

Supporting Information (SI)

Amino Siloxane Oligomer Modified Graphene Oxide Composite for the Efficient Capture of U(VI) and Eu(III) from Aqueous Solution

Donglin Zhao,[†] Lili Chen,^{†,‡} Mingwenchan Xu,[‡] Shaojie Feng,[†] Yi Ding,^{*†}

M. Wakeel,^{||} Njud S. Alharbi,[§] Changlun Chen^{*‡,§}

[†]*Key Laboratory of Functional Molecule Design and Interface Process, Anhui*

Jianzhu University, Hefei, 230601, PR China

[‡] *CAS Key Laboratory of Photovoltaic and Energy Conservation Materials, Institute*

of Plasma Physics, Chinese Academy of Sciences, P.O. Box 1126, Hefei 230031, PR

China

^{||} *Environmental Sciences, Bahauddin Zakariya University, Multan, 60800, Pakistan*

[§] *Department of Biological Sciences, Faculty of Science, King Abdulaziz University,*

Jeddah, 21589, Saudi Arabia

*Corresponding author: clchen@ipp.ac.cn (C.L. Chen), dyrqf@ahjzu.edu.cn (Y.

Ding),; Phone: +86-551-65592788. Fax: 86-551-65591310.

SI-1. Characterization

The scanning electron microscopy (FEI-JSM 6320F) image and the transmission electron microscopy (JEM-2010, Japan) image were obtained to investigate the morphologies and structures of the as-prepared materials. Fourier transform infrared spectroscopy (FTIR) was performed on a PerkinElmer IR-843 spectrometer (USA). The powder X-ray diffraction (XRD) were conducted by a Bruker D8-Advance X-ray diffractometer with Cu K α source ($\lambda=1.54178\text{\AA}$). Raman spectroscopy was obtained by a LabRam HR Raman spectrometry. TGA curves were examined by using a Shimadzu TGA-50 thermogravimetric analyzer in nitrogen atmosphere by heating the sample from room temperature to 800 °C at the rate of 10 °C/min with a nitrogen rate of 50 mL/min rate. The N₂ sorption-desorption isotherms were measured by a specific surface area and hole diameter analyzer (JW-BK132F, China) to evaluate their pore structures and N₂-BET surface area. The Zeta potential was measured at various pH with a Zetasizer Nano ZS instrument (Malvern Instrument Co., UK) at 25 °C. The particle size distributions were determined using a laser granulometer (Mastersizer Nano-ZS90ZS Malvern Instruments, UK) in liquid mode. The instrument is equipped with a liquid sample dispersion module that is suited for powders in suspension. Liquid dispersion was used for PAS-GO samples, where for each measurement approximately 20 mg of the powder was dispersed in 250 mL of solution. The X-ray photoelectron spectroscopy (XPS) was obtained by a Thermo Escalab 250 electron spectrometer.

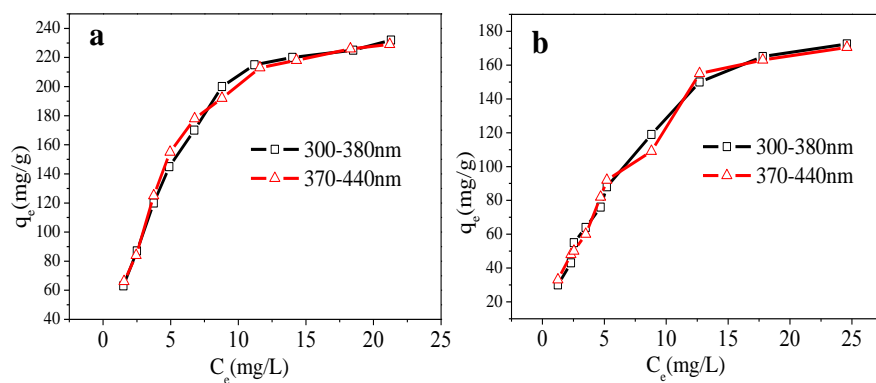


Figure S1. The sorption isotherms of U(VI) (a) and Eu(III) (b) on different sizes of PAS-GO ($T = 298$ K, $\text{pH} = 5.5 \pm 0.1$, $m/V = 0.1$ g/L.).

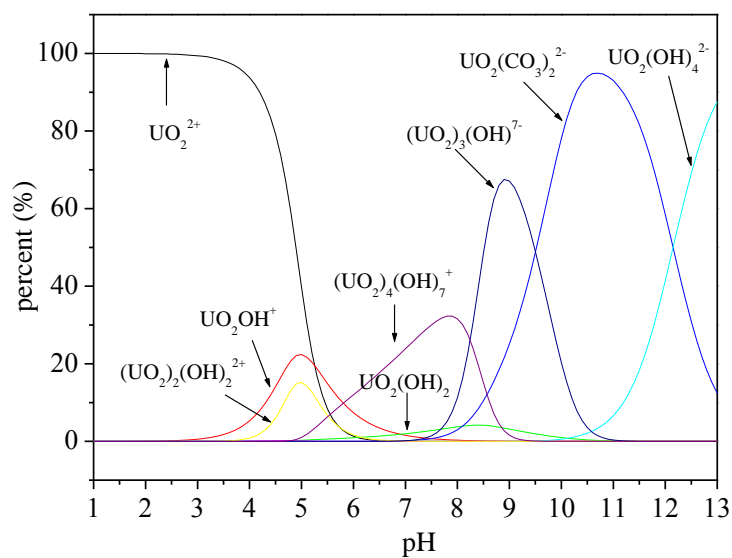


Figure S2. Distribution of aqueous U(VI) species as a function of pH values.

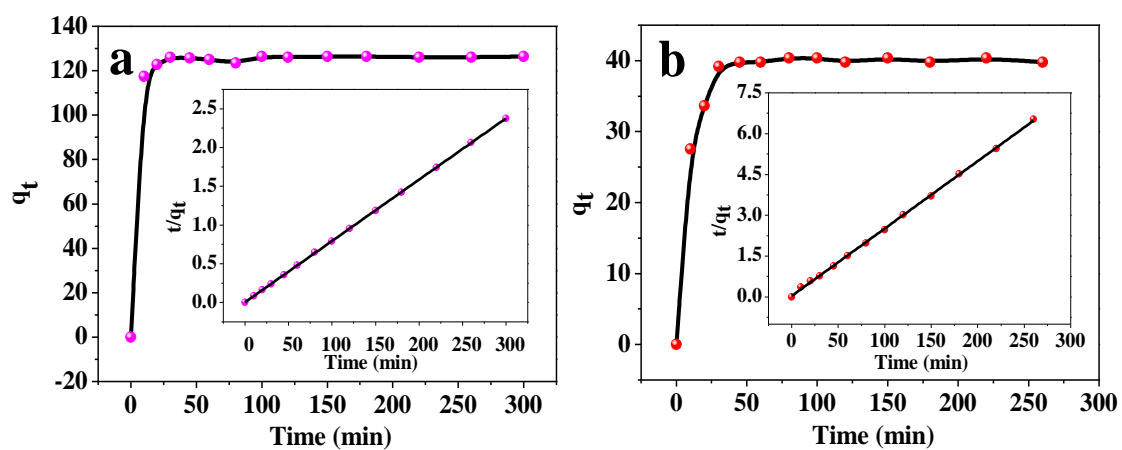


Figure S3. Effect of time on sorption of U(VI) (a) and Eu(III) (b), $\text{pH} = 5.5 \pm 0.1$, $T = 298 \text{ K}$, $m/V = 0.1 \text{ g/L}$, $C_0 = 16$ and 6 mg/L , respectively.

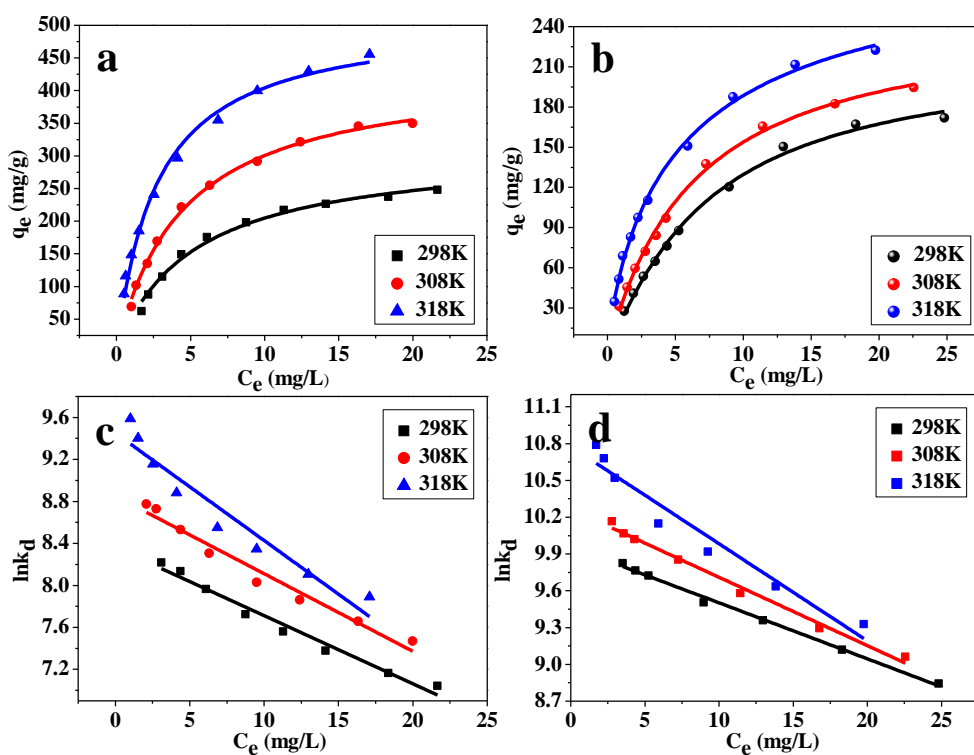


Figure S4. Sorption isotherms of U(VI) (a) and Eu(III) (b) sorption on PAS-GO; linear plot of $\ln K_d$ vs C_e for the sorption of U(VI) (c) and Eu(III) (d) on PAS-GO at 298, 308 and 318 K, $\text{pH} = 5.5 \pm 0.1$, $m/V = 0.1$ g/L.

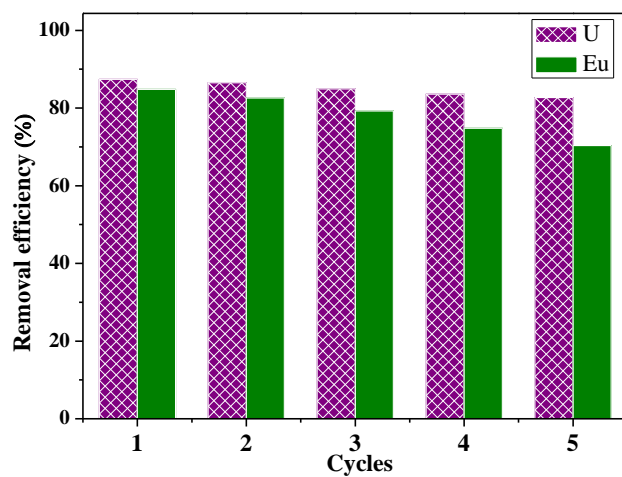


Figure S5. Recycling of PAS-GO for the removal of U(VI) and Eu(III), $T = 298\text{ K}$, $m/V = 0.1\text{ g/L}$, $\text{pH} = 5.5 \pm 0.1$, $C_0 = 16$ and 6 mg/L , respectively.

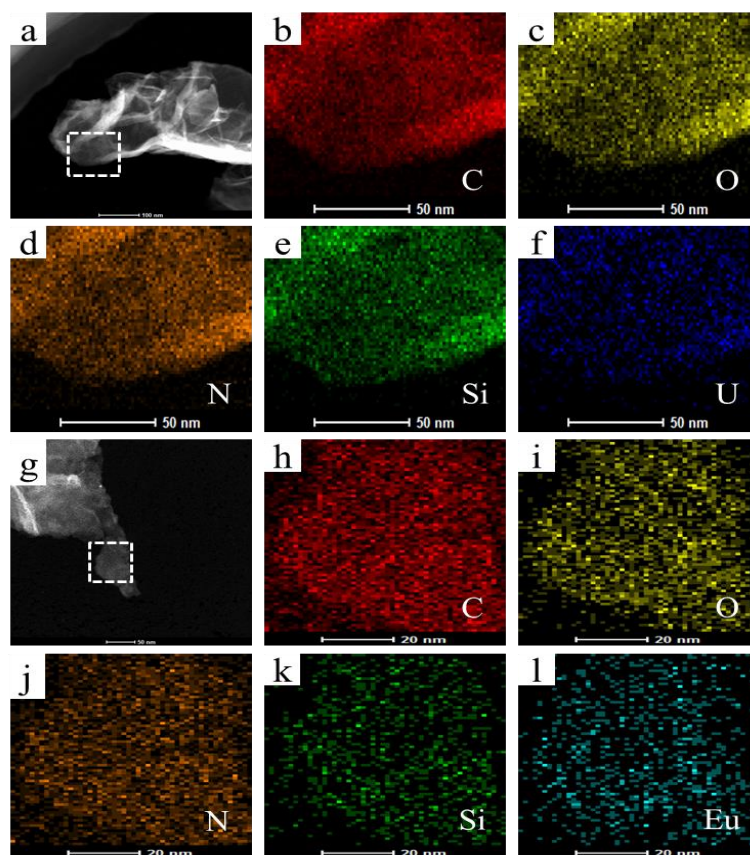


Figure S6. The elemental mapping images of PAS-GO samples after U(VI) (a-f) and Eu(III) (g-l) sorption calculated by TEM.

Table S1

The initial pH and final pH in the sorption system of U(VI) and Eu(III) onto PAS-GO.

	U(VI)								Eu(III)								
pH _{initial}	1.99	3.01	3.97	5.02	6.03	6.98	8.01	9.03	2.92	3.82	4.73	5.99	6.82	7.77	8.62	9.53	10.32
pH _{final}	2.42	3.39	4.12	5.55	6.26	7.04	7.78	7.93	3.21	4.09	4.92	6.19	6.88	7.51	7.76	8.24	8.43

Table S2

Kinetic Parameters of U(VI) and Eu(III) Sorption on the PAS-GO.

Elements	Pseudo-first order model			Pseudo-second order model		
	q_e (mg/g)	k_1 (g/mg/h)	R^2	q_e (mg/g)	k_2 (g/mg/h)	R^2
U(VI)	63.69	0.183	0.933	126.58	0.029	0.999
Eu(III)	29.24	0.08	0.810	40.32	0.017	0.999

Table S3

Isotherm Parameters for U(VI) and Eu(III) Sorption on the PAS-GO.

Elements	T (K)	Langmuir model			Freundlich model		
		q_{max} (mg/g)	K_L (mg/L)	R^2	n	K_F (mg ¹⁻ⁿ ·L ⁿ /g)	R^2
U(VI)	298	310.63	5.178	0.989	2.414	74.11	0.927
	308	434.50	4.460	0.997	2.341	105.50	0.951
	318	514.48	2.725	0.992	2.520	156.63	0.977
Eu(III)	298	243.90	9.414	0.993	1.612	4.274	0.972
	308	256.41	6.667	0.997	1.762	4.908	0.977
	318	263.16	3.658	0.997	2.014	5.887	0.970

Table S4

Comparison of the Maximum U(VI) and Eu(III) Sorption Capacity of on PAS-GO with Other Sorbents.

Sorbents	Elements	pH	T (K)	Sorption capacity (mg/g)	Ref.
PANI–CMK-3 composite	U(VI)	7.0	298	118.30	(1)
Oxy-HTCMs	U(VI)	4.5	298	183.40	(2)
GO/PPy composites	U(VI)	5.0	298	147.06	(3)
PAS-GO composite	U(VI)	5.5	298	310.63	This study
GtiP	Eu(III)	5.0	298	64.33	(4)
GO nanosheets	Eu(III)	6.0	298	175.44	(5)
Activated carbon	Eu(III)	4.5	298	46.50	(6)
PAS-GO composite	Eu(III)	5.5	298	243.90	This study

Table S5

Thermodynamic Parameters for U(VI) and Eu(III) Sorption on PAS-GO.

Elements	T(K)	ΔG^0 (kJ/mol)	ΔH^0 (kJ/mol)	ΔS^0 (J/mol. K)
U(VI)	298	-20.71		
	308	-22.66	42.90	123.27
	318	-24.99		
Eu(III)	298	-24.67		
	308	-26.29	32.08	190.17
	318	-28.49		

REFERENCE

- (1) Zhu, K.R.; Lu, S.H.; Gao, Y.; Zhang, R.; Tan, X.L.; Chen, C.L. Fabrication of hierarchical core-shell polydopamine@MgAl-LDHs composites for the efficient enrichment of radionuclides. *Appl. Surf. Sci.* **2017**, *396*, 1726–1735.

- (2) Li, F.Z.; Li, D.M.; Li, X.L.; Liao, J.L.; Li, S.J.; Yang, J.J.; Yang, Y.Y.; Liu, J. N. Microorganism-derived carbon microspheres for uranium removal from aqueous solution. *Chem. Eng. J.* **2016**, *284*, 630–639.
- (3) Pan, N.; Li, L.; Ding, J.; Li, S.K.; Wang, R.B.; Jin, Y.D.; Wang, X.K.; Xia, C.Q. Preparation of graphene oxide-manganese dioxide for highly efficient adsorption and separation of Th(IV)/U(VI). *J. Hazard. Mater.* **2016**, *309*, 107–115.
- (4) Li, C.R.; Huang, Y.; Lin, Z. Fabrication of titanium phosphate@graphene oxidenanocomposite and its super performance on Eu^{3+} recycling. *J. Mater. Chem.A* **2014**, *2*, 14979–14985.
- (5) Sun, Y.B.; Wang, Q.; Chen, C.L.; Tan, X.L.; Wang, X.K. Interaction between Eu(III) and graphene oxide nanosheets investigated by batch and extended X-ray absorption fine structure spectroscopy and by modeling techniques. *Environ. Sci. Technol.* **2012**, *46*, 6020–6027.
- (6) Gad, H.M.H.; Awwad, N.S. Factors affecting on the sorption/desorption of Eu(III) using activated carbon. *Sep. Sci. Technol.* **2007**, *42*, 3657–3680.