Supporting Information

Hierarchically Porous, Ultra-strong Reduced Graphene Oxide-Cellulose Nanocrystal Sponges for Exceptional Adsorption of Water Contaminants

Nariman Yousefi[1†], Kerwin Wong[1,2†], Zeinab Hosseinidoust[1,3], Henning Osholm Sørensen[4], Stefan Bruns[4], Yi Zheng[5], Nathalie Tufenkji[1]*

[1]Department of Chemical Engineering, McGill University, Montreal, QC, Canada

[2]Department of Chemical Engineering, University of Leuven, Leuven, Belgium

[3]Current affiliation: Department of Chemical Engineering, McMaster University, Hamilton, ON, Canada

[4]Nano-Science Center, Department of Chemistry, University of Copenhagen, Universitetsparken 5, 2100 Copenhagen Ø, Denmark

[5]Department of Physics, Technical University of Denmark, 2800 Kongens Lyngby, Denmark

* Corresponding Author. Phone: (514) 398-2999; Fax: (514) 398-6678; e-mail: nathalie.tufenkji@mcgill.ca

† Equal contribution

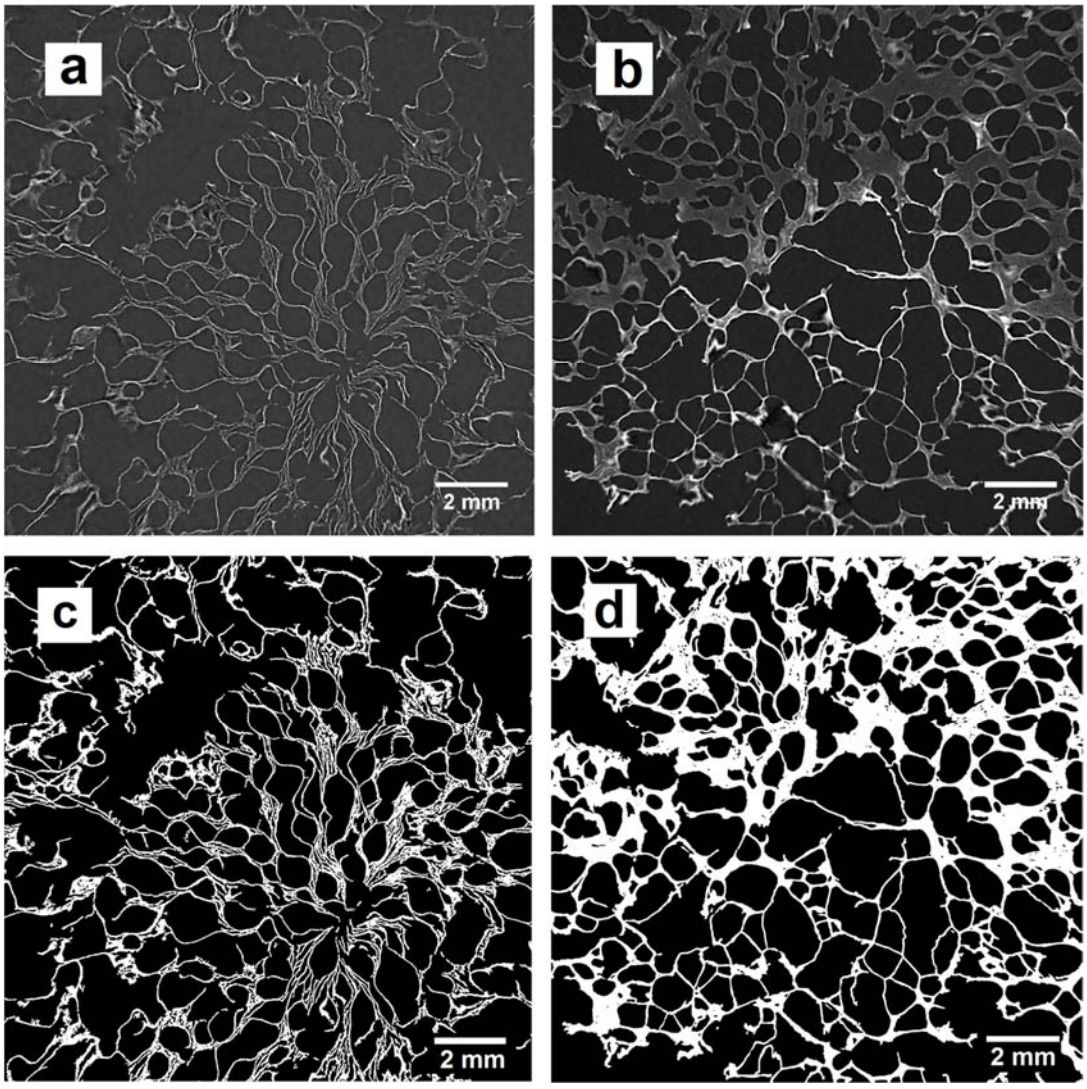

Figure S1. (a, b) As-obtained and (c, d) noise filtered X-ray tomography images of (a, c) rGO:VC 150 and (b, d) rGO:CNC 2:1 sponges.

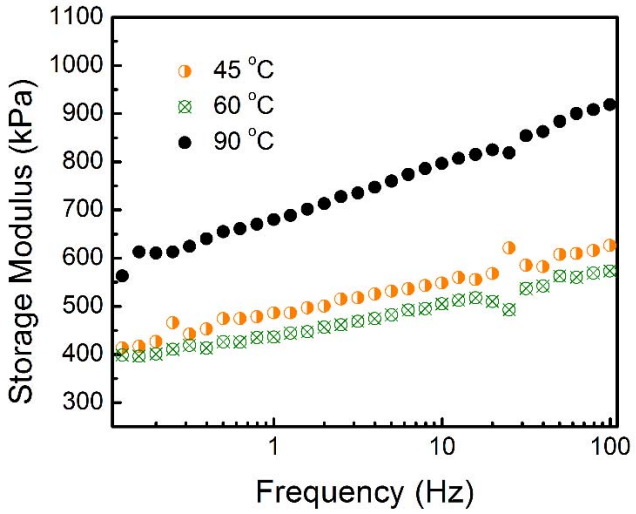

Figure S2. The storage modulus of rGO:CNC 2:1 sponge formed at low temperatures.

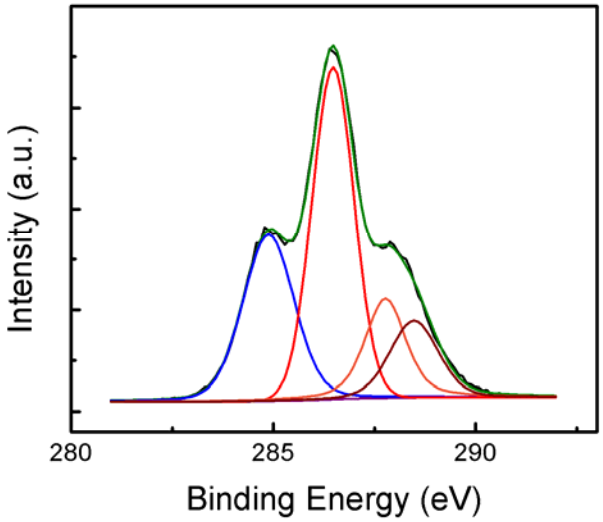

Figure S3. Deconvolution of C_{1s} XPS peak for bare CNC.

Table S1. Comparison between dynamic storage modulus of graphene oxide-based sponges reported in the literature and those prepared in this work

Material	G' at 10 Hz (kPa)	Measurement Method
GO[1]	1	Rheometry
GO[2]	0.3	Rheometry
rGO:VC 150 sponge*	139	Rheometry
GO/Polyacrilamide[3]	7	Rheometry
GO/hyaluronic acid[4]	2	Rheometry
GO (peroxidized)[5]	60	DMA
GO/Alginate/PAM[6]	30	Rheometry
GO/Sodium deoxycholate[7]	40	Rheometry
rGO:CNC 2:1 sponge*	800	Rheometry

* This work

Table S2. XPS C_{1s} peaks of rGO:CNC 2:1 sponge formed at low temperatures

Surface Composition	GO	GO:CNC 2:1 (45 °C)	GO:CNC 2:1 (60 °C)
% C=C (sp^2)	21.5	50.8	65.0
% C–C (sp^3)	12.1	18.2	10.4
% C–OH	11.3	14.6	11.8
% C=O/C–O–C	36.9	10.0	8.8
% C–OOH	18.2	6.4	4.0

Table S3. MB uptake for different graphene oxide-based materials reported in the literature

Material	q_{max} (mg·g^{-1})
rGO-based hydrogel[8]	8
Graphene/CNT hybrid[9]	82
Ni-doped Graphene/Carbon hydrogel[10]	150
Graphene hydrogel[11]	180
rGO hydrogel[5]	400
NORIT GAC*	400
rGO:CNC 2:1*	850
3D GO/Biopolymer gel[12]	1100

* This work

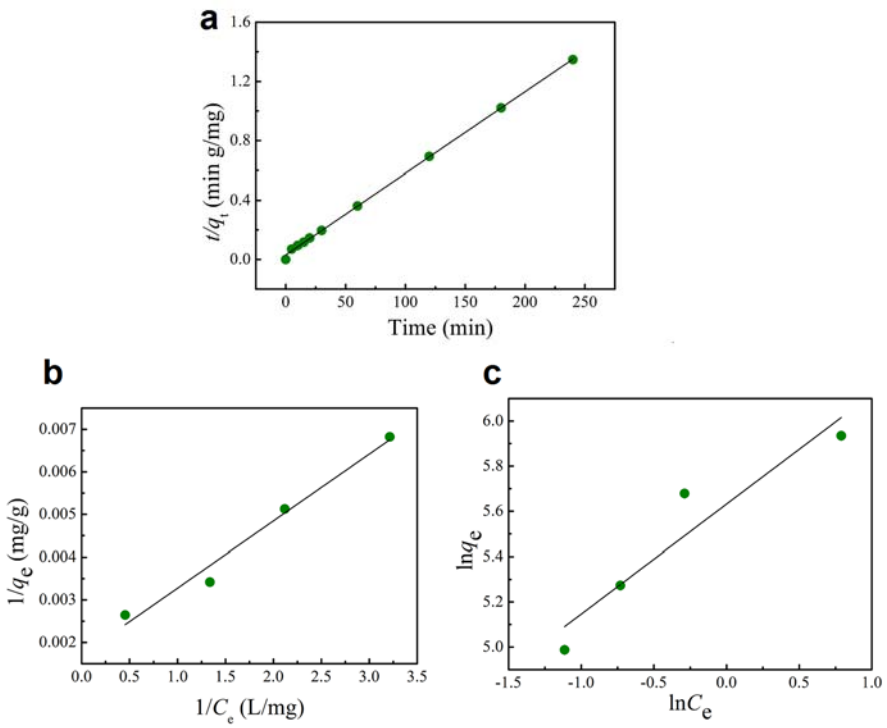

Figure S4. (a) MB batch uptake data (C_0 = 20 mg/L) could be fitted with the linearized form of pseudo-second order kinetic model. The adsorption isotherms were fitted with (b) Langmuir and (c) Freundlich adsorption isotherm models.

Table S4. Comparison between the adsorption capacities of rGO:CNC 2:1 sponges and those of granular activated carbon reported in the literature

Contaminant	Granular Activated Carbon (mg/g)	rGO:CNC 2:1* (mg/g)
MB	400 *	850
DCF	64 [13]	129
SMX	71 [14]	107
TC	455 [15]	149
EE2	20 [16]	117
Cu^{2+}	15 [17]	65
Cd^{2+}	5 [18]	76
Cyanotoxin	16–83 [19]	22

* This work

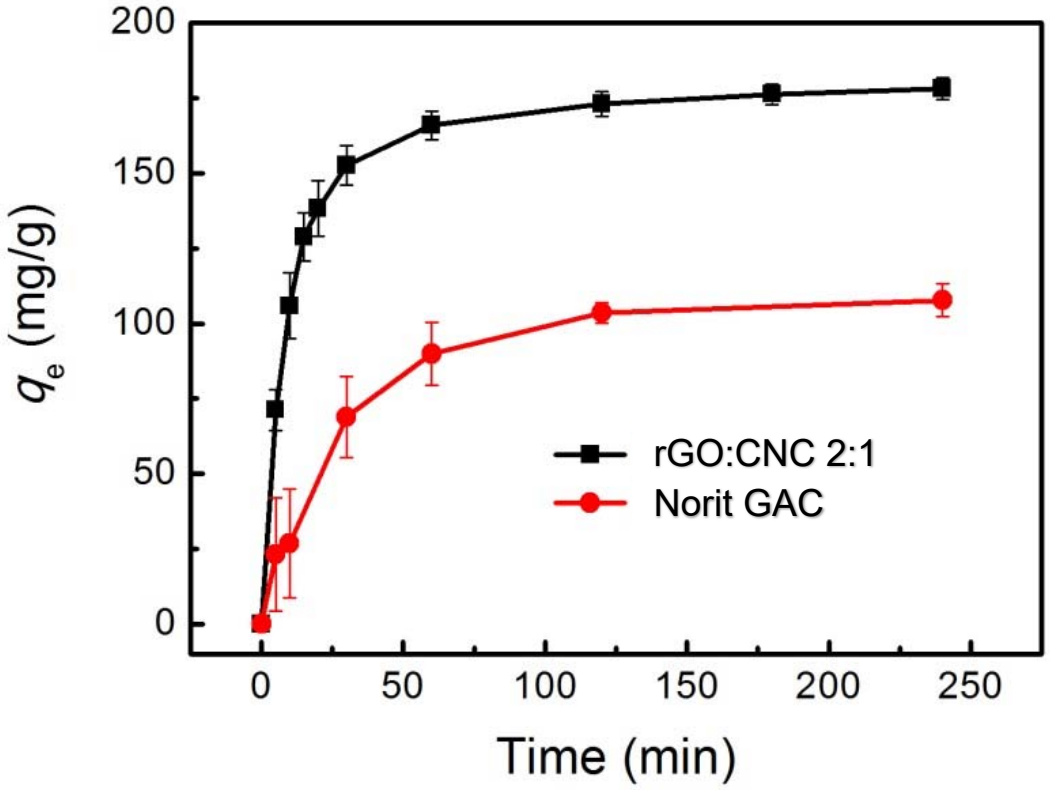

Figure S5. Kinetic comparison between MB adsorption capacity of rGO:CNC 2:1 sponge and commercially available Norit GAC830 granular activated carbon. The experimental conditions for measuring the adsorption capacity of the sponge and GAC were identical (refer to the Materials and Methods in the manuscript).

References

1. O. C. Compton, Z. An, K. W. Putz, B. J. Hong, B. G. Hauser, L. C. Brinson and S. T. Nguyen, *Carbon*, 2012, **50**, 3399-3406.
2. Q.-Y. Cheng, D. Zhou, Y. Gao, Q. Chen, Z. Zhang and B.-H. Han, *Langmuir*, 2012, **28**, 3005-3010.
3. S. Das, F. Irin, L. Ma, S. K. Bhattacharia, R. C. Hedden and M. J. Green, *ACS Appl. Mater. Interf.*, 2013, **5**, 8633-8640.
4. F. Song, W. Hu, L. Xiao, Z. Cao, X. Li, C. Zhang, L. Liao and L. Liu, *J. Biomater. Sci., Polym. Ed.*, 2015, **26**, 339-352.
5. J. Liu, C. Chen, C. He, J. Zhao, X. Yang and H. Wang, *ACS Nano*, 2012, **6**, 8194-8202.
6. J. Fan, Z. Shi, M. Lian, H. Li and J. Yin, *Journal of Materials Chemistry A*, 2013, **1**, 7433-7443.
7. Z. Tan, S. Ohara, H. Abe and M. Naito, *RSC Adv.*, 2014, **4**, 8874-8878.

8. J. N. Tiwari, K. Mahesh, N. H. Le, K. C. Kemp, R. Timilsina, R. N. Tiwari and K. S. Kim, *Carbon*, 2013, **56**, 173-182.
9. T. Wu, M. Chen, L. Zhang, X. Xu, Y. Liu, J. Yan, W. Wang and J. Gao, *J. Mater. Chem. A*, 2013, **1**, 7612-7621.
10. G. Wei, Y.-E. Miao, C. Zhang, Z. Yang, Z. Liu, W. W. Tjiu and T. Liu, *ACS Appl. Mater. Interf.*, 2013, **5**, 7584-7591.
11. J. Zhao, W. Ren and H.-M. Cheng, *Journal of Materials Chemistry*, 2012, **22**, 20197-20202.
12. J. Deng, B. Lei, A. He, X. Zhang, L. Ma, S. Li and C. Zhao, *J. Hazard. Mater.*, 2013, **263**, 467-478.
13. J. Sotelo, A. Rodríguez, S. Álvarez and J. García, *Chem. Eng. Res. Des.*, 2012, **90**, 967-974.
14. M. C. Tonucci, L. V. A. Gurgel and S. F. de Aquino, *Ind. Crops Prod.*, 2015, **74**, 111-121.
15. A. C. Martins, O. Pezoti, A. L. Cazetta, K. C. Bedin, D. A. Yamazaki, G. F. Bandoch, T. Asefa, J. V. Visentainer and V. C. Almeida, *Chem. Eng. J.*, 2015, **260**, 291-299.
16. A. Felebuegu, J. Lester, J. Churchley and E. Cartmell, *Environ. Technol.*, 2006, **27**, 1343-1349.
17. J. P. Chen, S. Wu and K.-H. Chong, *Carbon*, 2003, **41**, 1979-1986.
18. X. Huang, N.-y. Gao and Q.-l. Zhang, *J. Environ. Sci.*, 2007, **19**, 1287-1292.
19. P. Pendleton, R. Schumann and S. H. Wong, *J. Colloid Interf. Sci.*, 2001, **240**, 1-8.