

## Supporting Information

### **Fluorescent N doped carbon dots-hydrogel composite for concurrent selective detection and local hot spot promoted adsorption of uranium (VI)**

Qun Wang <sup>a\*</sup>, Tao Han <sup>a</sup>, Caiqin Miao <sup>a</sup>, Wei Qin <sup>b</sup>, Xiaohong Wu <sup>a\*</sup>

<sup>a</sup> School of Chemistry and Chemical Engineering, and <sup>b</sup> School of Materials Science and Engineering, Harbin Institute of Technology, Harbin, 150001, China

#### **Experimental Section:**

- 1. Luminescent Sensing of U(VI):** Table S1 was listed to compare with the previous reported detection limitation [1-15].
- 2. Selection of the precursors of the NCDs:** Fluorescence CDs containing graphene core and organic shell have excellent water solubility and various groups on the surface which play a crucial role in donating the reactive sites for  $\text{UO}_2^{2+}$  ions. Literature using levofloxacin, a kind of fluorinated quinolone derivatives, as a precursor of synthetic carbon dots had been reported [16]. On the one hand, Levofloxacin has a similar structure compared to norfloxacin compound which has nitrogen heterocyclic structure and various phosphorescent characteristics. On the other hand, uranyl ion had assessment based on the fluorescence quenching of Norfoxacin [17]. Norfloxacin is one kind of polluted compound in water which is excepted to removed or converted to environmental friendly CDs [18]. Therefore, we adopted norfloxacin with unique nitrogen heterocyclic structure as a precursor of synthetic NCDs for detection and adsorption of  $\text{UO}_2^{2+}$ .

**Table S1.** The detection limits comparison of the reported materials for  $\text{UO}_2^{2+}$ .

Materials	Detection limit	Ref.
CQDs	28 nM, 6.53 ppb	[1]
HNU-50	12 nM, 2.77 ppb	[2]
CMPAO	0.40 ppb	[3]
YTU-100	1.07 ppb	[4]
Tb-MOF/Tb-AG	0.0012 ppb	[5]
CDs	4.7 ppb	[6]
FPD	2100 ppb	[7]
QDCOF	28.6 ppb	[8]
CDs	710 ppb	[9]
Titanium electrode	24.5 ppb	[10]
Dual-color CDs	1899 ppb	[11]
Tb-FAP/agar	7.95 nM, 1.85 ppb	[12]
COF	6.7 nM, 1.56 ppb	[13]
small-molecule-based fluorescent probes	10 nM–1 $\mu\text{M}$ , 2.33 ppb–233 ppb	[14]
TAPM-DHBD	4.08 nM, 0.96 ppb	[15]
NCDs-CMH	8.4 nM, 1.94 ppb	This work

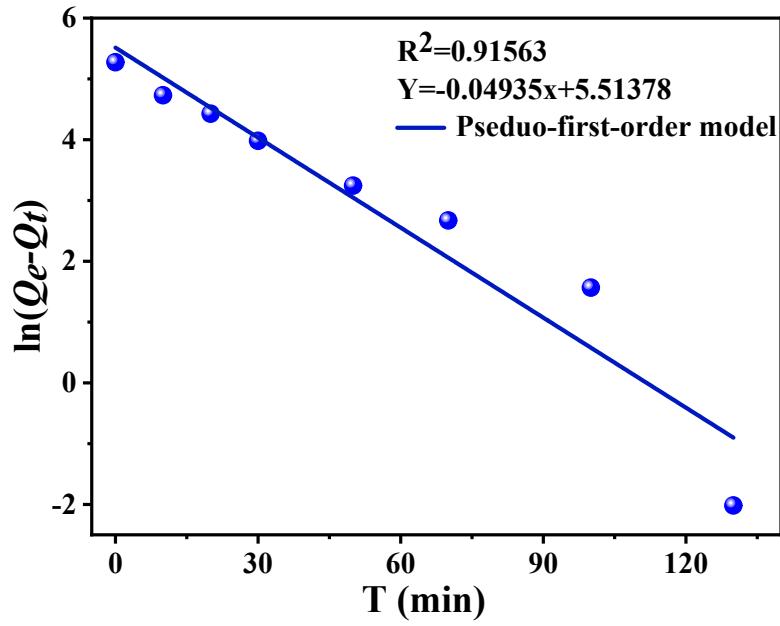


Fig.S1 the pseudo first order kinetic constant.  $q_e$ ,  $k_1$  can be calculated from the slope and intercept of the plot of  $\log (q_e-q_t)$  versus  $t$ .

Table.S2 The linear fitting results and kinetic parameters

Parameter	Pseudo-first-order model	Pseudo-second-order model
$C_0$ mgL <sup>-1</sup>	$k_1$ min <sup>-1</sup>	$R^2$
100	-0.04935	0.91563
		$k_2$ g(mgmin) <sup>-1</sup>
		0.00465
		$R^2$
		0.99872

Table.S3 The estimated thermodynamic parameters are summarized.

$T$ (K)	$K_d$ (mL/g)	$\Delta H^\theta$ (kJ/moL)	$\Delta S^\theta$ (J/moL)	$\Delta G$ (kJ/moL)
295	$1.90 \times 10^5$			-25.68
315	$1.939 \times 10^5$	28.35074	183.15742	-29.34
335	$1.974 \times 10^5$			-33.01

Table.S4 The comparison of adsorption capacity with previously reported work.

Absorption material	$C_0$ (mg/L)	$Q_e$	Ref.
PAA/CS hydrogel	500	290 mg/g	19
P(AAm/MA) hydrogels	1500	156 mg/g	20
Graphene (rGO) hydrogel	100	134 mg/g	21
Metal–phenolic network membranes	238	140mg/L	22
CS–Fe membranes	30	170 mg/g	23
CMH-NCDs	100	194 mg/g	<b>Our work</b>

Reference:

- [1]Z. Zhang, D. Zhang, C. Shi, W. Liu, L.H. Chen, Y. Miao, D.W. Juan, J.L. Li, S. Wang, 3,4-Hydroxypyridinone-modified carbon quantum dot as a highly sensitive and selective fluorescent probe for the rapid detection of uranyl ions, Environ. Sci. Nano, 2019, 6, 1457-1465.
- [2]X.D. Qin, W. Yang, Y.H. Yang, D.X. Gu, D.Y. Guo, Q.H. Pan. A Zinc Metal-Organic Framework for Concurrent Adsorption and Detection of Uranium, Inorg. Chem. 2020, 59, 9857-9865.
- [3]M.Y. Xu, T. Wang, P. Gao, L. Zhao, L. Zhou, D.B. Hua. Highly fluorescent conjugated microporous polymers for concurrent adsorption and detection of uranium, J. Mater. Chem. A, 2019, 7, 11214-11222.
- [4]M. Lei, Y.Y. Jia, W. Zhang, J. Xie, Z.J. Xu, Y.L. Wang, W. Du, W. Liu. Ultrasensitive and Selective Detection of Uranium by a Luminescent Terbium-Organic Framework, ACS Appl. Mater. Inter., 2021, 13(43), 51086-51094.
- [5]A.Qi. Cui, X.Y. Wu, J.B. Ye, G. Song, D.Y. Chen, Jie. Xu, Y. Liu, J.P. Lai, H. Sun. “Two-in-one” dual-function luminescent MOF hydrogel for onsite ultra-sensitive detection and efficient enrichment of radioactive uranium in water, J. Hazard. Mater., 2023, 448, 130864.
- [6] Q. Wang, H.Y. Zhang, D.M. Yu, W. Qin, X.H. Wu, Ultra-sensitive and stable N-doped carbon dots for selective detection of uranium through electron transfer induced  $\text{UO}_2^{2+}(\text{V})$  sensing mechanism. Carbon, 2022, 198, 162-170.

- [7] Z. Wang, C. Xu, Y.X. Lu, G.Y. Wei, G. Ye, T.X. Sun, J. Chen, Microplasma electrochemistry controlled rapid preparation of fluorescent polydopamine nanoparticles and their application in uranium detection[J]. *Chem. Eng. J.* 2018, 344, 480-486.
- [8] M.C. Zhang, Y. Li, L.J. Ma, X.H. Guo, X.F. Li, K. Li, X.Y. Wang, C.Q. Xia, S.J. Li, Crystalline quantum dots of covalent organic frameworks for fast and sensitive detection of uranium, *Chem. Commun.*, 2020, 56, 880-883.
- [9] Z. Wang, Y.X. Lu, H. Yuan, Z.H. Ren, C. Xu, J. Chen, Microplasma-assisted rapid synthesis of luminescent nitrogen-doped carbon dots and their application in pH sensing and uranium detection, *Nanoscale*, 2015, 7, 20743-20748.
- [10] V. Urbanova, M. Pumera, Uranium detection by 3D-printed titanium structures: Towards decentralized nuclear forensic applications, *Appl. Mater Today.*, 2021, 21, 100881.
- [11] J.J. Qian, N.N. Cao, J. Zhang, J.J. Hou, Q. Chen, C. Zhang, Y.D. Sun, S.J. Liu, L.F. He, K. Zhang, H.B. Zhou, Field-portable ratiometric fluorescence imaging of dual-color label-free carbon dots for uranyl ions detection with cellphone-based optical platform, *Chin. Chem. Lett.*, 2020, 31, 2925-2928.
- [12] H.J. Liu, X.L. Wang, T. Abeywickrama, F. Jahanbazi, Z.F. Min, Z.R. Lee, J. Terry, Y. B. Mao, Biomimetically synthesized luminescent  $Tb^{3+}$ -doped fluorapatite/agar nanocomposite for detecting  $UO_2^{2+}$ ,  $Cu^{2+}$ , and  $Cr^{3+}$  ions, *Environ. Sci. Nano.*, 2021, 8, 3711-3721.
- [13] W.R. Cui, C.R. Zhang, W. Jiang, F.F. Li, R.P. Liang, J.W. Liu, J.D. Qiu, Regenerable and stable  $sp^2$  carbon-conjugated covalent organic frameworks for selective detection and extraction of uranium, *Nat. Commun.*, 2020, 11, 436(1-10).
- [14] Y.Y. Fang, W. Dehaen, Small-molecule-based fluorescent probes for f-block metal ions: a new frontier in chemosensors, *Coord. Chem. Rev.* 2021, 427, 213524.
- [15] W.R. Cui, Y.R. Chen, W.Xu, K.Liu, W.B. Qiu, Y. Li, J.D. Qiu. A three-dimensional luminescent covalent organic framework for rapid, selective, and reversible uranium detection and extraction. *Sep. Purif. Technol.* 2023, 306 , 122726.
- [16] Tan J, Li Q, Meng S. Time-Dependent Phosphorescence Colors from Carbon Dots for Advanced Dynamic Information Encryption. *Adv. Mater.* 2021, 33, 2006781.
- [17] A. A. Elabd, O. A. Elhefnawy. Uranyl ion assessment based on the fluorescence quenching of Norfoxacin. *J. Radioanal. Nucl. Chem.* 2021, 329, 935-944.
- [18] P.C. Cai, J. Zhao, X.H. Zhang, T.Y., Zhang, G.M. Yin, S. Chen, C.L. Dong, Y. Huang, Y.Y. Sun, D.J. Y, B.S. Xing. Synergy between cobalt and nickel on NiCo<sub>2</sub>O<sub>4</sub> nanosheets promotes peroxymonosulfate activation for efficient norfloxacin degradation. *Appl. Cata. B: Environ.* 2022, 306, 121091.
- [19] J.R. He, F.L. Sun, F.H. Han, J.J. Gu, M.R. Ou, W.K. Xu, X.P. Xu, Preparation of a novel

- polyacrylic acid and chitosan interpenetrating network hydrogel for removal of U(vi) from aqueous solutions, RSC Adv., 2018, **8**, 12684-12691.
- [20] P. Akkas, O. Guven, Enhancement of uranyl ion uptake by prestructuring of acrylamide–maleic acid hydrogels, J. Appl. Polym. Sci., 2000, **78**, 284-289.
- [21] Y.R. He, S.C. Li, X.L. Li, Y. Yang, A.M. Tang, L. Du, Z.Y. Tan, D. Zhang, H.B. Chen, Graphene (rGO) hydrogel: A promising material for facile removal of uranium from aqueous solution, Chem. Eng. J., 2018, **338**, 333-340.
- [22] W. Luo, G. Xiao, F. Tian, J.J. Richardson, Y.P. Wang, J.F. Zhou, J.L. Guo, X.P. Liao, B. Shi, Engineering robust metal-phenolic network membranes for uranium extraction from seawater, Energy Environ. Sci., 2019, **12**, 607-614.
- [23] L.M. Zhou, H.B. Zou, Y. Wang, Z.R. Liu, Z.G. Le, G.L. Huang, T. Luo, A.A. Adesina, Immobilization of in situ generated Fe-0-nanoparticles in tripolyphosphate-crosslinking chitosan membranes for enhancing U(VI) adsorption, J. Radioanal. Nucl. Chem., 2017, 311, 779-787.