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Advances in carbon dots applications: From fluorescent ink and functional paper to the conservation of paper cultural heritage

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ABSTRACT

Paper and ink serve as important media for cultural inheritance and information transmission. With the development of civilization, the research on functional paper and ink has gradually gained attention. Carbon dots (CDs) have attracted widespread interest due to their excellent photoluminescence (PL) properties, strong ultraviolet (UV) absorption, electrical characteristics, and stability. This review summarizes the functionalization applications of CDs in ink and paper, and their applications progress in paper-based cultural relics conservation. To begin, the preparation methods of CDs are introduced. Subsequently, the PL, light absorption mechanisms, and electrical properties of CDs are discussed from the perspective of structure and classification. Then, we discuss the functional applications of CDs with unique optical or electrical properties in paper and ink, and also present the research progress of CDs in the field of paper conservation. Finally, the research development and potential challenges of CDs are presented. It is believed that in the near future, more properties of CDs will be developed, paving the way for the protection, development, and commercialization of paper and ink materials.

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1. Introduction

Paper and ink serve as the carriers of information, carrying the development of civilization and playing a crucial role in cultural inheritance and information transmission. With the passage of time, the paper-based cultural relics experience aging and degradation, so the functional research on paper, ink, and paper conservation has gradually gained more attention, including fluorescent ink [1,2], and functional paper [3,4], and multifunctional paper protection materials. These advancements have been accompanied by extensive exploration of new types of nanomaterials, such as carbon dots (CDs).

It is well known that CDs possess unique physical and chemical properties. Since the first report of fluorescent carbon nanoparticles in 2004 [5], numerous characteristics of CDs have been developed for diverse applications. As an emerging zero-dimensional carbon nanomaterial, owing to excellent biocompatibility, tunable fluorescence, strong ultraviolet (UV) absorption, high loading capacity, good conductivity, and ease of synthesis, CDs have attracted extensive attention from researchers in fields such as energy (catalysis [6–9], lasers [10], capacitors [11], light-emitting diodes [12–15]), optics (anti-counterfeiting [16,17], sensors [18,19]), and biomedicine (biological imaging [20], phototherapy [21]). CDs typically consist of a sp^2/sp^3 hybridized carbon core and an amorphous shell rich in O/N functional groups or polymer chains [22]. Compared to organic and inorganic fluorescent inks, CDs have low cost, environmentally friendly, unique photoluminescence (PL) properties and photostability, enabling versatile anti-counterfeiting printing by utilizing their fluorescence and phosphorescence properties. Moreover, many studies have demonstrated that CDs exhibit excellent electron acceptor/donor properties [23]. Their small size, large surface area, inherent structural defects, and abundant surface functional groups provide active centers or sites [24], thereby endowing paper with functionality [25].

The basic research and application of CDs have always been a hot topic in the fields of chemistry, materials, and other interdisciplinary fields. In recent years, owing to their environmental friendliness, PL properties, strong UV absorption, antioxidant properties, and photostability, CDs have demonstrated great potential in enhancing the functionality of paper and ink, leading to growing research interest in their applications. For instance, Wu *et al.* in-

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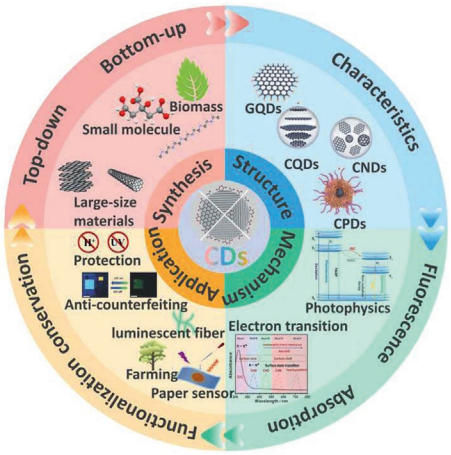


Fig. 1. Schematic diagram of the structure of this review.

troduced the optical properties of CDs and the anti-counterfeiting application of CDs inks based on optical properties and multiple stimuli [26]. Lu's team reviewed the luminescence mechanism and unique luminescent phenomena of CDs, and introduced the application of photoluminescent CDs in anti-counterfeiting [27]. Although many reviews have discussed the properties and applications of CDs, there remains limited comprehensive analysis of their functional application in paper and ink. The review on the role of CDs' excellent performance in paper cultural relics conservation has not been mentioned. Therefore, in this paper, we carefully review the synthesis methods of CDs (Section 2), the structure and classification of reported CDs, the PL mechanism, light absorption mechanism and electrical properties (Section 3), and then summarized functional applications in ink and paper from the perspective of photoelectrical properties (Section 4). Additionally, the latest research progress of CDs in the field of paper cultural relics conservation is described (Section 5). Finally, we present perspectives on the challenges and opportunities for CDs applications in the field of ink and paper (Section 6) (Fig. 1).

2. The synthesis methods of CDs

Achieving controllable synthesis of CDs is a prerequisite for studying their properties, and it is also the foundation for the practical application of CDs. In recent years, the physical and chemical methods for synthesizing CDs are usually classified as "top-down" and "bottom-up" methods.

2.1. Top-down

Top-down approaches involve fragmenting carbon sources (such as carbon fibers, carbon nanotubes, carbon rods, and graphene) into nano-sized carbon chips through various physical and chemical techniques, namely CDs (Fig. 2) [5,28–31] The approaches usually yield CDs with sp^2-hybridized carbon structure or graphite-like lattices. Physical methods include laser ablation, ion etching, arc discharge, and nanolithography by reactive ion etching [32–35]. However, physical techniques often require expensive equipment, which limits their practical applications [6]. One of the most common chemical "cutting" methods is the use of strong oxidizing acids, such as nitric acid and sulfuric acid, where the surface of the carbon chips generated during the chemical cutting process is modified by oxidation groups, thereby endowing the CDs with abundant oxygen functional groups [36–38]. In addition, chemical techniques include hydrothermal/solvothermal methods [39,40], in which graphene or similar materials are dispersed in

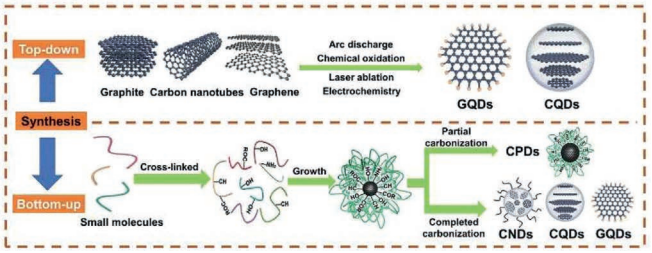


Fig. 2. Schematic illustration of the synthesis of CDs *via* top-down and bottom-up methods. Copied with permission [6]. Copyright 2021, Wiley Publishing Group.

solvents, and reacted under specific high-temperature and high-pressure conditions for a period of time, followed by purification to obtain the final product. Other top-down methods that have been used to synthesize CDs include electrochemical techniques [5,41], microwave/ultrasonic cutting techniques [29], *etc.* Generally, the top-down approaches have the advantages of easy operation, facile surface oxidation, and simple raw materials. However, they also face challenges such as controlling the size and morphology of the CDs.

2.2. Bottom-up

Bottom-up approaches involve further carbonization, dehydration and cross-linking of small molecules, polymers or biomass [42], and it is currently an effective and mainstream method for synthesizing fluorescent CDs (Fig. 2). Firstly, the precursors (small molecules or polymers or biomass) undergo dehydration and polymerization, followed by cross-linking to form carbon polymer dots (CPDs). With further carbonization, a graphite-like crystalline carbon core is produced, with surface attached functional groups or side chains [27]. Common dehydration and carbonization methods include pyrolysis in concentrated acids [43], combustion methods [44], microwave methods [45], hydrothermal/solvothermal methods [46,47], and microwave-hydrothermal methods [48]. Microwave methods enable rapid dehydration and carbonization of the carbon source through microwave treatment, and usually yields the product within a short period of time. The microwave power and reaction time affect the degree of carbonization, resulting in different sizes and emission spectra of the obtained CDs [49]. For example, Tian *et al.* prepared full-color solid-state luminescence (445–643 nm) CDs by controlling the reactant ratio and microwave power, demonstrating how microwave power affected graphitization and conjugated sp^2 domain [49]. The microwave method is simple and efficient, and is thus a commonly used method for kilogram-scale production [45,50]. Hydrothermal/solvothermal syntheses remain currently the most popular approaches. Typically, precursor carbon sources are mixed with solvents (*e.g.*, water, *N*,*N*-dimethylformamide (DMF), ethanol) and reacted at 100–260 °C for 2–36 h. Generally, higher temperatures and longer reaction times can enhance graphitization. Sun *et al.* precisely controlled pyrolysis temperature and reactant ratios to successfully obtain CDs that can cover the entire visible spectra. The increased temperature can promote the degree of citric acid carbonization, thereby causing a redshift in the spectrum [51]. Additionally, element-doped CDs (*e.g.*, N, S, B, F) can be easily prepared through precursors or solvents selection. Feng *et al.* employed carboxymethyl nanocellulose as the carbon source, adding urea, boric acid or glutathione to obtain CDs doped different elements [52]. After purification and post-treatment, CDs synthesized by hydrothermal/solvothermal methods typically exhibit uniform sizes, making them excellent models for the PL mechanism. How-

ever, these methods also suffer from long synthesis time and post-treatment time.

3. Structure and properties of CDs

The discovery of CDs has revolutionized the concept of photoluminescent materials and accelerated research on molecules and quantum dots. The composition and structure of CDs vary depending on precursor and synthesis conditions. However, their key structural features are similar, such as sp^2/sp^3-hybridized carbon cores, surface functional groups, and structural defects. These unique structural features are closely related to the optical properties and physical-chemical properties of CDs.

3.1. Structure and classification of CDs

CDs are a major class of carbon-based nanomaterials, distinct from others like carbon nanotubes or graphene. CDs are generally defined as fluorescent nanoparticles with sizes typically below 10 nm [35]. Structurally, CDs are primarily composed of sp^2/sp^3-hybridized carbon frameworks and may be functionalized with various surface groups, which commonly include O/N-containing groups but are not limited to them [53]. By adjusting synthesis conditions such as reaction temperature and the precursors used, the degree of carbonization and graphitization of the carbon core can vary from amorphous to graphitic crystalline structures [54], thereby controlling the size, crystallinity, and functional groups of CDs. The presence of sp^2 domains in the graphite core and the characteristic lattice of approximately 0.21 nm significantly contribute to the optical and electronic properties of CDs [55]. The surface shell of CDs contains various functional groups including carboxyl, hydroxyl, amino groups, or polymer chains, which heavily depend on the precursors used in the synthesis process. Among them, these diverse functional groups determine the functional performance, stability, solubility and surface modification potential of CDs [56]. Thus, this tunable surface chemistry enables property customization for specific applications by functionalizing the functional groups [57].

Currently, based on distinct formation mechanisms, microstructures of carbon cores, and physicochemical properties, CDs are mainly classified into carbon quantum dots (CQDs), graphene quantum dots (GQDs), carbonized polymer dots (CPDs), and carbon nanodots (CNDs) (Fig. 3a) [6,54,58]. These classifications can be interrelated by modifying graphene layers and carbonization degrees (Fig. 3b). GQDs are composed of disc-shaped single-layer/multi-layer graphene sheets and exhibit anisotropy properties with the lateral size (usually $<$10 nm) exceeding their height. They have feature graphene lattices with surface functional groups, and their photophysical phenomena are mainly determined by size (or emission π-domain dimensions) [39,54,59]. CQDs exhibit quasi-spherical morphology, and possess crystallinity derived from the graphene lattices and interlayer stacking. CQDs have functional groups on the surface, and exhibit photophysical characteristic depend on carbon core size [60]. CQDs can be distinguished from GQDs *via* atomic force microscopy (AFM) and high-resolution transmission electron microscopy (HRTEM) structural analysis [61]. CNDs are quasi-spherical and have a carbonized structure (lacking distinct crystallinity), with Surface chemical groups determine functionality. Finally, CPDs are quasi-spherical with unique core-shell nanostructure, consisting of a carbon-based core and functional group/polymer chain-rich shell [62,63]. CPDs were first proposed in 2018 based on the formation process, structure, and PL mechanism [64,65], and their optical properties mainly originate from the molecular state and crosslink-enhanced emission (CEE) effect [35,66].

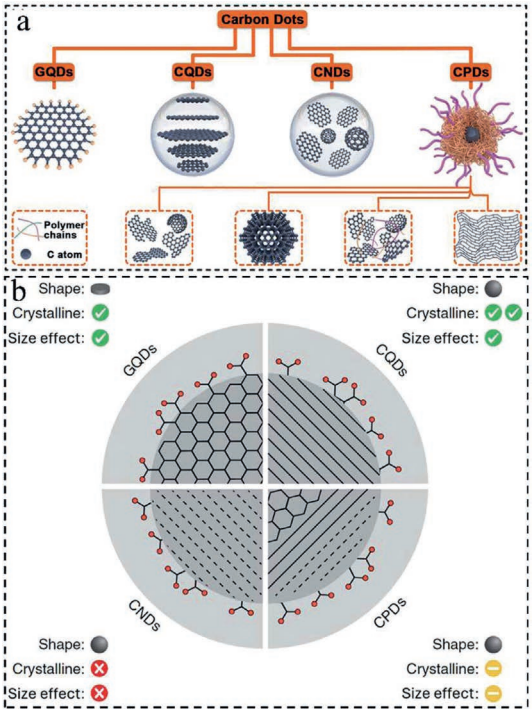


Fig. 3. (a) Schematic illustration of classifications and corresponding structures of CDs. Copied with permission [58]. Copyright 2021, Wiley Publishing Group. (b) Classification of CDs based on their physicochemical and photophysical properties of CDs. Copied with permission [54]. Copyright 2022, Springer Nature.

3.2. Photoluminescence properties

CDs have emerged as promising alternatives to traditional quantum dots containing toxic elements (*e.g.*, CdS and CdSe), with their optical properties being one of the most attractive features. The interesting photoluminescent properties are due to their structural diversity, which endows CDs with multiple optical properties [22]. The PL of CDs is generally classified into two fundamental forms, fluorescence and afterglow (including delayed fluorescence and phosphorescence) [27,67]. As clearly illustrated in Fig. 4a, the intrinsic mechanisms can be understood through the indicated electronic transitions (represented by arrows) between different energy states [68].

3.2.1. Fluorescence mechanism of CDs

Since the discovery of CDs, researchers have been actively speculating on their PL mechanisms. Currently, several theories have been proposed to explain the fluorescence mechanisms of CDs, namely molecular state emission, surface state emission, size effect and CEE effect, and these mechanisms occur during different formation processes of CDs (Fig. 4b) [27,69,70]. Firstly, during the process of small molecule polymerization to form polymers, the emission mechanism primarily tends to the molecular state. The molecular fluorophores attached to the surface or core of the carbon nucleus emit fluorescence. Sun's research group synthesized red emissive CDs from *o*-phenylenediamine and catechol system, and observed that the CDs exhibited the same PL mechanism and behaviors as the isolated 5,14-dihydroquinoxalino[2,3-*b*] phenazine (DHQP). Based on these, it is deduced that DHQP is remained or linked to the surface of CDs, indicating that the PL mechanism of the CDs originates from molecular state fluorescence (Fig. 4c) [71].

With the formation of CDs polymers structures, the CEE effect becomes increasingly prominent, a phenomenon first proposed by Yang *et al.* [72]. The CEE mechanism represents a luminescence enhancement effect induced by crosslinking, which operates

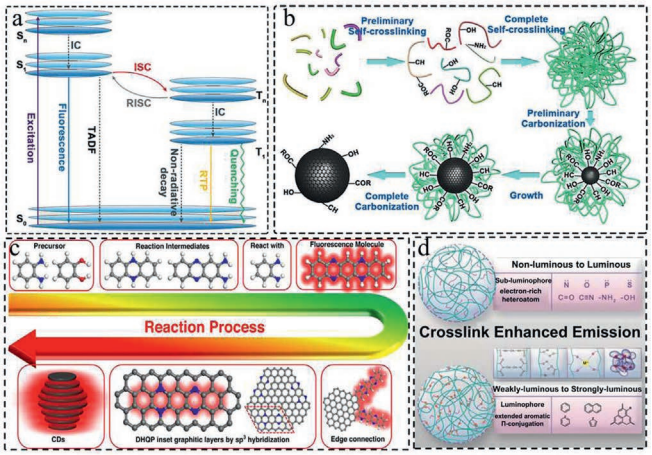


Fig. 4. (a) The simplified Jablonski diagram describes the key photophysical processes in CDs. Copied with permission [68]. Copyright 2020, Springer Nature. (b) The formation process of CDs in the bottom-up process. Copied with permission [27]. Copyright 2022, Elsevier. (c) The PL mechanism of the CDs: molecular state fluorescence. Copied with permission [71]. Copyright 2022, Springer Nature. (d) The PL mechanism of the CDs: CEE effect. Reproduced with permission [73]. Copyright 2020, Wiley Publishing Group.

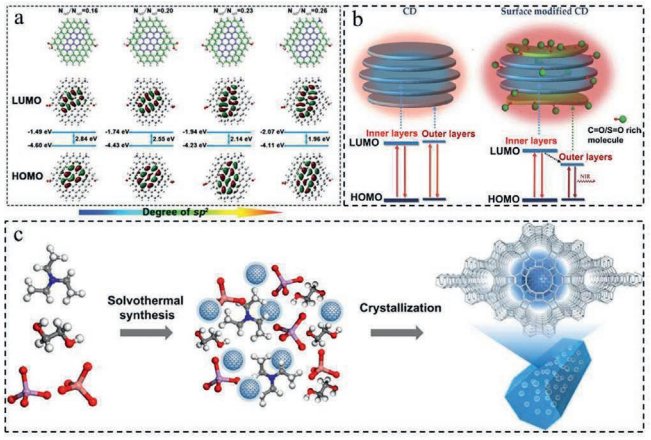


Fig. 5. (a) The PL mechanism of the CDs: sp^2/sp^3 hybridized domains. Copied with permission [75]. Copyright 2020, Wiley Publishing Group. (b) The PL mechanism of the CDs: surface state. Reproduced with permission [77]. Copyright 2018, Wiley Publishing Group. (c) Afterglow mechanism of CDs. Copied with permission [86]. Copyright 2017, AAAS.

through the combined processes of promoting radiative transitions and suppressing non-radiative transitions (Fig. 4d) [73]. Maser and co-workers conducted controlled polycondensation of citric acid and ethylenediamine, and combined with density functional theory (DFT) calculations, which demonstrated that the supramolecular interactions mediated by hydrogen bonds between polyamide chains are the fundamental origin of the universal blue luminescence observed in polymer CDs [74].

With increasing carbonization degree, carbon cores gradually form, and the fluorescence emission mechanism transforms into the size and surface state. The dimensions of the conjugated domains within the carbon cores influence luminescence bandgap, thereby determining the fluorescence emission properties of CDs. For example, Lu *et al.* developed a synthesis method to precisely control the fluorescence emission of CDs by tuning the sp^2/sp^3 hybridization ratio in the carbon cores under the guidance of DFT (Fig. 5a) [75]. The type and quantity of surface functional groups affect the electronic transition, the parameters of ground state and excited state, thereby changing the fluorescence emission properties of CDs [76]. Rogach and colleagues modified CDs with sulfonyl/carbonyl electron-accepting molecules through surface engineering strategy, thereby achieving near-infrared emission (absorption peak 715–724 nm, emission peak 750–760 nm). The functional groups bonded to the CDs surface and edges tune the luminescence bandgap, thereby promoting the electronic transition under near-infrared excitation (Fig. 5b) [77]. Additionally, both the carbon cores and surface states can jointly influence the fluorescence emission of CDs. Miao *et al.* reported a method to precisely tune emission wavelengths (430–630 nm) of CDs by adjusting the pyrolysis temperature and reactant ratios, thereby controlling the degree of graphitization and surface carboxyl functional group density, achieving full-spectra fluorescent CDs [51].

3.2.2. Afterglow mechanism of CDs

Both room-temperature phosphorescence (RTP) and thermally activated delayed fluorescence (TADF) belong to afterglow processes. RTP is a form of phosphorescence characterized by long-lived emission from the triplet state that remains observable at room temperature, following intersystem crossing (ISC) from the photoexcited singlet state [78]. If the energy gap between the singlet and triplet excited states is sufficiently small (<0.3 eV), the triplet excited state can undergo reverse intersystem crossing (RISC) to the excited singlet state by absorbing thermal energy, subsequently emitting delayed fluorescence when returning to the ground state [79,80]. Although fluorescence and TADF exhibit similar optical bandgaps, they display opposite temperature-dependent emission behaviors [81], allowing their differentiation by temperature-dependent spectroscopy. Generally, achieving RTP in CDs remains challenging. Current strategies focus on enhancing ISC efficiency, reducing non-radiative transitions by restricting the rotation or vibration between molecules through an external matrix, or limit the luminescent centers by self-crosslinking of CDs [68,82–84]. The realization of TADF requires a singlet-triplet energy gap typically <0.3 eV [85]. For example, Yu *et al.* proposed a zeolite-encapsulated CDs strategy to successfully prepare TADF materials with an ultra-long lifetime (350 ms). The energy gap of the CDs@zeolite composites is 0.22–0.23 eV, and the zeolites nanoconfined space effectively stabilize the triplet states of CDs, thereby achieving TADF behavior (Fig. 5c) [86].

3.3. Light absorption of CDs

CDs always exhibit different light absorption behaviors. However, they consistently demonstrate strong light absorption in the UV region and extend to the visible light and even near-infrared regions. Therefore, CDs are often explored as UV blockers in application fields. CDs are rich in conjugated π-electron systems, enabling them to absorb photoelectrons under UV light. The abundant surface groups of CDs can further enhance the UV absorption intensity and extend the visible light absorption ability. Fig. 6a illustrates the correlation between the absorption spectrum and electronic transition of CDs. The absorption band (<300 nm) belongs to the π-π^* transition of C=C bonds, and Band II (300–400 nm) can be attributed to the n-π^* transition of C=O bonds in the carbon core. Band III-V absorption bands result from surface-state transitions involving lone-pair electrons. Notably, some near-infrared-emitting CDs usually possess π-conjugated electrons in sp^2 domain or connected surface groups/polymer chains, which accounts for long-wavelength absorption in the 500–800 nm range [62].

The absorption properties of CDs are primarily influenced by the size of the π-conjugated domains, the presence of aromatic rings, the types and contents of surface functional groups, and variations in O/N content in the carbon core. The extent of conjugated aromatic rings in the carbon core determines the energy

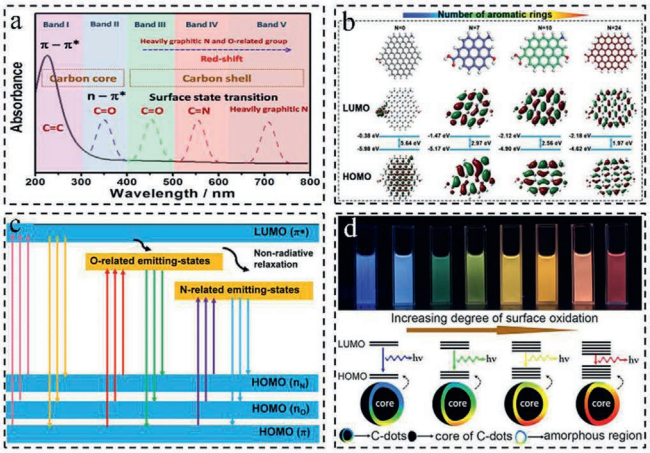


Fig. 6. The effect of CDs surface groups, aromatic rings, oxidation degree on absorption. (a) Schematic illustration of the relationship between electron transition and the absorption spectrum. Copied with permission [96]. Copyright 2020, Springer. (b) HOMO and LUMO states of the established model by increasing the aromatic rings. Copied with permission [75]. Copyright 2020, Wiley Publishing Group. (c) Influence of surface functional groups on the optical properties of CDs. Copied with permission [55]. Copyright 2025, Wiley Publishing Group. (d) Influence of different degrees of oxidation on the tunable PL of CDs. Copied with permission [90]. Copyright 2016, American Chemical Society.

gap between ground and excited state, and CDs with extended π-electron systems demonstrate reduced surface-state energy gaps through effective π-electron/surface-state coupling [87]. Through DFT calculations, Lu *et al.* demonstrated that with increasing number of benzene rings, the energy gap between the highest occupied molecular orbital (HUMO) and lowest unoccupied molecular orbital (LUMO) decreases, resulting in red-shifted absorption and emission spectra (Fig. 6b) [75]. Conversely, smaller CDs or those with fewer aromatic rings exhibit a larger HUMO-LUMO energy gap, leading to blue-shifted fluorescence [88]. Some studies have suggested that the surface modification through synthesis method, precursor selection, and element doping of CDs affect the types and quantities of surface functional groups, which directly influence the parameters of the ground state and excited state. For example, Ibrahim *et al.* using DFT calculations demonstrated that electron-donating groups (*e.g.*, hydroxyl and amino groups) can reduce the LUMO energy level, thereby narrowing the HUMO-LUMO energy gap. Notably, oxygen passivation at the edges of hexagonal nanodots was found to dramatically decrease the energy gap (Fig. 6c) [55,89]. Furthermore, enhanced surface oxidation effectively narrows surface-state bandgaps [87]. Xiong *et al.* prepared CDs with tunable PL properties and found that the carbon cores had similar particle size distribution and graphite structure. As the degree of surface-state oxidation increased, the band gap of CDs gradually decreased, and their absorption spectra also showed significant differences in the low-energy region (Fig. 6d) [90]. Other factors, such as structural or energy disorder, environmental effects, deprotonation, and exciton coupling, contribute minimally to absorption characteristics [91]. Therefore, through the mechanism of electron transition between the carbon core and surface functional groups, CDs can absorb specific wavelengths of light and convert it into other energy forms, making them valuable for anti-counterfeiting, UV-blocking materials, optical sensors, and photocatalytic fields [55,92–95].

3.4. Electrical properties of CDs

The conductivity of CDs is correlated with their structure, size, and surface functional groups. Generally, highly graphitized CDs exhibit superior conductivity because the conjugated structure formed by the sp^2-hybridized carbon atoms inside can facilitate rapid electron transfer [97]. In contrast, amorphous or non-crystalline CDs demonstrate relatively poorer conductivity. Notably, Surface functional groups and intrinsic defects can significantly reduce the conductivity of CDs. CDs possess remarkable charge transfer capabilities, displaying excellent electron transport characteristics in electrochemical reactions. The unique property has prompted researchers to integrate CDs with other highly conductive materials to enhance overall conductivity, thereby expanding their catalytic applications [98]. For instance, CDs with abundant functional groups on their surfaces can be used to fix Ru1CoP nanocrystals. After combination, they exhibit extremely high conductivity, activity and stability, thereby enhancing the efficiency of electrocatalytic hydrogen evolution [99]. Therefore, the electrical properties of CDs are often exploited to impart or enhance the functionality of materials.

4. Application of CDs in the functionalization of ink and paper

The absorption and emission properties of CDs can be precisely adjusted across a broad spectral range from UV to near-infrared. Through the selection of precursors, their emission types can range from fluorescence to phosphorescence, and even up-conversion. These diverse and fascinating properties of CDs make them highly attractive in the field of paper and ink materials. Currently, studies have reported the biocompatibility and environmental friendliness of CDs, confirming no threat to humans or the environment. This paper mainly summarizes the applications of CDs in paper and ink systems, including fluorescent ink [52], and functional paper.

4.1. Fluorescent ink

CDs exhibit typical PL properties, including fluorescence, RTP, and TADF. CDs are widely used as fluorescent inks, with anti-counterfeiting ink representing the primary direction. As early as 2017, Nuryantini *et al.* prepared CDs powder *via* pyrolysis, and combined it with polyvinyl alcohol and chitosan to obtain the best viscosity anti-counterfeiting ink, which could be used in commercial printers [100]. Li *et al.* fabricated a writable CDs anti-counterfeiting ink, where the CDs have the property of reversible oxygen cycling, with $Na_2S_2O_4$ solution as the quencher and oxygen as the restorer. Using the CDs solution, they created a landscape painting (Fig. 7a) with rhodamine-encrypted text “AAU FMBA”. Under UV light, a red landscape painting was displayed. While in $Na_2S_2O_4$ solution, the landscape painting disappeared, revealing the encrypted text. After reading the information, it was placed in an oxygen environment, and the quenched CDs were restored to fluorescence under the action of oxygen, achieving re-encryption of the information. This writable security ink demonstrates multiple anti-counterfeiting capabilities [101]. The afterglow properties of CDs enable diverse printing security applications [102]. Wei *et al.* first reported F, N-doped CDs with a long-lived triplet excited state, which is sensitive to pH value. Under acidic conditions, pH-responsive green RTP-CDs showed phosphorescence quenching, while the phosphorescence property is not affected under alkaline conditions. As shown in Fig. 7b, using fluorescent inks of different pH values to print graphics on a desktop inkjet printer, a blue error message “888” and a green leaf tree were displayed under UV light. when the UV light was turned off, a green correct message “609” and a bare tree were displayed. Additionally, QR code, text, and digital photos of buildings were printed on filter paper *via* using CDs ink, all with good resolution (Fig. 7c), indicating that the CDs inks have the potential for advanced anti-counterfeiting applications [103]. Ren *et al.* synthesized CDs with time-dependent phosphorescence colors using levofloxacin as the raw material by the

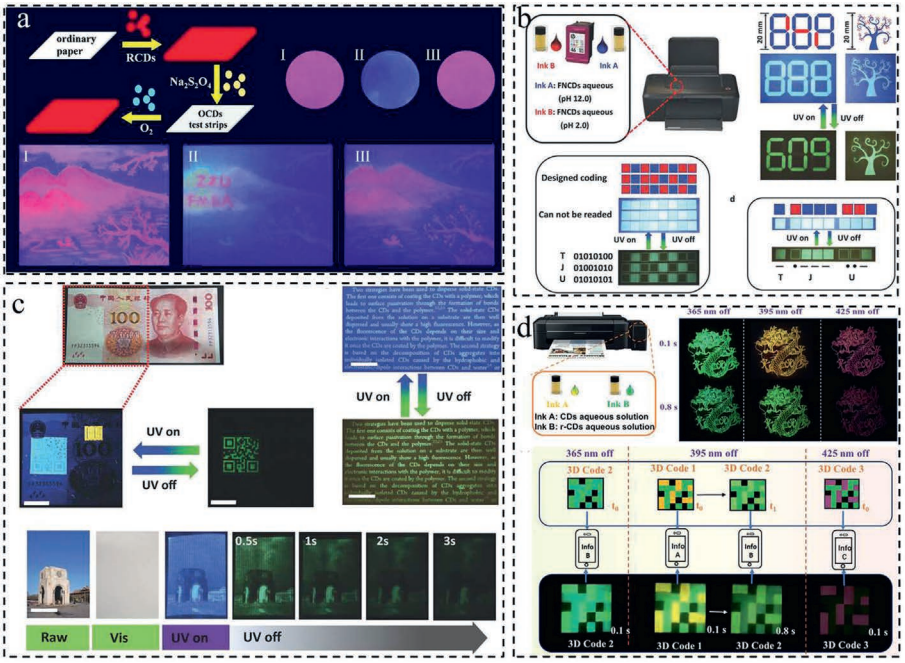


Fig. 7. Examples of applications of CDs as fluorescent ink. (a) The anti-counterfeiting application of writable CDs ink. Copied with permission [101]. Copyright 2021, Royal Society of Chemistry. (b, c) Multifunctional anti-counterfeiting applications of printable CDs ink Based on pH Response. Copied with permission [103]. Copyright 2018, Wiley Publishing Group. (d) Advanced dynamic information encryption application of printable CDs ink with time-dependent property. Copied with permission [16]. Copyright 2021, Wiley Publishing Group.

one-pot hydrothermal method. When this CDs ink was printed on paper (CDs@paper) and exposed to light of different wavelengths, the activated phosphorescent colors of CDs@paper were different. Based on the unusual phosphorescent properties of CDs, they are beneficial for advanced dynamic information encryption (Fig. 7d) [16].

Beyond anti-counterfeiting applications, CDs inks have demonstrated versatile utility in solvent-free printing, agricultural field, and daily life applications. For instance, Yang *et al.* were the first to report solvent-free processing of CDs *via* liquid crystallization, enabling the development of liquid crystal CDs (LC-CDs) emitting red, green, and blue primary colors. By grafting flexible alkyl chains onto CD surfaces *via* weak supramolecular van der Waals interactions, they induced a phase transition from crystalline to liquid crystal states. The breakthrough facilitated direct ink writing (DIW) using LC-CD inks. They also successfully produced bright fluorescent lines using designed printing system (injection pump, syringes with needles, and temperature control devices), and by further optimizing materials and parameters further improved resolution. This research paves the way for solvent-free security labeling and advanced manufacturing (Fig. 8a) [104]. Akbulut *et al.* designed a CDs ink formulation with tuned viscosity using polyethylene glycol and hydroxypropyl cellulose, and encapsulated the CDs ink in biodegradable shells *via* DIW. The results showed that the treated plants exhibited growth rates ~2.5-fold faster than the control group, which was attributed to the efficient photosynthetic efficiency and strong UV absorption ability of the CDs (Fig. 8b) [105]. Qu *et al.* synthesized CDs with a photoluminescence quantum yield (PLQY) close to 100% in aqueous solution *via* solvent-free dry pyrolysis and confirmed that the CDs have biological safety. The CDs aqueous solution was used as a fluorescent ink, which could well adsorb on various natural fibers, such as silk fibers, mouse hair, low-density artificial fibers, and cotton fibers. A practical demonstration featured CDs-stained silkworm cocoon covers paired with an operating blue LED chip, creating sustainable art pieces (Fig. 8c) [106].

4.2. Functional paper

CDs are currently a hot topic in the design of novel fluorescent probes. Combining CDs with paper to form CDs-based paper sensors allow for quantitative or semi-quantitative detection of targets through visual inspection or handheld devices, which are often applied in fields such as food safety testing, environmental monitoring, and medical diagnosis. For example, Liu *et al.* reported the *in-situ* synthesis of dual-emissive CDs on cellulose paper, thereby obtaining a fluorescent paper-based analytical device capable of detecting Cu^{2+} in drinking water, fruits, and vegetables. This fluorescent probe array exhibits high-sensitive detection performance for Cu^{2+} (limit of detection: 18.4 nmol/L), and the color changes of its emission spectra enable visual detection and quantitative measurement on smartphones (Fig. 9a) [107]. The concept of microfluidic paper-based analytical devices (μPADs), first introduced by Whitesides' team in 2007 [108], has gained significant research traction. Qian *et al.* developed a fully inkjet-printed CDs μPADs using three fluorescent CDs as sensors for simultaneous detection of food additives (vitamin C (AA), NO^{2-}, and sunset yellow (SY)), demonstrating excellent visual detection performance. During detection process, the obvious changes in fluorescence quenching meets the requirements of actual detection and can recognize the detection effect with the naked eyes. (Fig. 9b) [109]. Jiang *et al.* integrated the prepared blue light CDs and gold nanoclusters to develop a novel enzyme-free rapid paper sensor, and developed a portable smartphone platform integrated paper sensor for in-site visual quantitative detection of glyphosate (limit of detection 4.19 nmol/L). This approach was successfully applied to detect glyphosate (Fig. 9c) [110]. Lee *et al.* prepared Fe-doped CDs with high peroxidase enzyme activity by a microwave-assisted method, and fixed the CDs on the fiber nanofibrils through hydrogen bond interactions to produce a composite nanopaper. This composite nanopaper can be used for colorimetric detection of hydrogen peroxide and glucose (limit of detection: 0.93 and 1.73 μmol/L). The color changes of the

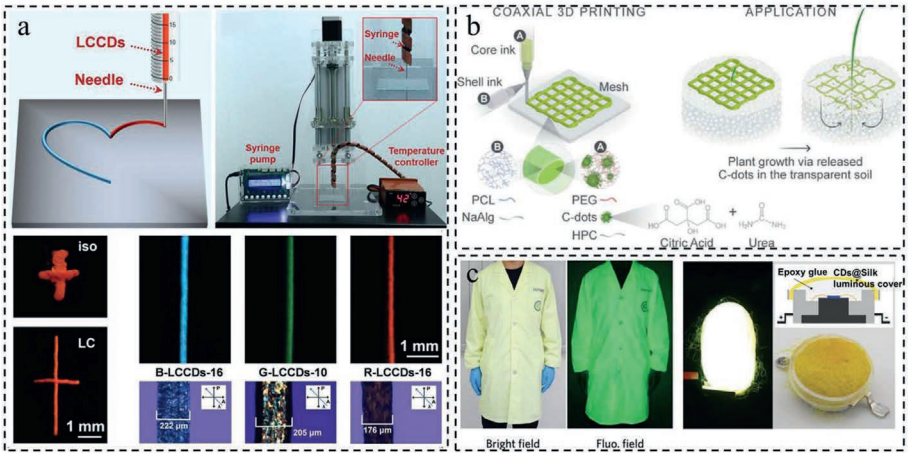


Fig. 8. Examples of applications of CDs as fluorescent ink. (a) The DIW method based on LC-CDs. Copied with permission [104]. Copyright 2023, Wiley Publishing Group. (b) CDs ink promotes crop growth. Copied with permission [105]. Copyright 2023, American Chemical Society. (c) Exquisite, eco-friendly handicraft with CDs ink. Copied with permission [106]. Copyright 2024, Wiley Publishing Group.

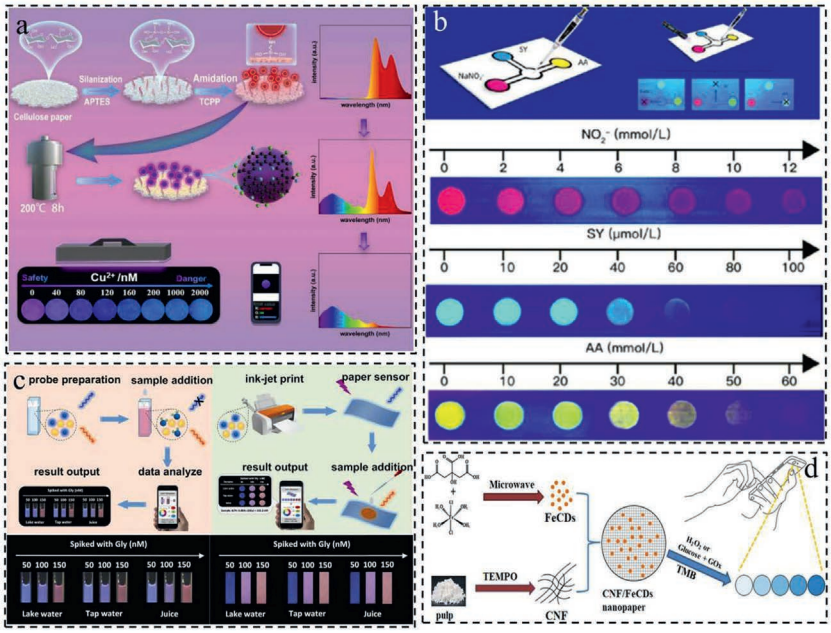


Fig. 9. CDs-based paper sensors: (a) Cu^{2+} paper sensor. Copied with permission [107]. Copyright 2025, Elsevier. (b) Food additive paper sensor. Copied with permission [109]. Copyright 2021, American Chemical Society. (c) Glyphosate paper sensor. Copied with permission [110]. Copyright 2022, Elsevier. (d) Hydrogen peroxide and glucose paper sensor. Copied with permission [111]. Copyright 2021, Elsevier.

nanopaper can be monitored by a smartphone, can be reused ten cycles, and has a shelf life of more than one month (Fig. 9d) [111].

CDs, as an information-encrypted ink, are printed onto paper substrates to endow the paper with anti-counterfeiting capabilities, which is conducive to the development of anti-counterfeiting labels in commercial applications. Qu *et al.* subjected levofloxacin to thermal treatment in melting boric acid at atmospheric pressure, and discovered that the resulting product exhibited RTP-TADF dual-mode afterglow. Furthermore, by printing CDs ink on paper substrates, the CDs@paper showed significant thermochromic properties. As the temperature increased from 193 K to 423 K, the persistence time of the afterglow pattern of CDs@paper shortened, the color changed from yellow to cyan, and then to blue. After being stored under ambient conditions for 2 months, the thermochromic behavior of the CDs@paper could still be reproduced. Based on the phosphorescence and TADF intensity ratios at different temperatures, CDs@paper demonstrated temperature sensing capability over a wide temperature range of 223–403 K. In addition, researchers used this dual-mode afterglow CDs and a single-mode phosphorescent CDs that had been reported as ink to print each area with 3D encoded patterns, ultimately presenting a printed three-dimensional dynamic code containing color and temperature information (Fig. 10a) [112]. The surface of CDs contains a large number of functional groups, which can also play an important role in electrochemical performance and devices. For example, Lu *et al.* anchored CDs on the reduced graphene oxide surface and synthesized an independent and flexible 3D carbon-based composite material (CDs@rGO paper). The CDs@rGO paper (Fig. 10b) can be used as a high-performance potassium-ion batteries anode. The unique 3D structure has efficient electron and ion transport channels, thereby enhancing the reaction kinetics. CDs provide oxygen-containing functional groups and abundant defects, which can improve the electrochemical performance. The results show that this flexible anode has the high capacity of 310 mAh/g at 100 mA/g, an

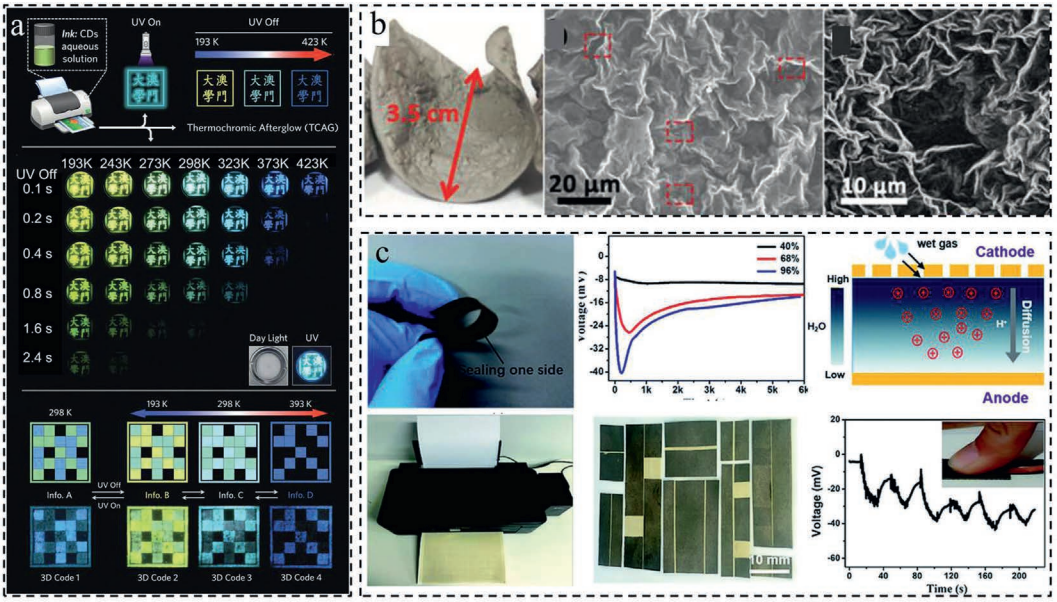


Fig. 10. Functional papers: (a) Advanced dynamic information encryption function paper. Copied with permission [112]. Copyright 2024, Wiley Publishing Group. (b) Prepared flexible, freestanding CDs@rGO film paper. Copied with permission [113]. Copyright 2020, Wiley Publishing Group. (c) Flexible moisture-electric energy transformation device. Copied with permission [25]. Copyright 2018, Royal Society of Chemistry.

extremely long cycle life (840 cycles with a capacity of 244 mAh/g at 200 mA/g), and excellent rate performance (undergo six consecutive currents changing from 100 mA/g to 500 mA/g, high capacity 185 mAh/g at 500 mA/g) [113]. In terms of fully exploiting the electrical properties of CDs, Zhang *et al.* reported a method of fabricating flexible moisture-electric energy transformation device by direct printing or dispersing CDs on paper. The team constructed an asymmetric structure on both sides of the porous paper to establish an ionic gradient. Under certain air humidity, the different water molecule adsorption on the two sides of CDs/paper led to the formation of an ionized carboxyl gradient, thereby generating a potential drop across the device. The results showed that the fabricated device could produce a continuous voltage of −40 mV at fixed relative humidity value for over 100 min. Furthermore, the device exhibited responsive behavior to direct contact with human fingers or breath, highlighting its potential applications in electronic skin and humidity sensors (Fig. 10c) [25].

5. Application of CDs in the conservation of paper cultural relics

In recent years, paper cultural relics have carried great historic and artistic value, leading to find out the effective protection materials and preventive methods for paper relics. Currently, paper cultural relics are experiencing aging and degradation during their long-term preservation, primarily due to the acidification and photo-induced degradation of cellulose [114]. People have explored the use of alkaline materials (*e.g.*, magnesium oxide, magnesium hydroxide, calcium hydroxide or amino silanes) for paper deacidification *via* the liquid-phase deacidification method [115]. However, for UV protection of paper, the variety of UV shielding materials applicable for paper protection and research on the protective effects are limited [93]. CDs are suitable for paper deacidification and preservation due to the tunability of their surface functional groups. Furthermore, CDs possess excellent UV absorption properties, environmental friendliness, and low toxicity. Therefore, they can be utilized as UV-blocking materials for paper, and are expected to offer a multi-functional protection effect for paper-based cultural relics. Yu *et al.* synthesized chitosan-based polymer dots through the hydrothermal method, which demonstrated good stability within the pH range of 4–12. The researchers applied them on Xuan's paper and found that the paper exhibited strong resistance to UV performance under UV aging [116]. To further explore the anti-UV protection effect of CDs on paper, our research group developed a novel type of CDs with high stability and strong UV absorption in 2024. The paper was immersed in the CDs solution, and the CDs were attached to the paper fibers. The accelerated aging experiment demonstrated that the CDs endowed the paper with the functions of acid resistance and UV protection (Fig. 11a). The paper coated with CDs exhibited green fluorescence and green RTP when exposed to 365 nm excitation. This not only demonstrated the binding of the CDs to the paper fibers, but also provided a broad application in anti-counterfeiting [117]. The unique photoluminescence properties endow CDs with practical utility and convenience as protective identifiers. As shown in Fig. 11b, CDs can serve as a structure-directing agent and be embedded into Mg-Al layered double hydroxide (LDH) materials *via* an *in-situ* growth strategy. This approach effectively thins the LDH while enhancing the UV absorption properties of the nanohybrids. The results demonstrated that the nanohybrids can slow down the acidification, oxidation, and yellowing degradation processes of paper under accelerated UV aging and dry-heat aging conditions. Furthermore, we also proposed that this nanohybrid material mainly achieved the effect of preventing paper from UV aging by absorbing UV and emitting low-energy light, thereby reducing the direct damage of UV to the paper fibers [93]. These studies have opened up new directions for the application of CDs in the conservation of paper cultural relics. Meanwhile, to meet the diverse preservation needs, our research group has also explored other methods using CDs for the preventive conservation of paper cultural relics. We proposed a highly safe and flexibly customizable method that utilized CDs films to prevent paper from UV damage. By compositing CDs with polyvinyl alcohol, the CDs films with highly tunable UV absorption and low visible light absorption was obtained. The content of CDs can be precisely adjusted to control the UV transmittance, thereby achieving personalized protection effects tailored to specific preservation and exhibition requirements (Fig. 11c) [118]. The structure of CDs determines their properties, and current stud-

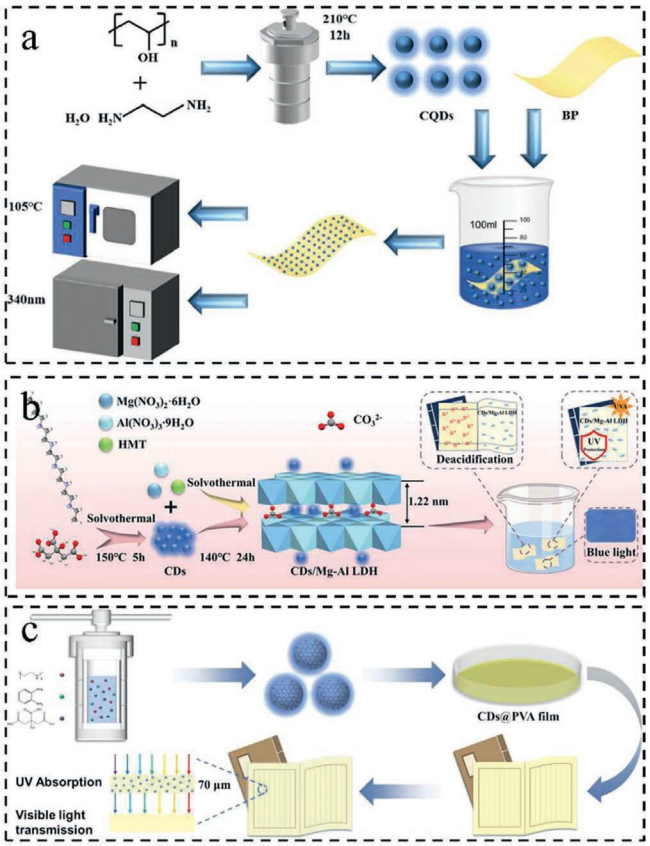


Fig. 11. Application of CDs in the conservation of paper cultural relics. (a) CDs solution for paper protection. Copied with permission [117]. Copyright 2024, American Chemical Society. (b) CDs-based composite nanosheets for paper protection. Copied with permission [93]. Copyright 2024, Wiley Publishing Group. (c) CDs films for paper preventive protection. Copied with permission [118]. Copyright 2025, Elsevier.

ies have demonstrated that their performances match well with the requirements for paper cultural relics conservation. However, given the various factors that cause paper aging and the initial stage of research on CDs' protection of paper, CDs still face issues and challenges such as long-term stability, biological and environmental safety, and the impact on the handwriting, ink, and surface state of the paper. It is believed that in the future, more researchers will pay more attention to and solve the related problems and challenges encountered in paper protection, and will also focus on modulating the structure of CDs to explore their multifunctional practical applications in the conservation of paper cultural relics.

6. Summary and perspectives

In recent years, several research teams have gradually conducted exploratory work on the functional applications of CDs in paper and ink systems. This review systematically summarizes the synthesis methods of CDs, including top-down (large-size materials) and bottom-up (small molecules, polymers, or biomass) strategies, aiming to provide options for industrial production of CDs. Then, starting from the classification and structure of CDs, we elucidate their fluorescence and afterglow mechanisms, light absorption properties, and relevant electrical characteristics. Finally, we highlight the functional applications of CDs based on the above optical and electrical properties in paper and ink materials, including anti-counterfeiting inks, paper-based sensors, the agricultural field, luminous handicrafts, and also emphasize the innovative research of CDs in the field of paper cultural relics conservation.

Although plenty of results have been achieved to date, research into CDs ink and CDs paper materials is still in its nascent stage. More discussions are needed regarding the future research and application trends.

(1) To meet the demands of industrial applications, large-scale production of CDs is essential. Currently, the synthesis methods of CDs still face several limitations, such as low yield and complex purification processes, which hinder the demand for industrial application of CDs. For large-scale production of CDs, primary considerations include: (i) scaling up production capacity and control costs through effective approaches, (ii) ensuring both high yield and quality of the produced CDs, and (iii) developing efficient purification protocols.

(2) To meet the needs of various practical applications, it is necessary to design the required structures (*e.g.*, size, crystallinity, defects, shape, functional group position and quantity) of CDs and study the reaction mechanism of CDs. Due to the different synthesis methods and pathways, the precise nucleation mechanisms and formation processes are unclear. It is imperative to develop *in situ* characterization techniques (such as *in-situ* Raman spectroscopy, *in-situ* Fourier Transform infrared spectroscopy) to elucidate CDs formation mechanisms, optimize reaction conditions, and systematically explore the influence of CDs synthesis on structure and performance.

(3) Due to variations in synthesis conditions and precursors materials, the PL mechanism of CDs remains controversial. Although multiple PL mechanisms of CDs have been reported, a deeper understanding of their structure-property relationships requires more advanced technical means (*e.g.*, spherical aberration-corrected electron microscopy, synchrotron X-ray radiation) and more research (including computational simulations and machine learning approaches) to conduct more in-depth theoretical studies and clarify the precise structure.

(4) Numerous studies have confirmed that CDs have the potential to be used in the next generation security and anti-counterfeiting inks. To achieve better practical applications, it is beneficial for information protection to establish secure and reliable security labels. This requires not only the stability of CDs, but also the ease of realization and guarantee of the stimulation conditions and guaranteed safety during their usage. Therefore, for anti-counterfeiting and data security applications, it is recommended to develop secure inks with specific excitation and emission, short response times, and high stability.

(5) The application of CDs in paper and ink products are exposed to people. Therefore, the safety during the use of CDs needs to be considered. Currently, most biological toxicity experiments have reported that low concentrations of CDs have no obvious toxicity to human cells and animals. However, people still worry about the safety of CDs. Standardized tests for the biological toxicity of CDs and guidelines for the usage dosage should be established to ensure the safety of their application.

(6) CDs possess excellent properties including UV absorption properties, stability, tunable luminescence, small size, large specific surface area, and modifiable surface functionality, making them highly promising for expanded applications in paper and ink materials. Our research group has leveraged the robust UV absorption capability of CDs to develop paper with integrated deacidification and UV-shielding functions. Furthermore, CDs exhibit additional advantageous characteristics such as antioxidant, antibacterial, and flame-retardant

properties. Exploiting these multifunctional features is expected to enable broader functional applications of CDs in advanced paper and ink systems.

We believe that with the continuous advancements by scientific researchers, controllable synthesis and large-scale production of CDS will be achieved. These advancements will significantly expand the application scope of CDs in paper and ink materials field.

CRediT authorship contribution statement

Jinchan Zhao: Writing – review & editing, Writing – original draft, Validation, Methodology, Investigation, Formal analysis, Conceptualization. **Zhenyu Dai:** Writing – review & editing, Investigation, Formal analysis, Conceptualization. **Bojun Zhang:** Writing – review & editing, Investigation, Formal analysis, Conceptualization. **Yizhuo Li:** Writing – review & editing, Investigation, Formal analysis, Conceptualization. **Mingliang Zhang:** Writing – review & editing, Investigation, Conceptualization. **Shiliang Mei:** Resources, Project administration, Investigation, Conceptualization. **Wanlu Zhang:** Investigation, Funding acquisition, Conceptualization. **Sinong Wang:** Writing – review & editing, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Conceptualization. **Ruiqian Guo:** Supervision, Project administration, Methodology, Investigation, Funding acquisition, Conceptualization.

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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