High-quality full-color carbon quantum dots synthesized under an unprecedentedly mild condition

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Highlights
- High-quality full-color CQDs was fabricated under unprecedentedly mild conditions
- Stable and narrow fluorescent emissions were performed with high quantum yields
- LEDs with different-color and white light emissions were fabricated
- Logic gate sensors that could distinguish multiple metal ions were developed

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High-quality full-color carbon quantum dots synthesized under an unprecedentedly mild condition

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SUMMARY
Carbon quantum dots (CQDs) are highly promising to be applied in light-emitting, chemosensing, and other cutting-edge domains. Herein, we successfully fabricate high-quality full-color CQDs under unprecedentedly low temperature and pressure (85 °C, 1.88 bar). Stable and narrow fluorescent emissions ranging from blue to green and red light were realized by simple amine engineering, which were further mixed into white-light CQDs with the absolute photoluminescent quantum yield reaching 19.2%. The average mass yield of the CQDs reached 69.0%. The optical performances demonstrated that the CQDs possessed uniform luminescent centers and dominant radiative decay channels. Component analysis further suggested that dehydrated condensation between carboxyl and amine groups directed the growth of the CQDs. By utilizing the CQDs, full-color light-emitting diodes and logic gate sensors were developed. This study paves an important step for promoting the application of CQDs by providing an energy-efficient, safe, and productive synthetic strategy.

INTRODUCTION
The discovery of room temperature light emission from quantum-sized carbon (<10 nm) in 2006 has triggered intensive research on light-emitting carbon quantum dots (CQDs) (Sun et al., 2006). The tunability of their photoluminescent spectra and their surface chemistry make them highly potential candidates to be used in light-emitting, chemosensing, and other domains (Wang et al., 2017, 2020; Jiang et al., 2015; Li et al., 2017). Moreover, CQDs also outperform many other fluorescent nanomaterials in terms of their high stability and environmental friendliness (Chung et al., 2020; Shi et al., 2021; Zhu et al., 2013; Huang et al., 2012; Yuan et al., 2019; Vasilopoulou et al., 2020).

Plenty of studies have been devoted to developing novel methods for the synthesis of high-quality photoluminescent CQDs, which are unexceptionally classified as “top-down” or “bottom-up” methods (Semeniuk et al., 2019; Nekoueian et al., 2019; Wareing et al., 2021; Briscoe et al., 2015). For a “top-down” method, CQDs are synthesized by breaking down larger carbon materials such as graphite that contain abundant sp²-hybridized carbon atoms, via arc discharge, laser ablation, and other energy-intensive strategies (Chung et al., 2021; Xu et al., 2013). On the other hand, high temperature, high pressure, and/or strong oxidative reagents are usually requested for the “bottom-up” synthesis of CQDs from small molecular precursors such as carbohydrates and amino acids, for the purpose of carbonizing the sp³ carbon to sp² carbon (Mosconi et al., 2015; Li et al., 2021; Ding et al., 2016; Baragau et al., 2020, 2021). Thus, hydrothermal and solvothermal reactions are frequently adopted in “bottom-up” methods. Besides the high energy consumption in these methods, the harsh synthetic conditions also raise the concerns on the safety of the methods and decrease the productivity by transforming the precursors partially to gaseous byproducts.

Up to now, the development of synthetic methods that are energy efficient, safe, and highly productive for CQDs is still on the way, which would be of great value for promoting the applications of CQDs in industry, by circumventing complicated production facilities and high energy input. Even the electrochemical methods are much greener and safer than their counterparts (Wang et al., 2014), and extra separation steps for obtaining optically homogeneous CQDs are required (Bao et al., 2011). Besides, the recently reported...
mild "bottom-up" methods by using phenols and ethylenediamine/diethylenetriamine as the precursors only produced blue and green CQDs (An et al., 2021; Liu et al., 2020; Zhang et al., 2018).

Herein, we develop a mild condensation strategy to produce high-quality full-color CQDs (Figure 1). 1,3,5-benzenetricarboxylic acid (BTCA) that contains only sp$^2$ carbon is chosen as the acid reagent, whereas diethylenetriamine (DETA), m-phenylenediamine (mPDA), o-phenylenediamine (oPDA), and p-phenylenediamine (pPDA) are used as the basic reagents. Different from the hydrothermal and solvothermal methods that relied on high temperature (180–250°C) and high pressure (up to 248 bar) (Baragau et al., 2020, 2021), our one-pot method was conducted at 85°C and nearly atmospheric pressure (about 1.88 bar). The fluorescent spectra of the obtained CQDs were successfully modulated over a wide range from blue to red by amine engineering. And white light emission was obtained by mixing the CQDs. The absolute fluorescence quantum yield (QYs) reached 19.2% for the white CQDs, and the average mass yield of the CQDs reached 69.0%. Moreover, owing to the high luminescence and the diversified color of the CQDs, light-emitting diodes (LEDs) with different-color and white light emission were fabricated, and logic gate sensors that could distinguish multiple metal ions with high accuracy were developed.

RESULTS AND DISCUSSION
Preparation and optical properties of the CQDs

Different from the traditional methods using carbohydrate, amino acids, or citrate as precursors, precursors containing only sp$^2$ carbon were first selected for the synthesis of CQDs in this study, by excluding the need for carbonizing the sp$^3$ carbon into sp$^2$ carbon. BTCA was selected as the acidic reagent, and mPDA, oPDA, and pPDA were chosen as the basic reagents. The reactions were performed by heating the mixture of BTCA and one of the selected bases in N,N-dimethylformamide (DMF) in glass bottles under 85°C for only 6 h, which was far below the temperatures in hydrothermal and solvothermal reactions (Wang et al., 2017, 2020; Ding et al., 2016; Qin et al., 2019; Meng et al., 2021; Liu et al., 2012; Yuan et al., 2018). Three CQDs that presented bright blue, green, and red light emission bands centered at about 440, 550, and 605 nm, respectively, were obtained (Figures 2A–2C). The three CQDs were termed m-bCQDs, o-gCQDs, and p-rCQDs, respectively.

For comparison, a range of amines containing sp$^3$ carbon were also selected as the basic reagents (Figure S1). Most of the as-prepared solutions emitted very weak blue fluorescence except for that of DETA, which emitted even the strongest blue light among all the blue light emitters (termed d-bCQDs, fluorescent band centered over 430 nm; Figures 2D and S2). Thus, DETA together with the other three phenylenediamines were used for the synthesis of CQDs in the followed experiment. It is notable that the use of DETA and another precursor containing only sp$^2$ carbon (p-aminophenol) in preparing bright blue CQDs under mild conditions also succeeded before (An et al., 2021).
To obtain the optimal reaction conditions, the effect of the solvent was first explored. It was observed that high-quality and diversified photoluminescence could only be achieved by using DMF as the solvent (Figure S3), which might be attributed to the higher basicity of DMF. Subsequently, the reaction temperature, time, and ratios of precursors were investigated thoroughly (Figures S4, S5 and S6, text S1.4). It was found that reacting at 85°C for 6 h was the optimal condition for d-bCQDs, m-bCQDs, and o-gCQDs and also produced satisfying p-rCQDs (Figure S4). At lower temperature, the growth of CQDs seemed to be much slower, whereas at higher temperature, the growth might be too fast to form the high-quality emitting centers of the CQDs. Besides, it was not commentary to significantly extend the reaction time, as the overgrowth might lead to products exceeding the quantum sizes.

As far as we know, full-color CQDs were scarcely prepared under 100°C by using any other “bottom-up” methods (Figure 2E) (Wang et al., 2020; Ding et al., 2016; Qin et al., 2019; Meng et al., 2021; Liu et al., 2012; Yuan et al., 2018). The temperature was even far below the boiling point of the solvent, which left the pressure estimated to be 1.88 bar (text S1.3). The low pressure could be borne by glass sample bottles, in sharp contrast to the high pressures in the hydrothermal and solvothermal processes that can only be borne by autoclaves. The relative low temperature and pressure are highly beneficial for the practical application of the CQDs, as complicated facilities and high energy input can be circumvented for their production.

Moreover, the average mass yield of the CQDs reached 69.0% (Table S1), which was comparable to previous work (Meng et al., 2021). The absolute photoluminescent quantum yield for d-bCQDs, m-bCQDs, o-gCQDs, and p-rCQDs was determined to be 35.3%, 0.6%, 16%, and 7.9% (Figure S7). And it was found the CQDs kept high photoluminescent stability under a long-term excitation (Figure S8) and presented good reproducibility among different batches, which also demonstrated the capacities of the CQDs for practical application.

Furthermore, the optical performances of the CQDs were investigated in details. The gradually redshifted fluorescent bands were consistent with the redshifted absorption bands, and the position of the excitation...
played an important role in the growth of these CQDs, as speculated in Figure S11. The dehydrate reactions might either be negligible or play little effects in the decay processes (Ding et al., 2016; Yuan et al., 2018).

Structure analysis
To further elucidate the compositions and structures of the CQDs (Figure S11), 1HNMR analysis was first adopted. Taking o-gCQDs as an example, in the as-synthetic product, the signals of excessive BTCA (8.61 ppm) and oPDA (6.35 ppm–6.55 ppm) were observed (Figure S12). Moreover, some of the characteristic proton signals of the benzimidazole were appeared (7.19 and 7.59 ppm); thus, benzimidazole moieties were supposed to be formed in o-gCQDs. Another two new singlet peaks appeared in high field, the peak with larger integration result was assigned to the condensed BTCA moieties (7.95 ppm), and the other peak was assigned to the imide moieties (8.20 ppm). Except for the characteristic proton peaks, some of the integral peak area of the proton signals presented good ratio relationship (Figure 2F), which further suggested the dehydrate condensation between the carboxyl and amine groups (Figure S12). Similar to o-gCQDs, imide groups were plausibly identified in d-bCQDs, m-bCQDs, and p-rCQDs (Figures S13, S14 and S15).

Moreover, these CQDs exhibited similar Fourier transform infrared (FTIR) spectra (Figure S16). The strong vibration bands of C=O were observed at 1510 cm−1 (Chen et al., 2021; Weetman et al., 2019). Bands at 3435, 1651, 1380, and 1100 cm−1 were ascribed to the stretching vibration of N-H, C=O, C-N, and C-O (An et al., 2021; Chen et al., 2021), respectively, which also demonstrated that the dehydrate condensation occurred between the carboxyl and amine groups. X-ray photoelectron spectroscopy (XPS) further verified the presence of carboxyl and amine in the CQDs (Figure S17) (Weetman et al., 2019). Moreover, the thermogravimetric analysis also indicated the containing of oxygen- and nitrogen-containing groups within the CQDs (Figure S18). When the temperature reached about 120°C, weight losses up to 8% were observed, which should be attributed to the evaporation of solvent (H2O and DMF). Subsequently, the weight losses of the CQDs above 120°C could be attributed to the decomposition of oxygen- and nitrogen-containing groups. Even being heated to 450°C, the weight losses were all less than 30%. The residues at 450°C should be attributed to the carbon cores of the CQDs. These data demonstrated that dehydrate condensation played an important role in the growth of these CQDs, as speculated in Figure S11. The dehydrate reactions other than extensive carbonization might be the reason for the succeeded synthesis of the CQDs under the mild reaction conditions. It is also highly notable that the mild synthetic method was beneficial for resolving the structures and condensation routes of the CQDs, as complex carbonization processes could be excluded.

In addition, as shown in Figure 3, all of these CQDs showed narrow size distribution with the average sizes of about 2.2 nm (d-bCQDs), 2.5 nm (m-bCQDs), 2.9 nm (o-gCQDs), and 3.4 nm (p-rCQDs), respectively. The high-resolution transmission electron microscopic (HRTEM) images further showed the in-plane lattice spacing of graphene (100 facet) in all the CQDs (Figures 3E–3H). X-ray powder diffraction (XRD) patterns around 25° corresponded to the (002) layer spacing of graphene in all the CQDs (Figure S19). AFM revealed that the heights were in the range from 1 to 2 nm, elucidating that they consisted of two–five layers of graphene. Besides, the matrix-assisted laser desorption/ionization-mass spectrometry (MALDI-MS) showed that the ions of CQDs were distributed in the mass-to-charge ratio range from 330 to 1500 (Figure S20). And from d-bCQDs, m-bCQDs, o-gCQDs, to p-rCQDs, the mass-to-charge ratios gradually increased, which was well consistent with the increasing of the overall sizes of the CQDs. The gradually increased sizes of the CQDs were consistent with the redshifted fluorescence and the redshifted absorption bands, which demonstrated the quantum confinement effect was probably embedded behind the redshifted spectra.
Figure 3. TEM and AFM characterization of the CQDs
(A) TEM image of d-bCQDs.
(B) TEM image of m-bCQDs.
(C) TEM image of o-gCQDs.
(D) TEM image of p-rCQDs.
(E) Highlighted lattice image of d-bCQDs.
(F) Highlighted lattice image of m-bCQDs.
(G) Highlighted lattice image of o-gCQDs.
(H) Highlighted lattice image of p-rCQDs.
(I) Statistically derived size distributions of d-bCQDs from its TEM image.
(J) Statistically derived size distributions of m-bCQDs from its TEM image.
(K) Statistically derived size distributions of o-gCQDs from its TEM image.
(L) Statistically derived size distributions of p-rCQDs from its TEM image.
(M) AFM image of d-bCQDs.
(N) AFM image of m-bCQDs.
(O) AFM image of o-gCQDs.
(P) AFM image of p-rCQDs.
Meanwhile, the emission spectra were also redshifted along with the increase of the C-N/C≡N ratio and the positive shifts of the Zeta potentials (Figures S17 and 4B), which indicated the surface states might also be involved in the emission efficiency. When triggered by different pH and different target ions, the photoluminescent intensities of the CQDs were changed, whereas the maximum emission wavelengths remained nearly constant (Figures S21 and S22). Thus, it was believed that the fluorescence of the CQDs origins from
the surface states and the energy gaps of the surface emissive sites were determined by both the sizes of the $\pi$-electron systems and the surface chemistries. Similar mechanism was also reported before (Bao et al., 2015; Liu et al., 2018). To further verify this proposal, $\text{H}_2\text{O}_2$ was mixed with the CQDs. It was observed that the emission intensities of CQDs decreased, but the emission wavelengths only shifted slightly (Figure S23). In such case, surface functional groups of CQDs were oxidized but the carbon cores were insusceptible to the damage.

Full-color CQDs preparation and application in LEDs

By mixing the solutions of d-bCQDs, o-gCQDs, and p-rCQDs at different volume ratios, various photoluminescent colors including blue, cyan, green, orange, red, and even white were created (Figures 4A and S24). The QY reached 19.2% for the white CQDs from this facile strategy, which is comparable to most of the reported CQDs that depended on other harsh preparation methods (Wang et al., 2020; Ding et al., 2016; Qin et al., 2019; Meng et al., 2021; Liu et al., 2012; Yuan et al., 2018). To further obtain solid-state CQDs, an electrostatic deposition method was used to deposit the CQDs on positively charged SiO$_2$ microparticles (modified with APTES), as the CQDs exhibited negative Zeta potentials that were attributed to the excessive carboxyl groups (Figures S16 and 4B). The value of the Zeta potential of the positively charged SiO$_2$ was $+21.1$ mV, whereas the Zeta potentials were $-32.9$, $-22$, $-25.6$, and $-15.4$ mV for d-bCQDs, m-bCQDs, o-bCQDs, and p-rCQDs, respectively. The SiO$_2$ microparticles were firstly well dispersed in the solution of the CQDs and then centrifuged. Almost all the CQDs were precipitated along with SiO$_2$ after centrifugation (Figure 4C). The dried powders exhibited the characteristic fluorescence emission bands of each CQDs. The solid-state CQDs displayed fluorescent emission spectra similar to the CQDs solution (Figures 4D–4G).
On the basis of the excellent fluorescent emission, the solid-state CQDs were successfully prepared into bright and uniform full-color light-emitting diodes (LEDs). Figures 5A–5C displayed the close-up view of the obtained LEDs with surface emission from blue and green to red emission of CQDs-based monochrome LEDs. The CIE coordinates of the emitted lights of these LEDs were (0.17, 0.1), (0.29, 0.6), and (0.61, 0.41) (Figure 5E), respectively. The emission spectra of the LEDs were shown in Figure 5F. By combining the d-bCQDs, m-bCQDs, and p-rCQDs with the ratio of 1:2:3 (V/V/V), the white LED with the CIE color coordinate (0.27, 0.31) has also been achieved (Figure 5D). The emission spectrum of the white LED covered the whole visible light region from 400 to 750 nm (Figure 5F). The full-color LEDs demonstrated that the CQDs were promising candidates for display and lighting.

Application in chemical sensing

It was found that the fluorescence of the CQDs could be tuned by pH and metal ions, as protonation/deprotonation and coordination sites, i.e. carboxyl groups and amino groups (Figures S21, S22 and 4B). Hence the CQDs could be further used for sensing pH and multiple metal ions (Figures 6, S21 and S22). The CQDs showed monotonous responses to pH in the range from 2.2 to 11 (Figure S21). The relative changes of the fluorescent intensities of d-bCQDs, m-bCQDs, and p-rCQDs after treated with ions were shown in Figure 6B. The different responding patterns demonstrated that the four CQDs could be used as a fluorescent array for distinguishing metal ions. The inner-filter effect was obviously insufficient to explain the responses (Figure S25), and the embedded mechanisms may need detailed studies in the future. Still, these disparate responses illustrated that the CQDs possessed rich response mechanisms, which provided a valuable chance to construct high-throughput devices for environmental analysis, medical diagnosis, food analysis, and other applications. Inspired by these responses to metal ions,

Figure 6. Application in chemical sensing

(A) Photos of d-bCQDs, m-bCQDs, o-gCQDs, and p-rCQDs treated with ions (1 mg L⁻¹) under UV light (365 nm).
(B) The biased ratios of the fluorescent intensities after treated with different ions (1 mg L⁻¹), respectively.
(C) The CQDs-based AND logic gates for identifying (I) Hg²⁺ and (II) Cd²⁺.
CQDs-based logic gate sensors could be further constructed. Take Hg\textsuperscript{2+} and Cd\textsuperscript{2+} as two examples, d-bCQDs, m-bCQDs, o-gCQDs, and p-rCQDs formed logic gates (Figure 6C).

CONCLUSIONS
In summary, developing energy-efficient, safe, and productive synthetic methods for full-color CQDs is crucial for their further success in LEDs, chemical sensing, and other applications. In this study, we report a new method in glass bottles under low pressure (1.88 bar) and with the reaction temperature as low as 85°C. The obtained CQDs possessed high QYs, high productivities, uniform luminescent centers, and dominant radiative decay channels. Besides, the mild reaction condition also made it possible to resolve the structures and condensation routes of the CQDs in depth. Full-color LEDs and logic gate sensors were developed based on the obtained high-quality CQDs. This study paves an important step for promoting the application of CQDs by providing an unprecedentedly mild synthetic strategy for full-color CQDs. This study also inspires the development of other mild synthetic methods that leave the structures of CQDs fully resolvable in the future, which is invaluable for building clear structure-property relationship and tuning photoluminescent properties at molecular accuracy.

Limitations of the study
Full-color CQDs were fabricated under mild conditions with the average mass yield reaching 69.0%. In the follow-up research, it is expected to further elucidating the growth mechanism by using sophisticated structural analytical techniques such as scanning tunneling microscopy (STM).

STAR METHODS
Detailed methods are provided in the online version of this paper and include the following:

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Supplemental information can be found online at https://doi.org/10.1016/j.isci.2022.104421.

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AUTHOR CONTRIBUTIONS
Y.-J. Tong prepared the materials, studied the fluorescent properties, and wrote the manuscript; L.-D. Yu, Y. Huang, and Q. Fu helped with the study on the fluorescent properties; N. Li, Y. Li, and Y. Ye helped with the characterizations of AFM, TEM, and FTIR; J. Xu designed the experiment and helped with the funding acquisition and supervision. F. Zhu, J. Pawliszyn, and G. Ouyang helped with project administration, review, and editing.
DECLARATION OF INTERESTS
The authors declare no competing interests.

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**STAR METHODS**

**KEY RESOURCES TABLE**

| REAGENT or RESOURCE | SOURCE | IDENTIFIER |
|---------------------|--------|------------|
| **Chemicals, peptides, and recombinant proteins** |        |            |
| 1,3,5-benzenetricarboxylic acid | Aladdin (Shanghai, China) | CAS: 554-95-0 |
| o-phenylenediamine | Aladdin (Shanghai, China) | CAS: 95-54-5 |
| m-phenylenediamine | Aladdin (Shanghai, China) | CAS: 108-45-2 |
| p-phenylenediamine | Aladdin (Shanghai, China) | CAS: 106-50-3 |
| N,N-dimethylformamide | Aladdin (Shanghai, China) | CAS: 68-12-2 |
| Zinc nitrate hexahydrate | Aladdin (Shanghai, China) | CAS: 10196-18-6 |
| Cadmium nitrate | Aladdin (Shanghai, China) | CAS: 10325-94-7 |
| Cobaltous nitrate hexahydrate | Aladdin (Shanghai, China) | CAS: 10026-22-9 |
| Chromium(III) nitrate nonahydrate | Aladdin (Shanghai, China) | CAS: 7789-02-8 |
| Copper nitrate trihydrate | Aladdin (Shanghai, China) | CAS: 10031-43-3 |
| Ferric nitrate nonahydrate | Aladdin (Shanghai, China) | CAS: 7782-61-8 |
| Diethylenetriamine | Makclin (Guangzhou, China) | CAS: 111-40-0 |
| Mercuric nitrate | Makclin (Guangzhou, China) | CAS: 10045-94-0 |
| Nickel(II) nitrate hexahydrate | Makclin (Guangzhou, China) | CAS: 13478-00-7 |
| Uranyl(VI) nitrate hexahydrate | Beijing HWRK (Beijing, China) | CAS: 13520-83-7 |
| **Other** |        |            |
| UV chip | Ruibaoguang Technology Company (Shenzhen, China) | NA |

**RESOURCE AVAILABILITY**

**Lead contact**
Further information and requests for resources should be directed to and will be fulfilled by the lead contact, Jianqiao Xu (xujq27@mail.sysu.edu.cn).

**Materials availability**
All unique/stable reagents generated in this study are available from the lead contact with a completed materials transfer agreement.

**Data and code availability**
All data are available in the paper and in supplemental information, and/or from the corresponding authors upon reasonable request. This paper does not report original code.

**EXPERIMENTAL MODEL AND SUBJECT DETAILS**
This work did not need any unique experimental model.

**METHOD DETAILS**

**Preparation of CQDs**
For the DETA originated blue CQDs (d-bCQDs), BTCA (0.015 g) and DETA (120 μL) were placed in a 20 mL glass bottle. DMF (12 mL) and H₂O (1.4 mL) were added. Then, the bottle was sealed with a TFPE septum lined steel cap, and heated up to 85°C and kept at this temperature for 6 h. Subsequently, the as-synthesized d-bCQDs solution was obtained with strong blue light emission under UV light (365 nm). Similarly, the other CQDs were prepared by replacing DETA with oPDA, mPDA, or pPDA. It should be noted, the masses of oPDA, mPDA, and pPDA were all controlled at 0.015 g. By referring to previous work, the as-synthesized solutions can be purified via silica column chromatography (Ding et al., 2016; Alaş and Genç, 2021; Jia et al.,
It was found the mixture components were more beneficial on the applications of sensing metal ions. The d-bCQDs, m-bCQDs, and o-bCQDs were directly used in following studies, but the treated p-rCQDs solution was diluted to 20–50 times before use.

**Preparation of CQDs with different ratios of precursors**

For the o-phenylenediamine (oPDA) originated o-bCQDs, BTCA (0.015 g) was separately mixed with oPDA (0.005, 0.010, 0.015, 0.020, 0.025 g) in five 20 mL glass bottle. DMF (12 mL) and H2O (1.4 mL) were added. Then, the bottle was sealed with a TFPE septum lined steel cap, and heated up to 85°C and kept at this temperature for 6 h. Subsequently, the as-synthesized o-bCQDs solution was obtained. Similarly, the other CQDs were prepared by replacing oPDA with different contente of DETA, mPDA, or pPDA.

**QY measurement**

The absolute QY values were measured from the ratios between photons emitted and absorbed by CQDs. The measurements were performed using an integrating sphere attached to Edinburgh FLS 1000. The excitation light was set at 360 nm with using a 450 W Xenon lamp. The CQDs solutions were placed in a UV quartz cuvette with a light path of 10 mm. The solvent DMF filled in the quartz cuvette was used as a blank sample for reference. The spectral correction curve that related to the sensitivity of the monochromator, detector, sphere coating, and optics to wavelength was provided by Edinburgh Instruments.

**LED fabrication**

All LEDs were fabricated with similar preparation process by referring previously reported work (Wang et al., 2020). Taking WLED as an example, 200 μL of the CQDs solution was mixed with the solution of polydimethylsiloxane precursors (Dow Corning 184 glue dissolved in cyclohexane, 0.1 g mL⁻¹). Then, the mixture was stirred for 10 min (1000 rpm) and added dropwise to the UV chip. Subsequently, the device was dried in an oven at 50°C for 30 min.

**Procedures for sensing metal ions**

The metal salt stock solutions were prepared in DMF (concentration 10 mg L⁻¹). The 100 μL of the aforementioned CQDs solution and an aliquot of metal salt solution (200 μL) were mixed. The mixture was diluted to a final volume of 700 μL by DMF. Subsequently, the photoluminescence spectra were collected under the maximum excitation wavelength.

**Reaction pressure calculation**

In this work, a series of CQDs was prepared in glass bottles with the reaction temperature at 85°C. We made a crude estimate of the reaction pressure. In a sealed glass bottle, the volumes of the gas and liquid in the bottle was considered unchanged as the reaction temperature far below the boiling point of the solvent (152.8°C for DMF and 100°C for H2O under 1 atm). The real pressure in the bottle was the sum of the gas pressure and vapor pressures under heating process.

First, the gas pressure was estimated using the equation:

\[
\frac{(P_1V_1)}{T_1} = \frac{(P_2V_2)}{T_2}
\]

where P, V, T denoted the pressure, volume and temperature for gas in the glass bottle, respectively. The initial P₁ was about 1 atm at 25°C (298.15 K). When the reaction temperature reached 85°C (358.15 K), the pressure in the glass bottle was calculated to be about 1.20 bar. Second, the vapor pressures were calculated using the Antoine equation (Thomson, 1946):

\[
\log P = A - B/T + C
\]

where A, B and C are the empirical constant. T and P denoted as the parameters of Celsius temperature (°C) and vapor pressure (mmHg), respectively. For H2O, the parameters of A, B and C are 7.96681, 1668.210 and 228.000 in the temperature range of 60–150°C, respectively. When the reaction temperature reached 85°C (358.15 K), the vapor pressure of H2O in the glass bottle was calculated to be 0.56 bar. For DMF, the parameters of A, B and C are 6.99608, 1437.840 and 199.830 in the temperature range of 60–350°C, respectively. When the reaction temperature reached 85°C (358.15 K), the vapor pressure of DMF in the glass bottle was calculated to be 0.12 bar. Therefore, the whole pressure in the reacting environment was calculated to be 1.88 bar.
QUANTIFICATION AND STATISTICAL ANALYSIS
Figures represent averaged or representative results of multiple independent experiments. Analyses and plots were performed with Origin.

ADDITIONAL RESOURCES
There are no additional resources needed to be declared in this manuscript, additional requests for this can be made by contacting the lead contact.