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Carbon quantum dots synthesis from waste and by-products: Perspectives and challenges

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ABSTRACT

Carbon dots (CDs) are a novel class of fluorescent nanomaterials, with properties such as photoluminescence, high solubility, low toxicity, and favorable biocompatibility. They are useful for applications in biomedicine, sensors, solar cells, and photocatalysis, among others. CDs synthesis using vegetal, animal, or industrial waste as a source has become a focus of interest among researchers. These waste materials are inexpensive and available at large, and the repurposing of natural resources has the potential to reduce pollutants and their environmental impacts. Residues from plant sources, such as peels, leaves, and flowers, are preferred over other sources, and the conversion to CDs is performed mainly through the hydrothermal method. However, some matters regarding this technology require further studies and elucidation, such as the increase CD's conversion yield from the raw material. Thus, we aimed to explore the use of waste and by-products in CDs synthesis, their potentials, and advantages, as well as present current challenges in the field of study.

1. Introduction

Carbon quantum dots (CDs) are nanomaterials with semiconductor properties discovered by accident in 2004, during the purification process of a single-walled carbon nanotube [1]. They are spherically symmetrical, with a size less than 10 nm, and a structure that can vary between amorphous and crystalline. Both amorphous and crystalline structures have interesting properties, such as photoluminescence and wavelength-dependent emission, high solubility, low toxicity, ease of functionalization, and biocompatibility [2].

CDs soon became a focus of interest among researchers due to their wide variety of technological applications in several fields, such as sensors, photocatalysis, bioimaging, drug delivery systems, solar cells, and LED devices [3–5]. In the years following the discovery of CDs, some review articles were published [4,6–10]. The reports mainly emphasize their synthesis processes and leading applications. Among these reported preparation processes, two synthesis approaches stand out, the top-down approach and the bottom-up approach (Fig. 1).

In the top-down approach, CDs are generated from relatively macroscopic carbon sources. Most natural products can be used as a matrix in this method. In the bottom-up approach, small molecules which –OH, –COOH, -C = O, and $–NH_2$ groups and polymers undergo dehydration and further carbonization to form the CDs, carbon nanodots, and polymer dots. CDs can also be formed in a bottom-up approach through the self-assembly of polycyclic aromatic hydrocarbon (PAH) [11]. Some studies combined the two approaches mentioned, such as Li et al. (2013), regarding the synthesis of fluorescent carbon nanomaterials [12].

Few articles address the use of waste and by-products for the synthesis of carbon dots (CDS), where the full potential for using these materials as a viable source for CD production deserves further analysis. Usually, reports are focused on the enhanced CD's photoluminescent properties, such as energy conversion rates in photocatalytic devices[13,14], solar cells [15], and LEDs [16,17]. However, the conversion yield of starting material into CDs is often omitted, with emphasis restricted on the synthesized material's quantum yield. Thus, we aimed to explore the use of waste and by-products from different sources in the CDs synthesis, their potentials, and advantages, as well as presenting current challenges in the field of study.

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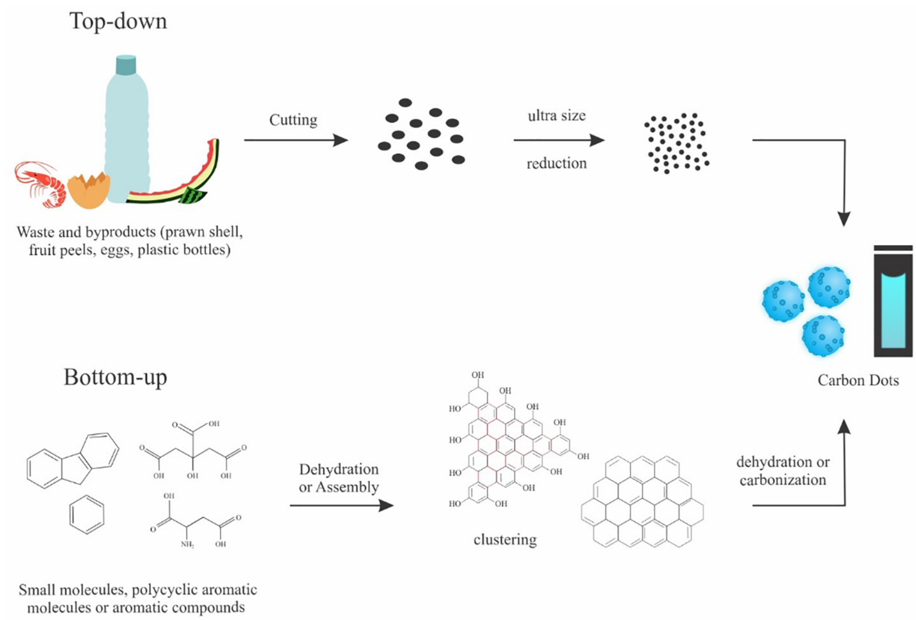

Fig. 1. Schematic representation of top-down and bottom-up synthesis approaches from waste and by-products.

2. CDs synthesis from waste and by-products: state-of-the-art

The use of natural or industrial waste and by-products emerges as a possibility for the transformation of materials with no aggregated value into nanomaterials with high aggregated value, with high potential for application in technological resources. Waste and by-products possess sufficient carbon content to be considered useful as starting materials for the synthesis of CDs, with the advantage of safety and biocompatibility, as well the facility to achieve the synthesis conditions [9]. Fig. 2 shows the CD's main sources, the most employed synthesis methods, and some relevant applications.

Natural sources such as human hair [18], lemon peels [19], plastic bottles [20,21], and manure [22,23] are rich in amino, sulfur, carboxyl, and hydroxyl groups. Depending on the synthesis method and the materials used as a source, these groups can remain on the CD's surfaces. These groups, when incorporated in a sufficient content, can provide solubility in water and additional possibilities for passivation and functionalization of the surface [13].

To provide a comprehensive reading of the CDs properties as a function of the different raw material used, the CDs produced were classified as a function of the waste type; vegetable waste and by-products, animal waste and by-products, and industrial waste and by-products, as well their limitations and perspectives.

2.1. Vegetable waste and by-products

Many articles discuss the use of vegetable waste as a source for CDs synthesis [9,24–26], with the main advantages to contribute to the reuse of vegetal by-products to produce materials with high aggregate value. Several vegetable parts can be used as a source for CDs, such as peels [27–29], fruits [30,31], flowers [29], and roots [15], as showed at Table 1.

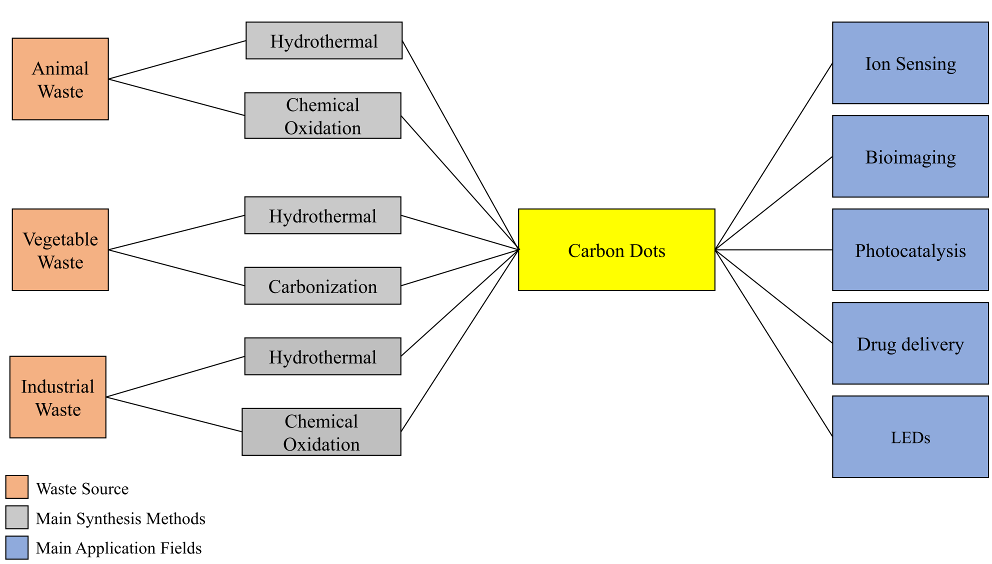

Fig. 2. Main raw waste materials, CDs synthesis methods, and CDs applications.

Table 1
Vegetable waste and by-products as a source for CDs and their related properties.

Carbon Source	Synthesis Method	Product Condition	Quantum Yield[1]	Reference
Bagasse	Combustion	60 °C/4 h	25,7%[2]	[24]
Bagasse	Hydrothermal	180 °C/12 h	11,8%	[42]
Bagasse pulp	Chemical oxidation	Toluene/24 h stirring	18,7%	[43]
Orange peels	Hydrothermal	200 °C/6 h	11,4%	[27]
Orange peels	Hydrothermal	180 °C/12 h	36,0%	[30]
Orange peels	Hydrothermal	200 °C/8 h	4,29%	[44]
Watermelon peels	Hydrothermal	220 °C/2 h	7,10%	[31]
Lemon peels	Hydrothermal	200 °C/12 h	14,0%	[19]
Mango peels	Hydrothermal	200 °C/4 h	No data	[45]
Onion peels	Carbonization	120 °C/15 lbs.	28,0%	[28]
Fruit/vegetable peels	Heating	150 °C/2 h	No data	[29]
Coconut shell	Hydrothermal	150 °C/3 h	No data	[25]
Papaya pulp	Pyrolysis	200 °C/15 min	23,7%	[46]
Rice residue	Hydrothermal	200 °C/12 h	23,5%	[47]
Platanus waste	Carbonization/laser ablation	600 °C/2 h	32,4%	[48]
Wheat straw	Hydrothermal	180 °C/4 h	13,0%	[49]
Wheat straw	Hydrothermal	250 °C/10 h	9,20%	[26]
Lotus roots	Hydrothermal	170 °C/6 h	No data	[50]
Carboxymethylcellulose	Hydrothermal	260 °C/2 h	44,0%	[33]
Banana pseudo-stem	Hydrothermal	120 °C/12 h	48,0%	[32]
Processed white rice	Carbonization	250 °C/24 h	41,0%	[51]
Wine lees	Carbonization	300 °C/3 h	6,80%	[52]
Waste tea	Hydrothermal	150 °C/6 h	7,10%	[53]
Food waste	Solvothermal	200 °C/2 h	0,26%	[16]

[1] Quinine Sulfate as reference.
[2] Rhodamine B as reference.

Vegetable waste presented the highest quantum yield values for CD production in comparison with animal and industrial waste, with values above 40% [32,33]. Both procedures adopted the hydrothermal synthesis under different reaction conditions. When the banana pseudo-stem was used as a source [32], the reaction condition was 120 °C for 12 h, while the carboxymethylcellulose [33] was transformed using 260 °C for 2 h. The role of the reaction condition by the hydrothermal method in the CD's quantum yield is still unclear. Somehow, a compromise between the time and temperature seems to be relevant, where with high temperature it is required a short time, and with lower temperatures required several hours to conversion, a detail that deserves further studies.

2.2. Animal waste and by-products

Animal waste and by-products can be classified in groups such as crustacean shells, human waste (hair and facial skin), animal manure, and milk-derived by-products, as showed at Table 2. Synthesis procedures include hydrothermal treatment (microwave-assisted or not), chemical oxidation, and carbonization. An environmental concern regarding these methods is the amount of residues, resulting in additional purification steps, increasing the use of reagents and overpricing the process.

CDs synthesized from animal waste and by-products are conducted mainly through the hydrothermal method due to its low cost and environmental-friendly routes. However, difficulties in controlling particle size were reported in the hydrothermal method [34–36].

From Table 2, the highest values for the quantum yield are derived from pigeon feathers and eggs obtained by pyrolysis[37] and from crab shells through microwave radiation [38], with values around 25% and 20% respectively. In this case, the microwave method provided a good quantum yield, reducing substantially the reaction time in comparison with the pyrolysis method [39].

2.3. Industrial waste and by-products

For industrial waste, their use as a source in the CDs synthesis presents as main vantage the possibility to reuse a material with no aggregated value and contribute to a reduction in waste products that pollute the environment. The industrial waste can be grouped into oil, paper and plastic waste, as showed at Table 3.

From Table 3, the highest values for quantum yield are derived from cat feedstocks [40] and petroleum coke [41], with values around 28% and 22% respectively. Both sources were converted into CDs by hydrothermal method under different reaction conditions. Plastics such as the one used in cat feedstocks, when submit-

Table 2
Animal waste and by-products as a source for CDs and their related properties.

Carbon Source	Synthesis Method	Product Condition	Quantum Yield[1]	Reference
Human hair	Hydrothermal	200 °C/24 h	10,7%	[18]
Crab shell	Hydrothermal/microwave-assisted	220 °C/10 min	19,8%	[38]
Crab shell	Sonochemical	Ultrasonication irradiation	14,5%	[54]
Prawn shell	Chemical Oxidation/Hydrothermal	200 °C/8 h	9,00%	[55]
Cow manure	Hydrothermal/microwave-assisted	250–300 °C/2 h180 W/20 min	No data	[22]
Cow manure	Chemical Oxidation	HNO_3 – 5 M	0,65%	[23]
Facial tissue	Chemical Oxidation	HNO_3	No data	[56]
Pigeon feathers, eggs, and manure	Carbonization	300 °C/3 h	24,9%	[57]
Expired milk	Heating	180 °C/2 h	8,64%	[58]
Whey	Thermal treatment	180–250 °C/10–40 min	11,4%	[59]

[1] Quinine Sulfate as reference.

Table 3
Industrial waste and by-products as a source for CDs and their related properties.

Carbon Source	Synthesis Method	Product Condition	Quantum Yield[1]	Reference
Waste paper	Hydrothermal	180 °C/10 h	10,8%	[60]
Waste paper	Hydrothermal	180 °C/12 h	No data	[61]
Plastic bottles	Hydrothermal	180 °C/ 12 h	5,20%	[20]
Plastic bottles	Ultrasonic-assisted chemical oxidation	H_2SO_4/HNO_3/700 W/2min	4,84%	[21]
Cat feedstocks	Hydrothermal	180 °C/ 24 °C	28,0%	[40]
Waste frying oil	Chemical oxidation	H_2SO_4/ 100 °C	3,66%	[62]
Kerosene soot	Chemical oxidation	HNO_3 – 5 M	3,00%	[63]
Waste oil	Ultrasonication	H_2SO_4/Ultrasonic waves	7,50%	[64]
Coal/petroleum coke	Hydrothermal	200 °C/2 h	21,9%	[41]

[1] Quinine sulfate as reference.

ted to the carbonization over a long period can result in CDs with increased quantum yield [40]. On the other hand, petroleum coke can be considered as a promising material, with satisfactory quantum yield using higher temperatures in a considerable shorter period [41]. As observed for animal and plant residues, the hydrothermal method was undeniable the method most used for the CDs synthesis.

3. Conclusions

The use of waste and by-products has made advances in CDs synthesis, due to the diversity in the applications for these materials. The varied composition of the raw materials reflected in the different values obtained for the quantum yields. The wide variety of reaction conditions and methods of synthesis raises questions about the route employed. Starting from the same raw material and applying the same synthesis method, the variation in synthesis parameters produces distinct effects on CD's quantum yield. Moreover, the different functional groups available as starting material improve the synthesized particles' properties.

The repurpose of vegetable waste for CDs synthesis stands out in comparison to animal or industrial wastes. Additionally, the hydrothermal method is the most recurrent in the literature because it is a low-cost technique. However, the advantages of this synthesis technique over others are not clear. Comparative studies based on a single synthesis matrix and employing different synthesis methods may help elucidate the synthesis method's role in conversion yield. Also, top-down approaches produce a large amount of residues, where the reduction of by-products remains a challenge in the field.

Future studies should focus on maximizing the conversion of the starting material into CDs. This would reduce the loss of raw material and the need to carry out various experiments to obtain a significant yield of these nanoparticles.

CRediT authorship contribution statement

Bruno Peixoto Oliveira: Conceptualization, Data curation, Investigation, Methodology, Writing - original draft. **Flávia Oliveira Monteiro Silva Abreu:** Conceptualization, Supervision, Formal analysis, Project administration, Writing - review & editing.

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.

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