

## Supplementary Information

### **Covalent Locking of Carbon Dots in Imprinted Chitosan Gel Beads for Reusable, Visual Tetracycline Detection**

Chaojie Yu,<sup>a</sup> Yuxin Sang,<sup>b</sup> Ziqi Liu,<sup>a</sup> Haihong Huang,<sup>a</sup> Zuwei Song,<sup>a</sup> Weina Li,<sup>\*a</sup>  
Qinxing Sun<sup>\*a</sup>

*a College of Chemistry and Pharmaceutical Sciences, Qingdao Agricultural  
University, Qingdao, 266109, China*

*b College of Materials Science and Chemical Engineering, Harbin Engineering  
University, Harbin 150001, China*

*\* Correspondence to: Qingdao Agricultural University, 700 Changcheng Road,  
Qingdao 266109, China*

*\*Corresponding author*

E-mail: wnli@qau.edu.cn (W. N. Li),

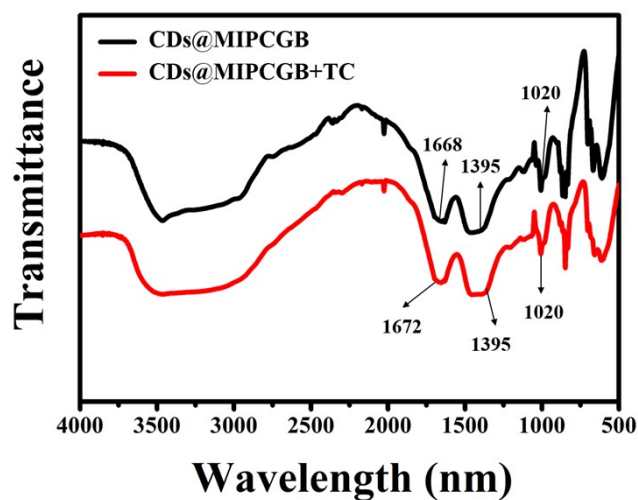


Fig. S1. FT-IR spectra of CDs@MIPCGB (black) and CDs@MIPCGB+TC (red).

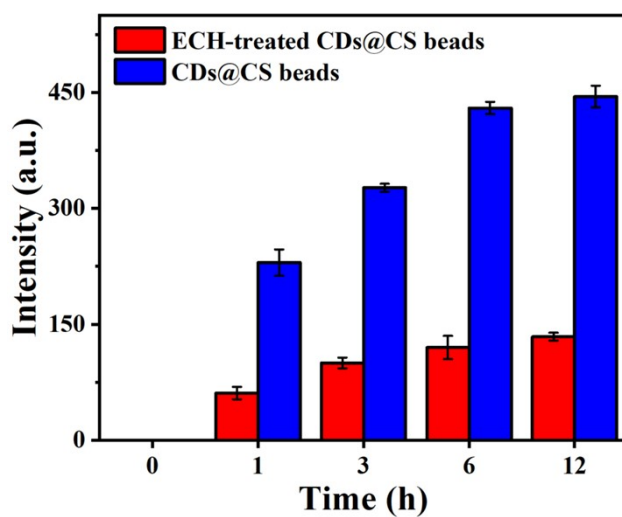


Fig. S2. Comparison of supernatant fluorescence between ECH-treated beads and a physically entrapped control under repeated washing/regeneration conditions.

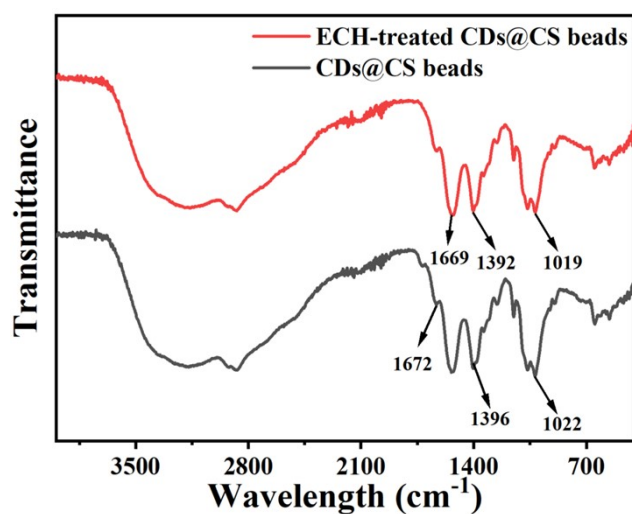
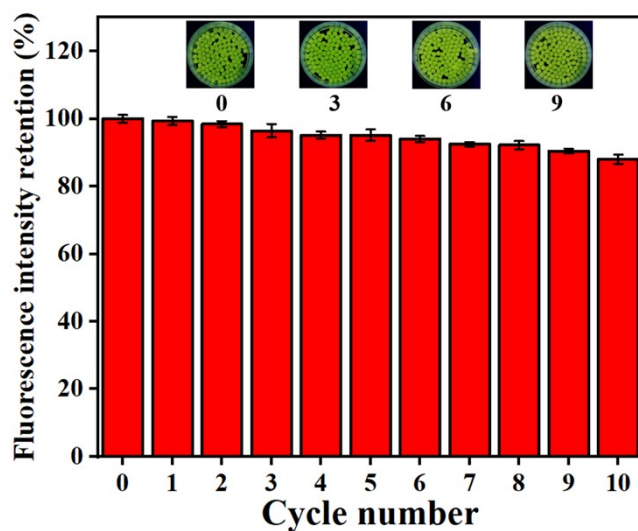
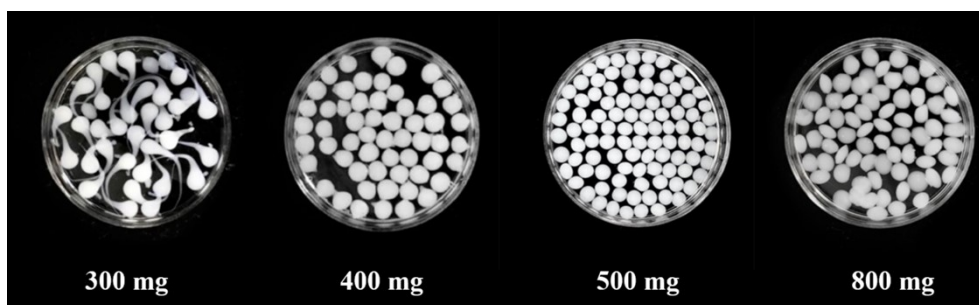


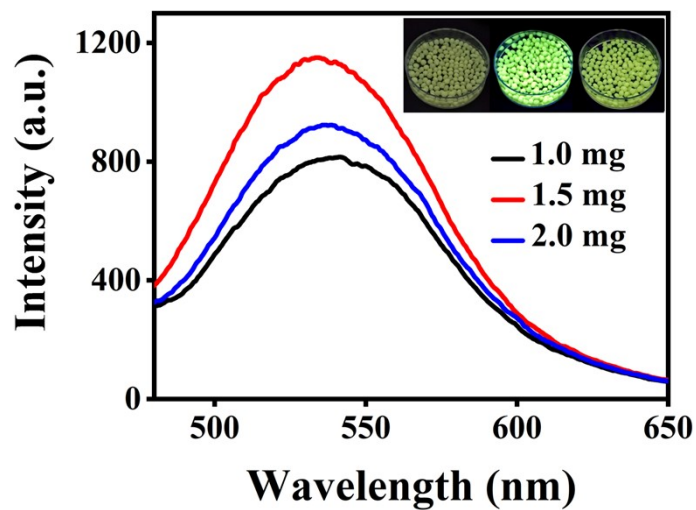
Fig. S3. Comparative FTIR spectra of CDs before and after ECH treatment.



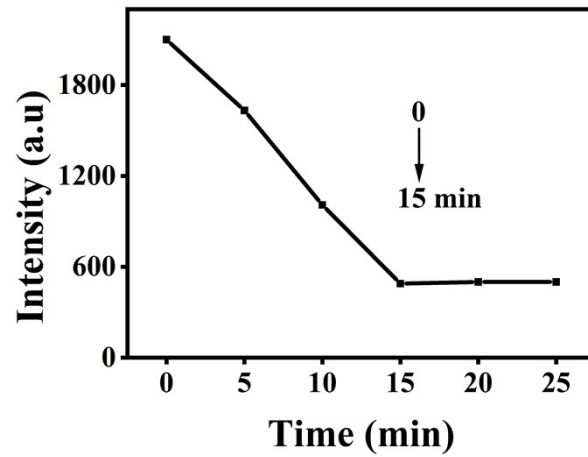
**Fig. S4.** Fluorescence response of CDs@MIPCGB to TC in 10 regeneration cycles and the corresponding fluorescent photographs of CDs@MIPCGB after 0, 3, 6 and 9 cycles.



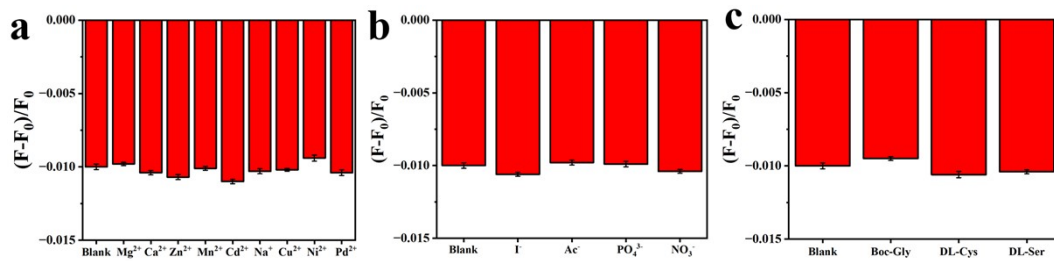
**Fig. S5.** Pellets morphology of chitosan with different concentrations.



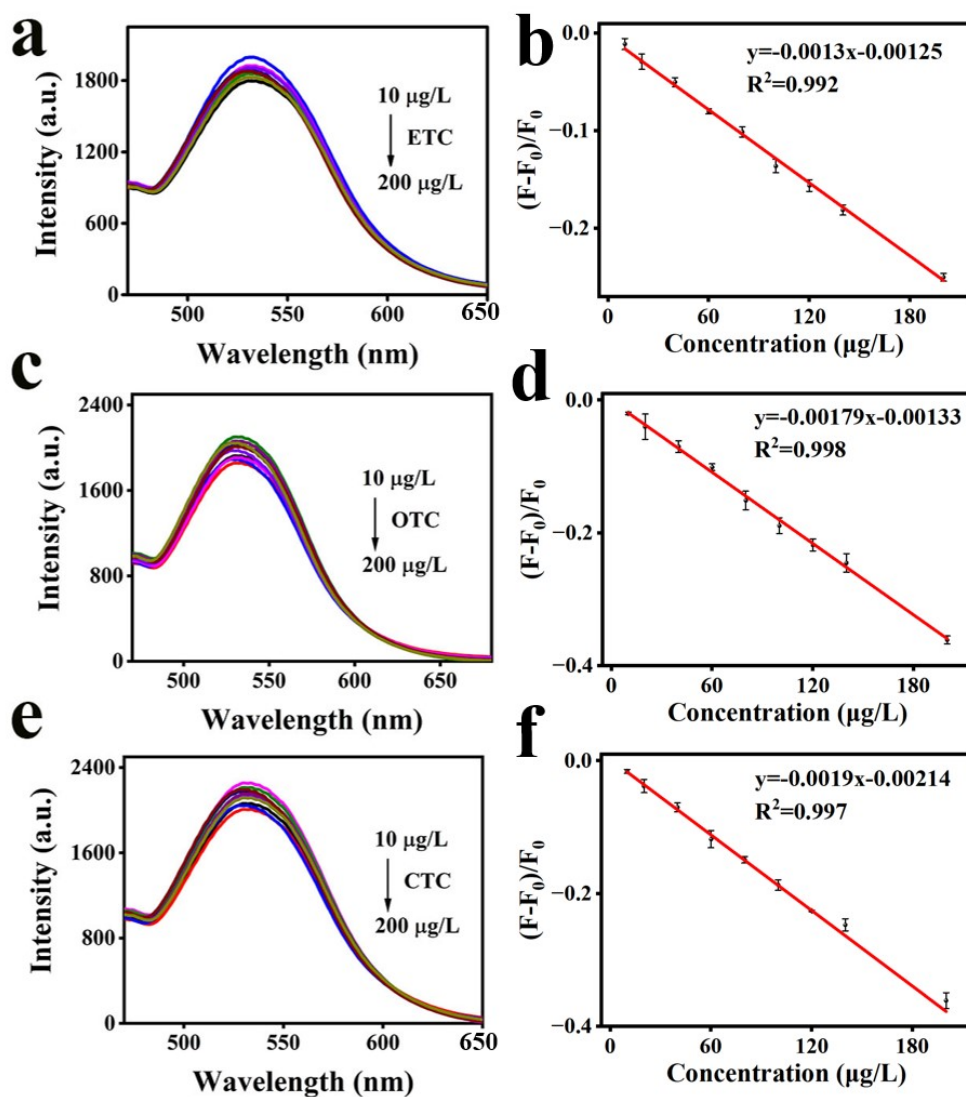
**Fig. S6.** Fluorescence spectra of the MIP chitosan gel beads with CDs doping amount varied from 1.0 mg to 2.0 mg. Inset: the corresponding photographs under UV light.



**Fig. S7.** The fluorescence intensity change of CDs@MIPCGB with incubation time as the addition of TC.



**Fig. S8.** Fluorescence responses of CDs@MIPCGB toward different (a) metal cations, (b) anions and (c) amino acids.



**Fig. S9.** (a) The fluorescence of CDs@MIPCGB with various concentrations of ETC. (b) The linear relationship between the fluorescence intensity change of CDs@MIPCGB and the concentration of ETC within the range of 10-200 µg/L. (c) The fluorescence of CDs@MIPCGB with various concentrations of OTC. (d) The linear relationship between the fluorescence intensity change of CDs@MIPCGB and the concentration of OTC within the range of 10-200 µg/L. (e) The fluorescence of CDs@MIPCGB with various concentrations of CTC. (f) The linear relationship between the fluorescence intensity change of CDs@MIPCGB and the concentration of CTC within the range of 10-200 µg/L.

**Table S1 Inter-day, intra-day and inter-batch recovery of CDs@MIPCGB at three levels of different concentrations TC with fluorescence method.**

Compounds	Spiked level (µg/L)	Intra-day			Inter-day			Inter-batch		
		Detection of TC (µg/L)	Recovery (%)	RSD (%)	Detection of TC (µg/L)	Recovery (%)	RSD (%)	Detection of TC (µg/L)	Recovery (%)	RSD (%)
TC	30	30.1	100.3		29.6	98.7		29.4	98.0	
		29.8	99.3	2.80	30.7	102.3	1.83	30.1	100.3	1.21
		31.4	104.7		30.2	100.7		29.9	99.7	
	80	81.3	101.6		80.2	100.2		80.7	100.9	
		79.3	99.1	1.26	82.1	102.6	1.39	79.3	99.1	1.64
		80.6	100.8		80.1	100.1		78.1	97.6	
		161.3	100.8		162.4	101.5		161.2	100.8	
	160	162.9	101.8	1.12	158.7	99.2	1.18	160.1	100.1	0.72
		159.3	99.6		159.9	99.9		158.9	99.3	

**Table S2 Inter-day, intra-day and inter-batch recovery of CDs@MIPCGB at three levels of different concentrations TC with smartphone (RGB) method.**










Compounds	Spiked level (µg/L)	Intra-day			Inter-day			Inter-batch		
		Detection of TC (µg/L)	Recovery (%)	RSD (%)	Detection of TC (µg/L)	Recovery (%)	RSD (%)	Detection of TC (µg/L)	Recovery (%)	RSD (%)
TC	30	30.1	100.3		29.2	97.3		30.4	101.3	
		29.5	98.3	3.02	29.9	99.7	2.85	30.8	102.7	2.02
		31.3	104.3		30.9	103.0		29.6	98.7	
	80	81.2	101.5		80.3	100.4		81.4	101.8	
		82.1	102.6	1.11	81.5	101.9	1.25	80.2	100.2	1.31
		80.3	100.4		79.5	99.4		79.3	99.1	
		162.2	101.4		161.4	100.9		160.2	100.1	
	160	159.6	99.8	0.85	160.1	100.1	0.44	158.1	98.8	1.07
		160.2	100.1		161.2	100.8		161.5	100.9	

1 **Table S3 Cross-device consistency of smartphone-assisted TC quantification using**  
 2 **CDs@MIPCGB under standardized imaging conditions (n = 3).**

Concentration of TC ( $\mu\text{g/L}$ )	Devices	Recovery (%)	RSD (%)
20	Huawei nova 13 Pro	$100.4 \pm 2.1$	2.1
	REDMI K80	$99.7 \pm 1.8$	1.8
	Vivo S20	$102.6 \pm 1.6$	1.6
80	Huawei nova 13 Pro	$98.7 \pm 2.4$	2.4
	REDMI K80	$97.6 \pm 3.1$	3.2
	Vivo S20	$101.4 \pm 2.4$	2.4
140	Huawei nova 13 Pro	$96.9 \pm 1.4$	1.4
	REDMI K80	$97.8 \pm 1.9$	1.9
	Vivo S20	$99.7 \pm 2.6$	2.6
200	Huawei nova 13 Pro	$102.1 \pm 3$	2.9
	REDMI K80	$100.6 \pm 2.9$	2.9
	Vivo S20	$98.5 \pm 1.4$	1.4

3

4 **Table S4 Determination of TC in milk samples using CDs@MIPCGB through fluorescence**  
 5 **intensity change, n=3.**

Spiked ( $\mu\text{g/L}$ )	Fluorescence spectra			Smartphone			
	Detection of TC ( $\mu\text{g/L}$ )	Recovery (%)	RSD (%)	Detection of TC ( $\mu\text{g/L}$ )	Images	Recovery (%)	RSD (%)
	48.6	97.3		49.2		98.4	
50	51.2	102.4	2.64	51.4		102.8	2.24
	50.3	100.6		49.9		99.8	
	99.6	99.6		101.7		101.7	
100	103.2	103.2	1.81	100.3		100.3	1.05
	102.1	102.1		102.4		102.4	
	154.6	103.1		149.1		99.4	
150	150.2	100.1	1.51	153.7		102.5	1.57
	151.3	100.9		150.4		100.3	

6

1 **Table S5 Comparison of this proposed method with previously reported methods for TC detection**  
 2 **in different samples.**

<b>Probe</b>	<b>Sample</b>	<b>Linear range (<math>\mu\text{g/L}</math>)</b>	<b>LOD (nM)</b>	<b>Detecti on time</b>	<b>References</b>
MIPs-AA/CQDs	water	444-26640	170	10 min	[1]
FeCu-MOF@MIP	milk	0-44.4	12.64	12 min	[2]
CdTe QD- SiO <sub>2</sub> @MIPs	milk and bovine serum	222-6660 and 222- 22200	140 and 190	2 h	[3]
ZIF-8&N- CDs@MIP	milk and egg	100-4000	101.35	30 min	[4]
QD/MS/MIP	serum	50-1000	33.78	10 min	[5]
MIP@paper	water	100-300	90.09	5 min	[6]
CDs@MIPCGB	milk	10-200	12.23	15 min	this work

3

## 1 References

- 2 [1] X. Wei, L. Lv, Z. D. Zhang and W. S. Guan, *J. Appl. Polym. Sci.*, 2020, 137, e49126.
- 3 [2] L. Zhang, M. P. Shen, Z. D. Li, F. X. Meng, J. K. Wang, A. Li, L. Wang, Y. N. Qin and M. W.
- 4 Zhang, *Sens. Actuators B Chem.*, 2025, 432, 137445.
- 5 [3] Y. Q. Yang, H. Niu and H. Q. Zhang, *ACS Appl. Mater. Interfaces*, 2016, 8, 15741-15749.
- 6 [4] J. N. Zhang, Y. Li, L. H. Teng, Y. C. Cao, X. L. Hu, G. Z. Fang and S. Wang, *Anal. Chim. Acta*,
- 7 2023, 1251, 341032.
- 8 [5] L. Zhang and L. Chen, *ACS Appl. Mater. Interfaces*, 2016, 8, 16248-16256.
- 9 [6] D. Barzallo, E. Palacio, L. Ferrer and M. D. T. Sotomayor, *Talanta*, 2025, 281, 126856.
- 10