Nanoscale Horizons

COMMUNICATION



View Article Online

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Cite this: DOI: 10.1039/c8nh00247a

Received 17th August 2018, Accepted 1st November 2018

DOI: 10.1039/c8nh00247a

Carbon dots produced *via* space-confined vacuum heating: maintaining efficient luminescence in both dispersed and aggregated states[†]

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Aggregation-induced quenching (AIQ) of emission is an obstacle for the development of carbon dots (CDots) for solid-state luminescent devices. In this work, we introduce a method to avoid AIQ and to produce highly luminescent CDots through a space-confined vacuum heating synthesis. In the presence of CaCl₂, a mixture of citric acid and urea forms an inflated foam under vacuum heating at 120 °C. Upon gradually increasing the heating temperature to 250 °C, blue emissive molecular species are first formed, and are then transformed into uniform-sized green emissive CDots through dehydration and carbonization processes taking place in the confined ultrathin spaces of the foam walls. The green luminescence of these CDots originates from conjugated sp² domains, and these CDots exhibit a high photoluminescence quantum yield (PLQY) of 72% in ethanol solution. Remarkably, due to the existence of only one type of recombination center in these nanoparticles, AIQ does not take place in CDot-based close-packed films, which show strong emission with a PLQY of 65%. Utilizing the differences in the emission properties of vacuum heating produced CDots, CDots synthesized through microwave-assisted heating, and commercial green fluorescent organic ink (namely, excitation-dependent vs. excitation-independent emission, and different stability against photobleaching), multilevel data encryption has been demonstrated.

Introduction

Strongly luminescent quantum dots (QDs), such as Cd-based chalcogenides, have distinct advantages for solid-state

Conceptual insights

Carbon dots (CDots), as a new family of luminescent carbon nanomaterials, have been explored extensively due to their distinct merits. However, the aggregation-induced quenching adherent to CDots in aggregated states has greatly hindered their applications in solid-state luminescent devices until now. The conceptual novelty of this work includes several innovative aspects. (1) For the first time, a space-confined vacuum heating method has been developed to synthesize CDots with efficient luminescence in both dispersed and aggregated states. (2) After purification, the resulting CDots possess uniform particle sizes, and just one kind of luminescent center, which makes them emit with a high PLQY of 72% in ethanol solution, while maintaining an equally high PLQY of 65% in close-packed films. (3) Utilizing the differences in the emission properties of vacuum heating produced CDots, CDots synthesized through microwave-assisted heating, and commercial green fluorescent organic ink (namely, excitation-dependent vs. excitationindependent emission, and different stability against photobleaching), multilevel data encryption has been demonstrated.

luminescent devices,^{1–3} such as light emitting diodes (LEDs) and television displays; however, the presence of heavy metals in these QDs hinders their practical application. Continuing efforts have been made towards exploiting other kinds of strongly emitting inorganic nanomaterials, which should be ideally nontoxic and environment-friendly, show high chemical stability, and be of low cost.^{4–7} Carbon dots (CDots) are luminescent materials satisfying most of these requirements, such as low cytotoxicity, strong resistance to photobleaching, high

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[†] Electronic supplementary information (ESI) available: Dissolution of 250 °C-heated foam through addition of water, results of the fitting of PL emission spectra of aqueous solutions of v-CDots synthesized at different temperatures, failed attempts to prepare CDots by mixing citric acid and CaCl₂, urea and CaCl₂, and citric acid and urea using the space-confined vacuum heating method, and drying a purified v-CDot ethanol solution in a glass tube. See DOI: 10.1039/c8nh00247a

photoluminescence quantum yields (PLQY) in the dispersed state, and ease of fabrication from low-cost precursors.^{8–25} However, aggregation-induced quenching (AIQ) of the CDot emission prevents their practical application in solid-state luminescent devices.^{26,27} To overcome or prevent AIQ, great efforts have been made towards developing suitable CDot-based composites. Starch, NaCl, BaSO₄, polyvinyl alcohol, polyhedral oligomeric silsesquioxanes and silica have all been used as matrices able to disperse and isolate CDots in order to prevent AIQ.^{26,28–34} However, reasonably high PLQYs are usually achieved in such composite materials at low CDot loading fractions only.²⁰

The excitation-dependent PL properties of CDots have been considered as their distinct characteristics,³⁵⁻⁴³ and have been ascribed to the presence of multiple luminescent centers in CDots, or a mixture of various luminescent substances, which could be not only CDots but also molecular fluorophores or their agglomerates.^{44–51} It is thus reasonable to assume that the main origin of AIQ in CDots is non-radiative recombination caused by energy or charge transfer among the different luminescent centers or substances in aggregates. Thus, it can be inferred that rational design of CDots possessing just one kind of luminescent center might be an effective approach for overcoming AIQ. In this work, we successfully produced CDots which show an efficient luminescence in both dispersed and aggregated states. This has been achieved through the spaceconfined vacuum heating approach, where a mixture of citric acid, urea and CaCl₂ forms an inflated foam under vacuum heating at 120 °C. Upon gradually increasing the heating temperature to 250 °C, blue emissive molecular species are first formed and then transformed into uniform-sized green emissive CDots (which we denote as v-CDots further below, with a "v" standing for "vacuum") through dehydration and carbonization processes taking place in the confined spaces of the thin foam walls. These CDots maintain very high PLQYs (65-75%) both in solution and in thin films.

Results and discussion

The vacuum heating synthesis was performed as follows. Citric acid (0.5 g), urea (1 g), and CaCl₂ (1 g) were dissolved in deionized water (3 mL) in a round-bottom flask. The solution was heated to 120 °C under vacuum, during which the water evaporated and the precursor materials gradually inflated into a spherical-shaped porous foam. Upon further increasing the heating temperature under continuously applied vacuum (1 Pa, maintained using a mechanical vacuum pump), highly luminescent foams with different emission colors were obtained (Fig. 1a). From the high-resolution transmission electron microscopy (HRTEM) images, it was difficult to distinguish crystalline nanoparticles in the samples synthesized below 200 °C, whereas crystalline nanoparticles (average diameter: 4.1 nm) were clearly observed in the samples synthesized at higher heating temperatures (such as the 250 °C sample shown in Fig. 1b). The lattice spacings of these particles were 0.21 and



Fig. 1 (a) Photographs of the inflated foams prepared by the vacuum heating method at different temperatures, taken under sunlight (upper panel) and UV light (lower panel). (b) HRTEM image of the v-CDots synthesized by the vacuum heating method at 250 °C. (c) Emission spectrum of a down-conversion WLED prototype based on the 250 °C powder. The insets of (c) show a photograph of the working device and its CIE color coordinates.

0.24 nm, which is consistent with the $(10\bar{1}0)$ and $(11\bar{2}0)$ crystallographic facets of graphitic carbon, respectively.^{52–54} By utilizing the 250 °C sample as a phosphor, down-conversion white LEDs (WLEDs) with Commission Internationale de l'Eclairage (CIE) chromaticity coordinates of (0.33, 0.32) could be easily fabricated by depositing the 250 °C sample on an InGaN LED chip (Fig. 1c), where the red-shift of the emission of the 250 °C sample in the WLEDs is mainly due to the progressive reabsorbance effect.^{30,55,56}

The CDot-containing foams can be easily ground into powders, which are readily soluble in water (Fig. S1 and Movie S1, ESI[†]). Absorption spectra of aqueous solutions produced from such powders synthesized at different temperatures consistently show two absorption peaks centered at 330 and 406 nm (Fig. 2a). The ratio of the absorption peak intensities at 330 and 406 nm gradually decreases in the samples produced at heating temperatures varying from 140 to 250 °C. Correspondingly, their PL spectra also differ, depending on the temperature used for the synthesis (Fig. 2b). The 140 °C sample has a single blue emission band located at 450 nm. For samples produced at higher temperature, this band gradually shifts to longer wavelengths, and another emission shoulder gradually emerges. The PL spectrum of the 250 °C sample finally exhibits just one green emission peak centered at 520 nm, indicating that all blue emissive species are transformed into green-emitting ones. For the samples exhibiting double PL peaks, the PL spectra could be well fitted by two Gaussianshaped profiles located at 450 and 520 nm (Fig. S2a, ESI⁺), pointing out the co-existence of blue and green emissive species.



Fig. 2 (a) UV-vis absorption spectra and (b) PL emission spectra of dilute aqueous solutions obtained by dissolving the foams synthesized by the vacuum heating method at different temperatures, as indicated. (c) Variations of the PL intensities of the two deconvoluted peaks (located at 450 and 520 nm) of the 220 °C-heated sample (whose PL spectrum is shown by a blue line in (b)) under UV light (15 W mercury lamp equipped with a 450 nm short wave pass filter) as a function of UV irradiation time.

As proposed in previous studies, the synthesis of CDots may result in the formation of species which emit due to the presence of either carbogenic particles or molecular fluorophores. 40,44,46,48,57,58 Molecular fluorophores often exhibit blue emission, and their structures are relatively fragile, leading to lower photostability. Carbogenic particles, on the other hand, should be chemically inert, and thus show better photostability than molecular species.48 With this consideration in mind, the resistance of the blue and green emissive species towards photobleaching was checked under ultraviolet (UV) light irradiation using a 15 W mercury lamp equipped with a 450 nm short wave pass filter (Fig. 2c and Fig. S2b, c, ESI[†]). After 60 min UV irradiation, the blue band at 450 nm was almost quenched in all the samples, while the intensity of the green emissive band at 520 nm only decreased by about 30% (Fig. S2c, ESI[†]). It is reasonable to assign the blue emissive band to molecular fluorophores, which form at a low heating temperature (140 °C), while the green emissive band originates from the v-CDots, which appear at higher heating temperatures (exceeding 220 °C) in accordance with HRTEM observations.

To reveal the possible occurrence of dehydration and carbonization processes taking place in the space-confined vacuum heating synthesis, C-13 nuclear magnetic resonance (¹³C NMR) spectra were collected from the different samples (Fig. 3a). For the sample prepared at 140 °C, there are five strong peaks, with the peak located at 162.5 ppm ascribed to the carbonyl carbon in urea, and the other four peaks located at 45.3, 75.8, 179.6 and 182.6 ppm to the aliphatic and carboxyl carbons in citric acid. Upon increasing the heating temperature from 140 to 200 °C, a new peak at 156.8 ppm assigned to the carbonyl carbon in the amide group appears, which is generated in the dehydration reaction between citric acid and urea.45 No characteristic peaks of aromatic carbon (sp² C) are found, indicating that no obvious carbonization takes place at the heating temperatures lower than 200 °C. Thus, the blue emissive species generated in the lower temperature range can be attributed to the amide-based fluorophores formed in the dehydration process between citric acid and urea. After increasing the heating temperature to 220 °C, multiple peaks in the range from 142.7 to 125.2 ppm appear in accordance with the typical range of aromatic carbon $(sp^2 C)$, while the peaks at 156.8 ppm of



Fig. 3 (a) 13 C NMR spectra in D₂O solutions, (b) XPS spectra and (c) O 1s XPS spectra of the v-CDot samples synthesized by the vacuum heating method at different temperatures, as indicated on frames. Fittings of the O 1s XPS spectra in (c) provide the assignment of C-OH/C-O-C and C=O vibrations.

amide groups and 162.5 ppm of carbonyl groups become weakened (Fig. 3a).⁴⁵ When the heating temperature increases to 250 °C, the sp² C signals are further enhanced. These emerging peaks of sp² C indicate that the carbonization process occurs at temperatures higher than 220 °C, resulting in the formation of green emissive v-CDots. The accompanying decrease of the amide signals indicates that the green emissive v-CDots are derived through carbonization of the blue emissive molecular species, in accordance with changes in their absorption and PL spectra (Fig. 2a and b).

The chemical compositions of the powdered samples obtained at different temperatures were studied by X-ray photoelectron spectroscopy (XPS) (Fig. 3b). XPS spectra show five peaks at 198.5, 284.0, 346.7, 400.0, and 531.5 eV, which are attributed to Cl $2p_{3/2}$, C 1s, Ca $2p_{3/2}$, N 1s, and O 1s, respectively. The decrease in oxygen content with increasing heating temperature is in accordance with thermal controlled dehydration and carbonization of the precursors. Fig. 3c presents the O 1s spectra of these samples, which can be fitted with two peaks at 531.7 and 533.0 eV.⁵⁹ The former is assigned to C–OH/C–O–C, and the latter is assigned to C=O. The ratio of C–OH/C–O–C and C=O related peaks gradually decreases upon increasing the heating temperature, which provides further evidence of the occurrence of the dehydration and carbonization processes.

We found that the simultaneous use of citric acid, urea and CaCl₂ is crucial for the formation of highly luminescent powders, which could not be obtained using just any two of these three precursors (Fig. S3, ESI†). With an increase of heating temperature, the contents of Ca and Cl elements in the samples gradually reduced, as evidenced in XPS spectra (Fig. 3b). The reduced amount of Cl can be attributed to the reaction between CaCl₂ and the carboxyl groups of citric acid, resulting in the formation of volatile HCl gas and sparingly soluble calcium carboxylate. The volatile HCl gas can be pumped out using a vacuum pump to generate an inflated foam structure, and the calcium carboxylate is capable of supporting the inflated foam structure. On the other hand, urea can be thermally decomposed to produce NH₃, which could take Ca²⁺ ions out in the form of CaCl₂·8NH₃ complexes under a vacuum pump.⁶⁰ These simultaneously generated gases allow for inflation of the dehydrated precursors into foams, which are further carbonized to form green emissive v-CDots at temperatures higher than 220 °C in the foam walls.

Based on the above data, a possible growth mechanism for the formation of v-CDots is illustrated in Scheme 1. Citric acid, urea and CaCl₂ are dissolved in water, where citric acid and urea assemble through the inter-molecular H-bonding between their carbonyl, hydroxyl and amine groups. After heating in a vacuum, volatile gases rapidly evolve and are pumped out to generate an inflated foam structure. Upon increasing the temperature from 140 to 160 $^\circ$ C, the precursors dehydrate and cross-link in the walls of the foam.40 These dehydrated and cross-linked molecules constitute blue emissive fluorophores (Scheme 1). Upon further increasing the temperature to 220 °C, green emissive v-CDots are gradually formed through the further carbonization of the blue emissive molecular species in the confined spaces of the foam walls (Scheme 1, Fig. 2a, b and Fig. S2a, ESI[†]). Upon heating to 250 °C, all blue emissive molecular species transform through carbonization to form uniform-sized green emissive v-CDots (Fig. 2b and Scheme 1). Such a formation process of v-CDots in the confined space of the foam effectively prevents further growth of their carbogenic cores, which is the reason for their small and uniform particle sizes. More importantly, the homogeneous structure of v-CDots leads to a uniform energy band gap, which helps to avoid energy transfer among v-CDots in aggregates and thus overcome the AIQ, as will be demonstrated below.

To obtain high quality v-CDot films, the 250 °C synthesized v-CDots were further purified to remove residual low weight molecules, salts and Ca^{2+} ions. Since Ca^{2+} can exist in the form



Scheme 1 Schematics of the growth mechanism of v-CDots synthesized via the vacuum heating method, which occurs through the formation of the blue emissive molecular species at a lower temperature (140 $^{\circ}$ C), followed by their carbonization and the formation of green-emissive carbon nanoparticles at higher temperatures (220–250 $^{\circ}$ C) in the confined-space foam.

of calcium carboxylate on the surfaces of the v-CDots, the powder was first dissolved in deionized water, and then the pH value of the resultant solution was adjusted to 3.2 by adding HCl, which is within the range of use of the dialysis membrane (pH 2-9). During the dialysis, the solution inside the dialysis membrane (*i.e.*, v-CDot aqueous solution) exhibits bright green emission under UV light excitation (Fig. S4b and c, ESI[†]). In contrast, the aqueous solution outside the dialysis membrane shows nearly no emission (Fig. S4b and c, ESI⁺), indicating that the substances outside the dialysis membrane mainly contained organic/inorganic salts and small molecular weight molecules after the hydrochloric acid treatment, while luminescent v-CDots are retained in the dialysis bag. The solution was dialyzed against deionized water for 10 days, and condensed by vacuum-rotary evaporation, with a production yield of purified v-CDots of 56%. The purified v-CDots were dispersed in ethanol for subsequent film coating. The optical properties of v-CDots in ethanol solution and in films produced thereof are illustrated in Fig. 4a. The absorption peak at 330 nm disappeared after dialysis (Fig. 2a and 4a), indicating the removal of any residual blue emissive molecular species. The v-CDots in ethanol solution show strong green emission with a PLQY of 72%. Importantly, the purified v-CDots can easily form strongly luminescent films, with an equally high PLQY of 65% (Fig. 4a and Fig. S5 and Movie S2, ESI†); their absorption and PL spectra in the film remain nearly identical to those of the ethanol solution, indicating that AIQ is conquered in v-CDots due to their uniform energy band gap. HRTEM study confirmed the presence of uniform-sized v-CDots with an

average diameter of 4.1 nm and lattice spacings of 0.21 and 0.24 nm (Fig. 4b and Fig. S6a, ESI⁺). Atomic force microscopy (AFM) showed well dispersed v-CDots with a uniform height of 1.7 nm (Fig. 4c and Fig. S6b, ESI⁺). An X-ray powder diffraction (XRD) pattern of the v-CDots film showed a diffraction peak centered at 3.2 Å (Fig. 4d), which is consistent with the (002) lattice planes of graphitic carbon. An energy-dispersive spectrum (EDS) of the purified v-CDots confirmed that most of the residual Ca species have been removed (Fig. 4e). As shown in Fig. 4f, the broad C=O stretching vibration of carboxylate in the 250 °C-heated sample turns into a narrow C=O stretching vibration of the carboxyl groups centered at 1713 cm⁻¹, and the amide I and II bands located at 1621 and 1556 cm^{-1} become more pronounced in the purified v-CDots. In addition, the O-H stretching vibration becomes stronger in the purified v-CDots. These results indicate the transformation of calcium carboxylate into carboxyl groups after purification of v-CDots. Moreover, abundant oxygen- and nitrogen-containing groups on the surfaces of the v-CDots could be another factor helping to avoid AIQ. Generally, the π - π stacking of the sp² domains in the carbon cores could give rise to the shortened energy band gaps and the energy transfer thereof, finally resulting in AIQ. Functional groups on the surfaces of v-CDots could prevent the π - π stacking of the sp² domain of the carbon core in aggregates and thus avoid AIQ.

As previously reported in the literature,³⁵ CDots synthesized from the same precursors citric acid and urea through microwaveassisted heating (we abbreviate them as m-CDots here) show excitation-dependent emission (Fig. 5b). Their PL maximum



Fig. 4 (a) UV-vis absorption spectra and PL emission spectra of the purified v-CDots in ethanol solution (black lines) and in the close-packed film (red lines). The insets show the corresponding photographs taken under sunlight and UV light, respectively. (b) HRTEM image, (c) AFM image, (d) XRD pattern, and (e) EDS pattern of the purified v-CDots. (f) FTIR spectra of v-CDots before (black line) and after (red line) purification.



Fig. 5 Excitation–emission maps of the (a) purified v-CDot ethanol solution and (b) m-CDot ethanol solution. (c) Time resolved PL decay curves of v-CDots (black) and m-CDots (red). (d) Fluorescence images of characters written on paper using v-CDots, m-CDots, and commercially available green fluorescent organic ink (see text for details).

gradually shifts from 495 to 555 nm when varying the excitation wavelength from 350 to 550 nm, implying the presence of different luminescence centers (due to the presence of CDots with different optical bandgaps in the ensemble) or radiative contributions from the surface states in m-CDots. Due to the presence of these multiple emissive centers, the emission of m-CDots is severely quenched in the aggregate state (Fig. S7, ESI⁺), due to the AIQ effect. In stark contrast, the emission of v-CDots remains almost unchanged when varying the excitation wavelength (Fig. 5a), indicating the existence of just one kind of luminescent center in these particles. To further prove this assumption, the PL decay curves of the films of v-CDots and m-CDots were compared (Fig. 5c). The m-CDot film shows a multi-exponential PL decay with a short average PL lifetime (1.1 ns), which is due to the energy or charge transfers between closely adjacent particles possessing multiple luminescent centers (Fig. 5c). In contrast, the v-CDot film has a PL decay curve very close to the monoexponential shape, and shows a much longer average PL lifetime of 5.7 ns.

The different optical characteristics of m-CDots and v-CDots can be utilized for encryption of luminescence information. Considering the much better photostability of CDots than those of organic dyes (Fig. S8, ESI[†]), multilevel encrypted characters have been composed on conventional paper using v-CDots, m-CDots, and green fluorescein sodium dye, as illustrated in Fig. 5d. These characters are invisible under daylight, while green fluorescent characters 8 8 8 are observed under blue light excitation (Fig. 5d). Under green-light excitation, the characters written with v-CDots and fluorescein sodium ink are invisible due to the excitation independent emissions of these two luminescent species (Fig. 5a), whereas only the red fluorescent characters 3 4 5 written with m-CDots appear (Fig. 5d), which is due to the excitation-dependent PL of the latter (Fig. 5b). Considering that v-CDots and m-CDots show much better photostability than the fluorescent organic ink (Fig. S8, ESI⁺), further encryption information can be extracted by photobleaching the characters written with fluorescein sodium ink. After continuous irradiation with UV light, its emission is completely quenched, whereas green fluorescent characters 9 9 9 written with v-CDots and m-CDots can still be clearly observed (Fig. 5d). The stability of v-CDots was evaluated by comparing their luminescence in ethanol solution before and after storage for 6 months; nearly unchanged PL spectra and PLQYs indicate good stability of v-CDots (Fig. S9, ESI⁺). These results demonstrate that CDots can be used as an encryption medium for long time luminescence information storage.

Experimental

Materials

Citric acid (99.5%), urea (99%), ethanol (99.7%), and anhydrous calcium chloride (99.9%) were purchased from Aladdin.

Hydrochloric acid (37%) and commercial fluorescein sodium dye solution (a kind of green fluorescent organic ink) were purchased from Beijing Chemical Factory. All chemicals were used without further purification. Water used in all experiments was purified through a Millipore system.

Synthesis of v-CDot powders

Citric acid (0.5 g), urea (1 g), and $CaCl_2$ (1 g) were dissolved in water (3 mL). The solution was dried at 120 °C under vacuum; the vacuum value of the system was about 1 Pa provided by a mechanical vacuum pump. After that, the heating temperature was increased to 140, 160, 200, 220 and 250 °C, respectively. By keeping the system at each heating temperature for 1 h, inflated foams with different emission colors were obtained, which could be easily ground into powders.

Purification of v-CDots

The 250 °C-heated v-CDot powder was dissolved in a dilute hydrochloric acid solution in water, and dialyzed against deionized water for 10 days using a Spectra/Por[®] Biotech Grade Cellulose Ester Dialysis Membrane (MWCO = 100–500 D), which was purchased from Spectrum Laboratories, Rancho Dominguez, CA. The purified aqueous solution of v-CDots was condensed in a vacuumrotary evaporator, and v-CDots were re-dissolved in ethanol.

Characterization

UV-visible absorption spectra were recorded on a Shimadzu UV-3101PC spectrophotometer. PL spectra were collected on a Hitachi F-7000 spectrofluorimeter. The absolute PLQYs of v-CDots in solutions and films were measured using a calibrated integrating sphere in an FLS920 spectrofluorimeter (excitation wavelength 400 nm). High resolution transmission electron microscopy (HRTEM) images were obtained on an FEI Tecnai-G2-F20 TEM at 200 kV. X-ray powder diffraction (XRD) measurements were carried out on a Siemens D5005 diffractometer. An Inca X-Max instrument (Oxford Instruments) was applied to measure energy dispersive spectra (EDS). Atomic force microscopy (AFM) images were obtained on a SA400HV microscope with a Seiko SPI3800N controller. X-ray photoelectron spectroscopy (XPS) was conducted on a VG ESCALAB MKII spectrometer with Mg KR excitation (1253.6 eV). Binding energy calibration was based on C 1s at 284.6 eV. ¹³C nuclear magnetic resonance (NMR) spectra were obtained on a Bruker Ultra Shield 500 MHz spectrometer. Fourier transform infrared (FTIR) spectra were obtained on a Nicolet AVATAR 360 FTIR instrument.

Conclusions

In summary, a space-confined vacuum heating method has been developed to synthesize CDots with equally efficient luminescence in both dispersed and aggregated states. During heating at temperatures lower than 200 °C, blue emissive molecular species are first formed through the dehydration process between citric acid and urea in the highly confined foam structure. After heating

at 250 °C, CDots with green emission are formed in the confined ultrathin spaces of the foam walls as a result of the carbonization of the blue emissive molecular species. After purification, the resulting CDots possess uniform particle size and emit green light with a high PLQY of 72% in ethanol solution, while maintaining an equally high PLQY of 65% in close-packed films, which indicates that due to the existence of only one type of recombination center in these nanoparticles, the AIQ process is efficiently prevented. Based on the differences between the luminescence properties of vacuum-heated CDots (excitation-independent emission), microwave-synthesized CDots (excitation-dependent emission), and commercial green fluorescent organic ink (excitation-independent emission, but low resistance against photobleaching), a multilevel data encryption concept has been realized. We believe that the space-confined vacuum heating method demonstrated here may be widely used in developing highly luminescent CDot solid-state materials from other precursors, and promote their applications in solid-state luminescence devices.

Conflicts of interest

The authors declare no conflicts of interest.

Acknowledgements

This work was supported by the National Natural Science Foundation of China (Project No. 51602304), the Jilin Province Science and Technology Research (Project No. 20160520008JH, 20150519003JH, 20170101191JC, 20170101042JC), and the Ministry of Education and Science of the Russian Federation (Grant 14.Y26.31.0028).

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