Supporting Information (ESI) for

N, S-co-doped carbon dots for rapid acid test paper and bioimaging

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Experimental section

Materials

All the reagents and chemicals were of analytical grade and used as received without further purification. Citric acid and soluble metal ion salts were supplied by Tianjin Jinko Institute of Refined Chemical Engineering (Tianjin, China). N-Methyl thiourea was purchased from Beijing Chemical Corp (Beijing, China). Quinine sulfate (98%, for FL standard reference) was obtained from Aladdin Ltd. (Shanghai China). Amino acids and glucose were obtained from Sigma-Aldrich Chemical Co. High purity water with a resistivity of 18.2 MΩ cm was obtained from the United States Milli-Q purification system (Millipore, MA, USA).

Preparation of N, S-CDs

Firstly, citric acid (1.92 g), N-Methyl thiourea (0.90 g) and 30 mL of distilled water were mixed in a 100 mL beaker. Under proper stirring and ultrasonic treatment, clear and colorless mixture solution was obtained. Subsequently, the mixture was transferred to the stainless steel autoclave lined with Teflon and reacted at 180 ^{o}C for 7 h. After natural cooling to room temperature, the gained dark brown suspension was centrifugated at 6000 rpm for 15 min, filtered by microporous membrane and dialysis (cutoff *Mn:* 1.0 kDa) for 48 h. Final N, S-CDs powders were collected by vacuum drying.

To obtain optimal preparation conditions, the fluorescent quantum yield (QY) was used to evaluate the as-prepared CDs. From the controlled experiments conducted, we prepared the N, S-CDs at 180 ^{o}C for 7 h with the reactant ratio of 1:1.

Characterization

X-ray powder diffraction (XRD) spectra were performed on a Shimadzu XRD-6100 spectrometer (Kyoto, Japan). The morphology of N, S-CDs was revealed on a JEOL JEM-2100 transmission electron microscope (TEM) with an accelerating voltage of 200 kV. Fourier transform infrared spectra (FT-IR) were accomplished using the KBr powder as the sample matrix on a Nicolet AVATAR 360 FT-IR spectrophotometer. X-ray photoelectron spectroscopy (XPS) analysis was performed on Thermo Fisher K-Alpha spectrometer (Thermo Fisher, USA). Fluorescence spectra were carried out at room temperature on a PC spectrophotometer (Shimadzu RF-5301) equipped with a xenon lamp. UV-vis absorption spectra were obtained by a Shimadzu UV-2550 spectrophotometer.

Quantum Yield Measurements

The relative fluorescence quantum yield (QY) of as-prepared N, S-CDs solutions were calculated by the typical procedure using following equation:

$$QY = Q_R \frac{I}{I_R} \frac{OD_R}{OD} \frac{n}{n_R^2}^2$$

Where QY is the quantum yield, I represents the measured integrated emission intensity (Emission wavelength: 360 nm), n is the refractive index, and OD is the optical density measured on a UV-Vis spectrophotometer (limited less than 0.05). The subscript R refers to the reference of quinine sulfate dissolved in 0.1 M H_2SO_4 (Q_R=0.546).

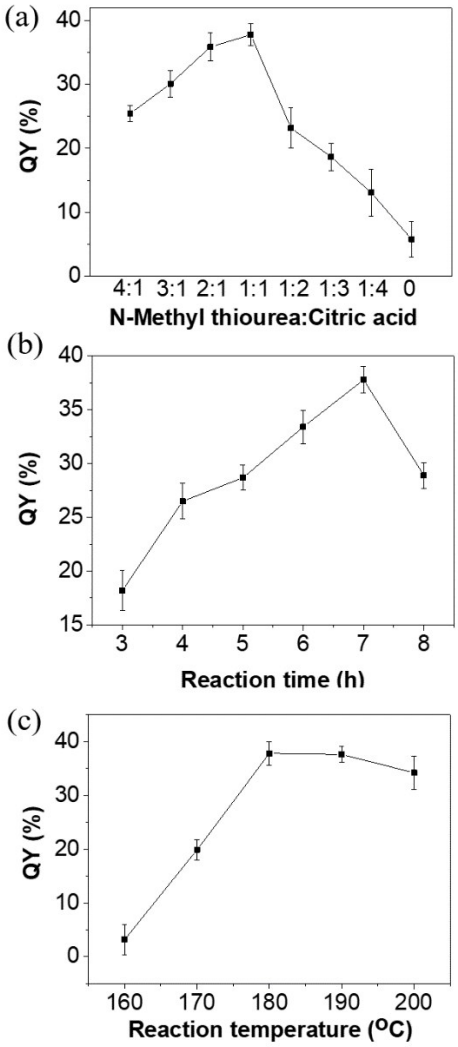

Fig. S1 The fluorescent quantum yield (QY) of the N, S-CDs at different preparation conditions. a) N, S-CDs of different starting material ratios, b) N, S-CDs of different reaction time, c) N, S-CDs of different reaction temperature.

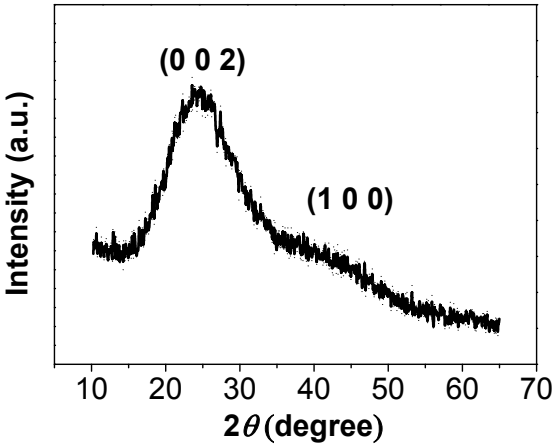

Fig. S2 X-ray diffraction (XRD) pattern of the N, S-CDs.

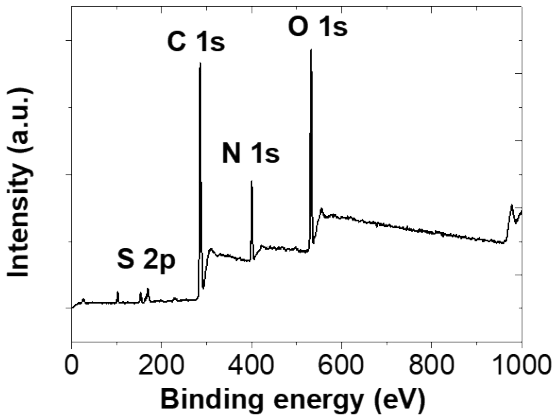

Fig. S3 XPS survey spectrum of the N, S-CDs.

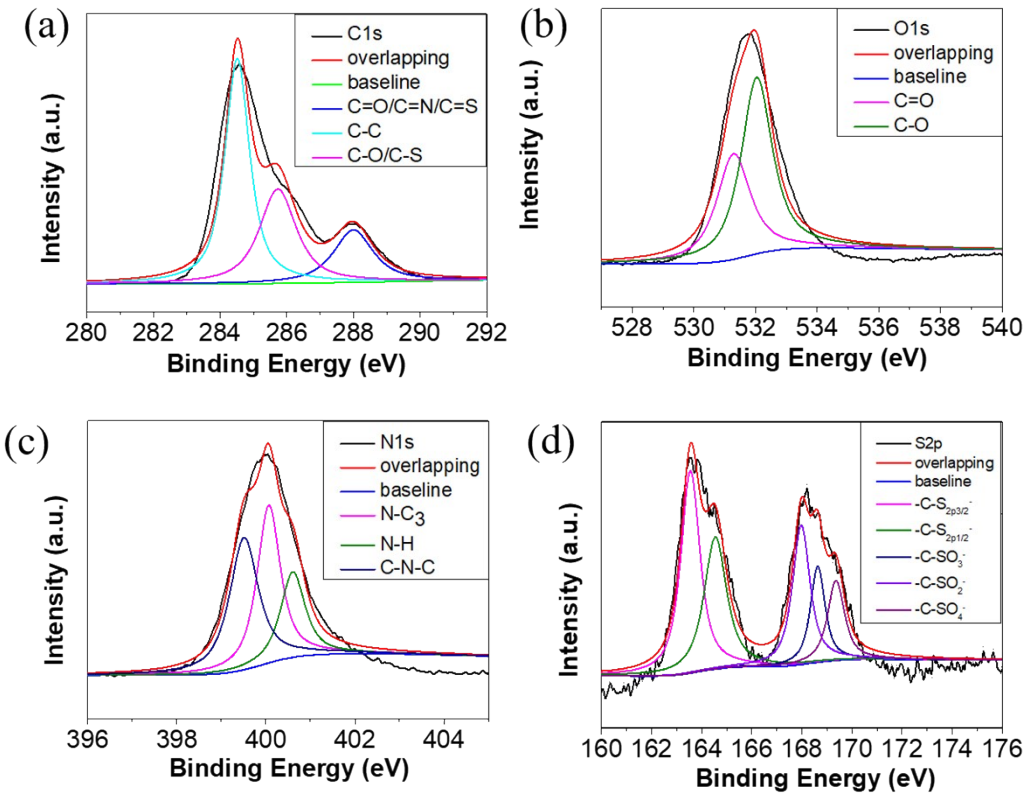

Fig. S4 High resolution XPS spectra of C1s (a), O1s (b), N1s (c) and S2p (d).

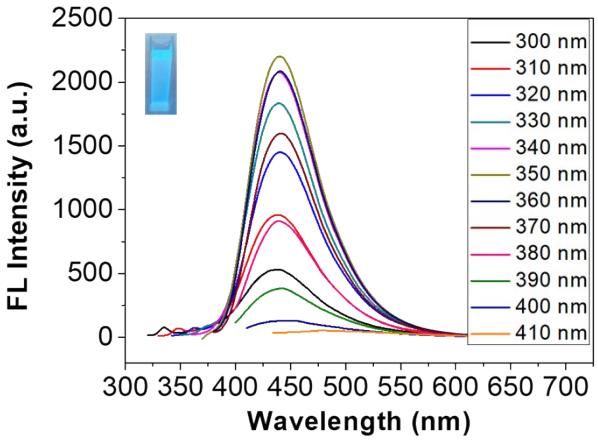

Fig. S5 Fluorescence emission spectra of N, S-CDs, excitation wavelength starts from 300 nm to 410 nm and increases in 10 nm increments. Inset: photographs of the N, S-CDs solution (pH=7.0) under a UV beam of 365 nm.

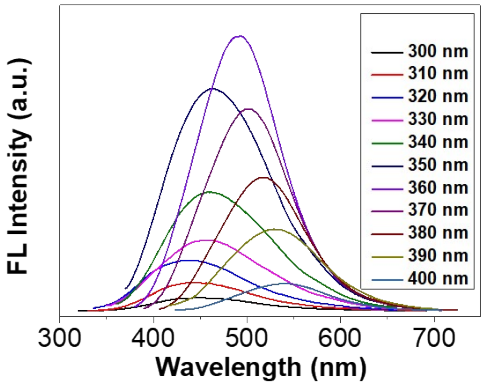

Fig. S6 Fluorescence emission spectra of CDs derived from citric acid, excitation wavelength starts from 300 nm to 400 nm and increases in 10 nm increments.

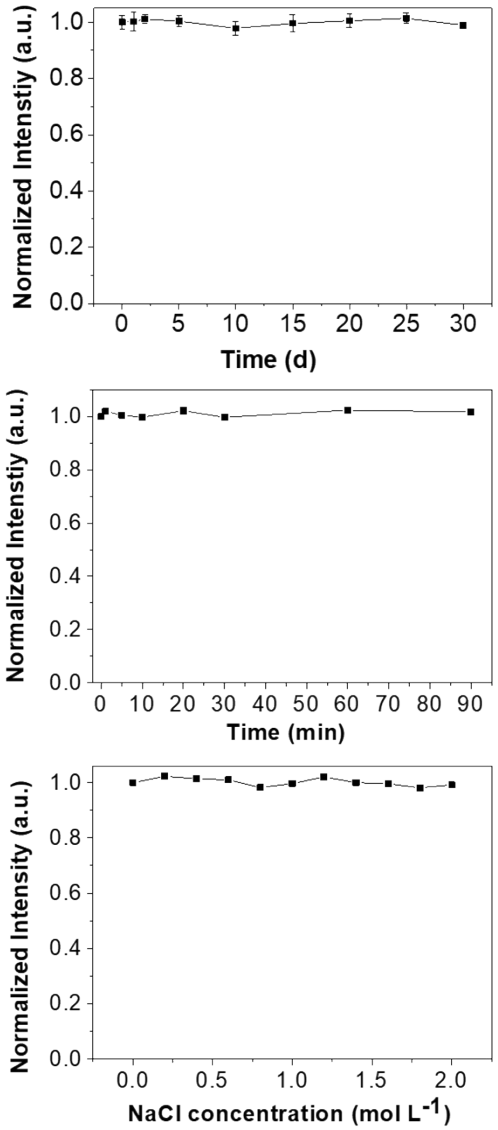

Fig. S7 The effects of long time storage (30 days) (a), continuous UV exposure (365 nm, 90 min) (b) and ionic strength (concentration of NaCl: 0-2.0 mol L^{-1}) on the fluorescence intensity of the N, S-CDs.

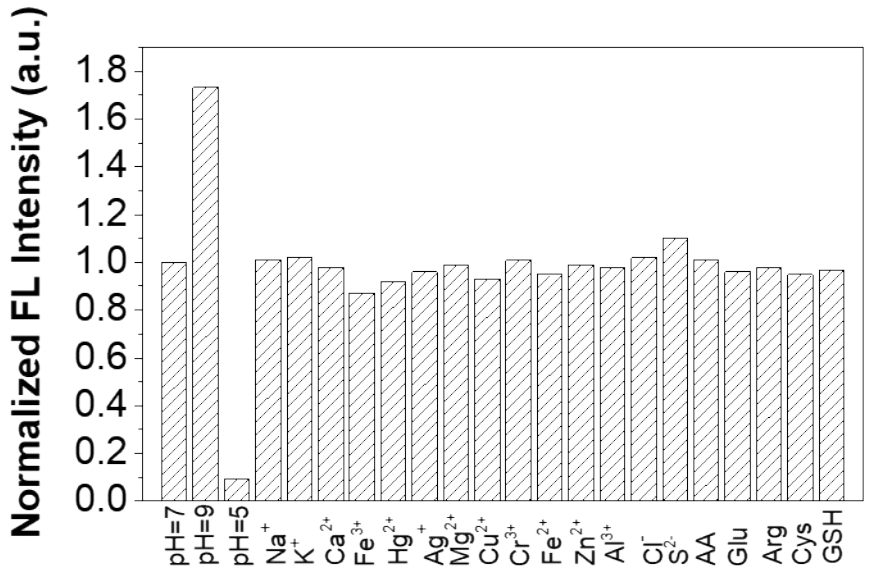

Fig. S8 Selectivity experiments of pure N, S-CDs at pH=5, 7, 9 and with other potential interfering species at pH=7.

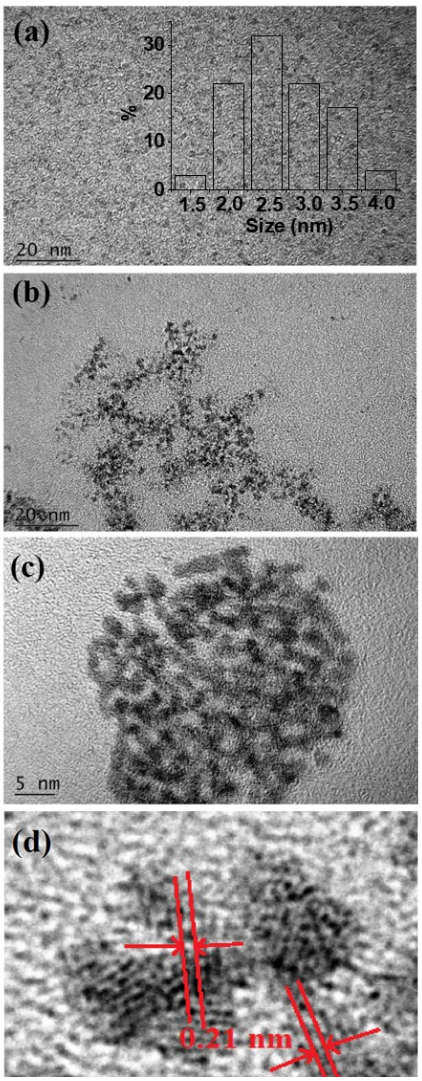

Fig. S9 TEM images of N, S-CDs solutions with different pH values, (a) pH=5; (b) pH=7; (c) pH=9; (d) high-resolution TEM micrograph of the N, S-CDs. Inset: size distribution of N, S-CDs.

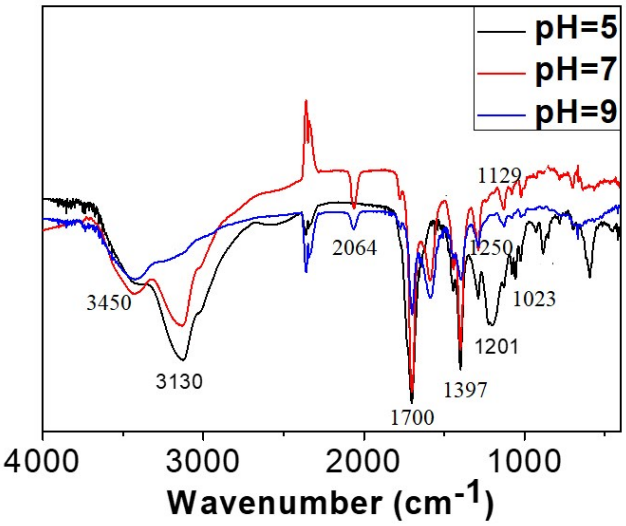

Fig. S10 FTIR spectra of N, S-CDs solutions with different pH values.

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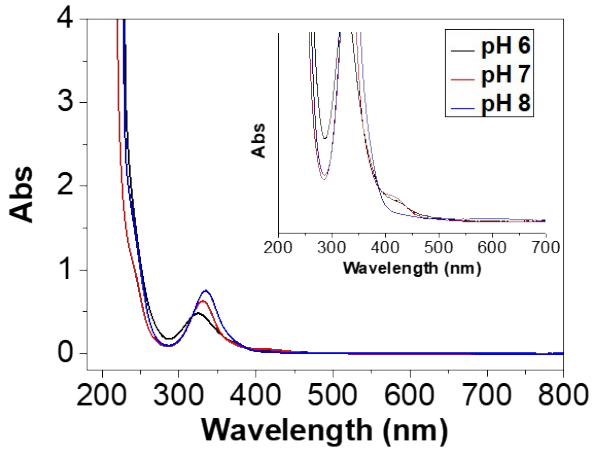

Fig. S11 The UV-vis spectra of N, S-CDs solutions with pH 6, 7, 8.

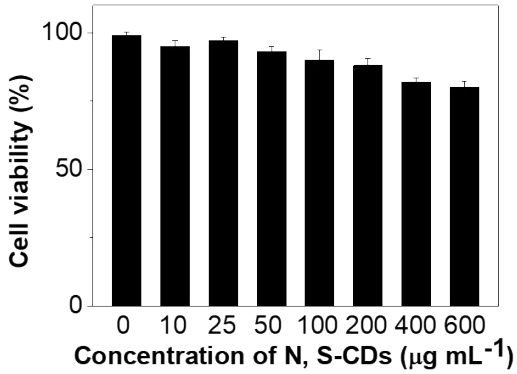

Fig. S12 Cell viability of HeLa cells with different concentrations of the as-prepared N, S-CDs.