CHEMISTRY
AN ASIAN JOURNAL

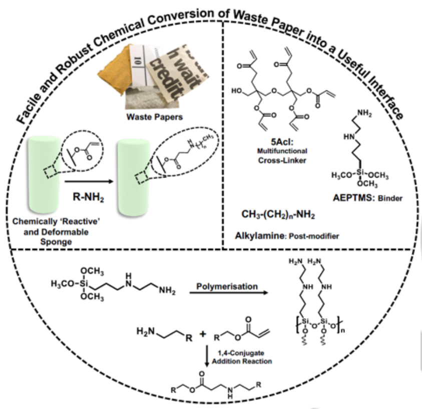
Supporting Information

# Design of a Waste Paper-Derived Chemically 'Reactive' and Durable Functional Material with Tailorable Mechanical Property Following an Ambient and Sustainable Chemical Approach

Arpita Shome, Adil M. Rather, Angana Borbora, Pillalamarri Srikrishnarka, Avijit Baidya, Thalappil Pradeep,* and Uttam Manna*

## Table of Contents

Facile conversion of waste paper into a deformable and chemically 'reactive' sponge. The covalent association of a binder with waste paper paved an avenue to tailor the mechanical property and the catalyst-free 1,4-conjugate addition reaction between the primary amine of the binder with a multi-functional cross-linker provided a facile basis to induce residual chemical 'reactivity'.




*Arpita Shome,* [a] *Adil M Rather,* [a], [b] *Angana Borbora,* [a] *Pillalamarri Srikrishnarka,* [c] *Avijit Baidya,* [c], [e] *Thalappil Pradeep** [c] *and Uttam Manna** [a], [d]

***Page No. – Page No.***

**Design of Waste Paper Derived Chemically 'Reactive' & Durable Functional Material with Tailorable Mechanical Property Following an Ambient & Sustainable Chemical Approach**

**Movie 1: Video graphic evidence of bouncing of a stream of water jet from the surface of the waste-paper derived superhydrophobic sponge**

**Movie 2: Demonstration of the absorption based selective separation of light oil (i.e. motor oil) from an oil/water interface using the waste-paper derived superhydrophobic sponge**

**Movie 3: Demonstration of the absorption based selective separation of model heavy oil (i.e. dichloromethane) from an oil/water interface using waste-paper derived superhydrophobic sponge**

**Movie 4: Demonstration of the gravity-driven filtration based selective separation of light oil (i.e. kerosene) from an oil/water mixture through the waste-paper derived superhydrophobic sponge**

**Movie 5: Demonstration of the gravity-driven filtration based selective separation of model heavy oil (i.e. dichloromethane) from an oil/water mixture through the waste-paper derived superhydrophobic sponge**

| Sample | Concentration of Waste Paper (wt%) | AEPTMS added (wt%) | Porosity (%) |
|---|---|---|---|
| $WPDS_{0.5}$ | 1.96 | 1.65 | 99.1 ± 1.2 |
| $WPDS_1$ | 1.96 | 3.25 | 98.4 ± 0.8 |
| $WPDS_2$ | 1.96 | 4.79 | 96.8 ± 1.6 |
| $WPDS_3$ | 1.96 | 6.29 | 95.1 ± 0.7 |

**Table S1** accounting for the concentration of waste paper and AEPTMS used for developing the waste paper derived sponges with varying porosities.

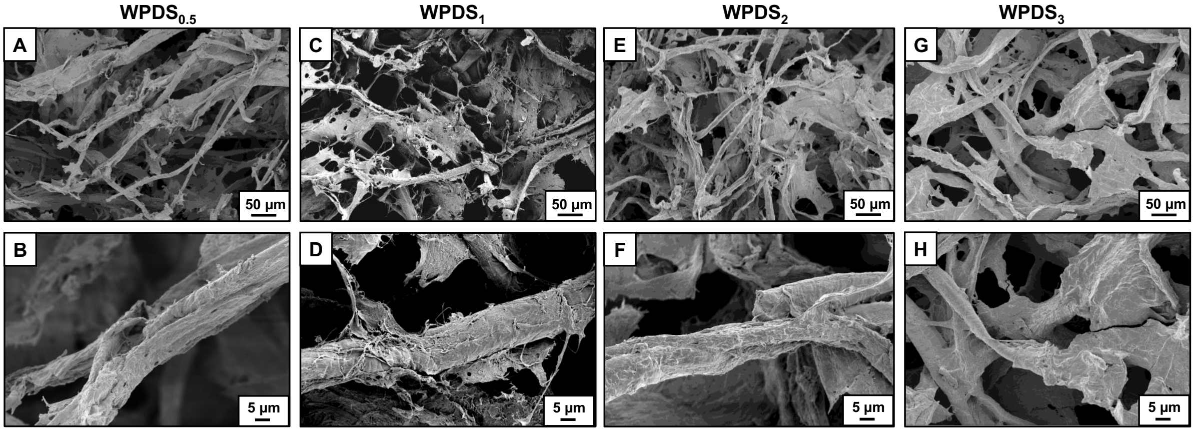



**Figure S1.** A-H) FESEM images at low (A, C, E, G) and high (B, D, F, H) magnifications exhibiting the interconnected, fibrous and porous morphology of the waste paper derived sponges doped with different concentrations of the binder, AEPTMS.

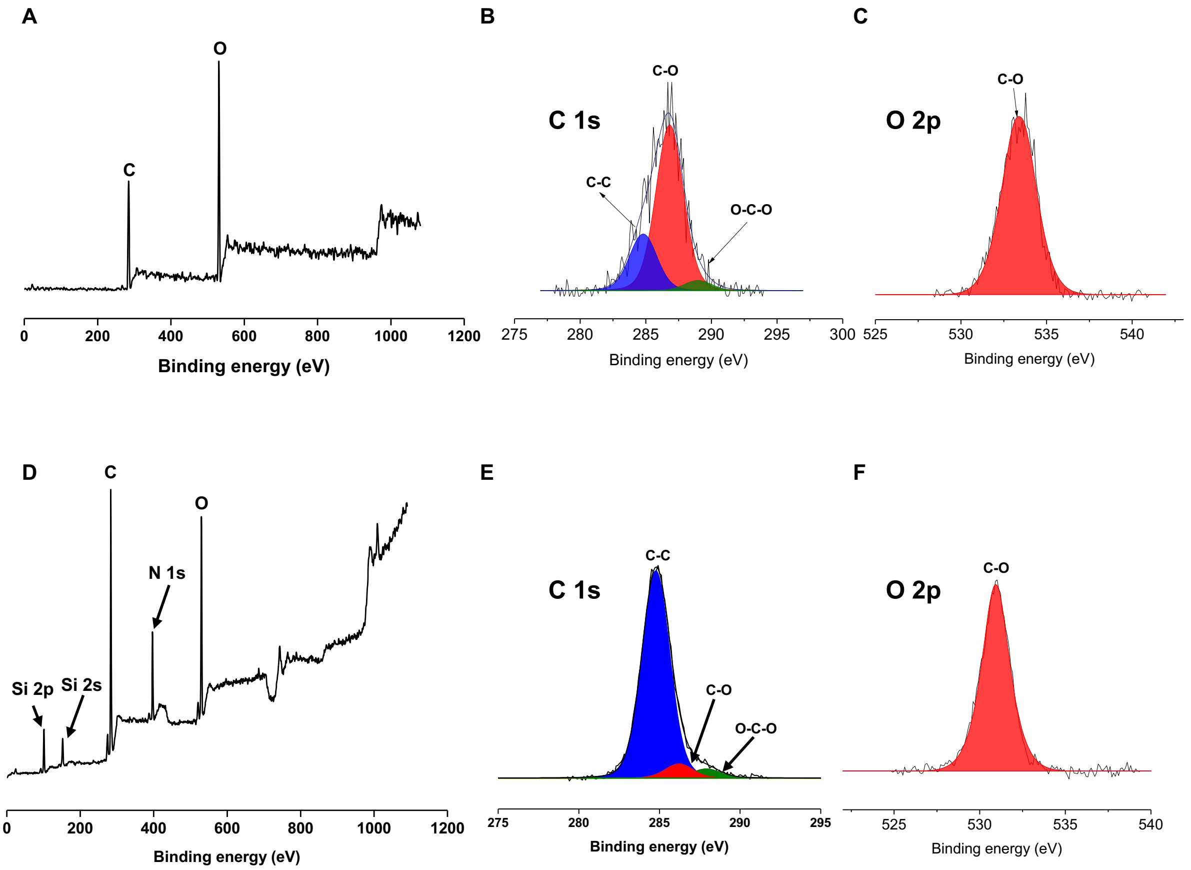



**Figure S2.** A) XPS spectra of waste paper derived sponge, $WPS_{control}$. B,C) De-convoluted XPS spectra of C 1s (B) and O 2p (C) of the waste paper derived sponge, $WPS_{control}$ respectively. D) XPS spectra of waste paper derived sponge,$WPDS_1$ E-H) De-convoluted XPS spectra of C 1s (E), O 2p (F), N 2p (G) and Si 2p (H) of the waste paper derived sponge,$WPDS_1$ respectively.

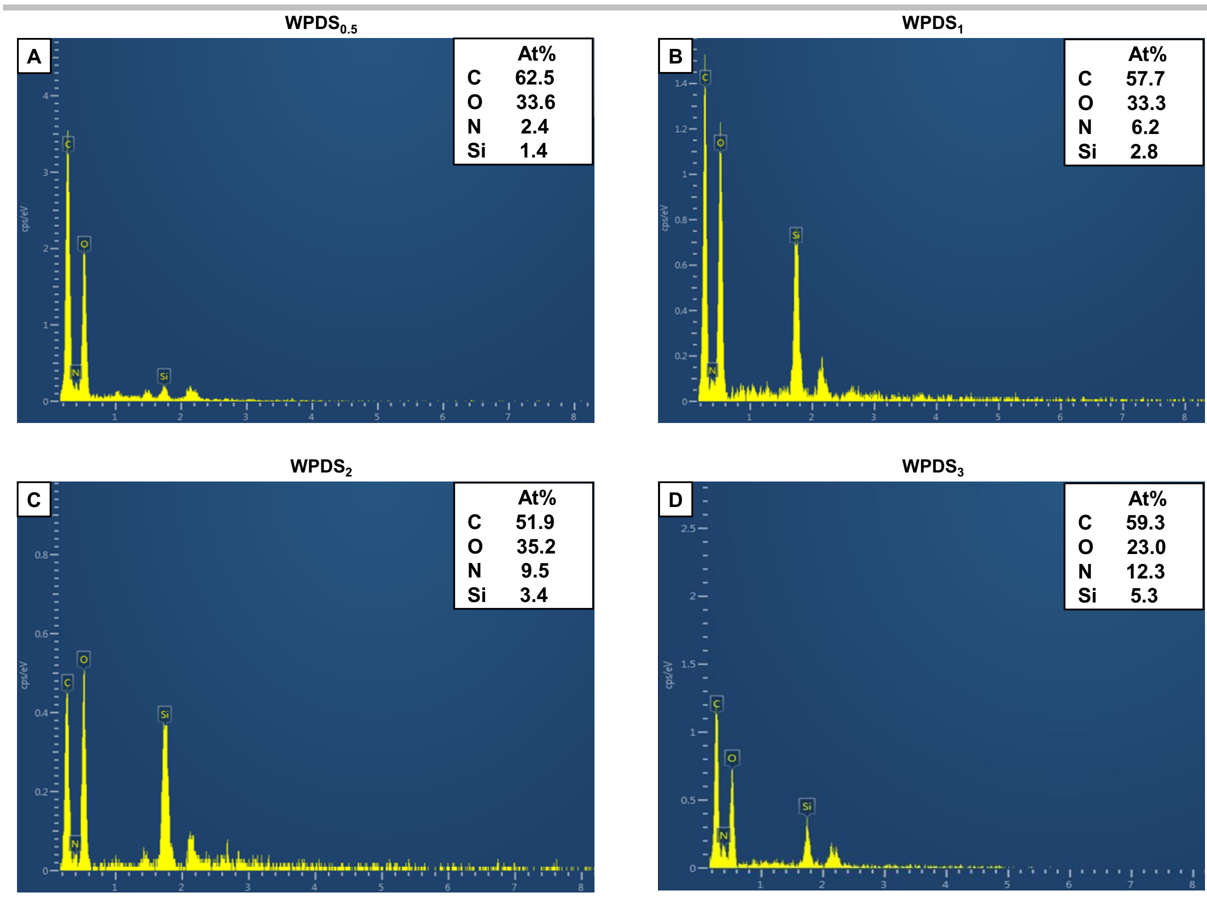



**Figure S3.** A-D) Energy dispersive X-ray spectral analysis of the waste paper derived sponges doped with different concentrations of the binder (AEPTMS) i.e. $WPDS_{0.5}$ (A), $WPDS_1$ (B), $WPDS_2$ (C) and $WPDS_3$ (D).

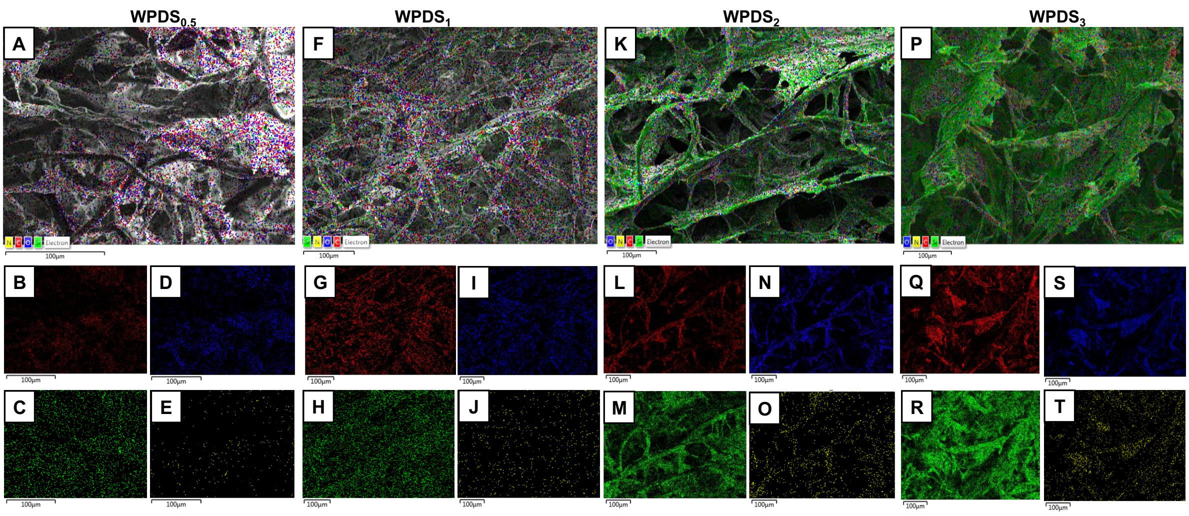



**Figure S4.** A-D) Energy dispersive X-ray mapping images of the waste paper derived sponges i.e. $WPDS_{0.5}$ (A-E), $WPDS_1$ (F-J), $WPDS_2$ (K-O) and $WPDS_3$ (P-T). The color codes for the elements are C (red), O (blue), Si (green) and N (yellow).

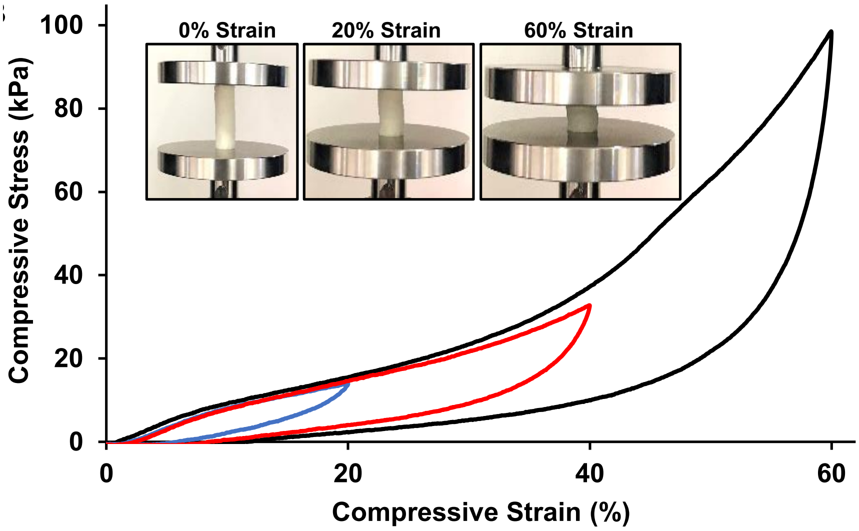



**Figure S5.** Plot illustrating the cyclic stress-strain analysis of $WPDS_1$ under different percentage of strain.

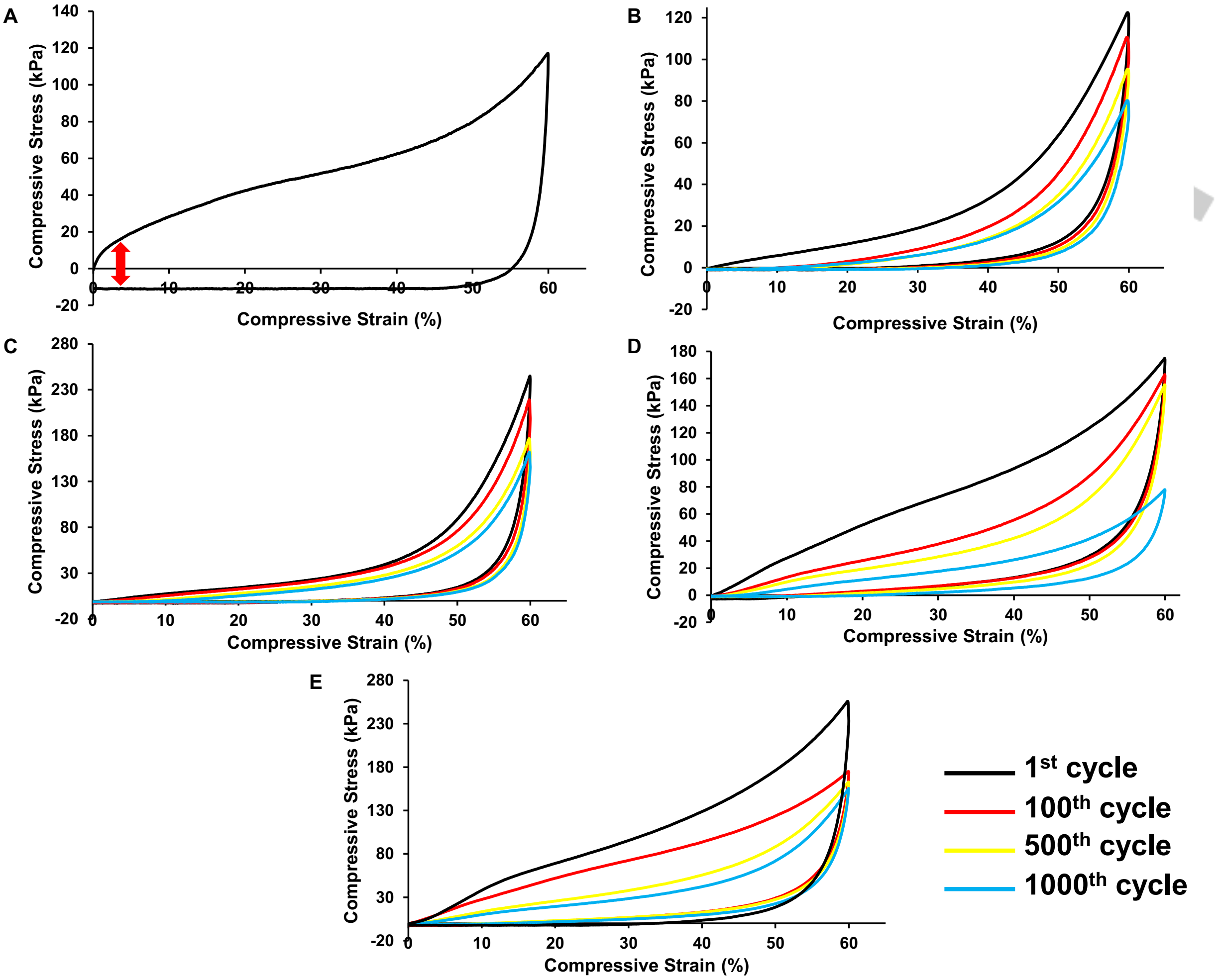



**Figure S6.** A-E) Plot accounting for the cyclic compression analysis of the waste paper derived sponge, $WPS_{control}$ (A) and the waste paper derived sponges doped with different concentrations of the binder (AEPTMS) i.e. $WPDS_{0.5}$ (B), $WPDS_1$ (C), $WPDS_2$ (D) and $WPDS_3$ (E).

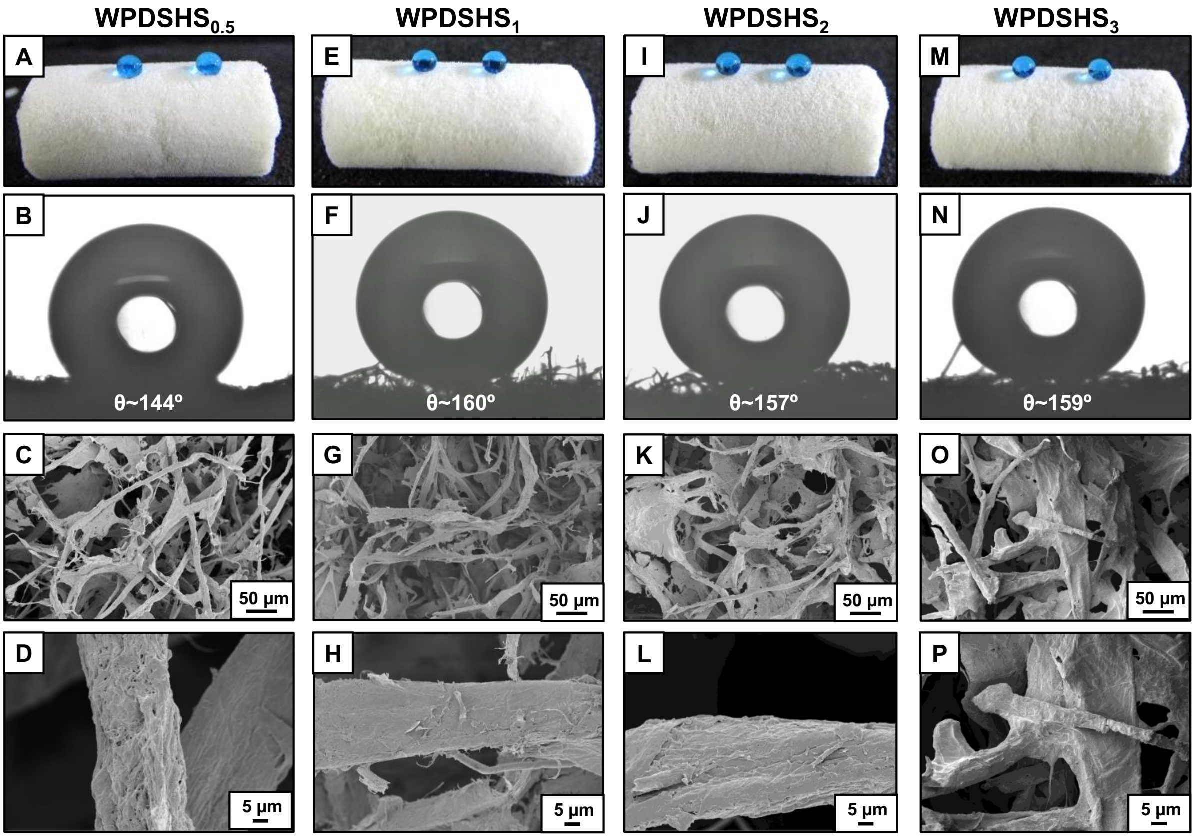



**Figure S7.** A-N) Digital images (A, E, I, M) and water contact angle images (B, F, J, N) of the beaded water droplet on the waste paper derived sponges that was post covalently modified with octadecylamine. C-P) FESEM images at low (C, G, K, O) and high (D, H, L, P) magnifications exhibiting the morphology of the waste paper derived sponges (doped with different concentrations of binder, AEPTMS) after post covalent modification.

| Sample | Advancing Contact Angle (°) | Contact Angle Hysteresis (°) | Density ($g/cm^3$) |
|---|---|---|---|
| $WPDSHS_{0.5}$ | 146.4±1.1 | 15.3±1.5 | 0.015 ± 0.9 |
| $WPDSHS_1$ | 159.4±1.2 | 7.3±0.4 | 0.017 ± 1.2 |
| $WPDSHS_2$ | 158.7±0.6 | 7.5±0.6 | 0.018 ± 0.6 |
| $WPDSHS_3$ | 157.2±0.6 | 7.6±1.4 | 0.022 ± 1.5 |

**Table S2.** Accounting for the advancing contact angle, contact angle hysteresis and density of the beaded water droplet on the waste paper derived sponges after post covalent modification.

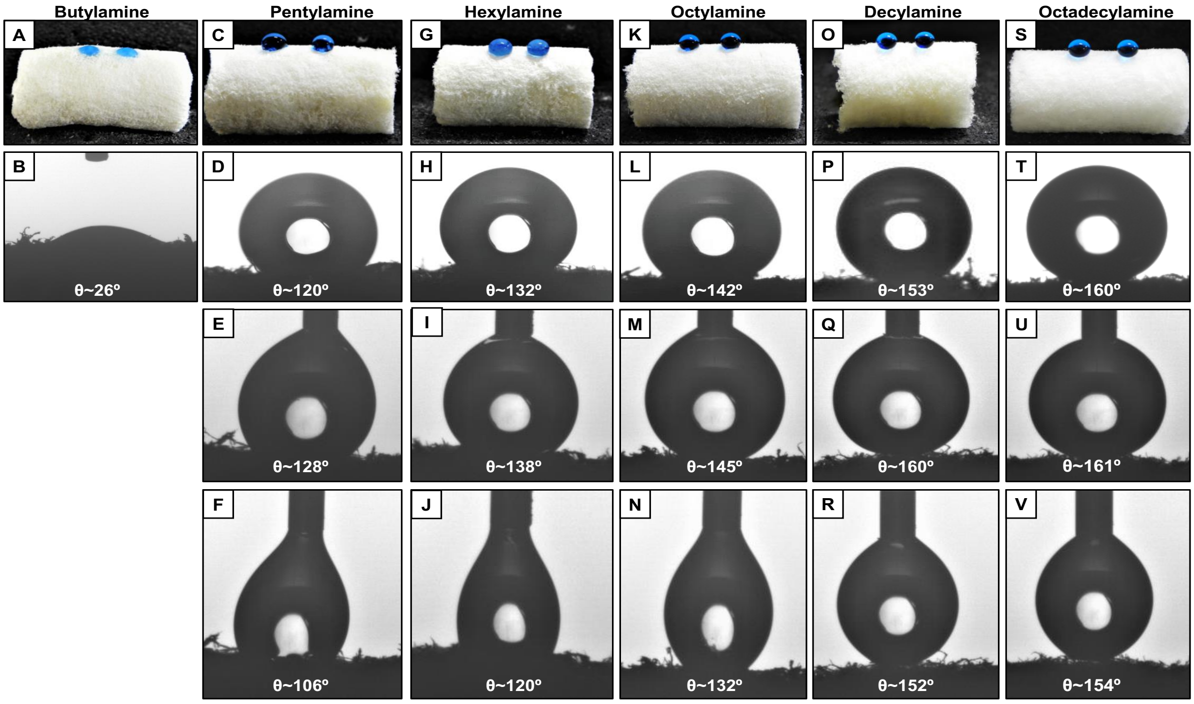



**Figure S8.** A-V) Digital images and contact angle images of the beaded water droplet on the waste paper derived sponge, $WPDS_1$ that was post covalently modified with butylamine (A-B), pentylamine (C-F), hexylamine (G-J), octylamine (K-N), decylamine (O-R) and octadecylamine (S-V).

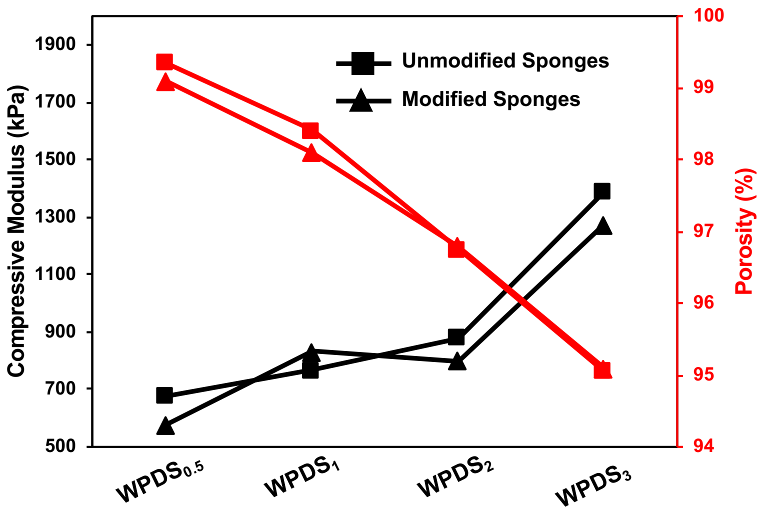



**Figure S9.** A) Plot illustrating the compressive modulus (black) and porosity (red) of the waste paper derived superhydrophobic sponges before and after post covalent modification to alter the wettability.

| Sample | Advancing Contact Angle (°) | Contact Angle Hysteresis (°) |
|---|---|---|
| **Office Paper** | 158.4 ± 0.6 | 7.5 ± 1.3 |
| **Newspaper** | 159.2 ± 1.1 | 7.3 ± 0.9 |
| **Cardboard Paper** | 157.8 ± 1.2 | 7.8 ± 1.1 |
| **Mixture of Papers** | 156.7 ± 0.8 | 8.21± 1.4 |

**Table S3** accounting for the advancing contact angle and contact angle hysteresis of the beaded water droplet on the different types of waste paper derived superhydrophobic sponges.

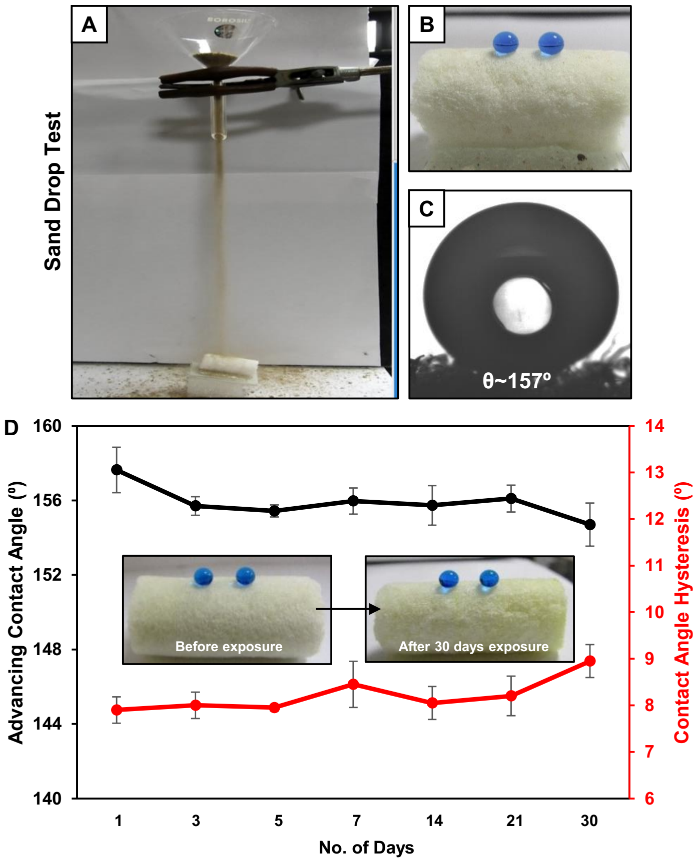



**Figure S10.** A-C) Digital image (A, B) and contact angle image (C) illustrating the sand drop test (A) followed by examination of water repellence after sand drop test (B-C) on the waste paper derived superhydrophobic sponge, $WPDSHS_1$. D) Plot accounting for the advancing contact angle (black) and contact angle hysteresis (red) of the beaded water droplet on the waste paper derived superhydrophobic sponge, $WPDSHS_1$ after 30 days exposure to UV radiation.

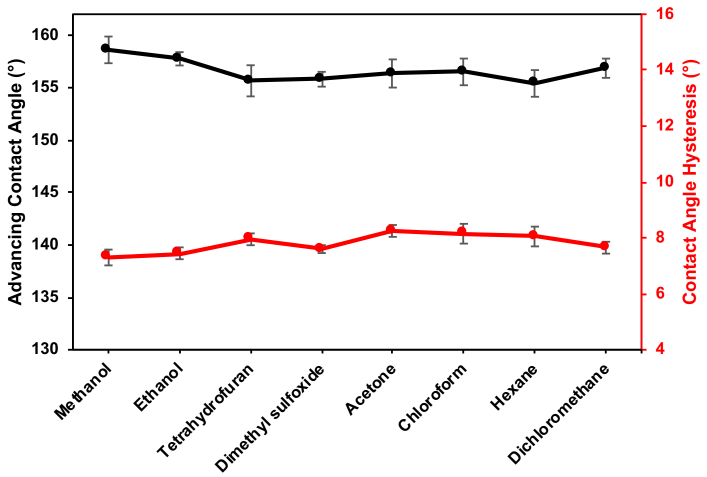



**Figure S11.** Plot accounting for the advancing contact angle (black) and contact angle hysteresis (red) of the beaded water droplet on the waste paper derived superhydrophobic sponge, $WPDSHS_1$ after 30 days exposure to different polar and non-polar organic solvents.

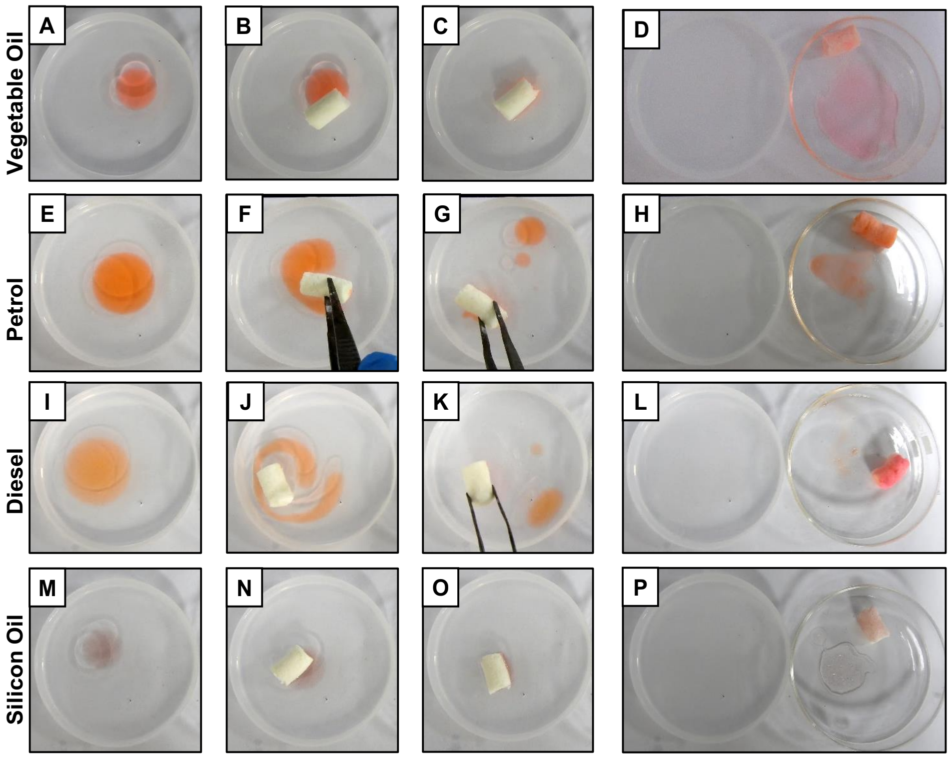



**Figure S12.** A-P) Digital images illustrating the absorption based selective separation of vegetable oil (A-D), petrol (E-H), diesel (I-L) and silicon oil (M-P) using the waste paper derived superhydrophobic sponge, $WPDSHS_1$ from an oil/water interface.

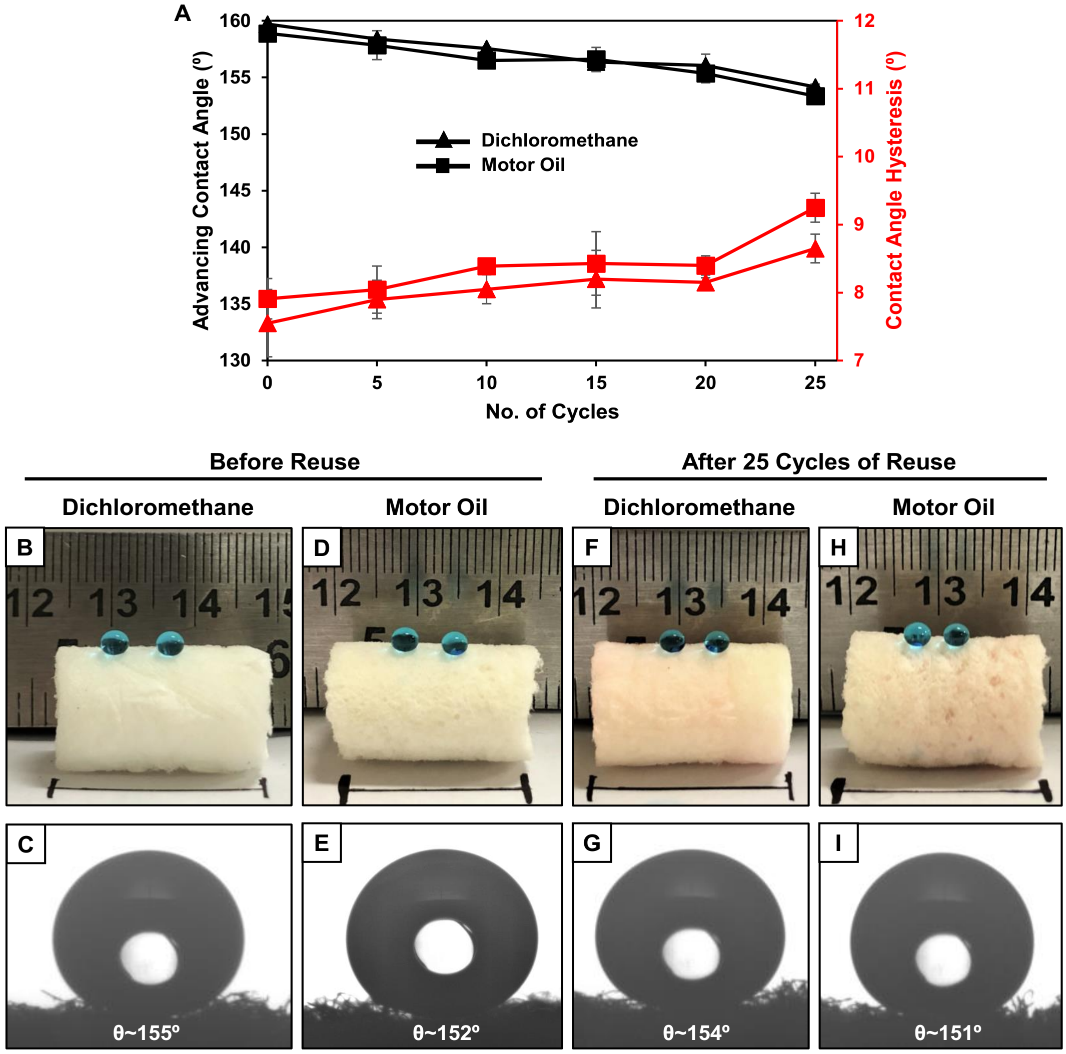



**Figure S13.** A) Plot accounting for the advancing contact angle (black) and contact angle hysteresis (red) of the beaded water droplet on the waste paper derived superhydrophobic sponge, $WPDSHS_1$ that was used repetitively for 25 times of selective absorption of both light (motor oil) and heavy (dichloromethane) oils from an oil/water mixture. B-I) Digital images (B, D, F, H) and contact angle images (C, E, G, I) of the waste paper derived superhydrophobic sponge, $WPDSHS_1$ before (B-E) and after (F-I) 25 times of use for selective absorption of light (motor oil) and heavy oils (dichloromethane) from an oil/water mixture.

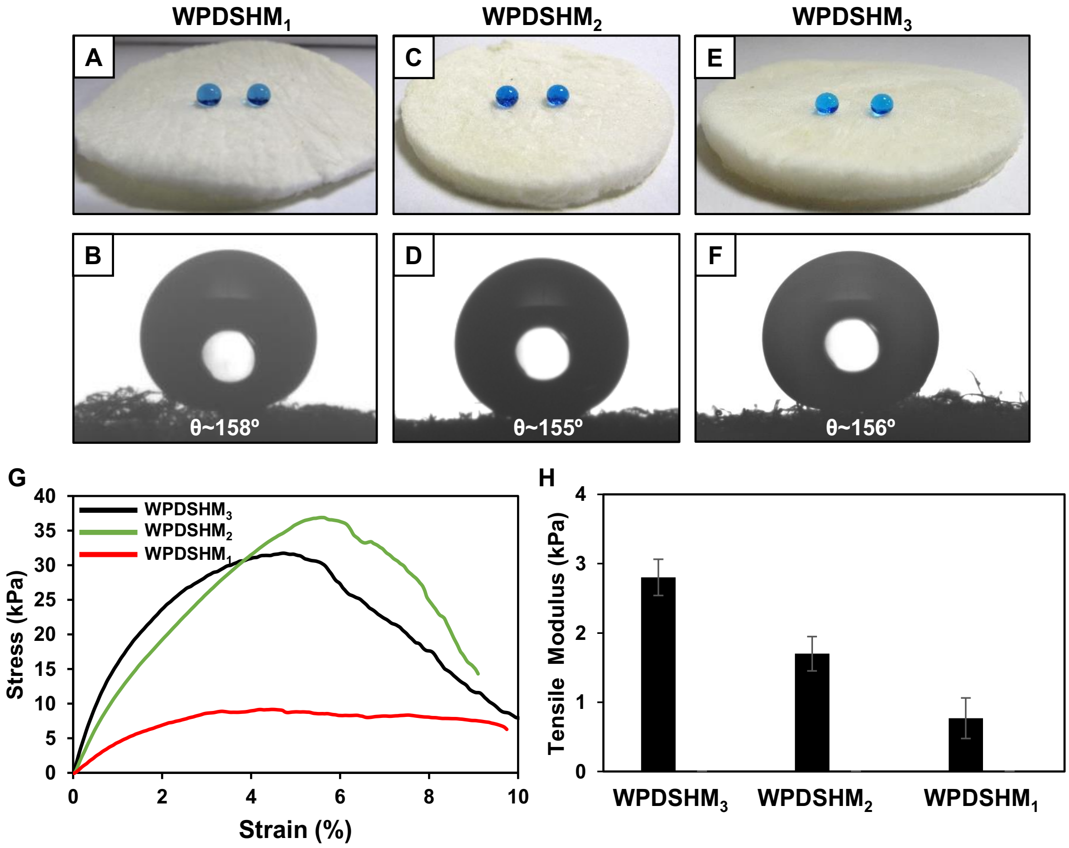



**Figure S14.** A-F) Digital images (A, C, E) and contact angle images (B, D, F) of the beaded water droplet on the waste paper derived superhydrophobic membranes that were developed using different concentrations of the silane binder (AEPTMS). G) Plot illustrating the tensile stress-strain analysis of the waste paper derived superhydrophobic membranes. H) Bar diagram accounting for the tensile modulus of the waste paper derived superhydrophobic membranes.

| Sample | Advancing Contact Angle (°) | Contact Angle Hysteresis (°) | Porosity (%) |
|---|---|---|---|
| **WPDSHM$_1$** | 158.4 ± 1.3 | 7.3 ± 0.7 | 99.3 ± 1.2 |
| **WPDSHM$_2$** | 156.6 ± 1.5 | 7.5 ± 0.8 | 97.5 ± 0.9 |
| **WPDSHM$_3$** | 157.9 ± 0.7 | 7.4 ± 1.1 | 96.1 ± 0.5 |

**Table S4** accounts for the advancing water contact, contact angle hysteresis and porosity of the waste paper derived superhydrophobic membranes that were developed with different concentrations of AEPTMS.

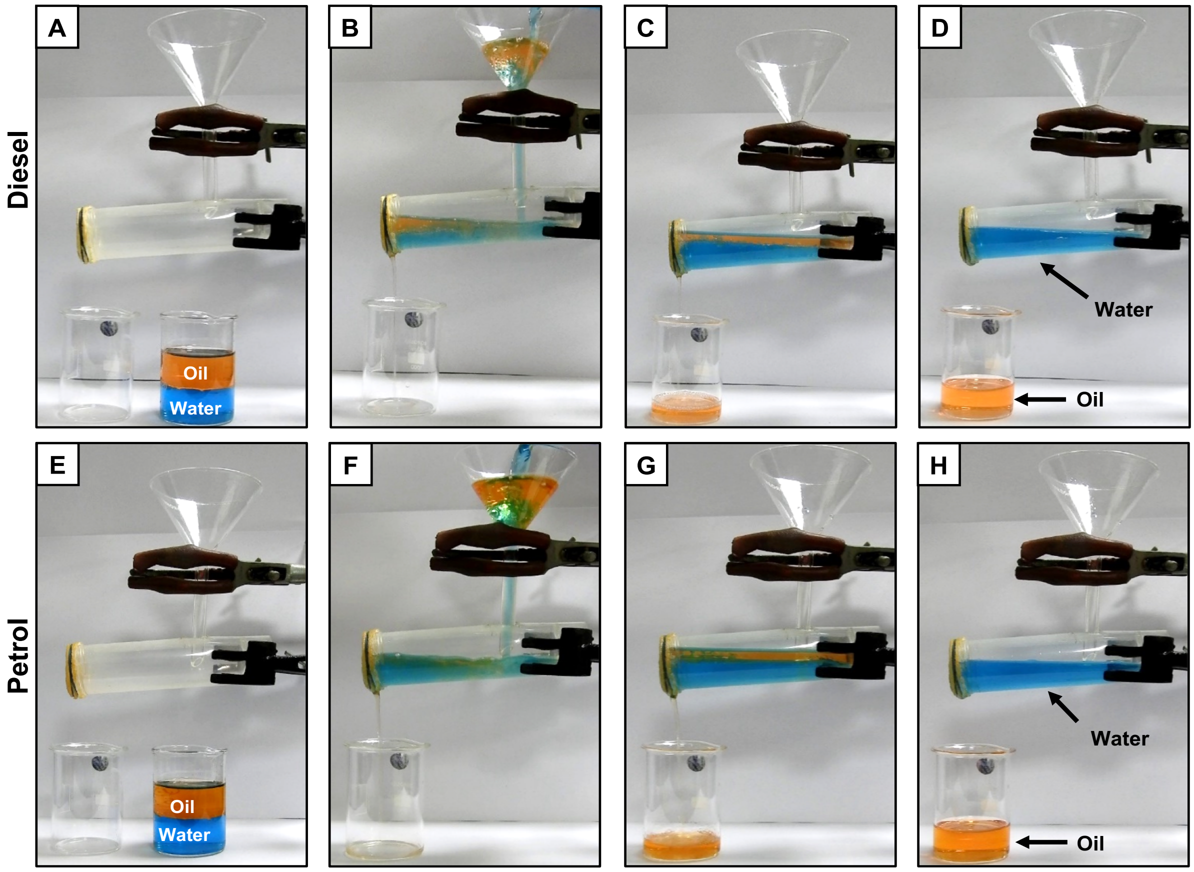



**Figure S15.** A-H) Digital images illustrating the gravity-driven filtration based selective separation of diesel (A-D) and petrol (E-H) by the waste paper derived superhydrophobic membrane, $WPDSHM_1$ from an oil/water mixture.

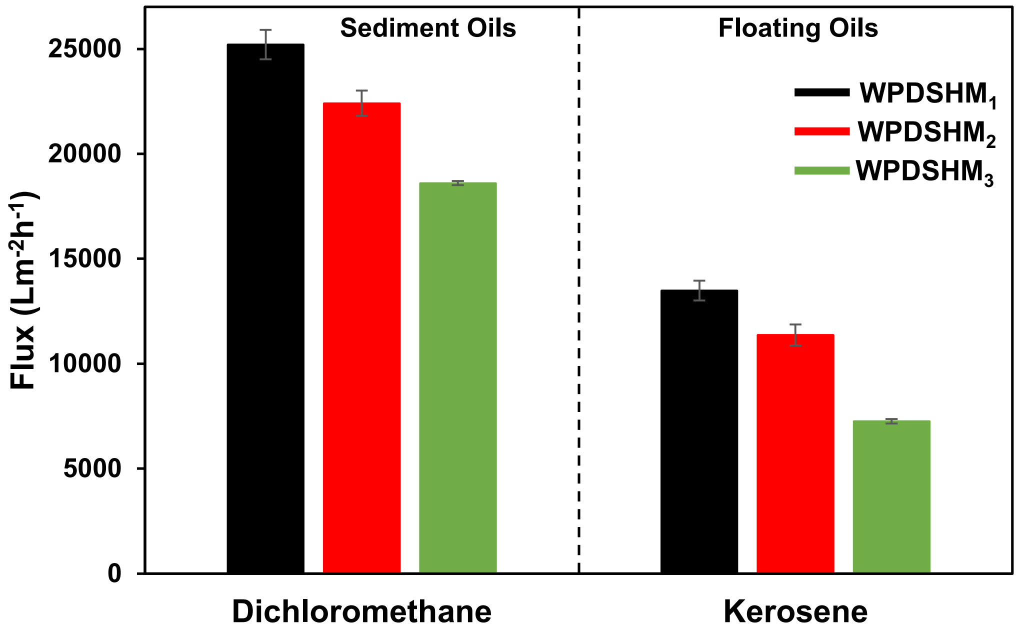



**Figure S16.** A) Plot accounting for the rate of flux of model sediment and floating oils through the waste paper derived superhydrophobic membranes with varying porosities.

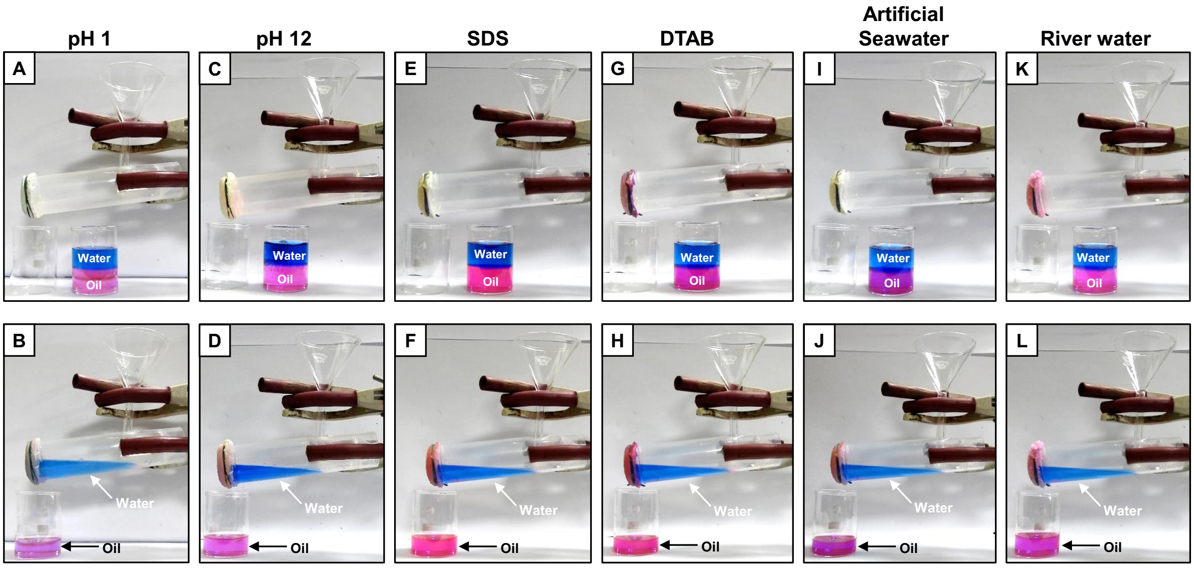



**Figure S17.** A-L) Digital images illustrating the gravity-driven filtration based selective separation of model heavy oil phase i.e. dichloromethane (dyed pink for visual inspection) by the waste paper derived superhydrophobic membrane, $WPDSHM_1$ from an oil/water mixture where the aqueous phase is chemically contaminated i.e. acidic water (A-B), basic water (C-D), surfactant contaminated (E-H), artificial sea water (I-J) and river water (K-L).

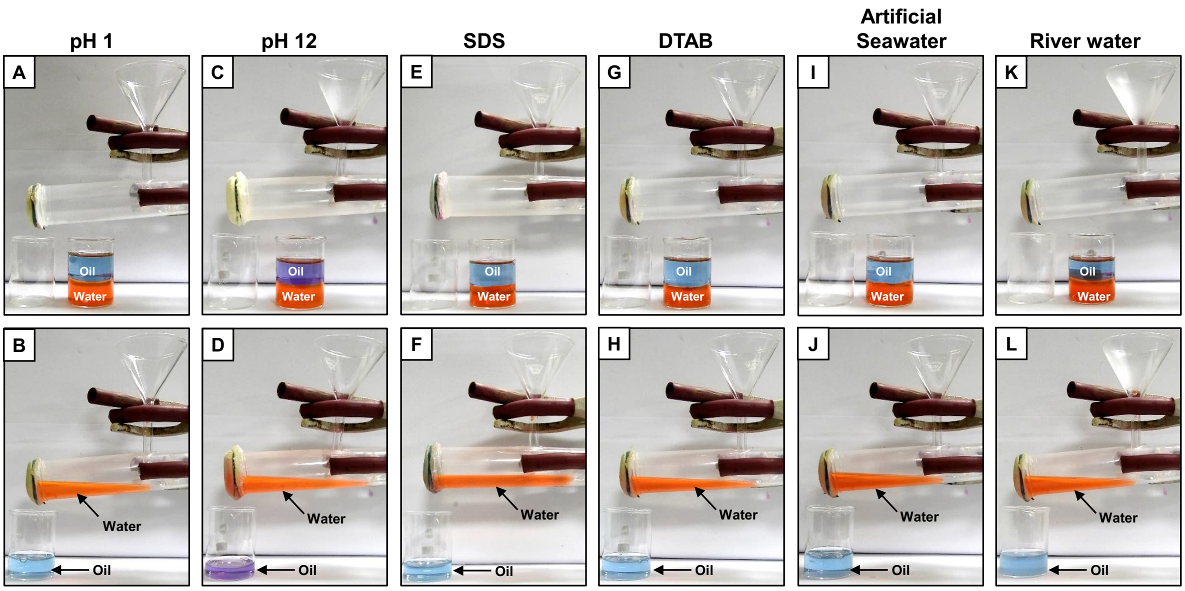



**Figure S18.** A-L) Digital images illustrating the gravity-driven filtration based selective separation of the light oil phase i.e. kerosene by the waste paper derived superhydrophobic membrane, $WPDSHM_1$ from an oil/water mixture where the aqueous phase is chemically contaminated i.e. acidic water (A-B), basic water (C-D), surfactant contaminated (E-H), artificial sea water (I-J) and river water (K-L).

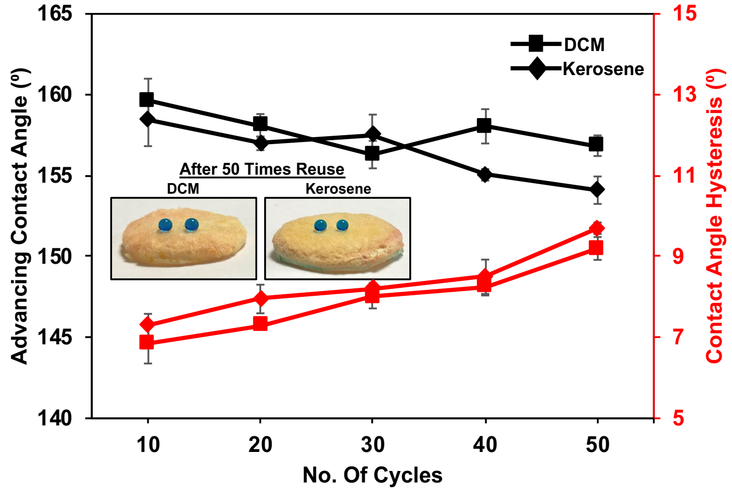



**Figure S19.** A) Plot accounting for the advancing contact angle (black) and contact angle hysteresis (red) of the beaded water droplet on the waste paper derived superhydrophobic membrane, $WPDSHM_1$ that was repetitively used (50 times) for separation of both light (kerosene) and heavy (dichloromethane) oils from an oil/water mixture. Inset images exhibit the intact water repellence property and physical integrity of the membrane after reuse.