Supplementary Information

Graphene electrochemical transistors decorated by Ag nanoparticles exhibiting high sensitivity for the detection of Paraquat over a wide concentration range

Hairui Wang[a], Enhui Hou[a], Na Xu[a], Ping Nie[a], Limin Chang[a*], Jianfeng Wu[b *], Xuelin Zhang[c*].

a. Key Laboratory of Preparation and Applications of Environmental Friendly Materials (Jilin Normal University), Ministry of Education, Changchun, 130103, China

b. State Key Laboratory of Toxicology and Medical Countermeasures, and Laboratory of Toxicant Analysis, Institute of Pharmacology and Toxicology, Academy of Military Medical Sciences, Beijing,100850, China

c. MEMS Center, School of Astronautics, Harbin Institute of Technology, Harbin, 150001, PR China

*Corresponding author: Tel.: +86 43181765057
E-mail address: aaaa2139@163.com (Limin Chang), ammswjf@hotmail.com (Jianfeng Wu), zhangxuelin@hit.edu.cn (Xuelin Zhang).

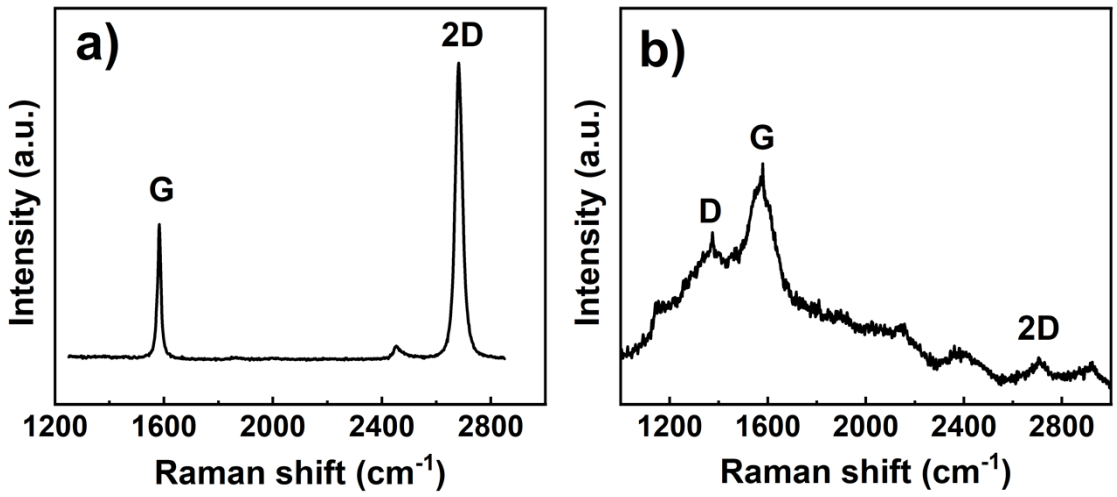

Fig. S1. Raman spectrum of the graphene film before (a) and after (b) Ag deposition.

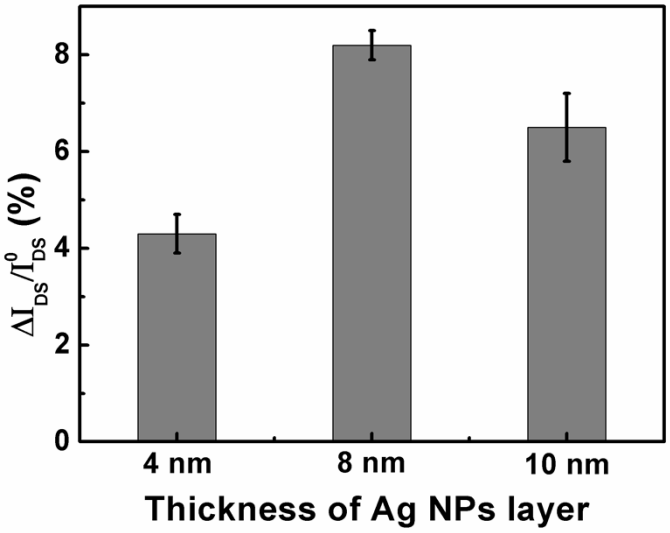

Fig. S2. Channel current response of the GECT functionalized by different thickness of Ag NPs measured in 0.1 M Na_2SO_4 solution with addition of 1 μM PQ. (V_{GS} = 0.7 V, V_{DS} = 0.05 V).

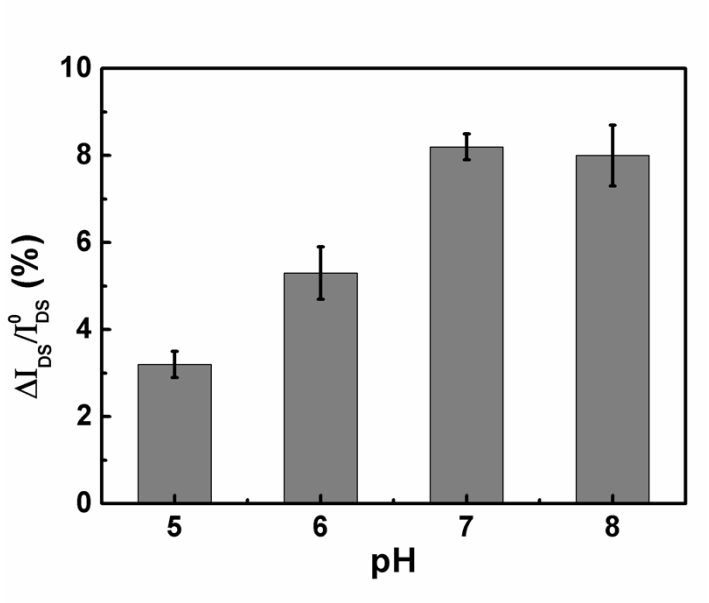

Fig. S3. Channel current response of the GECT functionalized by Ag NPs measured in 0.1 M Na_2SO_4 solution with different pH after adding 1 μM PQ. (V_{GS} = 0.7 V, V_{DS} = 0.05 V).

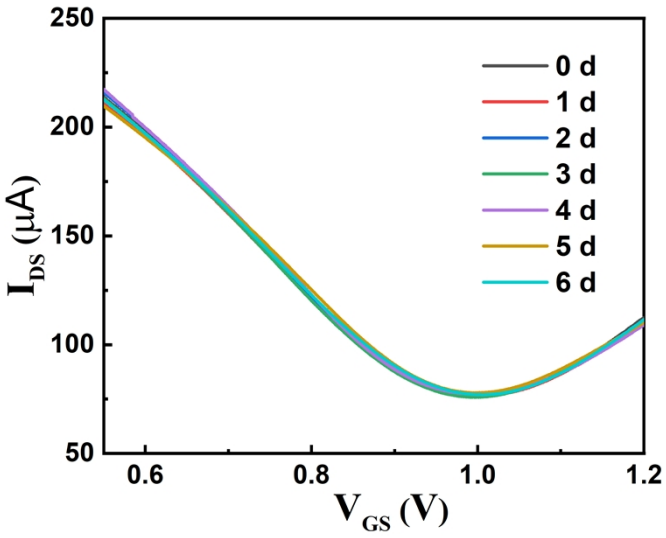

Fig. S4. Transfer curve (I_{DS} vs. V_{GS}, V_{DS} = 0.05 V) of the GECT after storage for different time.

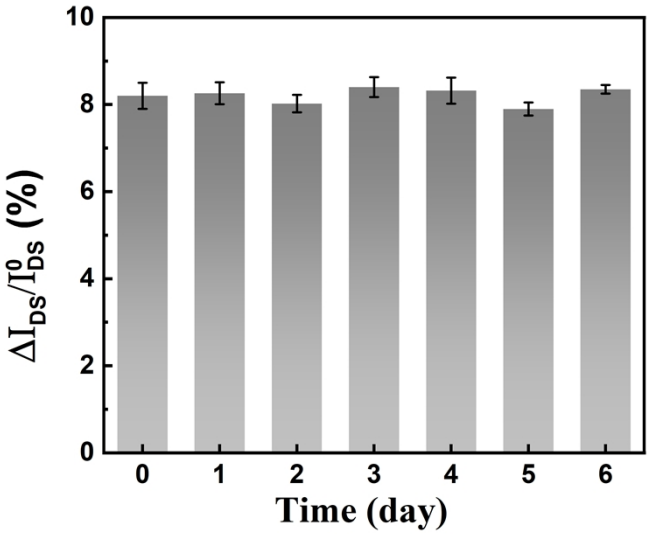

Fig. S5. Channel current response of Ag NPs-modified GECT after storage for different time (at 0.1 M Na_2SO_4 solution with presence of 1 μM PQ).

Table S1. Comparison of properties between the sensor prepared in this work and those prepared by traditional methods.

Electrode or matrix materials used in detection	Detection methods	Detection limit	Linear range	Ref.
CCDs/GCE	DPV	6.4 ×10^{-8} M	1.0×10^{-7} ~ 1.0×10^{-5} M	[S1]
Ag-CPE	DPV	2.0 ×10^{-8} M	1.0×10^{-7} ~ 1.0×10^{-3} M	[S2]
WP6@Ag@COF	DPV	1.4 ×10^{-8} M	0.01~50×10^{-6} M	[S3]
PyBTA/PSS	Fluorescent sensor	4 ×10^{-8} M	5 ~ 50 ×10^{-9} M	[S4]
CP[5]A-binding αHL nanopore	Biological nanopore-based techniques	2 ×10^{-9} M	\	[S5]
Fe_3O_4@Ag	SERS	1×10^{-10} M	\	[S6]
H-COF-SO_3H	SALDI-TOF MS	2.68 10^{-9} M	1.07×10^{-8}~1.61×10^{-6} M	[S7]
Ag NPs	GECT	1×10^{-10} M	1×10^{-10} M ~ 5×10^{-3} M	This work

Table S2. Recovery tests of Chinese cabbages samples with different PQ concentrations.

Added PQ amounts (μM)	Measured PQ amounts (μM)	Recovery (%)	Average recovery (%)	RSD (%)
1	1.031	103.1	102.867	7.148
	0.954	95.4		
	1.101	110.1		
10	9.871	98.71	94.566	4.658
	8.994	89.94		
	9.505	95.05		
100	101.657	101.657	101.339	3.944
	105.167	105.167		
	97.192	97.192		

References

[S1] H. Zhang, K.T. Huang, L. Ding, J. Yang, Y.W. Yang, F. Liang, Electrochemical determination of paraquat using a glassy carbon electrode decorated with pillararene-coated nitrogen-doped carbon dots, Chinese Chem. Lett., 2021, doi:10.1016/j.cclet.2021.09.002

[S2] A. Farahi, M. Achak, L.E. Gaini, M.A. El Mhammedi, M. Bakasse, Electrochemical determination of paraquat in citric fruit based on electrodeposition of silver particles onto carbon paste electrode J. Food Drug Anal., 23 (2015) 463-471, doi:10.1016/j.jfda.2015.03.003

[S3] X. Tan, Z. Zhang, T. Cao, W. Zeng, T. Huang, G. Zhao, Control Assembly of Pillar[6]arene-Modified Ag Nanoparticles on Covalent Organic Framework Surface for Enhanced Sensing Performance toward Paraquat, ACS Sustainable Chem. Eng. 7 (24) (2019) 20051-20059, doi:10.1021/acssuschemeng.9b05804.

[S4] H. Xu, K. Xiao, Q. Zhang, K. Huang, G. Song, Z. Yao, Rapid and visual detection of bipyridylium herbicides based on polyelectrolyte-induced

nanoassemblies of pyrenyl probes. ACS Sustainable Chem. Eng., 8 (17) (2020) 6861-6867, doi:10.1021/acssuschemeng.0c01870.

[S5] X. Jiang, M. Zang, F. Li, C. Hou, Q. Luo, J. Xu, J. Liu, Highly sensitive detection of paraquat with pillar[5]arenes as an aptamer in an alpha-hemolysin nanopore, Mater. Chem. Front., 5 (2021) 7032-7040, doi: 10.1039/d1qm00875g.

[S6] Ying Song a , Jinlei Chen a , Xiaobin Yang c , De Zhang a , Yanqiu Zou b , Dejiang Ni a , Jiaming Ye d , Zhi Yu a,* , Qiang Chen e,* , Shangzhong Jin b , Pei Liang, Fabrication of Fe_3O_4@Ag magnetic nanoparticles for highly active SERS enhancement and paraquat detection, Microchem. J., 173 (2022) 107019, doi.org/10.1016/j.microc.2021.107019

[S7] W. Tan, X. Xu, Y. Lv, W. Lei, K. Hu, F. Ye, S. Zhao, Sulfonic acid functionalized hierarchical porous covalent organic frameworks as a SALDI-TOF MS matrix for effective extraction and detection of paraquat and diquat, J. Colloid Interf. Sci., 603 (2021) 172-181, doi.org/10.1016/j.jcis.2021.06.077