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# **Electronic Supplementary Information**

# An Ultrafast and Sensitive Ratiometric Fluorescence pH Nanoprobe Based on Label-Free Dual-Emission Carbon Dots

Chao Xia,<sup>a</sup> Mengmeng Cao,<sup>a</sup> Jinfeng Xia,<sup>\*b</sup> Guohong Zhong,<sup>b</sup> Danyu Jiang,<sup>b</sup> Dafeng Zhang,<sup>c</sup> Jing Wang,<sup>d</sup> and Huili Li<sup>\*a</sup>

<sup>a</sup> Engineering Research Center for Nanophotonics & Advanced Instrument, Ministry of Education, Department of Physics and Materials Science, East China Normal University, Shanghai 200062, China.

<sup>b</sup> Key Laboratory of Transparent Opto-Functional Inorganic Materials, Shanghai Institute of Ceramics, Chinese Academy of Sciences, Shanghai 201899, China.

<sup>c</sup> School of Materials Science and Engineering, Liaocheng University, Liaocheng, Shandong 252000, China.

<sup>d</sup> Ministry of Education Key Laboratory of Bioinorganic and Synthetic Chemistry, State Key Laboratory of Optoelectronic Materials and Technologies, School of Chemistry, School of Materials Science and Engineering, Sun Yat-Sen University, Guangzhou 510275, China

#### \*Corresponding author:

Prof. Huili Li<sup>\*a</sup> *E-mail:* hlli@phy.ecnu.edu.cn; *Tel:* +86-21-62235465; *Fax:* 

+86-21-62235465.

Mrs. Jinfeng Xia<sup>\*b</sup> *E-mail:* xiajf@mail.sic.ac.cn; *Tel:* 86-21- 69987771; *Fax:* +86-21-69987769.

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Fig. S1 the excitation ( $\lambda_{em} = 580$  nm, dash line) and emission spectra ( $\lambda_{ex} = 365-500$  nm, solid lines with different colors) of as-prepared DECDs.



Fig. S2 The fluorescence quantum yield curves measured for the DECDs solution and reference sample at room temperature ( $\lambda_{ex} = 420 \text{ nm}$ ).



Fig. S3 The CIE chromaticity diagram of DECDs under 420 nm excitation.



Fig.S4 Normalized PL spectra of the control CDs under 365 nm excitation



Fig. S5 The XPS survey spectra of as-synthesized DECDs (a) and CDs4 (b)

	С	Ν	0
DECDs	61.62 %	7.37 %	31.01 %
CDs4	74.95 %	17.41 %	7.64 %

Table S1 The element contents of as-synthesized DECDs and CDs4, respectively.



Fig. S6 PL spectrum of the mixed system by CDs3 and CDs4 ( $\lambda_{ex} = 365$  nm).



Fig. S7 The UV-vis absorption spectra of control CDs.



Fig. S8 PL spectra of as-synthesized CDs with different dose of OA under 365 nm excitation.



Fig. S9 FT-IR spectra of DECDs.



Fig. S10 High-resolution XPS spectra of C 1s (a), O 1s (b) and N (1s) for DECDs.



Fig. S11 The intensity variation of 393 and 580 nm emissions with pH value.



Fig. S12 The pH-dependent CIE chromaticity diagram of DECDs under 365 nm excitation (A: pH=2.2, B: pH=3.0, C: pH=4.0, D: pH=5.0, E: pH=6.0, F: pH=7.0, G:

pH=8.0).



Fig. S13 The digital photos of DECDs solution under different pH values.



Fig. S14 Emission spectra of DECDs dispersed in different concentration of NaCl solution.



Fig. S15 Emission spectra of DECDs exposure to various cationic (a) and anionic (b) solutions with a 0.01 M concentration (The blank sample is refered to DECDs solution with pH=8.0)



Fig. S16 PL intensities of DECDs exposure to various cationic (a) and anionic (b) solutions with a 0.01 M concentration (The blank sample is referred to DECDs solution with pH=8.0)

Ions	Δ
Blank	0.11
$\mathbf{K}^+$	0.09
$Ca^{2+}$	0.18
$Na^+$	0.10
$Mg^{2+}$	0.11
$Ba^{2+}$	0.11
Ni <sup>2+</sup>	0.11
Fe <sup>2+</sup>	0.11
$\mathrm{Al}^{3+}$	0.64
$Cu^{2+}$	0.17
CO <sub>3</sub> <sup>2-</sup>	0.12
HCO <sub>3</sub> -	0.10
Cl-	0.11
CH <sub>3</sub> COO <sup>-</sup>	0.14
NO <sub>3</sub> -	0.09
SO4 <sup>2-</sup>	0.10
F	0.17
$\mathbf{H}^{+}$	17.45

Table S2 The emission intensity ratio (Δ) of 393 and 580 nm for DECDs exposure to various metal and H<sup>+</sup> cationic as well as anionic solutions with a 0.01 M concentration (The blank sample is referred to DECDs solution with pH=8.0)



Fig. S17 The PL response of DECDs at 393 and 580 nm (a) and  $\Delta$  (b) vs. immersion time under pH = 2.2.



Fig. S18 The Emission spectra (a) and  $\Delta$  (b) of DECDs under alternating conditions between pH = 2.2 and pH = 8.0 repeatedly for 5 cycles.



Fig. S19 Dependence of fluorescence intensity of DECDs on 365 nm UV irradiation time.



Fig. S20 Photographs under 365 nm lamp of two pieces of filter papers stained with DECDs solutions for pH detection: dipping 100  $\mu$ L NH<sub>3</sub>·H<sub>2</sub>O (a) and HCl (b), respectively.



Fig. S21 Characters written on filter paper by using DECDs-H (top) and DECDs (bottom) ink under 365 nm UV lamp.