, which has a crucial impact on the purity of synthetic products.

### 2.2.3. Template method

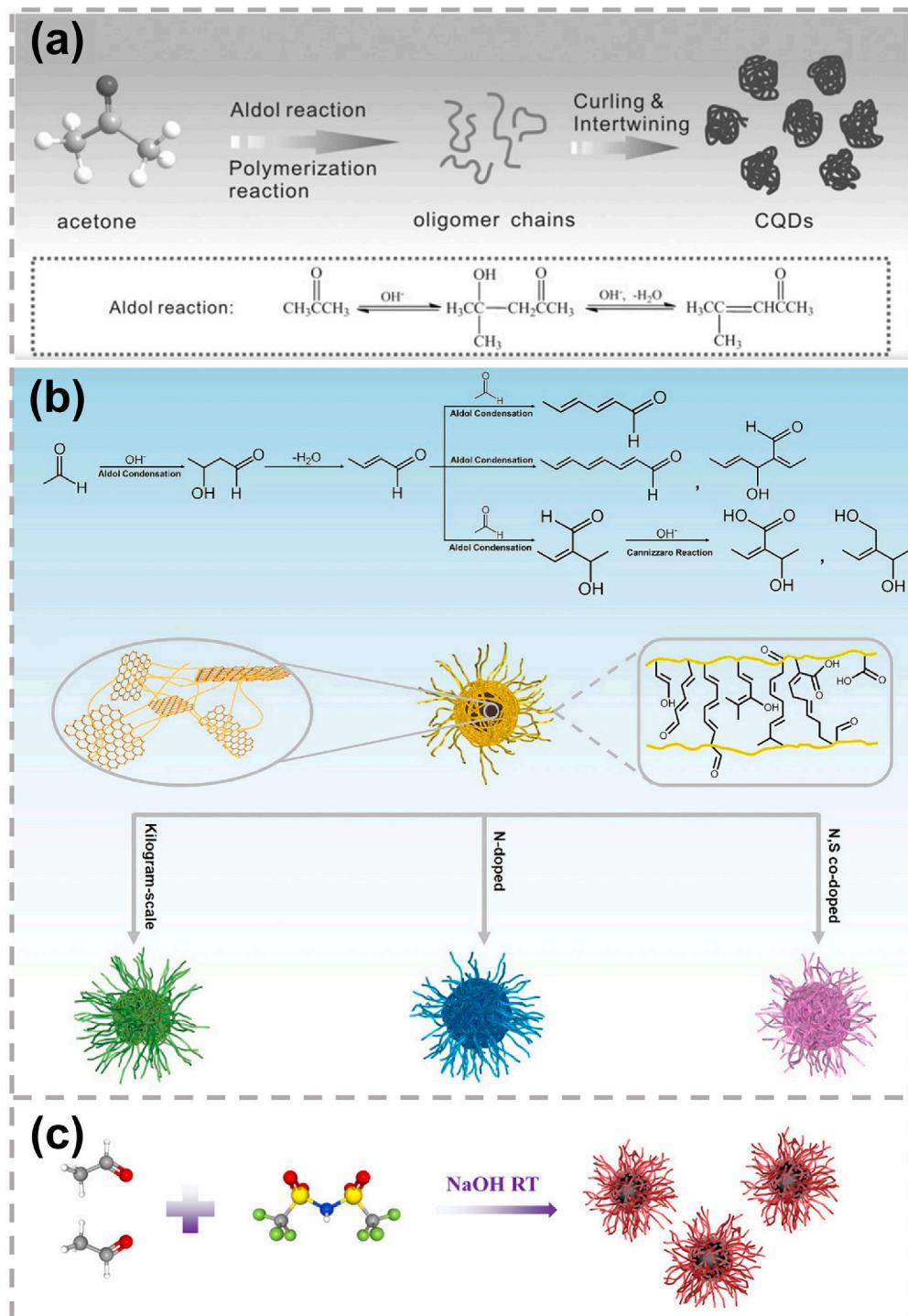
The template method generally takes a specific support material as the template. CDs are first synthesized on this material and then collected after removing the template by acid etching or other methods. CDs with an amorphous structure were synthesized using surfactant-modified silica spheres as carriers in a direct and novel synthesis route reported by Liu et al. [82]. Firstly, the satellite-like polymer/F127/silica composite was synthesized by utilizing amphiphilic triblock copolymer F127 functionalized silica colloidal sphere as carrier and phenolic resin as carbon precursor through aqueous route. Subsequent thermal processing and elimination of the silica carrier resulted in the formation of nano-sized CDs. Water-dispersible polychromatic photoluminescent CDs were ultimately obtained by conducting a treatment with acid and surface modification using a simple passivation method, as shown in Fig. 11a. The key of this method was to apply the silica nanospheres modified by surfactants as the carrier, which not only provided an anchor point for the polymerization of resols in solution, but also hindered



**Fig. 11.** A) The process for synthesizing multicolour photoluminescent CDs by template method. Reproduced with permission [82], Copyright 2009, Wiley-VCH; b) Scheme of CDs synthesized by MgO template pyrolysis. Reproduced with permission [83], Copyright 2020, Elsevier.

the clumping of CDs during the pyrolysis reaction. By utilizing this approach, the production of CDs exceeded 10%, the particle size was relatively uniform and the water dispersibility was satisfying, but the preparation process was complex. Template pyrolysis can also combine “bottom-up” and “top-down” methods to synthesize CDs [83]. Firstly, the MgO template was produced through a process involving extended precipitation of magnesium oxalate, followed by calcination at a temperature of 500 °C [84]. The template was positioned within a quartz tube bed reactor and exposed to a flow of either hexane or acetonitrile in

the presence of nitrogen gas for 15 min, while the reactor was heated to 700, 800 or 900 °C. The synthetic material underwent a reflux process using a 1:1 HCl/water solution, followed by filtration with distilled water utilizing a polypropylene membrane. After refluxing with nitric acid, the solution was neutralized by dropping 25% ammonia water. The products were obtained by centrifuging at 14000 rpm and dialysis with distilled water in a 3.5 kDa MFPI bag for 48 h, as shown in Fig. 11b. Their results showed that the colour of products ranged between green and orange, depending on the modifications made to the synthesis and



**Fig. 12.** A) The mechanism for the formation of CDs from acetone. reproduced with permission [38], Copyright 2015, Wiley-VCH; b) Schematic diagram of the synthesis route of CDs from acetaldehyde. Reproduced with permission [40], Copyright 2021, American Chemical Society; c) Schematic of the fabrication process for CDs-Li. Reproduced with permission [41], Copyright 2022, Wiley-VCH.

post-treatment conditions. Besides, reducing the oxidation time yielded smaller sized CDs, and the increase of synthesis temperature would result in a blue shift for photoluminescence of CDs, which was mainly because of the fewer defects formed in graphene layer at higher temperature. The presence of templates provides attachment sites for reaction substrates, which can regulate the growth of CDs. As a result, the synthesized CDs often have uniform sizes and water-dispersibility. However, the synthesis and final removal of templates make the entire synthesis process more complex, and the recyclability of template directly determines the efficiency of synthesis. Therefore, template method is not the preferred strategy for large-scale synthesis, but it possesses outstanding advantages in directing the structure of CDs.

#### 2.2.4. Aldol condensation polymerization

Our group invented this method to prepare CDs by blending acetone and sodium hydroxide in 2015 [38]. A few days later, the mixture became dark brown in the absence of any other treatment, and liquid acetone was converted into solid products. Subsequently, we proved that the high-yield products were CDs through some modern analysis methods. In the typical synthesis process, NaOH and acetone were mixed for 1 h under intense magnetic stirring, the mixture was subsequently exposed to ambient air, temperature, and pressure, and left for 120 h. A diluted HCl solution was added to achieve a neutral pH. The resulting product was separated via centrifugation and washed multiple times with deionized water. At last, CDs powder were collected after evaporating the final product at 100 °C for 12 h, as shown in Fig. 12a. Furthermore, we discovered that the reaction products obtained without undergoing washing or purification can be converted into 3D porous carbon structures, which exhibited exceptional rate performance and an ultra-long cycle life, rendering them ideal for utilization as anode materials in sodium-ion batteries. This method does not need harsh conditions such as heat treatment, high toxic reagents or special equipment. More attractively, it has a large yield and simple post-processing.

Under alkaline conditions, aldol reaction of acetone or aldehyde can take place to form unsaturated ketones or aldehydes. Next, polymer-like products with elongated carbon chains are formed through the polymerization of unsaturated ketones or aldehydes. These chains may twist and interconnect, resulting in the production of multiple nanodots. Dehydration and hydroxylation are potential side reactions that may occur under these highly alkaline conditions. As a result, the final product is likely a blend of oligomers and has been confirmed [38]. Furthermore, we replaced acetone with acetaldehyde to prepare CDs with the size of 3–5 nm [39]. The optimized process further improved the preparation efficiency of CDs. After further enhancement of the process, this method can reach the yield of kilogram in 2 h [40]. Based on the aldol reaction of acetaldehyde and sodium hydroxide to synthesize CDs, we added carbamide or cysteine to the reaction substrate to synthesize N-doped and N, S-co-doped CDs, respectively [40], as shown in Fig. 12b. Additionally, we also added bis(trifluoromethane)sulfonimide lithium salt to the initial aldol condensation system to synthesize Li-doped CDs and applied them as fillers for polymer electrolytes, successfully achieving high ionic conductivity and Li<sup>+</sup> migration number [41], as shown in Fig. 12c. It can be concluded that the aldol condensation method has great potential in the large-scale synthesis of CDs. Due to the diversity of raw materials, various structures and heteroatom-doped CDs can be conveniently designed.

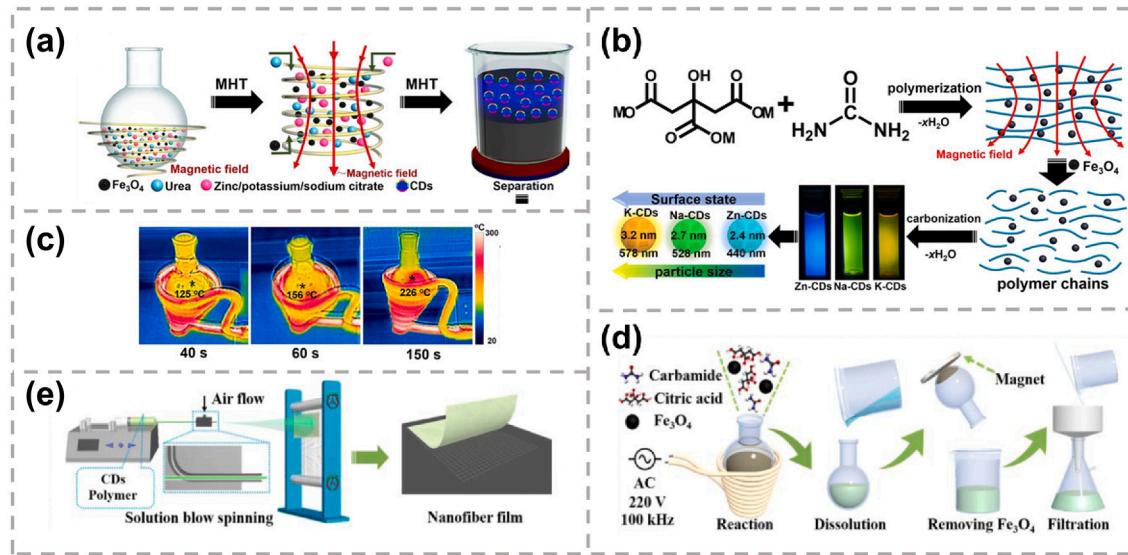
#### 2.2.5. Electromagnetic induction heating method

Electromagnetic induction heating or radio frequency heating of magnetic nanoparticles or conductive sensors has been widely applied [85–87]. The electromagnetic energy absorption/conversion (non-contact heating technology) on special materials can direct the heat to the place where it is needed. This technique serves not only as an alternative means of heating, but also as a potent tool to overcome the heat transfer restrictions that are typically associated with traditional “contact” heating reactors. This process happens virtually instantaneously on the

target sample, devoid of any notable thermal inertia, and boasts significantly superior heating efficiency [88]. Chen's team employed citrate and urea as precursors to produce fluorescent Zn<sup>2+</sup>, Na<sup>+</sup> and K<sup>+</sup> doped CDs on a large scale through a fast electromagnetic induction heating strategy for the first time [85]. This manufacturing route made full use of the magnetothermal effect to prepare CDs in one hour, and the output can reach 85 g. Compared with the traditional method, the yield of CDs was enhanced by a factor of approximately 160. The precursor was swiftly heated to about 230 °C with superparamagnetic Fe<sub>3</sub>O<sub>4</sub> nanoparticles in 3 min to finalize the reaction. When Fe<sub>3</sub>O<sub>4</sub> nanoparticles were placed in an alternating magnetic field, the magnetic moment will fluctuate with the increase of temperature. Consequently, the precursor underwent carbonization to form CDs, which were then dispersed in water and subjected to an external magnetic field after cooling. The product was separated into brown CDs suspension and Fe<sub>3</sub>O<sub>4</sub> solid by a magnet. After further ultrafiltration, the CDs dispersion was dried and collected, as shown in Fig. 13a-c. In addition, after a straightforward treatment, Fe<sub>3</sub>O<sub>4</sub> nanoparticles were capable of being reused for at least three cycles. During the synthesis procedure, the precursor was initially condensed and polymerized to form a cross-linked polymer chain. Subsequently, it was carbonized into a vast sp<sub>2</sub> domain. At temperatures below 192 °C, most of the precursors dehydrated and reacted to form polymer dots. As the reaction temperature increased, the cross-linked molecules carbonized and graphitized to form a typical CDs structure, with hydrophilic polymer chains surrounding its carbon nucleus. Conversely, elevated temperatures could result in excessive carbonization. Due to its complex functional groups, the synthesized CDs was applied as a nano-filler to enhance the mechanical properties of polycaprolactone nano-fiber scaffold derived from electronic spinning, which can improve the wound healing efficiency. Subsequently, their team conducted the same method to synthesize blue-green photoluminescent CDs with a yield of 25.37 g in a single batch in about 90 s, and fabricated polymer nanofiber membranes containing large-area CDs by utilizing solution blow spinning, exhibiting excellent fluorescence and improved mechanical characteristics [87], as shown in Fig. 13d-e. Electromagnetic induction heating has astonishing advantages in heating efficiency, greatly reducing reaction time and improving the uniformity of reaction products, which plays an important role in synthesizing CDs with uniform structure and size. However, in this method, magnetic substances cannot react with the reaction substrate and require certain recovery methods, so the system suitable for this method is relatively scarce. Developing inert and easily separable magnetic media may be a meaningful direction for improvement.

#### 2.2.6. Plasma method

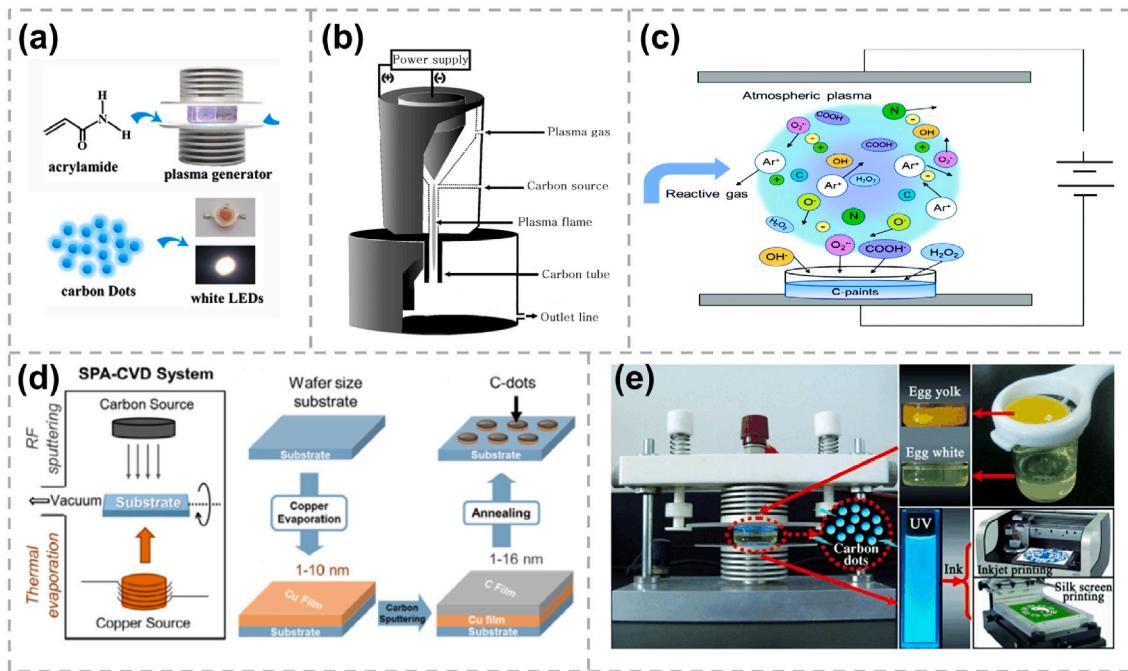
When a gas is subjected to high temperatures, it undergoes ionization, which involves the loss of one or more electrons from its atoms. This ionization process results in the formation of plasma. In plasma, positive and negative charges coexist and interact with each other, it has a very high temperature, which enables it to provide the energy required for carbonization, thus being applied in the synthesis of CDs. Jiang et al. used the submerged arc helium atmosphere plasma to generate free radicals, directly introduced ethylenediamine in the process of synthesizing CDs from benzene, and incorporated functionalization into the synthesis process to functionalized carbon nanoparticles [89]. High-speed centrifugation was employed to separate functional nanoparticles from the supernatant. The carbon nanoparticles without ethylenediamine were gathered and separated from the supernatant within 2 h, so there was no need for centrifugation. The supernatant was removed after separating the nanoparticles. To obtain the final product, they rinsed the nanoparticles with fresh benzene for five consecutive times to remove the unreacted ethylenediamine and by-products, and finally washed them with acetone for five times. Acrylamide has been employed as precursor to synthesize photoluminescent CDs through rapid plasma induction, which had a tight size range of 3–4 nm and exhibited a vibrant blue fluorescence, boasting a quantum yield of 6%



**Fig. 13.** a) Illustration of the large-scale manufacturing of CDs by electromagnetic induction heating technique. b) Illustration of proposed process to prepare CDs with citrate and urea. c) Infrared thermal photographs captured throughout the formation process of CDs. Reproduced with permission [85], Copyright 2020, Wiley-VCH; d) Flowchart outlining the process for producing CDs by electromagnetic induction heating method with citric acid and carbamide. e) Diagram illustrating the process of manufacturing large-scale nanofiber films embedded with CDs via solution blow spinning. Reproduced with permission [87], Copyright 2022, Elsevier.

[90]. The manufactured CDs were effectively utilized in white LEDs, as shown in Fig. 14a. Specifically, a glass dish containing acrylamide underwent a 10-minute air plasma treatment at a radio frequency power of 150 W. The obtained powder was ultrasonically dispersed in ethanol and the suspension was centrifuged to eliminate non-fluorescent deposits. This research also discovered that increasing the energy of the plasma treatment results in stronger photoluminescence of CDs. Atmospheric plasma treatment is a new technique for surface functionalization, with the aim of improving the optical properties and antibacterial

effectiveness of the synthesized CDs (AC-paints) [91]. Because of its potent fluorescence and minimal cytotoxicity, AC-paints can serve as a biological imaging agent emitting blue and green fluorescence. In this study, argon and oxygen or nitrogen in the plasma system were used as carrier gas and reaction gas respectively to produce various functional groups at CDs, thus improving fluorescence efficiency and antibacterial performance, as shown in Fig. 14c. When contrasted with the conventional technique of surface functionalization of CDs using additive passivator at high temperature, atmospheric plasma treatment is favourable



**Fig. 14.** a) Demonstration of the production of blue fluorescent CDs and their potential for use in high colour rendering index white LEDs. Reproduced with permission [90], Copyright 2013, Springer Nature; b) Schematic of the thermal plasma jet system for the production of GQDs. Reproduced with permission [92], Copyright 2014, American Chemical Society; c) Schematic of plasma reaction with AC-paints. Reproduced with permission [91], Copyright 2017, Royal Society of Chemistry; d) Demonstration of CDs fabrication using SPA-CVD. Reproduced with permission [93], Copyright 2019, Elsevier; e) Digital photographs of fluorescent carbon inks production using egg-derived CDs through plasma-assisted fabrication. Reproduced with permission [45], Copyright 2012, Wiley-VCH.

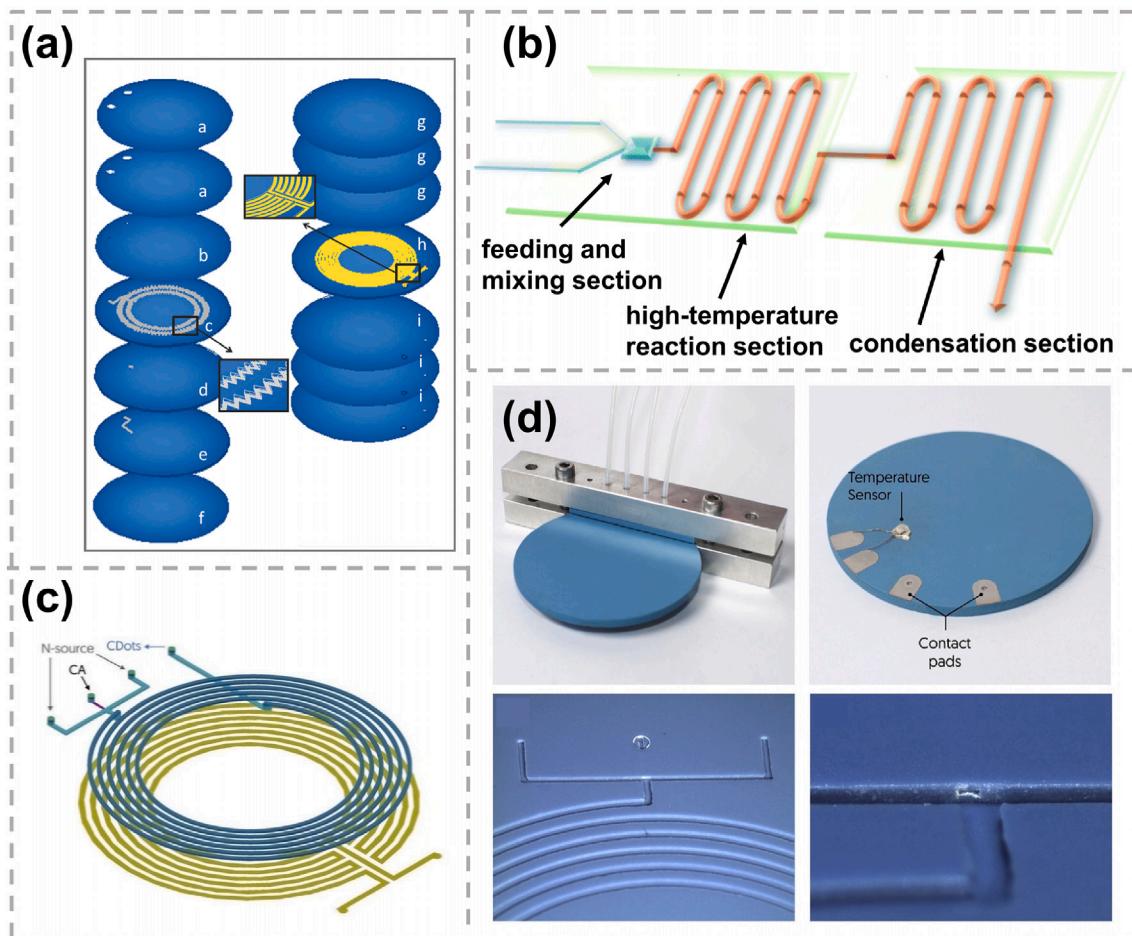
because it is easy to treat and does not need any additives at room temperature. Large-scale production of AC-paints can be accomplished through atmospheric plasma and is easy to process. Active oxygen can be generated on the surface at low cost. The use of thermal plasma jet is a manufacturing method of GQDs with controllable size and low cost [92]. In this process, a significant quantity of ethylene gas was injected continuously into the Ar plasma, producing a beam of carbon atoms. Subsequently, the carbon beam was diffused into a chamber through a carbon tube linked to the anode, as shown in Fig. 14b. Carbon materials, such as GQDs, are prepared by gas phase collision reaction and will not be dispersed in common solvents. By adjusting the length of the connected carbon pipe, the mean size of GQDs can be regulated, which is a process with controllable size, low cost and mass production. Plasma can serve as a reaction medium to complete the reaction inside, or input energy from the outside into the system to stimulate the reaction. More noteworthy is that changing the gas phase composition of the plasma can also achieve synchronous functionalization, reflecting the unique advantages of this method.

Similarly, Park et al. also proposed the growth of curved CDs in vacuum through a bottom-up method known as sputtered plasma assisted chemical vapor deposition (SPA-CVD) [93]. Through thermal annealing, copper nanoparticles (Cu-NP) were formed on the surface of a layer of copper, which was approximately 10 nm thick, along with an ultra-thin carbon film. This led to the creation of curved CDs on the surface, as shown in Fig. 14d. The hemispherical shape of nucleating Cu-NP controlled the size and shape of CDs. This strategy allowed for the growth of CDs with uniform thickness and adjustable sizes on a substrate like aluminium in the wafer size region without requiring a solvent. Through manipulation of the parameters for SPA-CVD sputtering and evaporation, it is possible to adjust both the size and concentration of CDs across a broad range. This method is possible to create a significant number of CDs rapidly. Besides, the process is conducted under conditions of ultra-high vacuum, thereby reducing the occurrence of impurities during the procedure. In addition, the edge of the synthesized CDs did not exhibit any functional groups that were incorporated during manufacturing, thereby enabling functionalization either in-situ or post-deposition. It is worth noting that the current methods for growing CDs tend to result in flat and strong luminescent GQDs. However, utilizing hemispherical Cu-NPs in SPA-CVD could result in more distorted and disorganized CDs. The employment of polymethyl methacrylate (PMMA) enables the transfer of the CDs layer onto the conductive electrode, exemplifying the utility of SPA-CVD CDs in memory devices. In order to ensure the final product remained both inexpensive and minimally toxic, cheap natural eggs were chosen as the initial material to obtain fluorescent stable CDs through a fast plasma induction method and further expanded its application as printing ink [45]. Specifically, between two quartz slides of the plasma generator, a glass dish containing either egg white or egg yolk was positioned. Subsequently, the sample was irradiated by a powerful and consistent plasma beam generated from the upper electrode for 3 min to produce dark black products. The CDs dispersion showed bright blue fluorescence under ultraviolet light, as shown in Fig. 14e. Plasma treatment provided CDs with amphiphilic property, which made it able to disperse in water and most organic solvents. High-temperature plasma furnished potent energy to initiate the pyrolysis of proteins containing nitrogen and oxygen functional groups, which facilitated the denaturation of proteins by rupturing the hydrogen bonds and causing an expansion in the polypeptide chains. At the same time, upon interaction with the active components in the plasma, the egg white underwent partial carbonization and oxidation. With continuous treatment, the long peptide chains underwent further decomposition into shorter chains, and finally formed CDs with rich point effects. This plasma-induced fluorescence CDs synthesis can be applied to different carbon sources. However, unlike gas-phase precursors fed together with plasma, the synthesis process of using plasma as external energy to initiate carbonization is difficult to achieve continuous operation, which will inevitably limit its

processing capacity. In addition, this method also imposes strict equipment requirements, thereby increasing production costs.

### 2.2.7. Microfluidic method

Micro-reactor is an effective tool for chemical research. In the micro-reactor, the synthesis and transformation take place in a transverse dimension usually less than 1 mm [94]. So far, micro-reactor controlled synthesis has gained significant attention across multiple disciplines, including organic synthesis, biology, and medicine [95,96]. Owing to the size reduction, the micro-reactor can obtain a series of unique characteristics, which cannot be achieved by the traditional batch reactor, including but not limited to a high ratio of surface area to volume, a short distance for diffusion, rapid transfer of heat and mass, a considerable reduction in the time needed for characterization, the ability to control each reaction parameter independently, and improved reproducibility for kinetic control. Lin et al. applied the microreactor synthesis system to the research of CDs for the first time [97]. A stock solution was prepared by dissolving glucose in formamide, afterward, the substance was injected into a Teflon capillary through a syringe, with a consistent speed maintained throughout the process. After heating to 180 °C in an oil bath, the product was gathered, subjected to vacuum distillation for concentration, and subsequently dialyzed to purify the CDs for characterization. With the application of micro-reactor, hundreds of reaction conditions had been selected, and the adjustable emission maximum with considerable photoluminescence (PL) quantum yield has been effectively achieved. By precisely controlling the reaction time, CDs at different development stages can be easily prepared. According to CDs at different reaction stages, they discovered the correlation between the PL properties of CDs and reaction parameters, and finally demonstrated the key factors that affected the PL properties in the synthesis process. It has been proved that the microreactor is not only an effective means for quickly evaluating CDs reaction conditions, but also has crucial value for theoretical research. The microfluidic system can achieve automatic synthesis of CDs [98]. As shown in Fig. 15a, the system consisted of two independent platforms, one was designated for thermal regulation, and the other for microfluidics. Uniform and monodisperse CDs measuring 3.3 nm in size were synthesized by utilizing ascorbic acid through thermal decomposition with dimethyl sulfoxide as the solvent and have been used for pH detection. Finally, the cytotoxicity and permeability of CDs were also studied to prove their applicability in biomedical applications through bioimaging. Besides, an effective microfluidic technique is able to continuously synthesize CDs emitting fluorescence across the full spectrum [99], as shown in Fig. 15b. The synthesis process can be accomplished in as little as 20 min, and the application of Fe<sup>3+</sup> precise detection and in vitro biological imaging has been realized. In a typical process, citric acid and urea were prepared to aqueous solution, which were pumped into the pipeline by two horizontal flow pumps. Finally, the sample was washed with acetone and methanol and subsequently subjected to freeze-drying to obtain the final product. CDs synthesized continuously exhibited a smaller and more uniform average diameter compared to those synthesized in an autoclave, reflected the uniform material and energy exchange properties in microreactors. A fully integrated low-temperature co-fired ceramic (LTCC) microreactor for synthesizing CDs was developed and described by Berenguel-Alonso et al. [100]. To ensure uniform surface chemical and physical properties, a monolithic all-ceramic device was used to integrate a jet, heating resistor, and optical window for fluorescence imaging of the reaction process within the micro-reactor, as shown in Fig. 15c-d. Different CDs were produced by hydrothermal strategy under high temperature and high pressure with citric acid and different nitrogen sources. The obtained CDs exhibited blue photoluminescence under UV irradiation, with quantum yield up to 77%, and were screened as metal nanoprobes and bioimaging contrast agents. The microfluidic reactor technology can not only enhance the synthesis process by providing better control over the formation and size consistency [101], but also reduce the usage of



**Fig. 15.** a) Microfluidic system with different layers for synthesis of CDs. Reproduced with permission [98], Copyright 2014, Royal Society of Chemistry; b) Schematic diagram of the uninterrupted process used to produce CDs with a complete range of spectral emissions. Reproduced with permission [99], Copyright 2019, Royal Society of Chemistry; c) 3D depiction of the microreactor channel and the heating element used in the synthesis of CDs; d) Top view of the microreactor, showcasing the inlet and outlet tubes; Bottom view of the microreactor, including the contact pads for the heating resistor and the integrated temperature sensor; Photographs showing the open microchannels, along with a detailed view of the middle inlet and two sheath flow channels. Reproduced with permission [100], Copyright 2019, Elsevier.

energy, amount of reagent required, and waste production [102]. However, one of the primary drawbacks hindering the wider application of LTCC devices is their opacity, as it impedes the visualization of reaction process, which is very important for understanding the behaviour of fluid flow.

### 3. Conclusion and outlook

Numerous uncomplicated, inexpensive, and effective synthesis pathways for CDs have emerged following the advent of CDs in 2004. This represents a significant divergence in comparison to the arduous and costly chemical reactions needed for the production of other fluorescent nanoparticles [103]. As the earliest employed method, the fluorescence of CDs synthesized by arc discharge method is outstanding, but the particle size is uneven, and the output is extremely limited, making it unsuitable for large-scale manufacturing. Laser ablation has the capability to yield nanoparticles that possess not only comparable composition and structure to the target, but also entirely distinct chemical traits and structural attributes [49]. The yield of CDs produced through laser ablation is acceptable, but the particle size lacks uniformity, and the equipment requirements are comparatively strict, rendering it unsuitable for industrial manufacturing. Among these techniques, chemical oxidation stands out as a practical and efficient approach for mass production that does not require sophisticated

equipment. Numerous research groups employ this method extensively to generate carbon nanoparticles [54]. Moreover, the carbon sources necessary for this process are varied and straightforward to acquire [55]. Electrochemical approach offers the benefits of easy operation and affordability, rendering it one of the comparatively low-cost, uncomplicated, and environmentally friendly methods for synthesizing CDs [58]. In addition, CDs produced by this strategy have high yield and uniform particle size. However, it requires special equipment and there are few reactions suitable for electrochemical method. Despite the fact that the output of CDs produced through pyrolysis is quite limited, the operation is simple, the conditions are controllable, and the raw materials are easy to obtain. It had been reported that a large amount of biomass waste was used for pyrolysis to prepare carbon CDs [59]. Solvothermal method is very popular among most of strategies for the synthesis of CDs. It can be used to decompose macromolecules into small molecules at high temperature or polymerize small molecules into large molecules. With the assistance of microwave, solvothermal method can overcome the problem of uneven heating in the process of using individually, which is more conducive to the production of CDs with uniform size. The yield of CDs prepared by template method is more than 10% [82], while the size of the particles is uniform and they exhibit exceptional water dispersibility, the manufacturing procedure is excessively complicated. In contrast, the aldol condensation method can prepare CDs on a large scale, which has a promising industrialization

prospect. Besides, heteroatom doping can be realized by changing the type of reaction substrate to control the composition of CDs directionally. At present, the strategies of CDs synthesis continue to rely on pyrolysis and hydrothermal/solvothermal techniques. Obviously, these traditional methods are both time-consuming and energy-intensive, rendering them inadequate for meeting the requirements of practical large-scale manufacturing. The advent of more efficient synthetic methodologies like ultrasonic, electromagnetic induction heating and microfluidics has the potential to expedite the progress of CDs from laboratory preparation to industrial preparation to meet the demand of large-scale industrial applications. The synthesis strategy still requires further research and development to enable it to be implemented in future industrial scale production. To conclude, the benefits and drawbacks of different techniques are outlined in the table below.

Currently, there existed misunderstandings regarding the dispersion system properties of CDs. Numerous papers suggested that CDs are water-soluble or can be dissolved in specific solvents. While considering the size of carbon dots, their dispersion form in solution is more similar to that of colloids, rather than existing in molecular form like a solution. This can be well verified by light scattering. The Tyndall effect can be clearly observed in the dispersion system of CDs [104], further indicating that CDs are not dissolved in solvents, but dispersed within them. The dispersibility in solvents and potential application of CDs are intricately linked to the nature and extent of functional groups present on their surface. How to precisely control the surface functionalization of CDs in the synthesis process is a very meaningful research direction. Therefore, having a grasp of the mechanism behind every synthesis method is crucial. Regrettably, the functional groups pivotal in influencing the properties of CDs may not be universally suitable under all circumstances. Most CDs exhibit limited thermal stability. Typically, when exposed to temperatures exceeding 200 °C, certain surface functional groups start to lose activity, which is even more severe in the presence of catalysts, especially for some CDs obtained from biomass. At higher temperature, nearly all surface functional groups on CDs are gradually eliminated, and the internal carbon nucleus structure undergoes alterations, including aggregation and an increased level of graphitization [105,106]. This will have a significant impact on the optical performance of CDs, almost all surface photoluminescence excitation will be invalid, which hinders the original application of CDs. However, the transformation is not meaningless, and the transformation of the internal structure of carbon nucleus will also bring additional properties, such as the construction of special carbon structures and the generation of new optical properties. Reasonable utilization of these characteristics will also enrich the application fields of CDs.

Among the steps involved in producing CDs, the separation and purification of these compounds from the complex mixture generated during the synthesis process stands out as particularly critical. CDs exhibit a low density and possess a significant number of hydrophilic functional groups, which makes this process more challenging. In some cases, due to the diversity of size and surface functional groups, it is still difficult to meet the high standard of practical application after removing large particles by filtration or centrifugation and removing small molecules by dialysis [107]. So, it is particularly important to update effective separation and purification technology. Meanwhile, it is urgent to develop innovative synthetic routes and combine existing synthetic routes to synthesize more efficiently. Regardless, there remains ample opportunity for enhancing the synthesis of CDs. Not surprisingly, in consideration of its low cost, scalability, remarkable chemical stability, biocompatibility and low toxicity, CDs will continue to be active in the forefront of material science and continue to make contributions to human development.

#### Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence

the work reported in this paper.

#### Data availability

The authors do not have permission to share data.

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