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Enhanced photoluminescence and characterization of multicolor carbon dots using plant soot as a carbon source

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ABSTRACT

Carbon dots (C-dots) are a class of novel fluorescent nanomaterials, which have drawn great attention for their potential applications in bio-nanotechnology. Multicolor C-dots have been synthesized by chemical nitric acid oxidation using the reproducible plant soot as raw material. TEM analysis reveals that the prepared C-dots have an average size of 3.1 nm. The C-dots are well dispersed in aqueous solution and are strongly fluorescent under the irradiation of ultra-violet light. X-ray photoelectron spectroscopy characterization demonstrates that the O/C atomic ratio for C-dots change to from 0.207 to 0.436 due to the chemical oxidation process. The photo bleaching experiment reveals that the C-dots show excellent photostability as compared with the conventional organic dyes, fluorescein and rhodamine B. The fluorescence intensity of the C-dots did not change significantly in the pH range of 3–10. To further enhance the fluorescence quantum yield, the C-dots were surface modified with four types of passivation ligands, 4,7,10-trioxa-1,13-tridecanediamine (TTDDA), poly-L-lysine (PLL), cysteine and chitosan and the fluorescence quantum yields of the TTDDA, PLL, cysteine and chitosan passivated C-dots were improved 1.53-, 5.94-, 2.00- and 3.68-fold, respectively. Fourier-transform infrared (FTIR) spectra were employed to characterize the surface groups of the C-dots. The bio-application of the C-dots as fluorescent bio-probes was evaluated in cell imaging and *ex vivo* fish imaging, which suggests that the C-dots may have potential applications in biolabeling and bioimaging.

1. Introduction

Carbon nanomaterials including carbon nanotubes, nanoparticles, nanofibers, graphene oxide and carbon dots (C-dots), have attracted great attention due to their wide applications in solar cells [1], bioimaging [2], photocatalysts [3], nanoelectronic devices [4], photodynamic therapy [5] and gene delivery [6]. Among them, carbon dots are a new class of functional nanomaterials exhibiting their inherently unique photoluminescent properties, such as wavelength-tuneable emission, excellent solubility, good biocompatibility and chemical inertion [7–10]. Unlike the semiconductor quantum dots derived from heavy metal precursors incurring long term toxicity [11], C-dots are environmentally friendly and lack of known cytotoxicity [12,13]. The spectroscopic studies have shown that the mixture of C-dots of different sizes with the ability to emit in different color at different excitation wavelength [14]. The multicolor property of C-dots has made them promising "nano-lights" that can emit tuneable photoluminescence for various applications. Particularly, considerable research effort has been focused on their potential applications in biological field [12,15–17].

Though the C-dots have shown some unique properties, the easy synthesis from an easily available carbon source of highly fluorescent C-dots is still a challenge. The mechanism of C-dots associated photoluminescence is not clear completely. The suggested reason behind the luminescence might be the radiative recombination of the energy-trapping sites on the C-dots surface[7]. In order to enhance the quantum yield (QY) of carbon dots, much effort has been directed toward the use of surface passivation method to achieve a strong photoluminescence [13]. Doping C-dots with inorganic salt ZnS or SiO_2 is able to enhance the QY of C-dots [10]. Enhancing the luminescence of C-dots with a reduction pathway is also reported recently [18]. Surface passivation with polyethylene glycol (PEG) has been thought as an effective way to improve the fluorescent properties of the C-dots [7]. Other organic molecule, such as 4,7,10-trioxa-1,13-tridecanediamine (TTDDA), has also been used in surface passivation of C-dots derived from commercial activated carbon [9]. However, the large scale preparation and surface passivation of C-dots are still expected to provide highly fluorescent C-dots by using economical and facile approaches.

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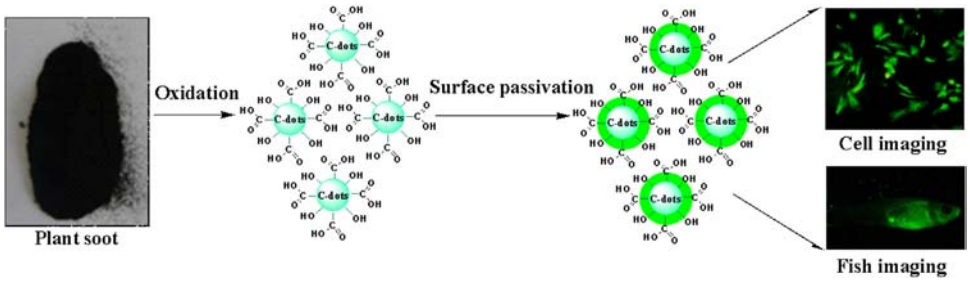

Scheme 1. Synthesis scheme of carbon dots derived from plant soot.

Up to now, various carbon sources have been used to prepare C-dots such as laser ablation of graphite [19,20], electrochemical treatment of graphite [21], commercial activated carbon by bottom-up method [9], heating carbohydrate, watermelon peels or PEG with microwave pyrolysis [22–24]. The prepared C-dots are highly water-soluble, nano-sized and multi-colourful. However, all these above-mentioned methods suffer from more or less drawbacks such as complex and time-consuming processes, severe synthetic conditions, and high cost, which limit their wide applications [25]. Thus, a low-cost and facile synthesis of C-dots from an economical carbon source is highly desired [2,9].

On the other hand, intense research has focused on the development of facile methodologies for preparing nanomaterials using natural soot widely distributed in nature to replace conventional chemicals [26–30]. For example, carbon nanoparticles have been prepared from corn stalk soot and used as stationary phase of hydrophilic interaction chromatography, [28] or from castor oil soot using acid treatment for electrochemical applications [30]. Multicolor C-dots have been prepared by the acid oxidation of candle soot [26]. However, it is an impossible task to produce candle soot for a large amount. To the best of our knowledge, no attention has been paid to producing multicolor C-dots from the combustion soot of plants (instead of candles).

In the present study, a simple and low-cost strategy toward water-soluble, carbon-rich, photoluminescent multicolor C-dots was developed by refluxing the plant soot in nitric acid for the first time (Scheme 1). The obtained C-dots are highly water-soluble and strongly photoluminescent under ultra violet (UV) light. The C-dots exhibit similar photoluminescence characteristics as compared to those from candle soot [26] or natural gas soot [27]. We further demonstrate that the surface passivation of the C-dots by using various surface passivation agents may be an effective strategy to improve the fluorescent properties. Interestingly, these multicolor C-dots may also used as unique nanostructural probes for biomedical imaging applications.

2. Experimental

2.1. Materials and instrumentation

Plant soot was collected from burned plants and sieved to remove big particles. The smaller size of the plant ash (<100 mesh, 150 μm) was used for C-dots preparation. Nitric acid (HNO_3, 69.8%,) was purchased from Fisher Co., Dialysis bags with a molecule weight cutoff of 1000 Da were purchased from Shanghai Green Bird Science &Technology Development Co., China. De-ionized water was prepared with a Milli-Q-Plus system (18.2 MΩ). 4,7,10-trioxa-1,13-tridecanediamine (TTDDA, $M_w=220$) was purchased from J&K Scientific Co., China. poly-L-lysine (PLL; MW 10 k) was kindly provided by Prof. Zhibo Li (Institute of Chemistry Chinese Academy of Sciences). Cysteine was purchased from Shanhai Yuanju Biotechnology Science &Technology Co., China. Chitosan with M_w of 30 kDa was degraded from raw material (Yuhuan Ocean Biomaterials Corporation, Zhejiang, China) in our laboratory. All other reagents were of analytical grade and were from J&K Scientific Co., China.

2.2. Preparation of C-dots

Multicolor carbon dots were prepared by using a modified procedure reported in the literature.[27] Typically, in a 250 mL flask, 0.5 g of plant soot was dispersed in 100 ml of HNO_3 (1.5 mol/L) aqueous solution and sonicated for 5 min. The mixture was refluxed at 110 °C for 20 h. After the reactant solution cooled down to room temperature, a brownish supernatant was obtained by filtration to remove the water insoluble impurities. The excessive acid of the products solution was neutralized by Na_2CO_3 aqueous solution and the resulting C-dots product was dialyzed against deionized water through a dialysis membrane for 2 days. The bare C-dots were lyophilized and stored at 4 °C for further characterization and use.

2.3. Surface passivation of C-dots

TTDDA, poly lysine (PLL), cysteine or chitosan passivated C-dots: To 100 mL of bare C-dots solution (about 0.5 mg/mL), 1.0 g of surface passivation agent was added (pH=7.0) and sonicated for 5 min. The mixture was heated to 110 °C and reflux for 72 h. The TTDDA or cysteine passivated C-dots were obtained by removing the excessive TTDDA or Cysteine by dialysis against deionized water through a dialysis membrane with a molecule weight cutoff of 500 Da. The PLL passivated C-dots were obtained by ultrafiltration. The chitosan passivated C-dots were obtained by filtration to remove the excessive precipitated chitosan in ethanol.

2.4. Reduction of C-dots

Reduction of C-dots: 2.0 g of $NaBH_4$ was added into the bare C-dots (about 0.5 mg/mL, 100 mL) and stirred at room temperature for 24 h. After reaction, the yellow supernatant was purified by dialyzing against deionized water through a dialysis membrane (MWCO 1000) to remove the extra $NaBH_4$. The reduced C-dots were lyophilized and stored at 4 °C for further characterization and use.

2.5. Characterization

Transmission electron microscopy (TEM) was performed using a FEI Tecnai G^2Spirit at an acceleration voltage of 120 kV. Nano ZS90 Zetasizer (Malvern Instruments, Malvern, U.K.) was used to determine the Zeta potential. Absorption and fluorescence spectra were recorded at room temperature on UV-2550 UV–vis spectrophometer (Shimadzu, Japan) and Luminescence Spectrometer 55 (Perkin-Elmer) respectively. Fourier-transform infrared (FTIR) spectra were recorded on a Burker Vector 22 spectrometer. XPS

spectra were used to characterize the chemical composition using an Escalab 250 Xi X-ray photoelectron spectrometer (Thermo Scientific).

2.6. Fluorescence quantum yield measurements

The relative fluorescence quantum yield (Φ) of the C-dots was measured according to the procedure of a reported method [9] and calculated using the equation of $\Phi_x = \Phi_{std}\ I_x A_{std} \eta_x^2 / I_{std} A_x \eta_{std}^2$. The optical densities were measured on Shimadzu UV-2550 UV–vis spectrophometer. In the equation, I_x and I_{std} are the fluorescence intensities of the C-dots and the standard, and A_x *and* A_{std} are the optical densities of the C-dots and the standard, respectively. Quinine sulfate in 0.1 M H_2SO_4 was chosen as a standard with a quantum yield Φ_{std}=0.54 at 350 nm. η_x *and* η_{std} are the refractive index of the C-dots and the standard, respectively. The absorbencies of all the samples in 1.0 cm cuvette were kept under 0.1 at the excitation wavelength to minimize re-absorption effects.

2.7. Cell imaging

Chinese Hamster Ovary (CHO) cell line 1640 medium with 10% Fetal Bovine Serum was trypsinized and seeded in 24-well tissue culture plates at a density of 1×10^5 cells/well. The fluorescent PLL passivated C-dots with a concentration of 1 mg/mL were added to each well. After 2 h incubation with glass slide inside humidified 5% CO_2 incubator for cell attachment, the glass slide was washed thoroughly with PBS buffer and the cells were fixed with formalin. Cell-imaging was performed on an inverted Nikon TE-2000U microscope with an excitation filter of 380–420 nm and 450–490 nm for blue and green color respectively.

2.8. Ex vivo guppy fish imaging

Ex vivo imaging was carried out with a CRi Meastro Ex *in vivo* imaging system (Caliper Life Sciences Inc. U.S.A.). The adult guppy fish was put in the water mixed with the passivated C-dots (~5 mg/ml) and the control fish were put in fresh water. After 10 min, the fish was thoroughly rinsed with distilled water and imaged on the *in vivo* imaging system. Spectral fluorescence images were obtained using the appropriate filters for CNPs (excitation: 455 nm; emission: 515 nm long-pass filter; acquisition settings: 500–750 nm in 10 nm steps). Exposure times were automatically calculated and the acquisition setting was 1500 ms.

3. Results and discussion

Plant soot containing small carbon particles is a residue powder left after the combustion of plants parts like bark, wood, leaves, woody debris, and other plant debris, which is usually taken as a waste. In this study, the plant soot was collected from the bottom of a boiler. It can be produced in very large amount from the plant residue powder compared with the candle soot. Therefore, the easily available plant soot could be an economical precursor for large scale production of the multicolor C-dots. By using a modified nitric acid oxidation method, strong fluorescent C-dots were prepared. The formation of C-dots was first confirmed by TEM measurements. Fig. 1a shows a representative TEM micrograph of the C-dots. It can be seen that the C-dots are mostly of spherical shape and dispersed rather evenly on the TEM grid surface. The average size of the C-dots was found to be 3.1 nm based on statistical analysis of 150 dots. Moreover, the corresponding particle size distribution histogram in Fig. 1b suggests that the particle size is narrowly distributed with diameters in the range of 2.0–4.3 nm. As shown in the inset of Fig. 1b, the C-dots aqueous solution was yellowish and a blue emission was observed when these C-dots were excited with a 321 nm ultraviolet (UV) lamp. The results demonstrated that the fluorescent C-dots in nanoscopic dimension can be prepared via a one-step method of nitric acid refluxing with plant soot.

The chemical composition of the obtained C-dots was characterized by X-ray photoelectron spectroscopy (XPS) techniques. As shown in Fig. 2, the XPS survey of the C-dots shows two peaks at 498.0 and 1072.0 eV originated from Na_{Auger} and Na_{1s}, respectively. Another peak at 979.7 eV associated with O_{Auger} was also found. As for the XPS spectrum of plant soot, there are a predominant graphic C_{1s} peak at *ca.* 284 eV, a pronounced N_{1s} peak at *ca.* 400 eV and an O_{1s} peak at *ca.* 532 eV. These results indicate that the plant soot contains carbon, nitrogen and oxygen in a weight ratio of 79.54:4.00:16.46. As we know, the plant soot is hydrophobic and insoluble in aqueous solution. The O/C atomic ratio for plant soot was about 0.207, which changed to 0.436 due to the chemical transformation after chemical oxidation with nitric acid. More oxygen component produced during the synthesis processes may indicate that even more hydroxyl, carboxyl groups were formed on C-dots. The emerging peak associated with Na ions in the XPS spectrum of C-dots has proved this point. Furthermore, the high water solubility of C-dots might be due to the presence of $-$COONa groups.

Good photostability of a fluorescent material is significant for their application in bio-medical filed. The photostability of the prepared C-dots was investigated by using the traditional dyes as

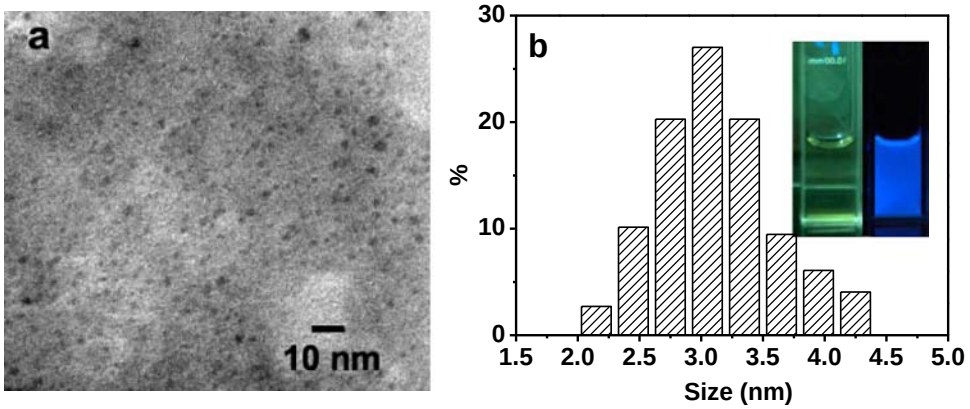

Fig. 1. TEM image (a) of the C-dots derived from plant soot and the size distribution (b) of the C-dots determined by TEM, where 150 C-dots were measured. The average size of the bare C-dots is about 3.1 nm. The inset in Fig. 1b shows the bright filed photograph and photoluminescent image of the C-dots excited with a 321 nm ultraviolet (UV) lamp.

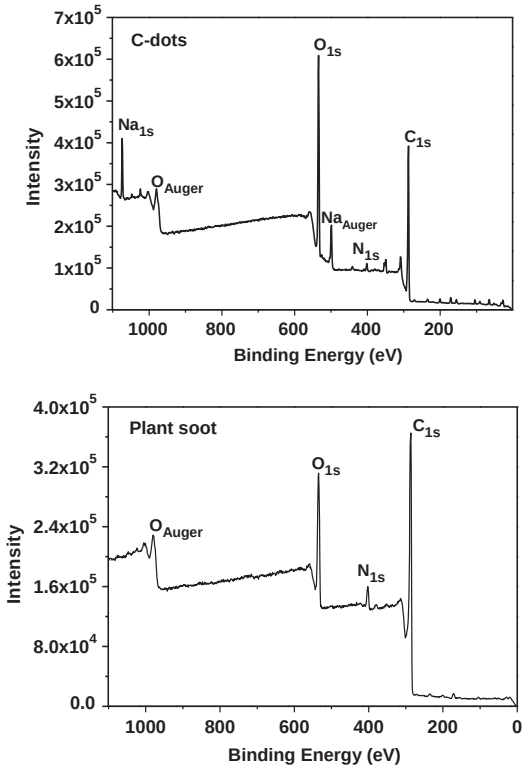

Fig. 2. X-ray photoelectron spectroscopy (XPS) spectra of the plant soot and the C-dots.

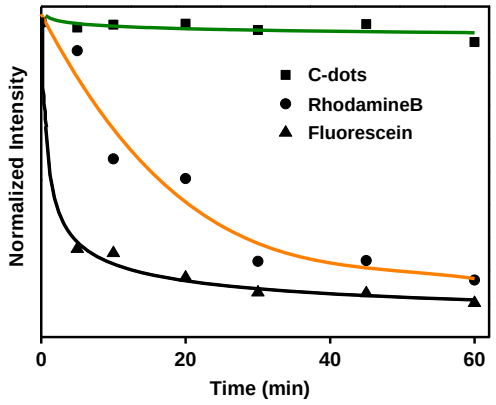

Fig. 3. Photo-bleaching experiment of the C-dots, rhodamine B and fluorescein in aqueous solution with 60 W filament lamp as an excitation source.

control. As shown in Fig.3, the bare C-dots showed excellent photostability as compared with the conventional organic dyes, fluorescein and rhodamine B. The fluorescence intensity of the bare C-dots remained unchanged even after being irritated with a 40 W incandescent lamp for 60 min, whereas the fluorescence intensity of rhoamine B and fluorescein decreased 82% and 89% respectively at the same period. This property makes the C-dots good candidates as fluorescent probes for live cells imaging, which may have a potential for microscopy and analytical applications.

Fig. 4 shows the effect of pH effect on the fluorescence intensity of C-dots. From the results we found that the fluorescence intensity of the C-dots was slightly changed in the range of pH 6.0–10.0 and decayed gradually with pH decreasing from 5.0 to 3.0. This is quite different from that of the C-dots derived from candle soot, which fluorescence intensity decreased significantly (by 40–89%) upon changing from a neutral to either an acidic or a basic solution[26]. Even though this mechanism is not fully

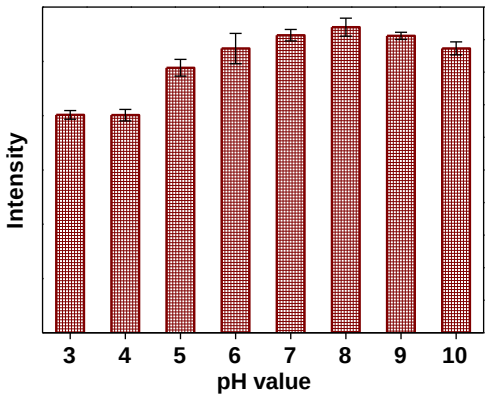

Fig. 4. The effect of pH values on fluorescence intensity of bear C-dots. (a) λ_{ex}=280 nm, and (b) λ_{em}=413 nm.

understood, the pH effect indicates that the fluorescent species in the dots have nearly neutral sites because of the fluorescence intensity not being significant quenched in the acidic media [31]. The radiative recombination of the energy-trapping sites on the C-dots surface is not significantly affected with varying the pH value from alkaline to acidic, which corresponds with the protonation of the carboxyl groups of the CDs. The C-dots derived from plant soot might have potential applications in a wide range of pH values in the nano-biological fields.

UV–vis absorption and photoluminescence (PL) spectra of C-dots, the surface passivated C-dots as well as the reduced C-dots were displayed in Fig. 5. For the UV–vis absorption spectrum of the prepared C-dots, no appreciable absorption peak was observed for the C-dots, which was consistent with the C-dots derived from commercial activated carbon.[9] The corresponding emissions of C-dots covered the UV/Vis wavelength range, from violet (~430 nm) to yellow-green (~550 nm). A strong emission spectrum centered at 430 nm was observed when it was excited at 320 nm. The emission peak position from C-dots shifted to longer wavelengths and the intensity gradually decreased with increased excitation wavelengths. By selecting quinine sulfate in 0.1 M H_2SO_4 as the standard, the measured relative fluorescence quantum yield (QY) of the bare C-dots was 0.72% (Table 1). This value was comparable to that of the carbon dots derived from candle soot [26] and was greater than that of C-dots from natural gas soot [27]. As we know that the luminescence mechanism of the CDs was still not clearly understood, the proposed reason might be the rediative recombination [7]. The green luminescence of the CDs might be attributed to surface energy traps [7], and the blue emission of the C-dots might be attributed to zig-zag sites [31,32] due to the grapheme molecules embedded in the C-dots.

Surface passivation was thought to be essential for the C-dots to enhance the luminescence of the C-dots. Therefore, four types of amine-terminated compounds, TTDDA, PLL, cysteine and chitosan, were used as surface passivation agents to improve QY of the C-dots in this study. After surface passivation, the passivated C-dots were strongly photoluminescent under a UV excitation (images were not included here). The UV–vis absorption and photoluminescence emission spectra of passivated C-dots were shown in Fig. 5. Note that the absorption spectra did not change much after the passivation process. However, the maximum emission wavelengths showed a red-shift due to the introduction of the passivation agents. For example, the TTDDA passivated C-dots displayed a 12-nm red shift of the emission wavelength. The relative fluorescence QYs of the first batch TTDDA, PLL, cysteine and chitosan passivated C-dots were improved 1.53-, 5.94-, 2.00- and 3.68-fold, respectively (Table 1). The PLL-passivated C-dots showed the highest relative QY of 4.28%. The zeta-potentials after

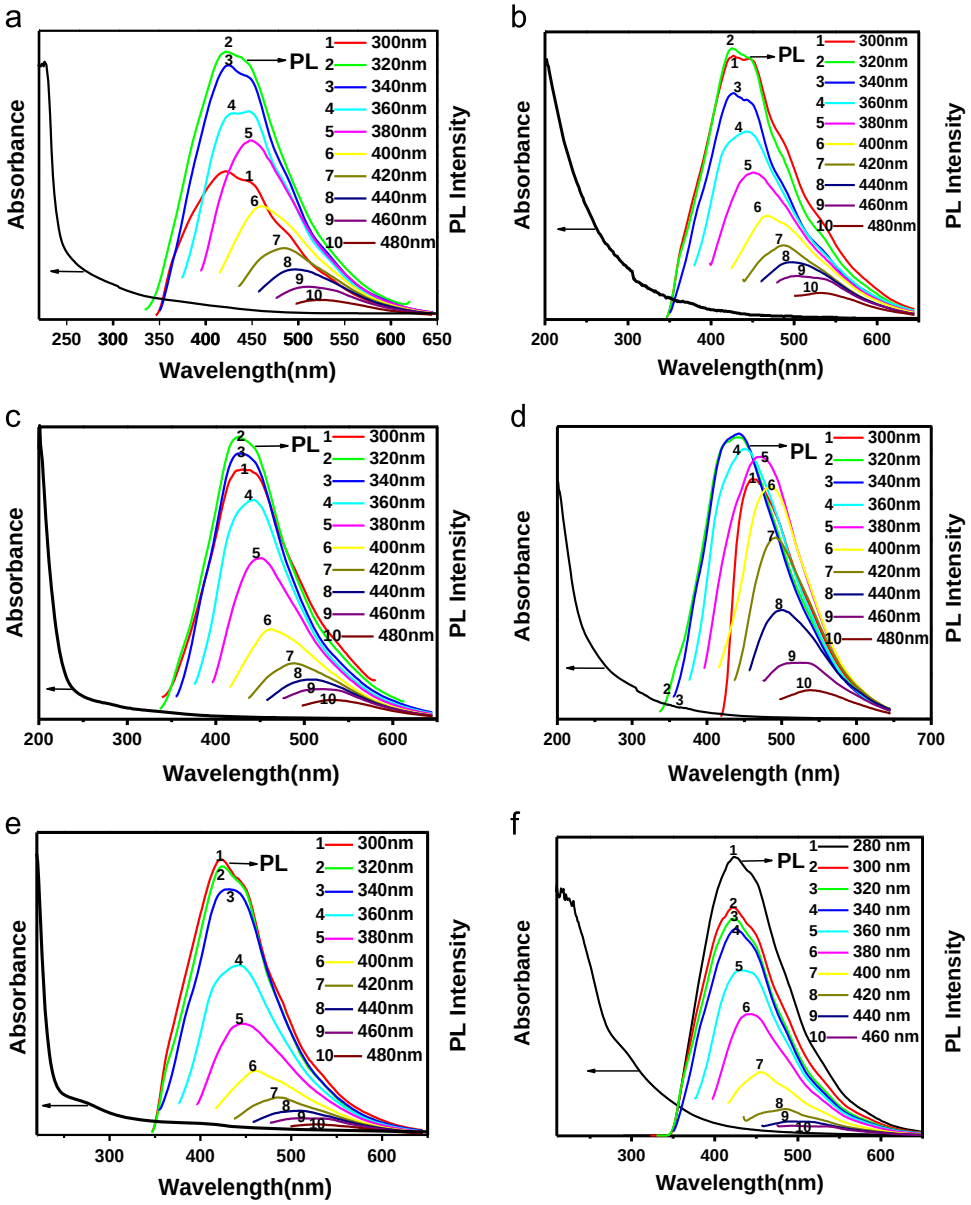

Fig. 5. UV–vis absorption and photoluminescence (PL) emission spectra for C-dots (a), TTDDA C-dots (b), PLL C-dots (c), cysteine C-dots (d), chitosan C-dots (e) and reduction C-dots (f) excited at wavelengths from 280 nm to 500 nm with 20 nm increment. The inset is the normalized emission spectra intensity.

Table 1
Physicochemical parameters of the C-dots.

Materials	Max. emission (nm)	[a]FWHM (nm)	[b]QY(%) /two batches	Zeta-potential (mV)
C-dots	422	120	0.72/0.75	−13.83
[c]TTDDA-dots	434	120	1.10/1.26	−8.06
[d]PLL-dots	433	110	4.28/4.13	0.69
[e]Cys-dots	457	137	1.44/1.14	−20.63
[f]Chi-dots	430	115	2.65/2.94	0.04
[g]R-dots	423	120	2.36/2.27	−16.03

[a] FWHM: the full width at a half maximum.
[b] QY, relative quantum yield of two batches samples.
[c] TTDDA-dots: 4,7,10-trioxa-1,13-tridecanediamine passivated C-dots.
[d] PLL-dots: α-poly-L-lysine passivated C-dots.
[e] Cys-dots: cysteine passivated C-dots.
[f] Chi-dots: chitosan passivated C-dots.
[g] R-dots: reduction C-dots.

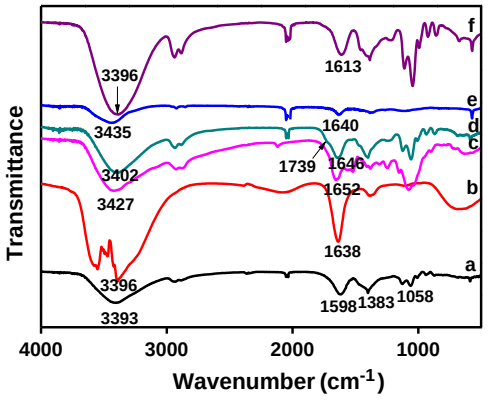

Fig. 6. FTIR of C-dots (a), TTDDA-C-dots(b), α-PLL-C-dots(c), cysteine C-dots (d), chitosan C-dots (e) and reduction C-dots (f).

passivation varied from −20.63 to 0.69 mV, which highly depended on the passivation agents.

The C-dots after reduction with sodium borohydride exhibited strong luminescence and their relative QY increases 3.28-fold after reduction treatment (Fig. 5f and Table 1). No obvious change of emission wavelength was found after reduction with sodium borohydride. In the previous study carried out by Zheng and co-workers [18], the relative QY of the reduced state carbon dots was also greatly enhanced by reducing carbon dots with $NaBH_4$. The proposed mechanism for the enhanced luminescence of reduced C-cots could be ascribed to the formation of surface hydroxyl groups [18]. The role of hydroxyl groups was verified by treating the C-dots with hydroxyl group-selective oxidant like $KMnO_4$ and the luminescence of the C-dots was greatly quenched, demonstrating that the luminescence enhancement of the C-dots is due to the formation of surface hydroxyl groups.

The fourier-transform infrared (FTIR) spectra were used to identify the surface groups of the bare C-dots. As shown in Fig. 6, the characterized absorption peaks of the broad 3393 cm^{-1} O–H stretching vibration and the 1622 cm^{-1} C=O stretching vibration were observed. The zeta-potential measurement in Table 1 indicated that the bare C-dots had negative value of −13.83 mV at pH 6.5. All the results revealed that the carboxyl groups of the C-dots were formed by treatment of plant soot with

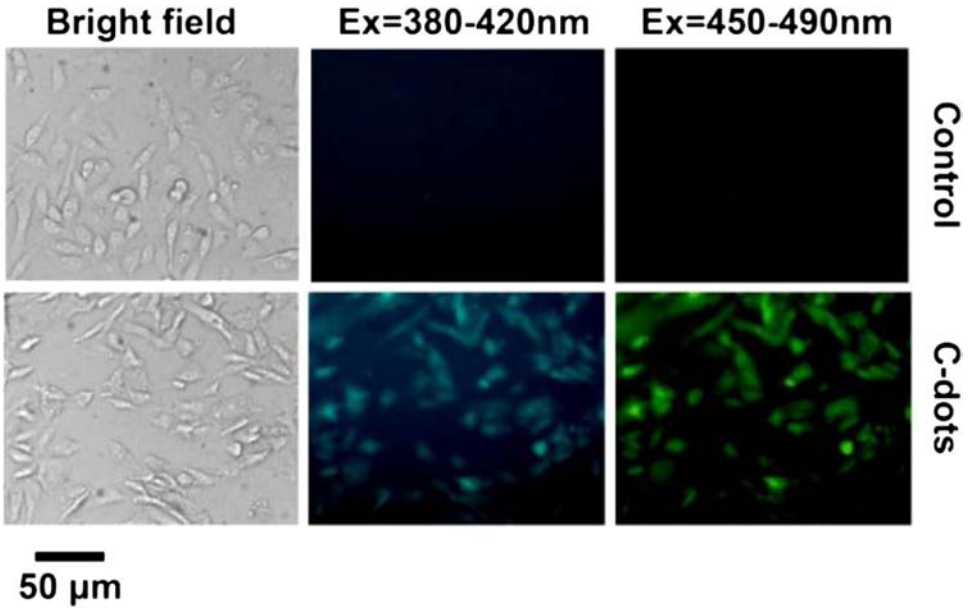

Fig. 7. Bright filed and fluorescence microphotographs of CHO (Chinese Hamster Ovary) cells incubated with PLL-passivated C-dots (C-dots) and without C-dots (Control) with an excitation filter of 380–420 nm and 450–490 nm for blue and green color respectively.

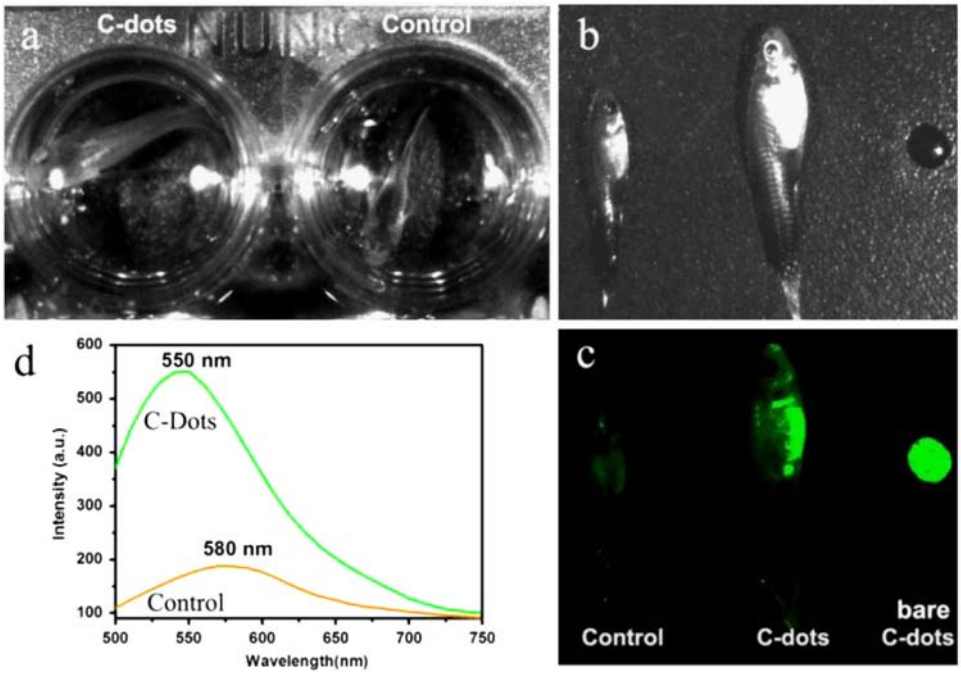

Fig. 8. *Ex vivo* guppy fish imaging. Photograph of the C-dots labeled guppy fish (C-dots) and its control (control) (a), brightfield image (b) and fluorescence image (c) of the C-dots labeled guppy fish, its control and bare C-dots measured with CRi Meastro imaging system. Fluorescence spectra comparison of the C-dots and its control (d). Exposure time was 1500 ms.

nitric acid reflux. The FTIR spectra of the passivated C-dots (Fig. 6b–d) clearly showed the presence of the passivation agents. As for the reduced C-dots, FTIR analysis (Fig. 6f) showed the absorption band of C=O at 1598 cm^{-1} shifted to 1613 cm^{-1}. The zeta-potential of the reduced C-dots decreased from −13.83 mV to −16.03 mV (Table 1). Hence these results indicated the transformation of the surface groups for the reduced C-dots.

The potential applications of the fluorescent C-dots derived from plant soot were evaluated in Chinese Hamster Ovary (CHO) cells and guppy fish imaging, respectively. The CHO cells incubated with PLL passivated C-dots in Fig. 7 become bright when exited at the wavelength of 380–420 nm and 450–490 nm, respectively. This indicated that the PLL passivated C-dots were able to label CHO cells after a simple incubation. Moreover, no quenching effect was observed in the continuous excitation for 15 min cell imaging. This demonstrates that the fluorescent C-dots derived from plant soot have a good photostability, which is essential for observation of the cell imaging for a long time.

For organisms imaging, small guppy fishes were selected because they are small aquatic craniate animals, in which the fluorescence signal can be easily observed with a Meastro *in vivo* imaging system. As shown in Fig. 8, the C-dots labeled guppy fish clearly showed enhanced luminescence as compared with its control with a signal-to-noise (S/N) ratio of 2.95. Fluorescence spectra analysis confirmed that the fluorescence signal was coming from the passivated C-dots. This result suggested that the passivated C-dots might have potential for small aquatic craniate animals imaging.

4. Conclusions

In summary, we have demonstrated a facile synthesis of fluorescent C-dots from cheap and easily obtained plant soot. These C-dots show multi-color fluorescence, excellent photostability and suitability for their use in various pH conditions. After surface passivation or reduction with sodium borohydride, the relative QYs of the C-dots can be further improved. These novel C-dots have been successfully used for cellular and fish imaging. The tunable multicolor emission, excellent photostability, good pH adaptablility and low cost production of these C-dots derived from cheap plant soot make them good candidates for more practical biomedical applications.

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References

[1] D.J. Bindl, M.Y. Wu, F.C. Prehn, M.S. Arnold, Nano Lett. 11 (2011) 455–460.
[2] S. Chandra, P. Das, S. Bag, D. Laha, P. Pramanik, Nanoscale 3 (2011) 1533–1540.
[3] L. Cao, S. Sahu, P. Anilkumar, C.E. Bunker, J.A. Xu, K.A.S. Fernando, P. Wang, E.A. Guliants, K.N. Tackett, Y.P. Sun, J. Am. Chem. Soc. 133 (2011) 4754–4757.
[4] X.J. Yang, D. Austin, M.A. Guillorn, V.I. Merkulov, A.V. Melechko, D.H. Lowndes, M.L. Simpson, Nanotechnology 3 (2003) 342–345.
[5] B. Tian, C. Wang, S. Zhang, L.Z. Feng, Z. Liu, Acs Nano 5 (2011) 7000–7009.
[6] C.J. Liu, P. Zhang, X.Y. Zhai, F. Tian, W.C. Li, J.H. Yang, Y. Liu, H.B. Wang, W. Wang, W.G. Liu, Biomaterials 33 (2012) 3604–3613.
[7] Y.P. Sun, B. Zhou, Y. Lin, W. Wang, K.A.S. Fernando, P. Pathak, M.J. Meziani, B.A. Harruff, X. Wang, H.F. Wang, P.J.G. Luo, H. Yang, M.E. Kose, B.L. Chen, L.M. Veca, S.Y. Xie, J. Am. Chem. Soc. 128 (2006) 7756–7757.
[8] S.T. Yang, L. Cao, P.G.J. Luo, F.S. Lu, X. Wang, H.F. Wang, M.J. Meziani, Y.F. Liu, G. Qi, Y.P. Sun, J. Am. Chem. Soc. 131 (2009) 11308–11311.
[9] Z.A. Qiao, Y.F. Wang, Y. Gao, H.W. Li, T.Y. Dai, Y.L. Liu, Q.S. Huo, Chem. Commun. 46 (2010) 8812–8814.
[10] R.L. Liu, D.Q. Wu, S.H. Liu, K. Koynov, W. Knoll, Q. Li, Angew. Chem. Int. Ed. 48 (2009) 4598–4601.
[11] W.C.W. Chan, S.M. Nie, Science 281 (1998) 2016–2018.
[12] L. Cao, X. Wang, M.J. Meziani, F.S. Lu, H.F. Wang, P.J.G. Luo, Y. Lin, B.A. Harruff, L.M. Veca, D. Murray, S.Y. Xie, Y.P. Sun, J. Am. Chem. Soc. 129 (2007) 11318.
[13] S.T. Yang, X. Wang, H.F. Wang, F.S. Lu, P.J.G. Luo, L. Cao, M.J. Meziani, J.H. Liu, Y. F. Liu, M. Chen, Y.P. Huang, Y.P. Sun, J. Phys. Chem. C 113 (2009) 18110–18114.
[14] J.C. Vinci, I.M. Ferrer, S.J. Seedhouse, A.K. Bourdon, J.M. Reynard, B.A. Foster, F. V. Bright, L.A. Colón, J. Phys. Chem. Lett. (2013).
[15] L. Cao, S.T. Yang, X. Wang, P.J.G. Luo, J.H. Liu, S. Sahu, Y.M. Liu, Y.P. Sun, Theranostics 2 (2012) 295–301.
[16] P.J.G. Luo, S. Sahu, S.T. Yang, S.K. Sonkar, J.P. Wang, H.F. Wang, G.E. LeCroy, L. Cao, Y.P. Sun, J. Mater. Chem. B 1 (2013) 2116–2127.
[17] Y. Xu, M. Wu, Y. Liu, X.Z. Feng, X.B. Yin, X.W. He, Y.K. Zhang, Chem. Eur. J. 19 (2013) 2276–2283.
[18] H.Z. Zheng, Q.L. Wang, Y.J. Long, H.J. Zhang, X.X. Huang, R. Zhu, Chem. Commun. 47 (2011) 10650–10652.
[19] H.T. Li, X.D. He, Z.H. Kang, H. Huang, Y. Liu, J.L. Liu, S.Y. Lian, C.H.A. Tsang, X.B. Yang, S.T. Lee, Angew. Chem. Int. Ed. 49 (2010) 4430–4434.
[20] W. Sun, Y.X. Du, Y.Q. Wang, J. Lumin. 130 (2010) 1463–1469.
[21] H.T. Li, H. Ming, Y. Liu, H. Yu, X.D. He, H. Huang, K.M. Pan, Z.H. Kang, S.T. Lee, New J. Chem. 35 (2011) 2666–2670.
[22] A. Jaiswal, S.S. Ghosh, A. Chattopadhyay, Chem. Commun. 48 (2012) 407–409.
[23] J.J. Zhou, Z.H. Sheng, H.Y. Han, M.Q. Zou, C.X. Li, Mater. Lett. 66 (2012) 222–224.
[24] H. Zhu, X.L. Wang, Y.L. Li, Z.J. Wang, F. Yang, X.R. Yang, Chem. Commun. (2009) 5118–5120.
[25] S. Liu, J.Q. Tian, L. Wang, Y.W. Zhang, X.Y. Qin, Y.L. Luo, A.M. Asiri, A.O. Al-Youbi, X.P. Sun, Adv. Mater. 24 (2012) 2037–2041.
[26] H.P. Liu, T. Ye, C.D. Mao, Angew. Chem. Int. Ed. 46 (2007) 6473–6475.
[27] L. Tian, D. Ghosh, W. Chen, S. Pradhan, X.J. Chang, S.W. Chen, Chem. Mater. 21 (2009) 2803–2809.
[28] Y.Y. Li, L. Xu, T. Chen, X.Y. Liu, Z.G. Xu, H.X. Zhang, Anal. Chim. Acta 726 (2012) 102–108.
[29] W. Zhang, G.F. Wang, N. Zhang, C.H. Zhang, B. Fang, Chem. Lett. 38 (2009) 28–29.
[30] K.S. Prasad, M.C. Chuang, J.A.A. Ho, Talanta 88 (2012) 445–449.
[31] D.Y. Pan, J.C. Zhang, Z. Li, C. Wu, X.M. Yan, M.H. Wu, Chem. Commun. 46 (2010) 3681–3683.
[32] D.Y. Pan, J.C. Zhang, Z. Li, M.H. Wu, Adv. Mater. 22 (2010) 734–736.