

Supplementary materials

1. Cl-contained graphene quantum dots (Cl-GQDs)

Cl-GQDs were purchased from the Nanjing XFNANO Materials Tech Co. Ltd.,
China ([Http://www.xfnano.com](http://www.xfnano.com)).

Table 1S Essential information

Items	Content
Synthesis method	Hydrothermal method
Standard	Emitting blue light
Purity	~ 80% quantum dots
Concentration	Standard concentration: 1 mg/mL, Maximum concentration: 20 mg/mL
Packaging	Minimum package is 100 mL
Quantum yield	< 5%

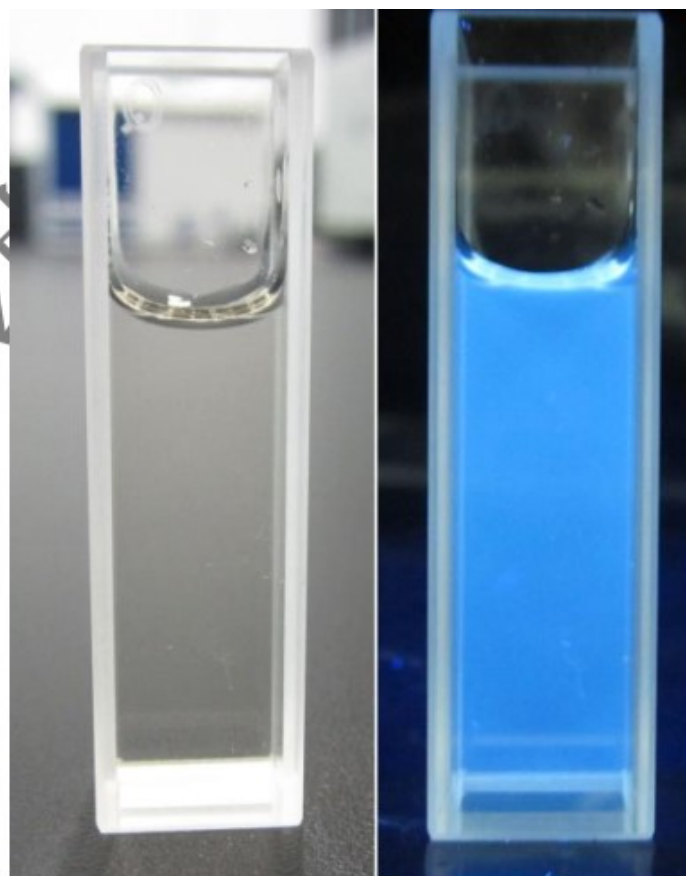


Fig. 1S Photos, photo under the natural light (left), photo under 365 nm UV light
(right) of Cl-GQDs

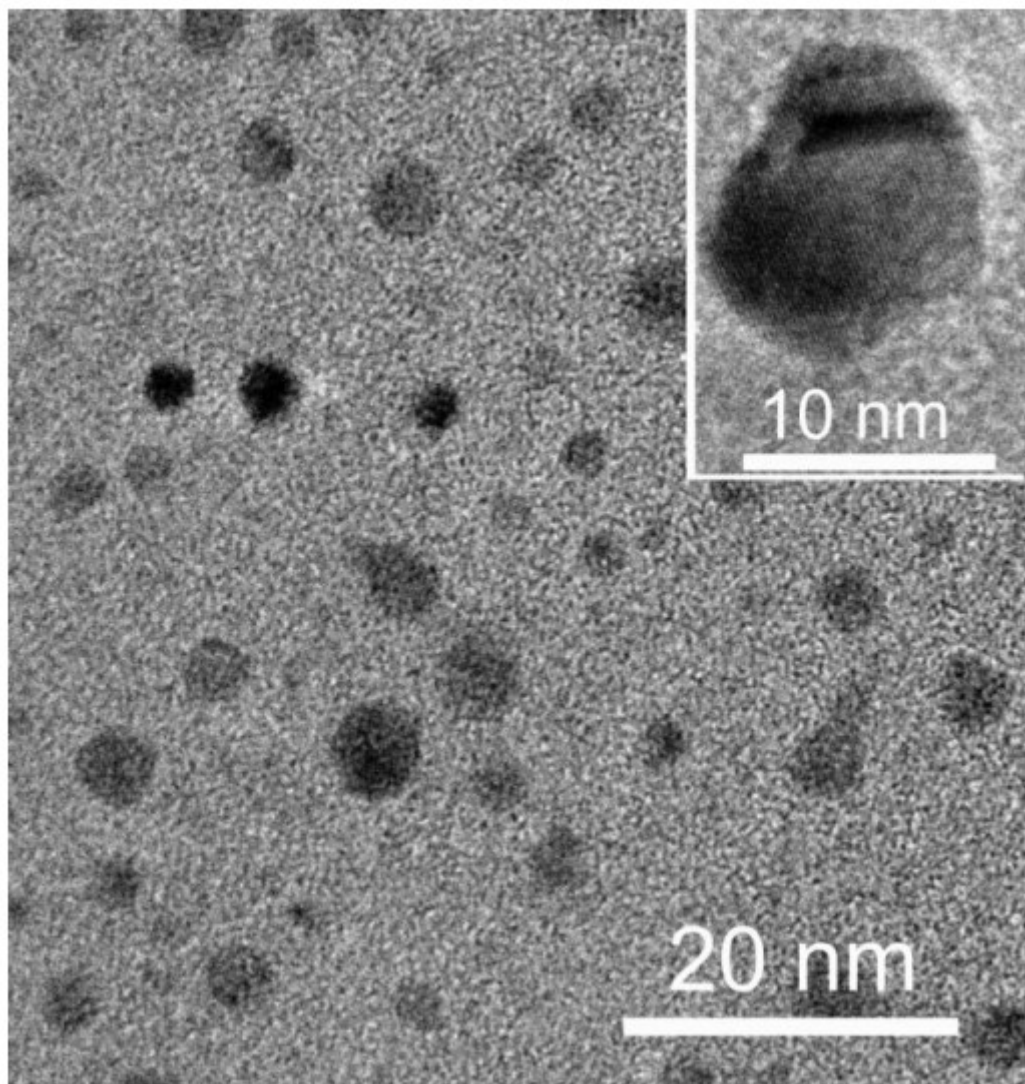


Fig. 2S SEM image of Cl-GQDs

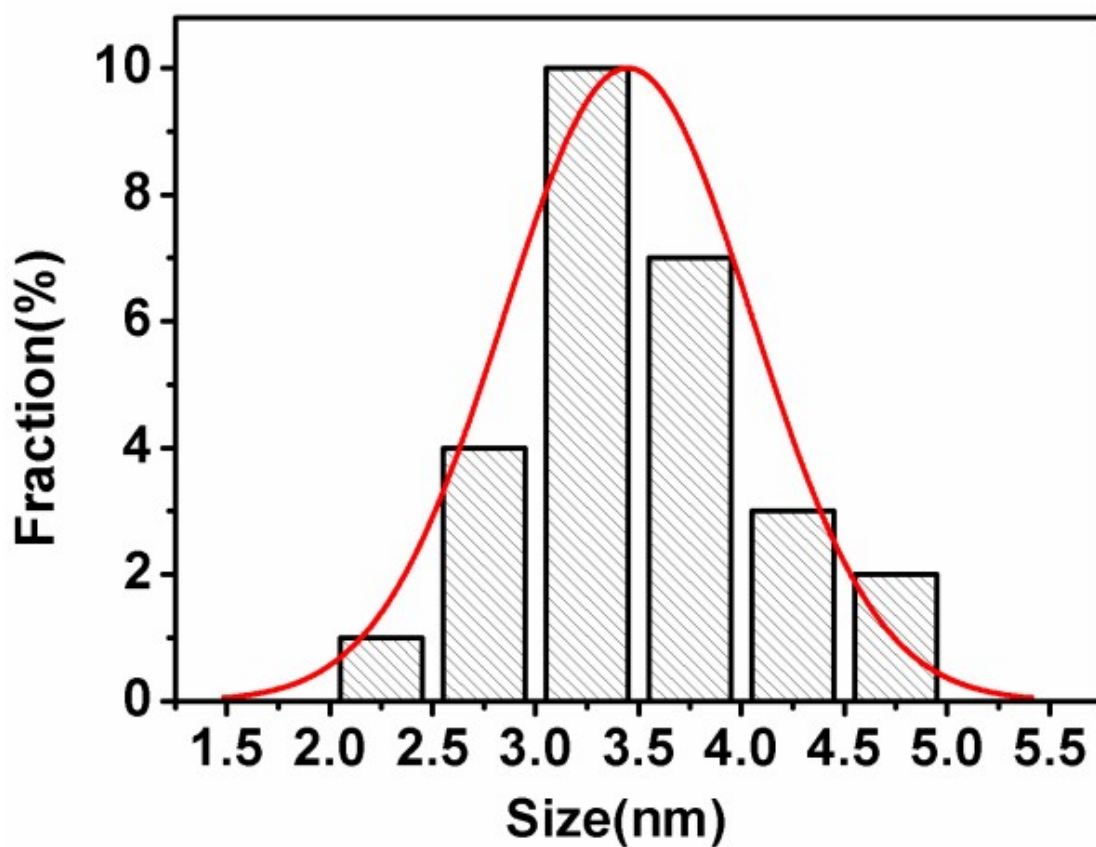


Fig. 3S Size distribution of Cl-GQDs

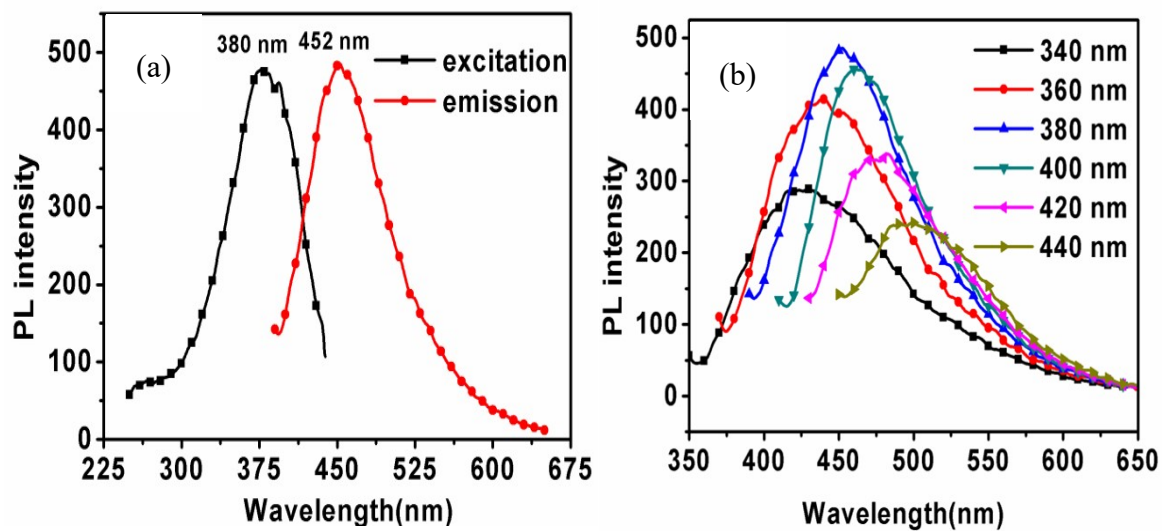


Fig. 4S PLE&PL spectra of Cl-GQDs: (a) PLE&PL spectra, (b) PL spectra excited at different excitations

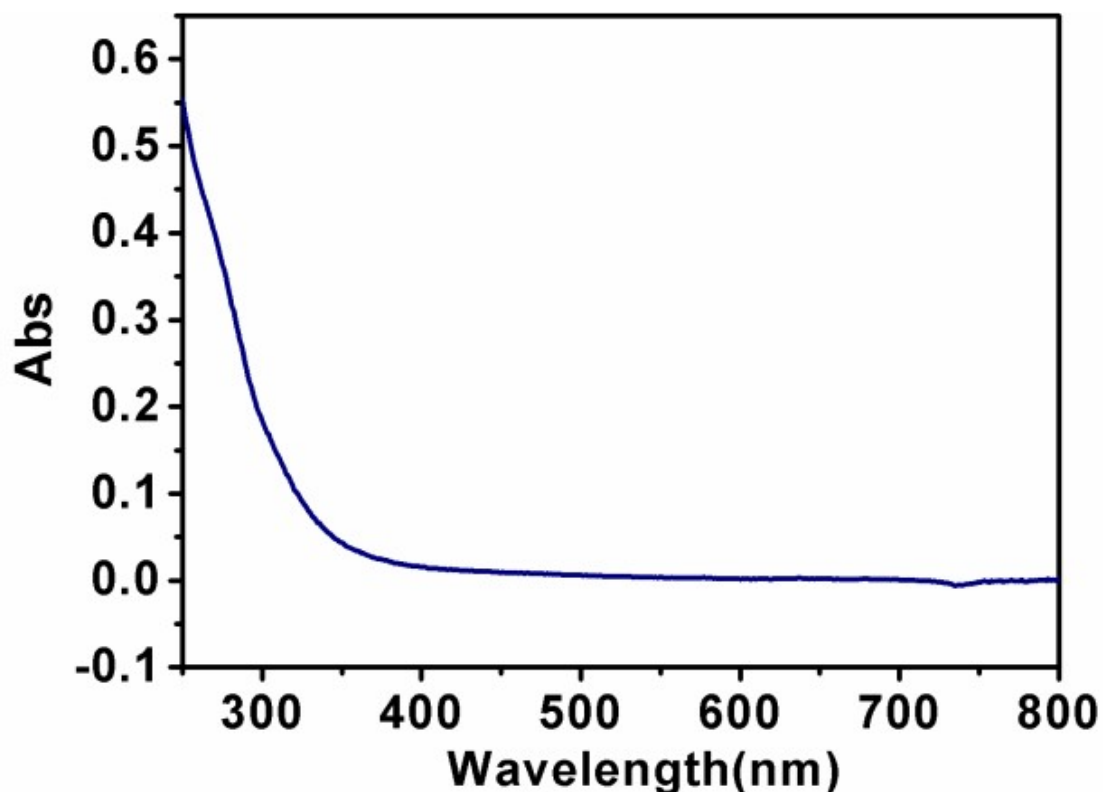


Fig. 5S UV-Vis absorption spectrum of Cl-GQDs

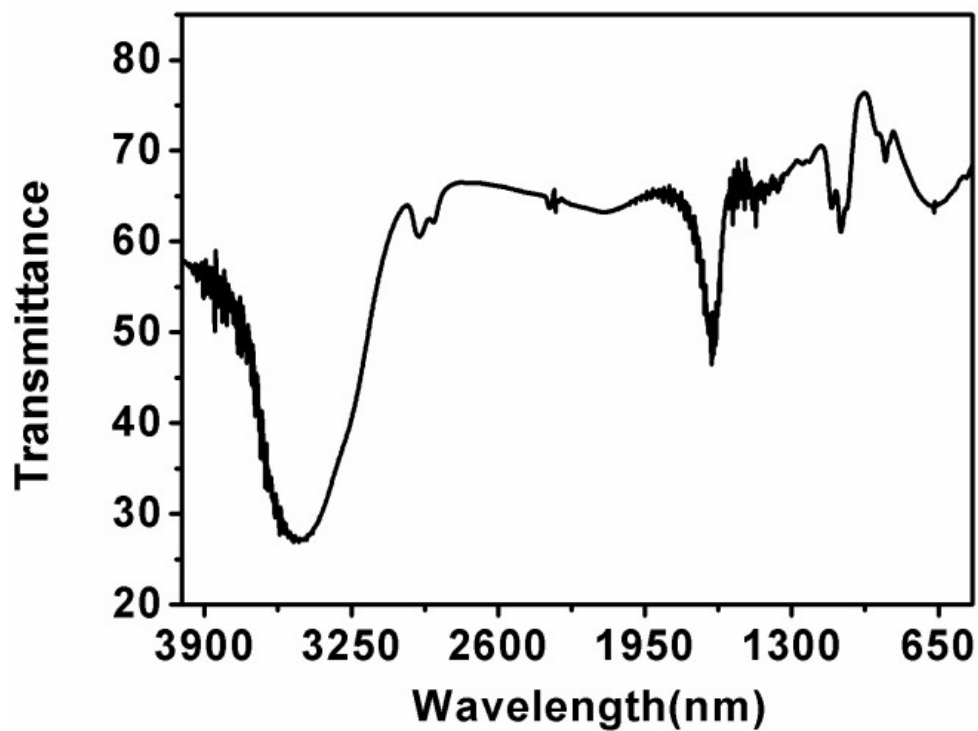


Fig. 6S FT-infrared spectrum of Cl-GQDs

2. Experimental procedure and methods

2.1. Reagent and apparatus

Cl-contained graphene quantum dots (Cl-GQDs) were purchased from the Nanjing XFNANO Materials Tech Co. Ltd., China, and used without further purification. All chemicals were reagent-grade pure and purchased from the Sinopharm Chemical Reagent Co. Ltd., China.

The elemental contents of sample were determined by using a Perkin-Elmer

Optima 3100 RL inductively coupled plasma atomic emission spectrometry (ICP-AES). Powder X-ray diffraction (XRD) was performed at a scanning rate of 5 °/min from 5 to 70° for 2θ at room temperature by using a Rigaku D/max 2500 V diffractometer. It was equipped with a graphite monochromator by utilizing monochromatic CuKα radiation ($\lambda = 0.154178$ nm). Excitation and emission (PLE&PL) spectra were recorded at room temperature by a Shimadzu RF-53001 spectrophotometer equipped with a xenon lamp as the excitation source, the morphology was observed by SEM, and the elements were analyzed by EDS (EDS, Oxford INCA300, England). Fourier Transform infrared spectroscopy (FTIR) spectra were measured by a Nicolet IS 50 with the KBr pellet technique.

2.2. Preparation of the samples

2.2.1 $KGdF_4:xEu^{3+}$ ($KGF:xEu^{3+}$)

The co-precipitation method was used to prepare a serious sample, and took $KGdF_4:0.20Eu^{3+}$ as an example. The preparation processes were as follows: (i) With the molar ratio of F: K: Gd: Eu = 6.0: 6.0: 0.8: 0.2, 3.2 mL of solution $Gd(NO_3)_3 \cdot 6H_2O$ (2.5 mol/L), 2.0 mL of solution $Eu(NO_3)_3 \cdot 6H_2O$ (1.0 mol/L) and 15 mL deionized water were mixed, and stirred for 30 minutes to mix evenly; (ii) 10 mL $KF \cdot 2H_2O$ (6 mol/L) was dropwise dropped into the mixed solution, and stirred 30 minutes, then stood for 24 hours before the suction filter; (iii) The precipitates was collected by washing with deionized water and absolute ethanol and filtering, and dried at 120 °C for 2 h to obtain

$\text{KGdF}_4:0.20\text{Eu}^{3+}$ ($\text{KGF}:0.20\text{Eu}^{3+}$). The preparation method of a series of $\text{KGF}_4:x\text{Eu}^{3+}$ ($x = 0.05 - 0.25$) samples is the same as that of $\text{KGF}:0.20\text{Eu}^{3+}$.

2.2.2 $\text{KGdF}_4:0.20\text{Eu}^{3+}, y\text{Yb}^{3+}$ ($\text{KGF}:0.20\text{Eu}^{3+}, y\text{Yb}^{3+}$)

The co-doped processes of sensitizer Yb^{3+} are similar to those of $\text{KGF}_4:0.20\text{Eu}^{3+}$. The $\text{Yb}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (1 mol/L) solution is needed to be measured with the $\text{Gd}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ and $\text{Eu}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ solution together before stirring, and following steps are the same as that of $\text{KGF}:0.20\text{Eu}^{3+}$.

2.2.3 $\text{KGdF}_4:0.20\text{Eu}^{3+}, 0.002\text{Yb}^{3+}@GQDs_z$ ($\text{KGF}:0.20\text{Eu}^{3+}, 0.002\text{Yb}^{3+}@GQDs_z$)

A certain amount of GDQs solution must be mixed with $\text{Yb}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (1 mol/L), $\text{Gd}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (2.5 mol/L) and $\text{Eu}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$ (1 mol/L) solution together before stirring, and following steps are the same as that of $\text{KGF}:0.20\text{Eu}^{3+}$.

3. Characterization of sample

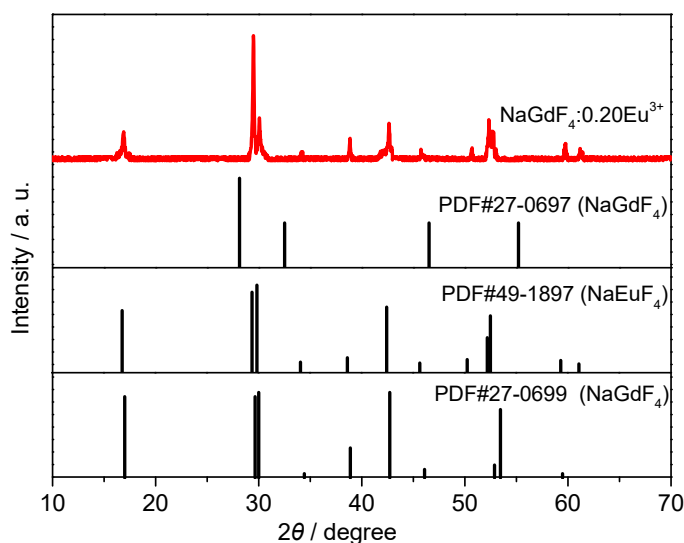


Fig. 7S XRD patterns of $\text{NaGdF}_4:0.20\text{Eu}^{3+}$