Supporting Information

Nano-scaled surface modification of polydimethylsiloxane using carbon nanotubes for enhanced oil and organic solvent absorption

Chong Cheen Ong[†], Satisvar Sundera Murthe [†], Norani Muti Mohamed[†,‡], Veeradasan Perumal[‡,¶], Mohamed Shuaib Mohamed Saheed[†,‡]*

[†]Department of Fundamental & Applied Sciences, Universiti Teknologi PETRONAS, 32610 Seri Iskandar, Perak Darul Ridzuan, Malaysia.

[‡]Centre of Innovative Nanostructures & Nanodevices (COINN), Universiti Teknologi PETRONAS, 32610 Seri Iskandar, Perak Darul Ridzuan, Malaysia.

[¶]Department of Mechanical Engineering, Universiti Teknologi PETRONAS, 32610 Seri Iskandar, Perak Darul Ridzuan, Malaysia.

*Corresponding Author

Email: shuaib.saheed@utp.edu.my

Telephone: +605-3687653

FESEM analysis

Figure S1 shows the FESEM analysis of various concentration of CNT, 0.2mg/ml (PC0.2), 0.4mg/ml (PC0.4), 0.6mg/ml (PC0.6), 0.8mg/ml (PC0.8mg/ml) and 1mg/ml (PC1) disperse in the sugar mixture before adding PDMS pre-polymer.

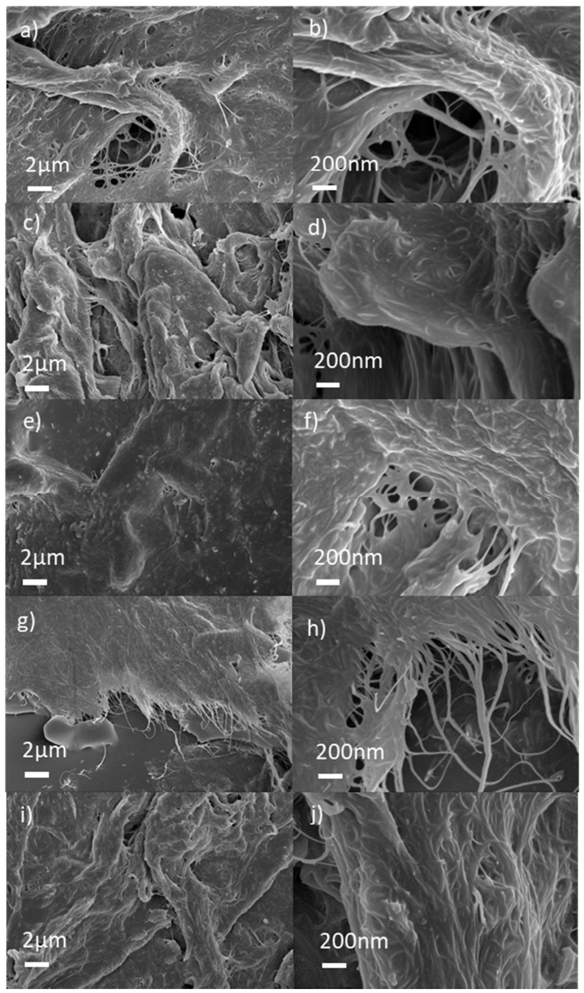

Figure S1 FESEM image of cross-section of (a,b) PC0.2 (c,d) PC0.4 (e,f) PC0.6 (g,h) PC0.8 (i,j) PC1

Surface Wetting Mechanism

Surface roughness is essential part to improve hydrophobicity can be proven by;

Wenzel's equation.

$$cos\theta_{rough} = rcos\theta_{flat} \quad (1]$$

Cassie-Baxter equation

$$cos\theta_{rough} = \phi_s cos\theta_{flat} - (1 - \phi_s) \quad (2)$$

where r is surface roughness factor (actual surface area: geometrical one) and ϕ_s is fraction of solid surface that contacts water. Knowing the fundamental of surface roughness influence greatly in developing highly hydrophobic surface, nanostructure CNTs was used to enhance the surface asperities in a nano-scaled structure.

XPS Analysis

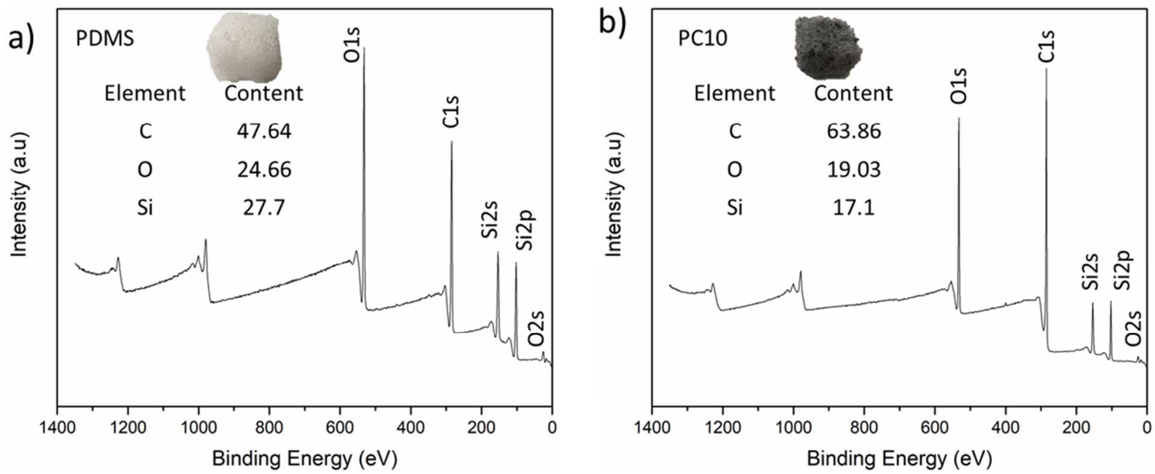

Figure S2: a) XPS spectrum of PDMS b) XPS spectrum of PC10

Contact Angle and Young's Equation

To determine whether the material have both property, water contact angle (CA) θ was used which can be calculated by Young's equation.

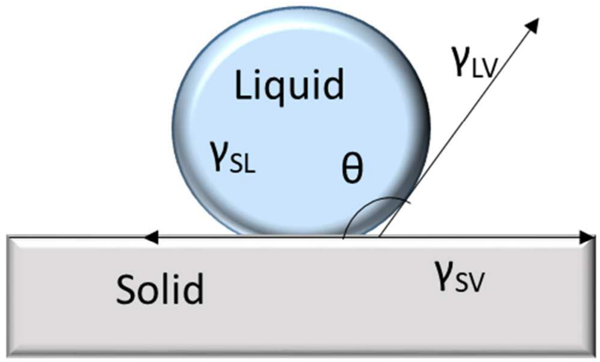

Figure S3: Schematic illustration of liquid on solid surface

$$\cos\theta = \frac{\gamma_{SV} - \gamma_{SL}}{\gamma_{LV}} \quad (3)$$

Where γ_{SV} is interfacial tension between solid and vapor, γ_{SL} is interfacial tension between solid and liquid, and γ_{LV} is interfacial tension between liquid and vapour. From this equation, for an ideal absorbent to have high CA, surface energy of the solid need to be lowered.

Table S1: Relation of water contact angle to surface hydrophobicity

Water Contact Angle	**Hydrophobic/Hydrophilic**
<90°	Hydrophilic
90° to 150°	Hydrophobic
>150°	Superhydrophobic

If contact angle is less than 90°, the surface is considered hydrophilic. For hydrophobic surface, the contact angle should be between 90° to 150°. To improve hydrophobicity, surface roughness of material can be manipulated. This is because surface roughness and low surface energy material can create an artificial hydrophobic surfaces[1].

Comparison of Performance

Table S2: Comparison of absorbent material performance of previous work

Absorbent Material	%	Absorbate	Authors/Year
Carbon nanotube sponges[2]	10000	Mineral Oil Vegetable Oil Diesel Oil Octane Ethyl Acetate	Gui *et al*. 2011
Polydimethylsiloxane Sponge[3]	1100	Chloroform Dichloroform 1,2-dichlorobenzene Silicone Oil Motor Oil N,N-dimethylmethane Toluene Transformer Oil Methanol Ethanol Acetone	Choi *et al*. 2011
Magnetic Carbon nanotubes[4]	5600	Diesel Oil Gasoline	Gui *et al*. 2013
Carbon Nanotubes-Graphene Hybrid Aerogel[5]	13000	Pump Oil Vegetable Oil Diesel Gasoline Ethyl Acetate	Hu *et al*. 2014
Graphene coated cotton[6]	500	Chloroform Hexane Acetone Ethanol Methanol Pump Oil Rap Oil Ethyl Acetate	Ge *et al*. 2014
Hard Template PDMS[7]	1301	Dichloromethane n-Hexane Petrol Diesel Oil Toluene	Zhao *et al*. 2014

Functionalized Graphene Aerogel[8]	10000	Chlorobenzene Chloroform Dimethylformamide Toluene Pump Oil Tetrahydrofuran Methanol Ethanol Acetone	Hong *et al*. 2015
Electrospun carbon-silica nanofiber sponge[9]	14000	Toluene Tetrahydrofuran Dimethylformamide Isopropyl Alcohol Benzyl Alcohol Isooctane Pump Oil Silicone Oil Olive Oil	Tai *et al*. 2015
Polyurethane@Fe_3O_4@SiO_2@fluoropolymer sponges[10]	4450	Petrol Crude Oil Toluene Soybean Oil n-hexane ethanol Tetrachloromethane Chloroform Petroleum ether 1,2-dichlorobenzene	Wu *et al*. 2015
Fe_3O_4 nanoparticles decorated 3D graphene aerogels[11]	2000	Gasoline Engine Oil Cyclohexane Octane Xylene	Li *et al*. 2016
Nitrogen-doped graphene sponge[12]	20000	Diesel Kerosene Engine Oil Vegetable Oil Gasoline	Yang *et al*. 2016
Porous and Wrinkle Graphene[13]	5300	Pump Oil Bean Oil Lubricating Oil Olive Oil Ethanol	Fu *et al*. 2016

PDMS-CNT (current work)	3100	Chloroform Dichloromethane Chlorobenzene Vegtable Oil Engine Oil Toluene Diesel Oil Petrol Cyclohexane	Ong *et al*. 2018

REFERENCE

(1) Gupta, S.; Tai, N.-H. Carbon materials as oil sorbents: a review on the synthesis and performance J. Mater. Chem. A **2016**, 4 (5), 1550–1565.

(2) Gui, X.; Li, H.; Wang, K.; Wei, J.; Jia, Y.; Li, Z.; Fan, L.; Cao, A.; Zhu, H.; Wu, D. Recyclable carbon nanotube sponges for oil absorption *Acta Mater.* **2011**, *59* (12), 4798–4804.

(3) Choi, S.-J.; Kwon, T.-H.; Im, H.; Moon, D.-I.; Baek, D. J.; Seol, M.-L.; Duarte, J. P.; Choi, Y.-K. A Polydimethylsiloxane (PDMS) Sponge for the Selective Absorption of Oil from Water *ACS Appl. Mater. Interfaces* **2011**, *3* (12), 4552–4556.

(4) Gui, X.; Zeng, Z.; Lin, Z.; Gan, Q.; Xiang, R.; Zhu, Y.; Cao, A.; Tang, Z. Magnetic and highly recyclable macroporous carbon nanotubes for spilled oil sorption and separation *ACS Appl. Mater. Interfaces* **2013**, *5* (12), 5845–5850.

(5) Hu, H.; Zhao, Z.; Gogotsi, Y.; Qiu, J. Compressible Carbon Nanotube-Graphene Hybrid Aerogels with Superhydrophobicity and Superoleophilicity for Oil Sorption *Environ. Sci. Technol. Lett.* **2014**, *1* (3), 214–220.

(6) Ge, B.; Zhang, Z.; Zhu, X.; Men, X.; Zhou, X.; Xue, Q. A graphene coated cotton for oil/water separation *Compos. Sci. Technol.* **2014**, *102*, 100–105.

(7) Zhao, X.; Li, L.; Li, B.; Zhang, J.; Wang, A. Durable superhydrophobic/superoleophilic PDMS sponges and their applications in selective oil absorption and in plugging oil leakages *J. Mater. Chem. A Mater. energy Sustain.* **2014**, *2*, 18281–18287.

(8) Hong, J. Y.; Sohn, E. H.; Park, S.; Park, H. S. Highly-efficient and recyclable oil absorbing performance of functionalized graphene aerogel *Chem. Eng. J.* **2015**, *269*, 229–235.

(9) Tai, M. H.; Gao, P.; Yong, B.; Tan, L.; Sun, D. D.; Leckie, J. O. A self-assembled superhydrophobic electrospun carbon-silica nanofiber sponge for selective removal and recovery of oils and organic solvents *Chem. Eur. J.* **2014**, 21(14), 5395-5402.

(10) Wu, L.; Li, L.; Li, B.; Zhang, J.; Wang, A. Magnetic, durable, and superhydrophobic polyurethane@Fe_3O_4@SiO_2@fluoropolymer sponges for selective oil absorption and oil/water separation *ACS Appl. Mater. Interfaces* **2015**, *7* (8), 4936–4946.

(11) Li, Y.; Zhang, R.; Tian, X.; Yang, C.; Zhou, Z. Facile synthesis of Fe_3O_4 nanoparticles decorated on 3D graphene aerogels as broad-spectrum sorbents for water treatment *Appl. Surf. Sci.* **2016**, *369*, 11–18.

(12) Yang, W.; Gao, H.; Zhao, Y.; Bi, K.; Li, X. Facile preparation of nitrogen-doped graphene sponge as a highly efficient oil absorption material *Mater. Lett.* **2016**, *178*, 95–99.

(13) Fu, C.; Wang, Z.; Liu, J.; Jiang, H.; Li, G.; Zhi, C. Large scale fabrication of graphene for oil and organic solvent absorption *Prog. Nat. Sci. Mater. Int.* **2016**, *26* (3), 319–323.