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Synthesis and application of carbon quantum dots for document security: current status and future outlook

Zhang Zhe, Guo Tianbao*, Wang Jie

Graduate School of China People's Police University, Langfang, Hebei, China
z2391025688@163.com
**Corresponding author*

Abstract: *This paper mainly summarizes the research progress of the preparation, modification and application of carbon quantum dots in the field of certificate anti-counterfeiting, and focuses on its application in anti-counterfeiting ink, fluorescent labeling, doping materials, barcode and anti-counterfeiting film. In addition, the article also analyzes the problems and future challenges such as the urgent need to optimize the preparation process of carbon quantum dots, the need for future development in combination with multidisciplinary development, and the damage of the best fluorescence performance conditions to the certificate itself. In general, as a new type of nanomaterial, carbon quantum dots have broad application prospects in the field of document anti-counterfeiting.*

Keywords: *Carbon quantum dots; Luminescent properties; Anti-counterfeiting applications*

1. Introduction

Carbon quantum dots (CQDs) are a class of nanomaterials composed of carbon elements, with a particle size range of 1-10 nm, and have excellent optical properties, chemical stability, and biocompatibility [1]. In recent years, CQDs have become a hot research direction and have been widely used in various fields, such as bioanalysis, fluorescence imaging, and document anti-counterfeiting [2]. This article will review the preparation methods and modification techniques of CQDs, focus on the application of CQDs in the field of entry-exit document anti-counterfeiting, and look forward to their future development.

1.1 Concept of carbon quantum dots

Carbon quantum dots are nano-scale structures formed by tens to hundreds of carbon atoms through chemical synthesis or natural formation. CQDs can be regarded as a hard-core soft-shell structure consisting of a core carbon atom cluster protected by an external surface modification layer composed of oxygen, nitrogen, sulfur, and other elements[3]. CQDs have a large specific surface area and excellent optical properties[4], making them a broad application prospect in biomedicine and optoelectronic devices.

1.2 Classification of carbon quantum dots

CQDs have various classification methods, which can be classified according to different characteristics such as source, preparation method, particle size, surface functional group, etc. According to the source, they can be divided into two categories: biomass-based carbon quantum dots and non-biomass carbon quantum dots [5]. Biomass-based carbon quantum dots mainly use biomass as raw materials for preparation, such as tofu residue [6], lignin [7], etc. Non-biomass carbon quantum dots are especially chemically synthesized from carbon sources. According to the preparation method, the standard CQDs preparation methods mainly include top-down and bottom-up preparation methods. The top-down preparation method specifically adds a carbon source and surfactant in the solution and then obtains CQDs through a high-temperature carbonization reaction, while the bottom-up preparation method first prepares polymer-wrapped CQDs precursors and then gets CQDs through processes such as pyrolysis and oxidation-reduction. According to the particle size, CQDs are usually divided into two categories: large CQDs (> 8nm) and small CQDs (< 8nm)[8]. In addition, CQDs can also be classified according to different surface functional groups, such as hydroxylation, carboxylation, amination, etc., which will affect the properties and applications of CQDs [9]

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1.3 Luminescent characteristics of carbon quantum dots

The luminescence mechanism of CQDs may involve various complex physical and chemical phenomena, which have not yet been determined. However, the fluorescence resonance energy transfer mechanism (FRET) and surface defect state mechanism are considered to be the two main luminescence mechanisms[10].

Fluorescence resonance energy transfer mechanism (FRET): When CQDs absorb external excitation light, they form an excited state, then transfer energy to surrounding molecules through fluorescence resonance energy transfer, resulting in luminescence. This mechanism usually requires high concentration to work.

Surface defect state mechanism: Some imperfect chemical bonds and non-planar structures on the surface of CQDs produce local energy bands and unpaired electrons. Subsequently, spontaneous radiation occurs and emits fluorescence. This mechanism usually produces luminescent effects at low concentrations, and its fluorescent properties are more stable.

In summary, the luminescence mechanism of CQDs may involve various complex physical and chemical phenomena. At present, the research on the luminescent performance of CQDs is still in the stage of continuous exploration and development. Different preparation methods, material sources, post-processing methods, etc., will affect the luminescent characteristics of CQDs[5].

2. Preparation and modification of carbon quantum dots

2.1 Preparation methods of carbon quantum dots

CQDs can be prepared by various methods, which can be classified into top-down and bottom-up approaches according to the preparation process[11].

The top-down approach is to add a carbon source and surfactant in the solution and obtain CQDs by high-temperature carbonization reaction. Typical surfactants usually include SDS, PVP, etc. This method can produce CQDs with high fluorescence efficiency and good biocompatibility, but it also has some problems, such as difficulty in controlling the preparation conditions and easy to be contaminated[12].

The bottom-up approach is first to prepare polymer-wrapped CQDs precursors and then obtain CQDs by pyrolysis, oxidation-reduction, and other processes[7]. Common polymer template materials include PVA, gelatin, etc.[13], and biomass materials such as tofu residue and lignin can also be used as carbon sources. This method can produce CQDs with smaller particle sizes and more uniform surface modification, and the preparation conditions are also relatively simple, but it requires high temperature or strong oxidation-reduction conditions[14], and the prepared CQDs usually need to be purified several times to obtain high-quality products[5].

2.2 Modification methods of carbon quantum dots

The principle of carbon quantum dots modification mainly includes changing the surface properties and electronic structure. The change of surface properties is primarily achieved by introducing different functional groups or compounds on the surface of CQDs. The electronic structure change is achieved by doping other atoms or molecules into CQDs[15]. Standard modification methods of carbon quantum dots include the surface modification method, doping method, and synthesis method[16]. CQDs' optical and chemical properties can be modified by doping metal or non-metal elements. Among them, doping metal elements mainly include Cu[17], Zn[18], Mg[19], Mn[20], etc., while doping non-metal aspects is a more mainstream modification method. Doping metal elements can adjust the fluorescence emission wavelength and intensity of CQDs and improve the light stability and fluorescence quantum yield of CQDs. The processes for doping metal elements mainly include deposition-reduction, co-precipitation, etc. Zhang X et al., in their study, used copper hydroxide to modify the surface of CQDs and prepared an infrared fluorescent CQD-CuO composite (a reference not inserted). Doping non-metal elements can change the fluorescence properties of CQDs and improve their fluorescence quantum yield and lifetime [21]. Common non-metal factors include N, S, etc.[22]. Sneha Gupta et al. introduced hydroxyl functional groups on the surface of CQDs, which enhanced their fluorescence intensity and stability [23].

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3. Applications of 3-Carbon Quantum Dots in Anti-Counterfeiting

Carbon quantum dots (CQDs) are a new type of nanomaterial with excellent fluorescence properties, high photostability, and chemical stability. CQDs have broad application prospects in anti-counterfeiting and can be used as anti-counterfeiting inks, fluorescent labels, doping materials, barcodes, and anti-counterfeiting films [22].

3.1 Application of CQDs in Anti-Counterfeiting Inks

CQDs can be used to prepare anti-counterfeiting fluorescent inks to enhance their anti-counterfeiting performance. Different colors of CQD anti-counterfeiting fluorescent inks can be prepared by changing the surface functional groups and particle size. Y. Hu and Z. Gan doped various colored fluorescent CQDs into ink, which made the printed products show bright color fluorescence under ultraviolet-visible light, thus enhancing their anti-counterfeiting effect and achieving high fluorescence intensity and stability [24]. In addition, CQDs also have good light, water, and chemical corrosion resistance, which can improve the durability and stability of printed products[25]. Prepare RTP materials by modifying CQDs for preparing phosphorescent anti-counterfeiting inks or phosphorescent-fluorescent multiple anti-counterfeiting inks:

CQDs can be modified to obtain RTP (room temperature phosphorescence) materials [26], which show phosphorescence characteristics and are used to prepare phosphorescent anti-counterfeiting inks or phosphorescent-fluorescent multiple anti-counterfeiting inks [27]. However, RTP anti-counterfeiting inks may have a problem with RTP material quenching. The possible causes of quenching include the solvent effect: some organic or inorganic compounds (such as nitrites, heavy metal ions, etc.) interact with carbon quantum dots causing fluorescence quenching; aggregation effect: carbon quantum dots self-aggregate phenomenon leads to the reduced intermolecular distance between them resulting energy transfer between multi-level excited states quenching luminescence; photochemical reaction: carbon quantum dots react with some oxidants or reductants after being exposed to light causing fluorescence quenching [27]. The following measures can be taken to avoid the quenching phenomenon: that is, the material undergoes non-radiative relaxation losing phosphorescence characteristics. To solve this problem following measures can be taken: select suitable RTP materials: selecting materials with longer phosphorescence lifetime can reduce the possibility of non-radiative relaxation.

Optimize preparation process: by adjusting solvent pH value temperature, other factors optimize the CQDs preparation process and reduce the risk of RTP material quenching.

Prepare phosphorescent-fluorescent multiple anti-counterfeit inks: combine CQDs and other fluorescent dyes from composite markers to improve security performance and reduce the possibility of quenching.

Select suitable solvent: selecting suitable solvent key avoiding carbon quantum dot quenching Generally speaking, non-polar solvents (such as heptane xylene, etc.) should select to avoid high concentration cations anions aqueous solution affecting carbon quantum dots.

Surface modification: surface modification enhance carbon quantum dot stability and liquid ant-quench ability. For example, introducing hydroxyl amine to other functional groups gives the carbon quantum dot surface electrostatic attraction and reduces self-aggregation, thus improving fluorescence stability.

In summary, avoiding carbon quantum dot quenching liquid requires careful consideration of various factors, including selecting suitable solvent surface modification, adding an ant-quench agent, optimizing the preparation method, etc.

Doping Fe ions preparing magnetic fluorescent security ink: CQDs doped Fe ions preparing magnetic fluorescent security ink. However, ion doping may affect CQD fluorescence performance leading to decreased fluorescence intensity .To solve this problem following measures can take to optimize doping conditions adjust doping Fe ion concentration, doping time other parameters, optimize doping conditions, reduce impact CQD fluorescence performance, coat protective layer particle surface coating protective layer reduces ion impact CQD fluorescence performance at the same time, improve security performance choose appropriate CQD synthesis method also not bad way solve problem appropriate synthesis method ensures better fluorescence performance higher stability thus reducing impact ion doping

In summary, carbon quantum dots are a new type of nanomaterial widely used security ink modifying different colors. CQD security fluorescent ink preparing phosphorescent security ink phosphorescent-fluorescent multiple security ink doping Fe ions preparing magnetic fluorescent security ink further improve its security performance application process needs pay attention optimizing protecting CQD reduce impact fluorescence performance choosing suitable methods measures solve related problems [28]

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3.2 Anti-counterfeiting applications of CQDs as fluorescent labels

Carbon quantum dots (CQDs) are novel nanomaterials with excellent fluorescence and chemical stability widely used in anti-counterfeiting fields. CQDs can achieve anti-counterfeiting purposes as fluorescent labels through their unique fluorescence characteristics.

Due to the high photostability and chemical stability of CQDs, their fluorescence intensity can be maintained for a long time, and they will not lose their fluorescence characteristics even after long-term storage or use.

In addition to single CQDs fluorescent labels, CQDs can be combined with other fluorescent dyes to form composite brands, further improving the anti-counterfeiting performance. Shen L. et al. combined CQDs with fluorescent dyes, which can produce multiple fluorescence signals, thereby increasing the complexity and anti-counterfeiting performance of the labels. At the same time, due to the excellent solubility and controllability of CQDs, they can be surface-modified or functionalized to enhance their fluorescence and anti-counterfeiting performance further.

3.3 Anti-counterfeiting applications of CQDs as doping materials

CQDs are novel nanomaterials with excellent fluorescence and chemical properties, widely used as doping materials in various materials. Li, H et al. added CQDs as doping materials to plastics, textiles, ceramics, and other materials [29] to enhance their anti-counterfeiting performance and achieve anti-counterfeiting applications. The specific implementation steps are to add CQDs to plastic products, which can show unique anti-counterfeiting marks by special fluorescence effects. These marks are formed due to the excellent fluorescence properties and controllable surface functional groups of CQDs, which can interact with plastic molecules and embed into them. When stimulated by external factors, CQDs will emit unique fluorescence signals, thus achieving anti-counterfeiting protection for plastic products. CQDs can also be doped into other materials, such as textiles and ceramics. Adding CQDs to fabrics can give them special fluorescence effects, thus enhancing their anti-counterfeiting performance. Doping CQDs into ceramics can give them a special fluorescence effect similar to banknotes, improving the anti-counterfeiting performance of ceramic products without affecting their appearance and texture.

3.4 Anti-counterfeiting applications of CQDs combined with barcodes

CQDs, as a new type of anti-counterfeiting material, can achieve a higher level of anti-counterfeiting protection when combined with traditional barcode technology. This technology has many application prospects in fields such as cultural relic protection, currency anti-counterfeiting, etc.

Li et al. printed barcodes on paper and added CQDs to form a novel composite label. This label can be verified by machine scanning and using human eyes to observe the fluorescence effect of CQDs. When excited by external stimuli, CQDs emit unique fluorescence signals, which provide a more intuitive and reliable means for barcode authentication. At the same time, due to the excellent chemical stability and photostability of CQDs, their fluorescence intensity can be maintained for a long time, and they will not lose their fluorescence characteristics even after long-term storage or use.

Besides paper, combining CQDs and barcode technology has application prospects in other materials. Wu et al. added CQDs to plastic products and printed barcodes on their surface, which can increase the difficulty of counterfeiting and improve security. In addition, combining CQDs with other fluorescent dyes to form multiple fluorescent labels can further enhance the anti-counterfeiting effect.

3.5 Application of CQDs in anti-counterfeiting films

CQDs, as a new type of nanomaterial, are widely used in anti-counterfeiting. Adding CQDs to anti-counterfeiting movies can achieve a higher level of anti-counterfeiting protection, which has significant application value.

Preparing CQDs nanofilms is a standard application method. The film consists of CQDs and other polymers or inorganic substances, forming a multilayer structure. By modifying the surface of CQDs, the fluorescence performance and stability of the film can be effectively controlled, thus realizing the function of authenticity identification. Liu et al. prepared CQDs nanofilms for cultural relic protection, which can recognize and record the fluorescence characteristics of the surface of cultural relics to prevent theft or damage. Similarly, adding CQDs to document anti-counterfeiting films can also increase the difficulty of counterfeiting and improve security.

In addition, the anti-counterfeiting performance of CQDs nanofilms can be further improved by

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controlling their thickness and composition. For example, in document anti-counterfeiting films, CQDs nanofilms can be combined with other nanomaterials, such as metal nanowires and two-dimensional materials, to form composite structures to enhance the anti-counterfeiting effect. At the same time, by preparing multilayer structures of CQDs nanofilms, they can have good chemical stability and anti-counterfeiting performance under different conditions.

In summary, CQDs, as a new type of nanomaterial, have a wide range of application prospects in anti-counterfeiting. By adding CQDs to applications such as anti-counterfeiting ink, fluorescent labels, doping materials, barcodes, and anti-counterfeiting films, the anti-counterfeiting performance can be significantly improved, and new ideas and methods for the development of the field of anti-counterfeiting can be provided.

Among them, CQD anti-counterfeiting films use different preparation methods and doping substances to achieve multiple anti-counterfeiting effects and become one of the representatives of new types of anti-counterfeiting materials [31]. To avoid the quenching phenomenon in solution for carbon quantum dots RTP materials, appropriate solvents, surface modification, adding quenching inhibitors, optimizing preparation methods, etc., measures can be chosen.

4. Challenges and future development of CQDs in anti-counterfeiting

Although CQDs have many advantages in the field of anti-counterfeiting, there are still some challenges and difficulties at present. Such as how to realize large-scale preparation and industrial production and solve the matching problem of CQDs with other materials. Here are some problems that need to be solved urgently in the future development of CQDs and some solutions summarized by previous researchers.

4.1 Preparation process needs to be optimized

At present, the preparation process of CQDs still has problems, such as difficult-to-control preparation conditions, easy to be polluted, etc., which need to further optimize the preparation process and improve its controllability and stability. Standardization of raw materials: With the continuous development of anti-counterfeiting technology, standardization of anti-counterfeiting materials also becomes increasingly important. Therefore, it is necessary to establish a complete set of standards for anti-counterfeiting materials for unified management and supervision. The current method for preparing carbon quantum dots is single, low yield, and separation steps are troublesome mainly due to the following reasons: Preparation methods are not mature enough: The current methods for preparing carbon quantum dots mainly focus on traditional methods such as chemical reduction method and electrochemical method, which have a serious difficulty, complex operation, and limited yield. Material sources are limited: Preparing high-quality carbon quantum dots requires high-quality raw materials and fine-processing technology. However, the current material sources are still subject to certain limitations, such as high cost or unstable yield of natural materials or synthetic materials. Separation steps are troublesome: A lot of separation and purification work is required during the preparation process of carbon quantum dots, which requires special equipment and technology, and will cause some environmental pollution.

To solve these problems, we can consider taking the following measures in futural development to improve the efficiency and quality of carbon quantum dot preparation: Explore new preparation methods: For example, using microwave-assisted synthesis, laser ablation, combustion synthesis, and other new methods can reduce the difficulty of preparation, shorten reaction time, improve yield and quality of carbon quantum dots. Choose suitable raw materials: To improve the yield and quality of carbon quantum dots, we need to choose suitable raw materials and preparation conditions, such as using cheap and easy-to-obtain carbon sources, controlling reaction temperature and time, etc. Simplify separation work: Optimizing separation and purification steps, adopting more environmentally friendly and economical processes, and minimizing environmental impact [32].

4.2 The future development of carbon quantum dots requires interdisciplinary integration

In addition, the development of carbon quantum dots anti-counterfeiting materials also needs to be integrated with other disciplines and fields. For example, by drawing on the fluorescent probe technology in biology, the preparation technology in chemical engineering, and the photoelectric property research in physics, more ideas and methods can be provided for the innovation of carbon quantum dots anti-counterfeiting materials. Moreover, while promoting national policies and industrial capital investment, carbon quantum dots anti-counterfeiting materials will also face more opportunities and challenges.

4.3 The best fluorescence performance external conditions of carbon quantum dots damage to the document itself

Carbon quantum dots generally have the best fluorescence performance in neutral or weak alkaline environments. When applied to document anti-counterfeiting, it should also be considered whether the document anti-counterfeiting paper is tamper-proof and extremely sensitive to external pH. For carbon quantum dots that cannot show their best fluorescence performance under neutral conditions, to ensure their fluorescence performance under non-optimal pH conditions, the following measures can be taken: Surface modification: Surface modification of carbon quantum dots, such as adding appropriate functional groups or organic molecules, can adjust their surface charge density and pH responsiveness. This can make CQDs maintain good stability and fluorescence performance in different environments. Nanocomposite materials: CQDs can be combined with other nanomaterials (such as titanium dioxide or zinc oxide) to form a new anti-counterfeiting marking material. These nanomaterials have their special properties, combined with CQDs' optical properties, which can achieve a higher anti-counterfeiting effect. Choose a suitable solution environment: For some pH-sensitive carbon quantum dots, their fluorescence performance can be maintained by choosing a suitable solution environment. For example, using buffer solutions during preparation can make CQDs maintain good fluorescence performance under non-optimal pH conditions [33].

In short, by utilizing surface modification, nanocomposite materials and choosing a suitable solution environment, etc., CQDs' fluorescence performance under non-optimal pH conditions can be improved so that they can be more widely used in fields such as anti-counterfeiting ink.

4.4 Selecting appropriate solvents for preparing carbon quantum dot anti-counterfeiting ink

The problem of clogging caused by slow solvent evaporation: Slow solvent evaporation will cause CQDs and stabilizers to condense and precipitate in print heads resulting in clogging print heads. Currently, there are only methods such as adjusting solvent volatility, increasing print head cleaning frequency, or using higher quality print heads [5] to solve this problem.

In addition, with the continuous development of science and technology, people's demand for anti-counterfeiting materials is constantly increasing; Carbon quantum dot anti-counterfeiting materials will continue to innovate and upgrade. For example, more precise diversified preparation methods and complex multi-level anti-counterfeiting schemes can be explored further, enhancing anti-counterfeiting materials' reliability and security. At the same time, practical applications need to strengthen supervision management over anti-counterfeiting materials and establish improved anti-counterfeit systems to protect consumer rights interests and maintain market order.

In summary, carbon quantum dots are promising anti-counterfeit material with wide application prospects and commercial value; Future research development will inject new vitality into field countermeasures while promoting the application range further expand deepen.

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