Supporting Information

Facile Optimization of Hierarchical Topography and Chemistry on Magnetically-active Graphene Oxide Nanosheets

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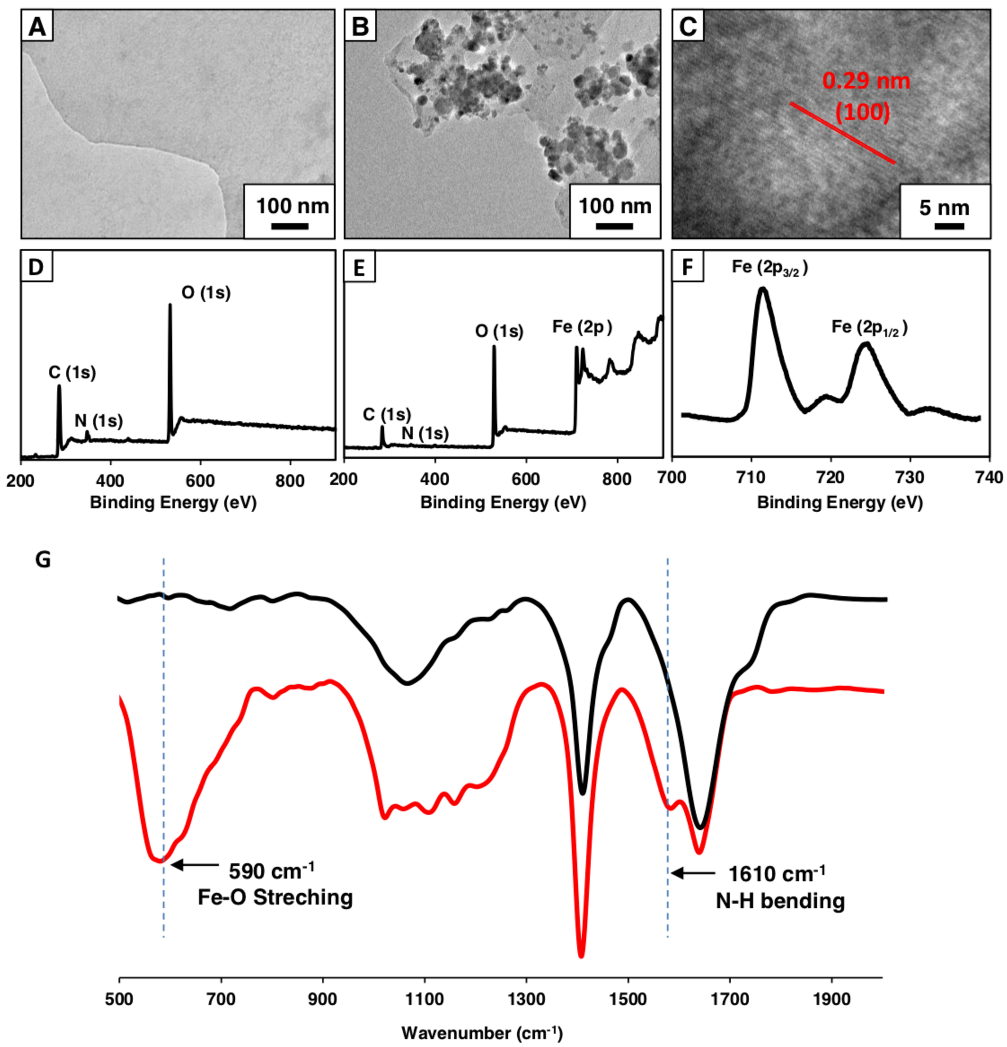

Figure S1. A-B) TEM images of nitro-graphene oxide (NGO; A) and magnetically active graphene oxide (MAGO; B). C) HRTEM images confirms the deposition of Fe_3O_4 nanoparticles on MAGO. D-F) XPS spectra of NGO (D) and MAGO (E-F). G) FTIR spectra of NGO (black) and MAGO (red).

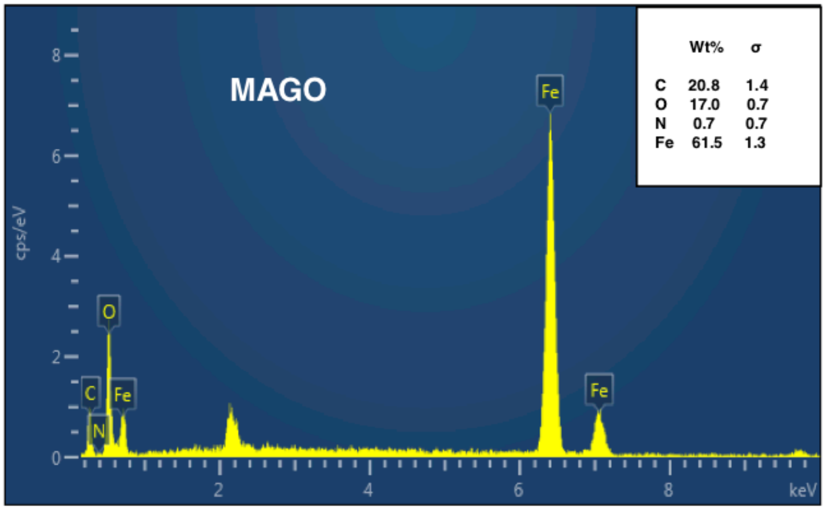

Figure S2. EDX spectra of MAGO.

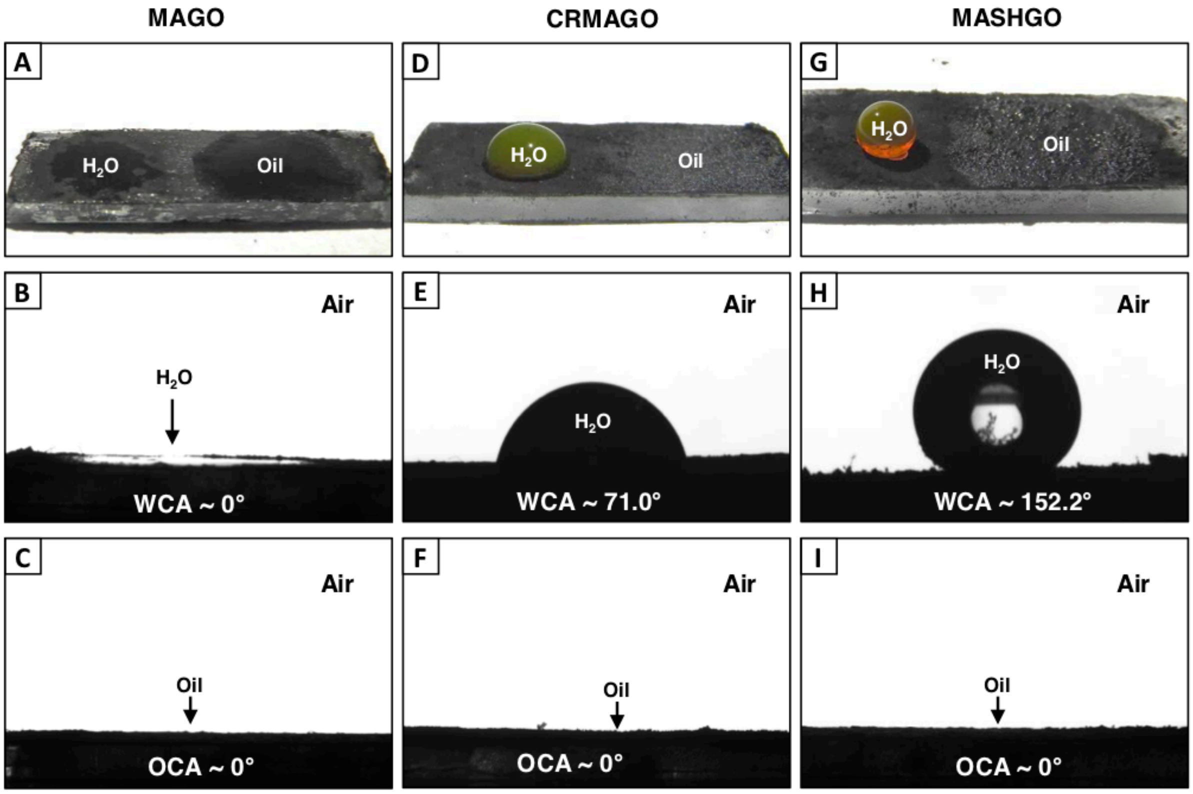

Figure S3. A-I) Digital images (A,D,G) and static contact angle images (B-I) of beaded water (in air) and oil (in air) droplets on MAGO (A-C), CRMAGO (D-F), MASHGO (G-I).

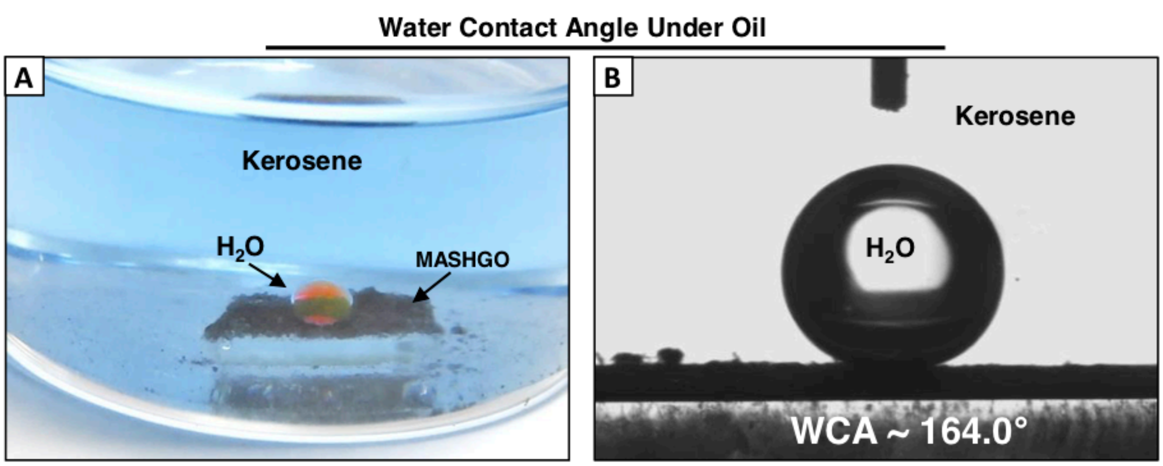

Figure S4. A-B) Digital image (A) and static water contact angle image (B) of beaded water droplets on MASHGO, under oil phase (kerosene).

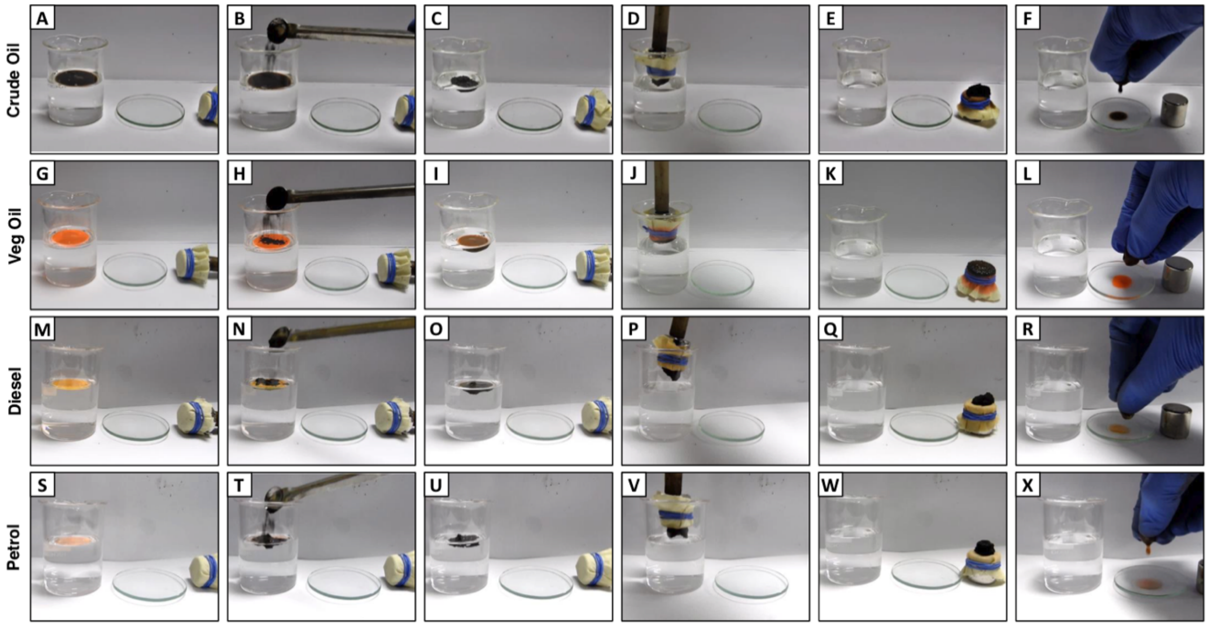

Figure S5. A-X) Digital images demonstrating the separation and collection of different floating oils (crude oil (A-F), veg oil (G-L) and diesel (M-R), petrol (S-X)) from water/air interface by using MASHGO and external magnet, where floating oil phases (A, G, M, S) were exposed to MASHGO (B,H,N,T), and immediately respective oil phases were absorbed by MASHGO (C,I,O,U). D-F, J-L, P-R, V-X) The oil-absorbed MASHGO was collected using fabric wrapped external magnet.

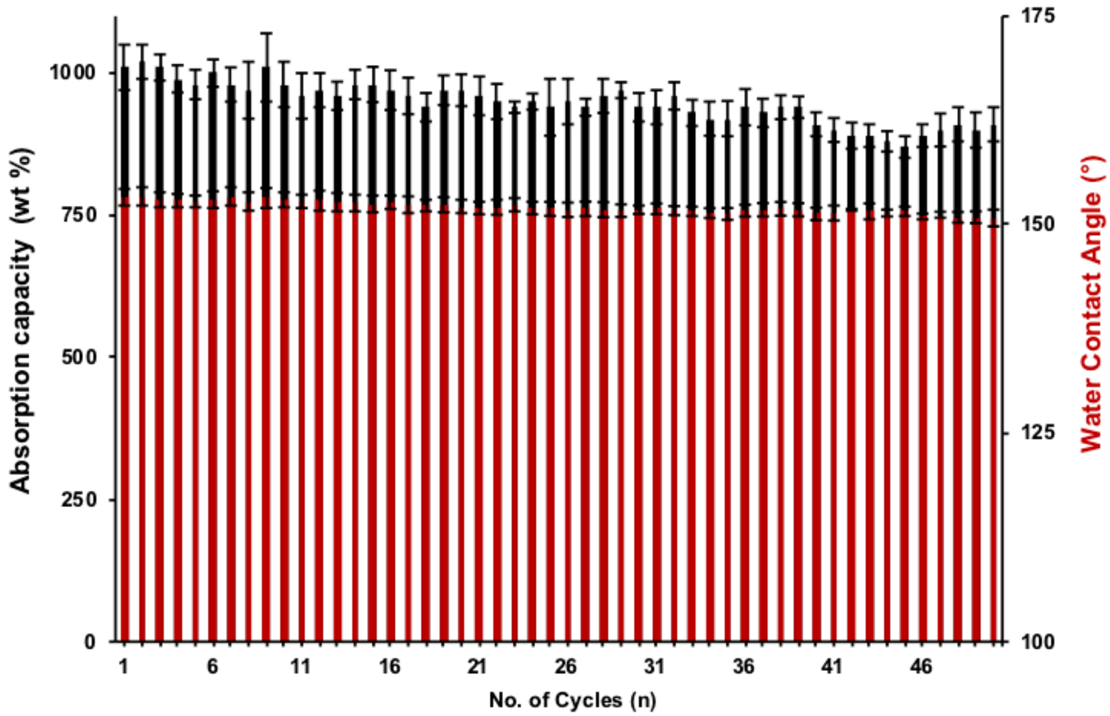

Figure S6. Bar graphs accounting crude oil absorption capacity(black) and water wettability (red) of MASHGO after the repetitive (up to 50 cycles) uses.

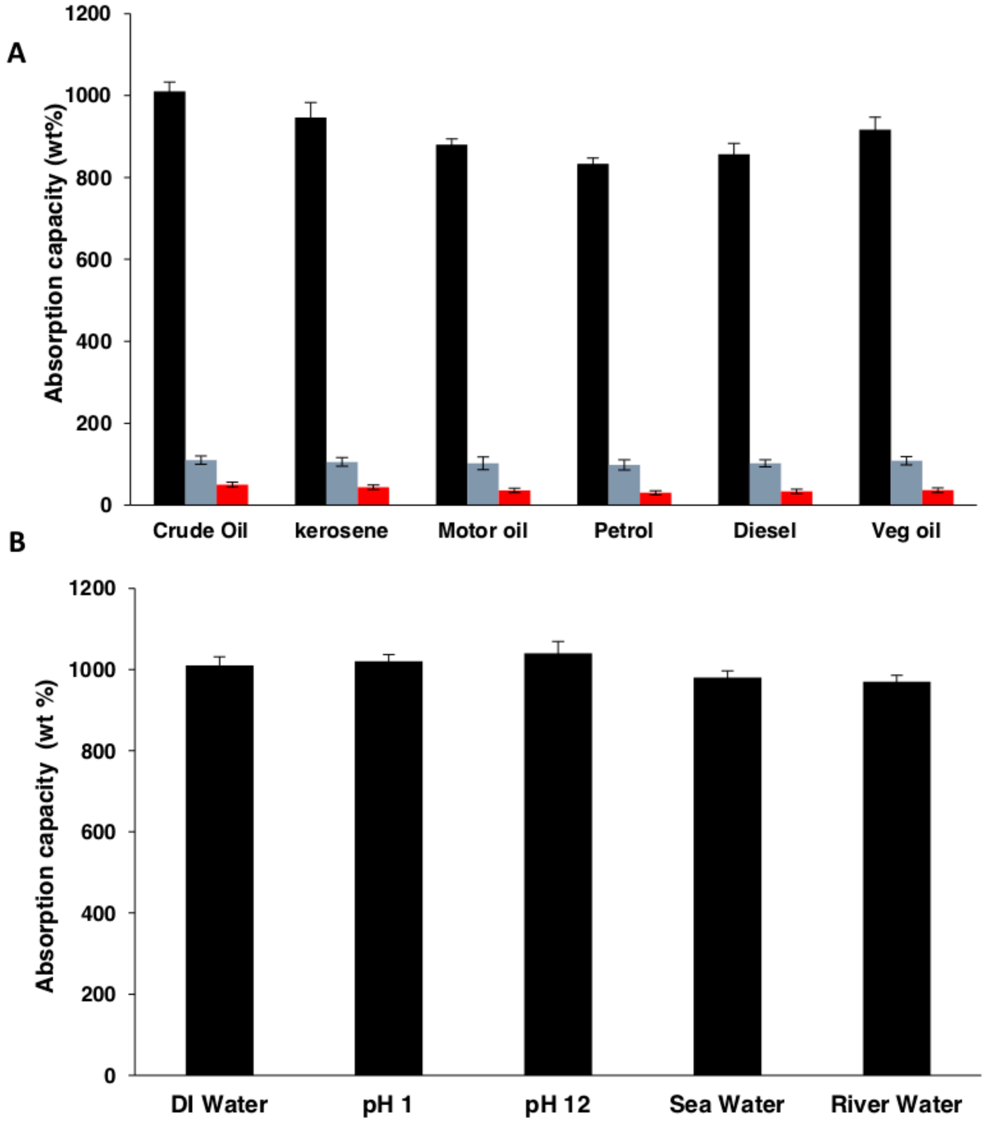

Figure S7. A) The plot comparing the oil-absorption capacity (wt.%) for various oils by MAGO (red), CRMAGO (blue), MASHGO (black). B) The plot accounts the crude oil-absorption capacity (wt. %) in presence of different and complex aqueous phases, including DI water, highly acidic, alkaline, artificial seawater to river water.

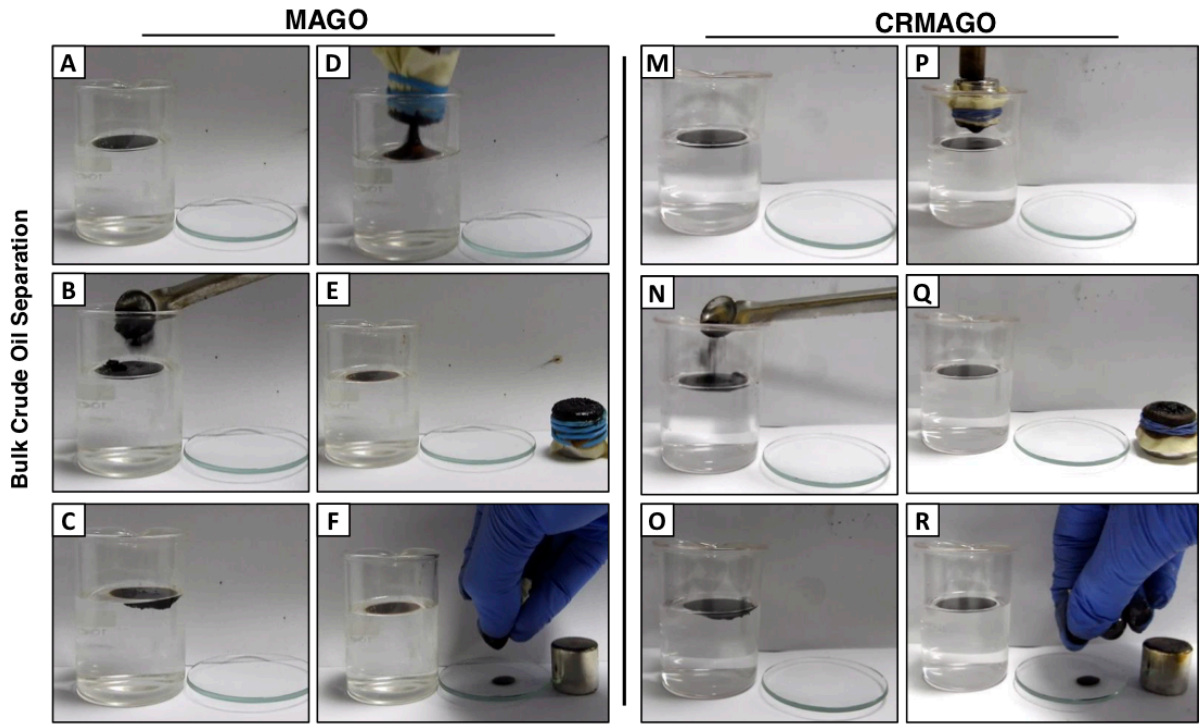

Figure S8. A-R) Digital images describing the failure in separating the floating crude oil from water/air interface by using MAGO (A-F) and CRMAGO (M-R).

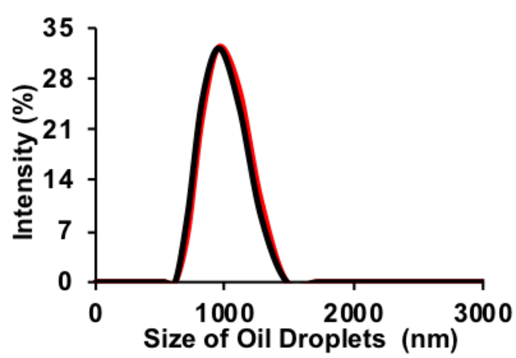

Figure S9. DLS study accounting the stability of 2% crude-oil-in-water emulsion(black line) after 24 h (red line) waiting period.

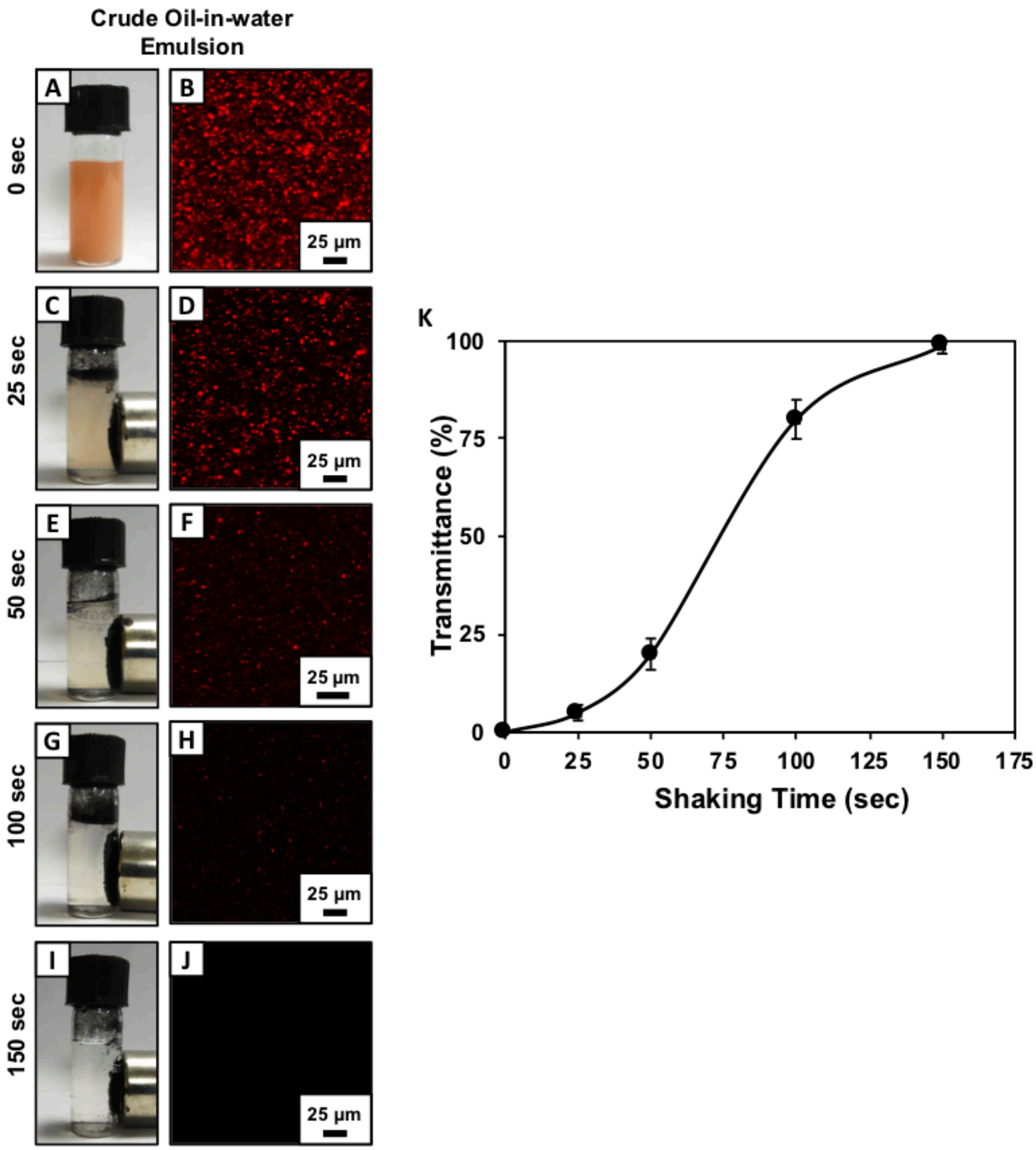

Figure S10. A-J) Digital images (A-I) and fluorescence microscopy images (B-J) accounting the progress of separation of crude oil-in-water emulsion by MASHGO. K) Plots showing the change in transmittance (%) of the crude oil-in-water emulsion solution with time of the separation process.

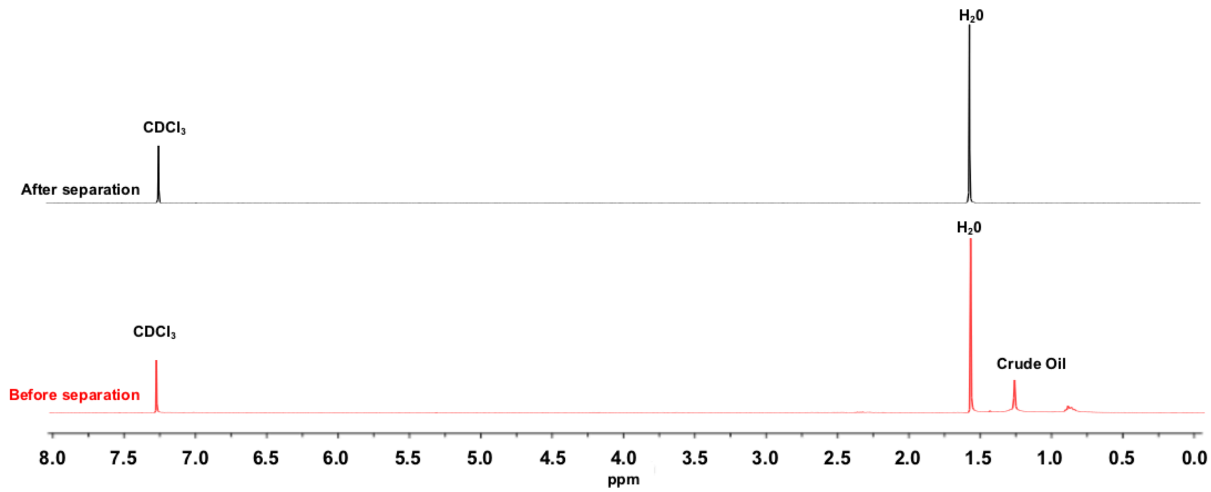

Figure S11. ^{1}H-NMR spectra in $CDCl_3$ of crude oil-in-water emulsion solution before (red) and after performing the oil/water separation (black) process using MASHGO.

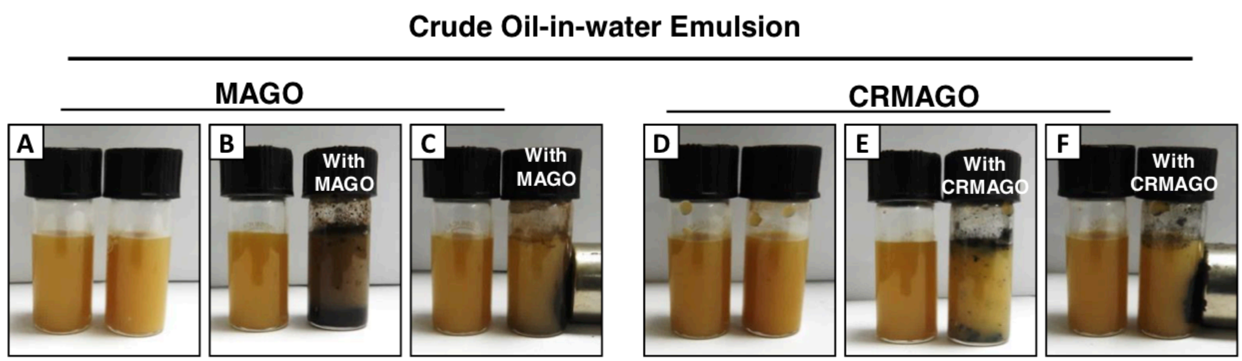

Figure S12. A-F) Digital images illustrating the failure of crude oil-in-water emulsion separation by both MAGO (A-C) and CRMAGO (D-F).

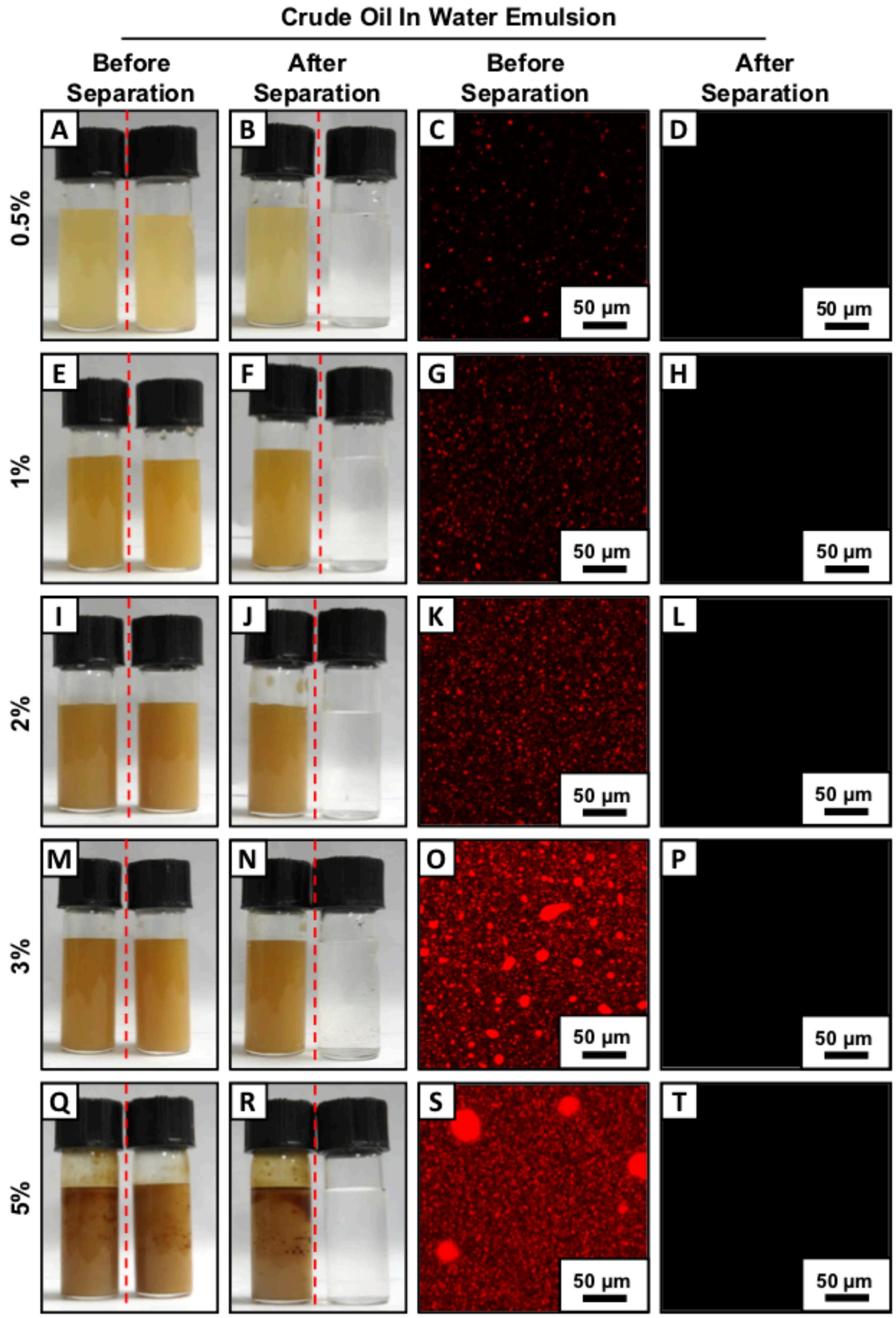

Figure S13. A-T) Digital images (A-B, E-F, I-J, M-N, Q-R) and fluorescence microscopy images (C-D,G-H, K-L, O-P, S-T) of different v/v % (0.5%; A-D, 1%; E-H, 2%; I-L, 3%; M-P, 5%; Q-T) of crude oil-in-water emulsion before (A,C,E,G,I,K,M,O,Q,S) and after (B,D,F,H,J,L,N,P,R,T,) performing oil/water separation using MASHGO. There are two separate vials of the same crude-oil-in-water emulsion solution were agitated both in presence (right-side vial) or absence (left-side vial) of added MASHGO for separating respective emulsion solutions.

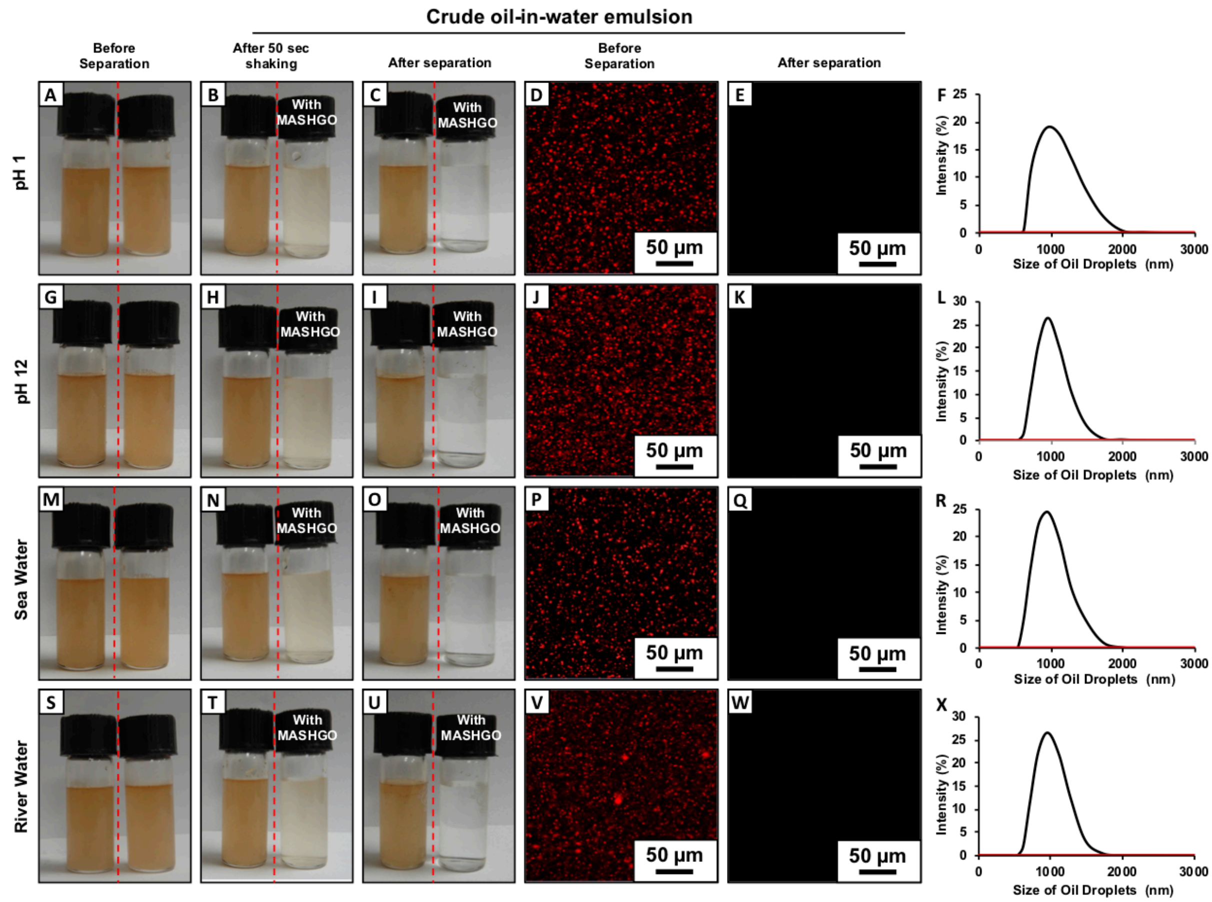

Figure S14. A-C, G-I, M-O, S-U) Digital images of crude oil-in-water emulsion before (A,G,M,S), during (50 sec shaking; B,H,N,T) and after performing the oil/water separation (C,I,O,U) using MASHGO in different practically relevant conditions, including highly acidic (pH 1; A-C) water, highly alkaline (pH 12; G-I) water, sea water (M-O) and river water (S-U). There are two separate vials of the same crude-oil-in-water emulsion solution in presence (right-side vial) or absence (left-side vial) of added MASHGO. D-E, J-K, P-Q, V-W) Fluorescence microscope images of crude oil-in-water emulsion before (D,J,P,V) and after separation (E,K,Q,W) of oil/water using MASHGO in different aqueous environment (pH 1 (D-E), pH 12 (J-K), sea water (P-Q), river water(V-W)). F,L,R,X) Accounting DLS study on crude oil-in-water emulsion before (black) and after oil/water separation (red) by MASHGO in different aqueous environment (pH 1 (F), pH 12 (L), sea water (R), river water(X)).

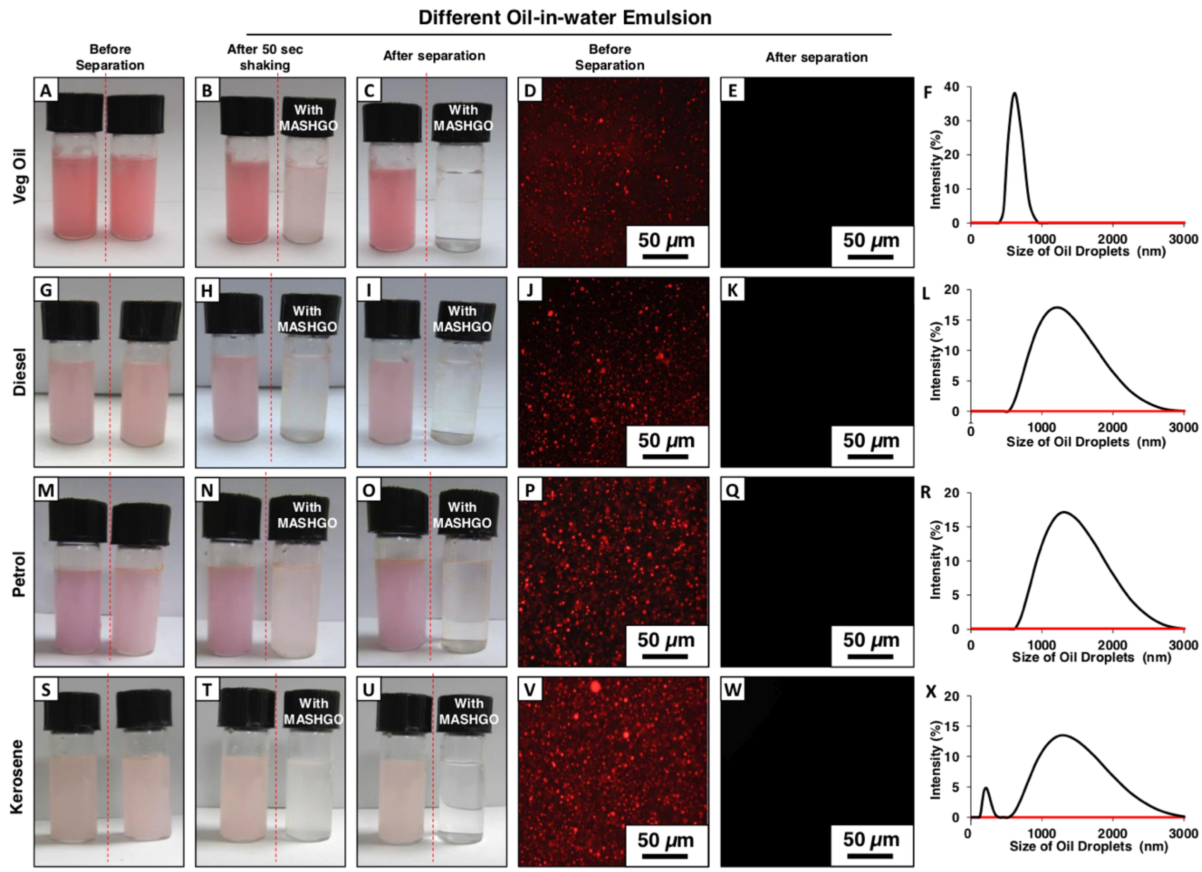

Figure S15. A-X) Digital images (A-C, G-I, M-O, S-U), fluorescence microscope images (D-E, J-K, P-Q, V-W) and DLS plots (F,L,R,X) accounting successful separation of oil-in-water emulsions that were prepared using various oils (veg oil: A-F; Diesel: G-L; petrol: M-R; kerosene: S-X), through selective absorption of respective oil droplets by MASHGO. The left-sided and right-sided vials of the dotted red lines denoted crude oil-in-water emulsion without and with MASHGO, respectively. D-E, J-K, P-Q, V-W) Fluorescence microscope images of various oil-in-water emulsion before separation (D,J,P,V) and after separation (E,K,Q,W) by MASHGO. F,L,R,X) DLS plots of various oil-in-water emulsions before (black) and after (red) oil/water separation by MASHGO.

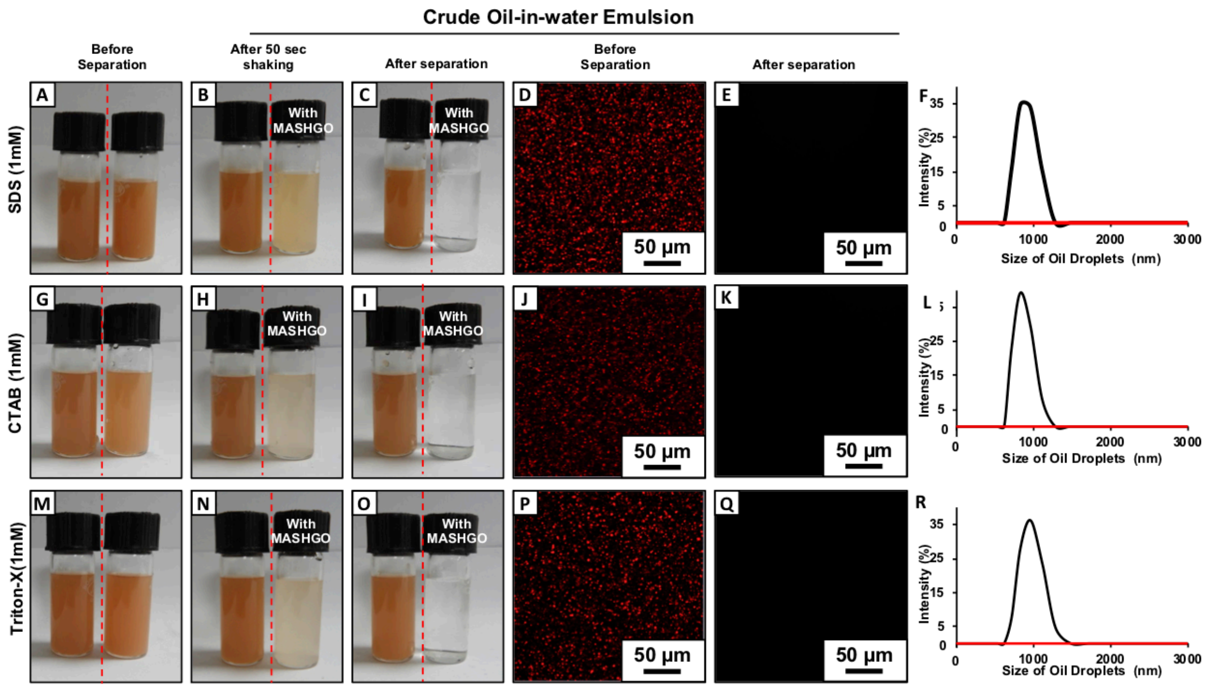

Figure S16. A-C, G-I, M-O) Digital images of crude oil-in-water emulsion before (A,G,M), during (after 50 sec; B,H,N,) and after (C,I,O) oil/water separation by MASHGO in presence of anionic (SDS; A-F), cationic (CTAB; G-L) and neutral (Triton-X; M-R) surfactants. The left-sided and right-sided vials of the dotted red lines denoted crude oil-in-water emulsions without and with MASHGO, respectively. D-E, J-K, P-Q) Fluorescence microscope images of crude oil-in-water emulsion before (D,J,P) and after (E,K,Q) oil/water separation by MASHGO in presence of different surfactants. F,L,R) DLS plots of crude oil-in-water emulsion before (black) and after (red) oil/water separation by MASHGO in presence of different surfactant.

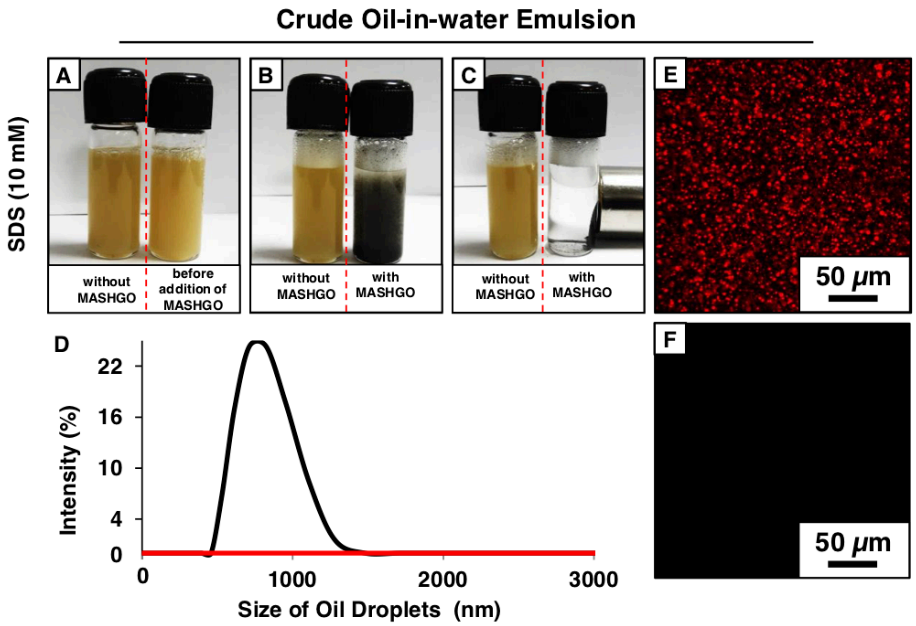

Figure S17. A-C) Digital images of crude oil-in-water emulsion before (A), during (after 50 sec; B) and after (C) oil/water separation by MASHGO in presence of anionic (SDS; A-C) surfactants having concentration (10 mM) above its CMC . The left-sided and right-sided vials of the dotted red lines denoted crude oil-in-water emulsions without and with MASHGO, respectively. D) DLS plots of crude oil-in-water emulsion before (black) and after (red) oil/water separation by MASHGO in presence of the surfactant. E-F) Fluorescence microscope images of crude oil-in-water emulsion before (E) and after (F) oil/water separation by MASHGO in presence of SDS (having concentration of 10mM).

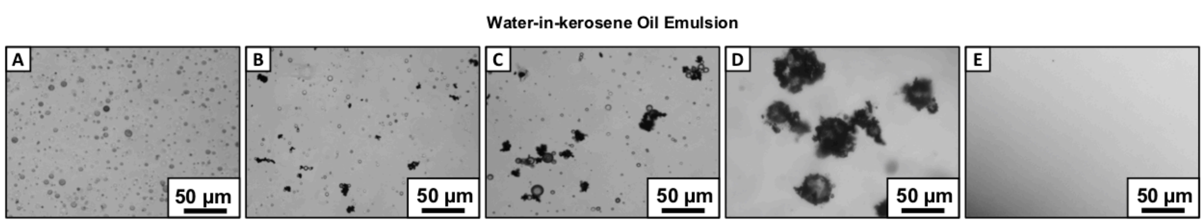

Figure S18. A-E) Bright-field microscopy images of water-in-oil (kerosene) emulsion in presence of MASHGO. With agitation, the water droplets are started coalescence on MASHGO in the kerosene phase and finally formed a 'Pickering-type' droplet, where the aqueous phase is wrapped with MASHGO in kerosene. On application of external magnet, the 'Pickering-type' droplets are separated and provided a water-free fuel phase (E).

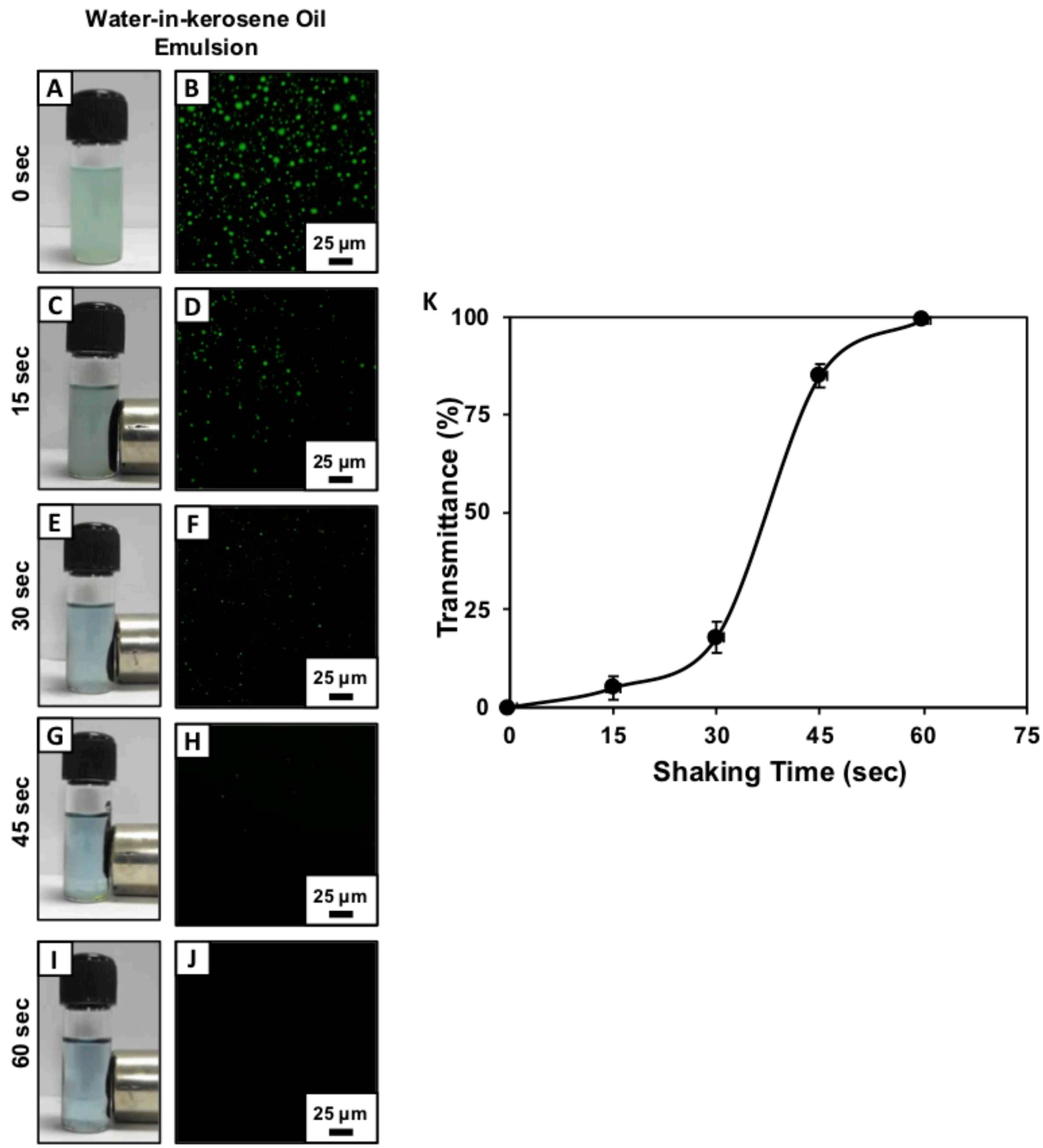

Figure S19. A-J) Digital images (A-I) and fluorescence microscopy images (B-J) depicting the separation of water-in-oil emulsion by MASHGO. K) Plots showing the change in transmittance (%) of the crude water-in-oil emulsion with time of the separation process. The oil phase was kerosene.

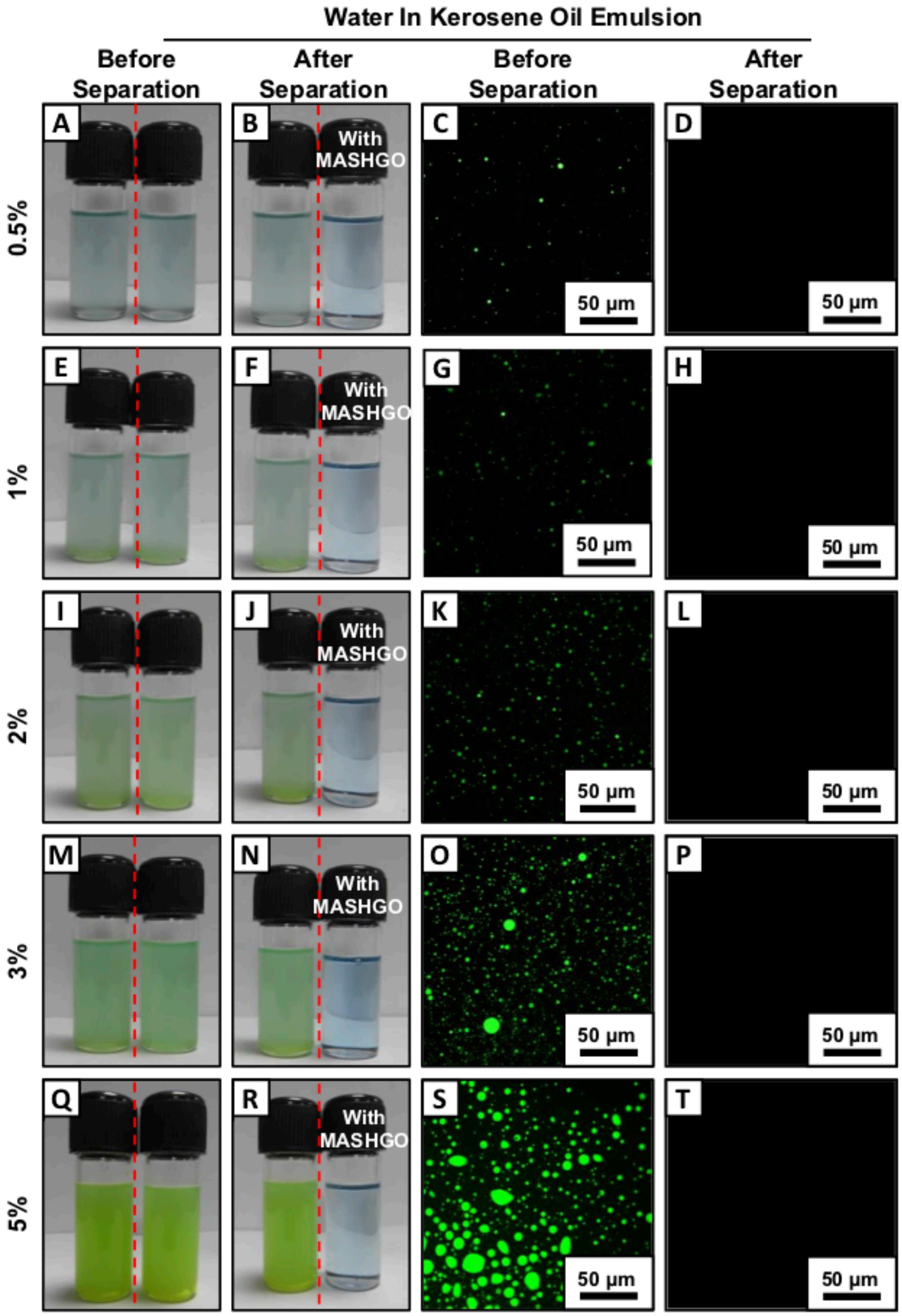

Figure S20. A-T) Digital images (A-B, E-F, I-J, M-N, Q-R) and fluorescence microscopy images (C-D,G-H, K-L, O-P, S-T) of different v/v % (0.5%; A-D, 1%; E-H, 2%; I-L, 3%; M-P, 5%; Q-T) water-in-kerosene oil emulsions, before (A,C,E,G,I,K,M,O,Q,S) and after (B,D,F,H,J,L,N,P,R,T,) oil/water separation by MASHGO. Aqueous phases were labeled with water soluble fluorescein dye.

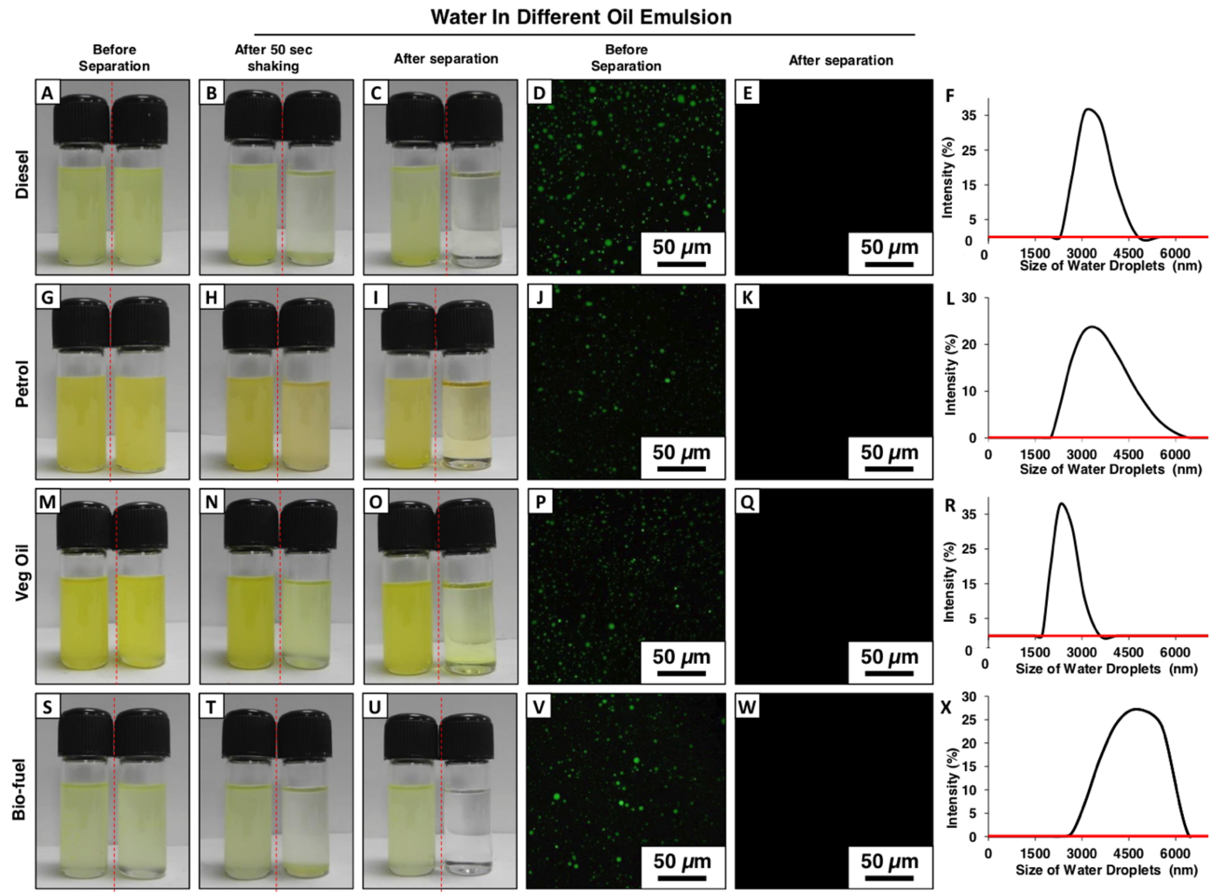

Figure S21. A-C, G-I, M-O, S-U) Digital images of water-in-oil emulsions that were prepared using various oils (Diesel; A-C, petrol; G-I, Veg oil; M-O, bio-fuel; S-U), before (A,G,M,S), during (after 50 sec; B,H,N,T) and after (C,I,O,U) oil/water separation by MASHGO. The left-sided and right-sided vials of the dotted red lines denoted water-in-oil emulsion without and with MASHGO, respectively. D-E, J-K, P-Q, V-W) Fluorescence microscope images of various water-in-oil emulsions ((Diesel; D-E, petrol; J-K, Veg oil; P-Q, bio-fuel; V-W)) before (D,J,P,V) and after oil/water separation (E,K,Q,W) by MASHGO. F,L,R,X) DLS plots of various water-in-oil emulsion (veg oil; F, Diesel; L, petrol; R, kerosene; X) before (black) and after (red) separation by MASHGO (Diesel; F, petrol; L, Veg oil; R, bio-fuel; X).

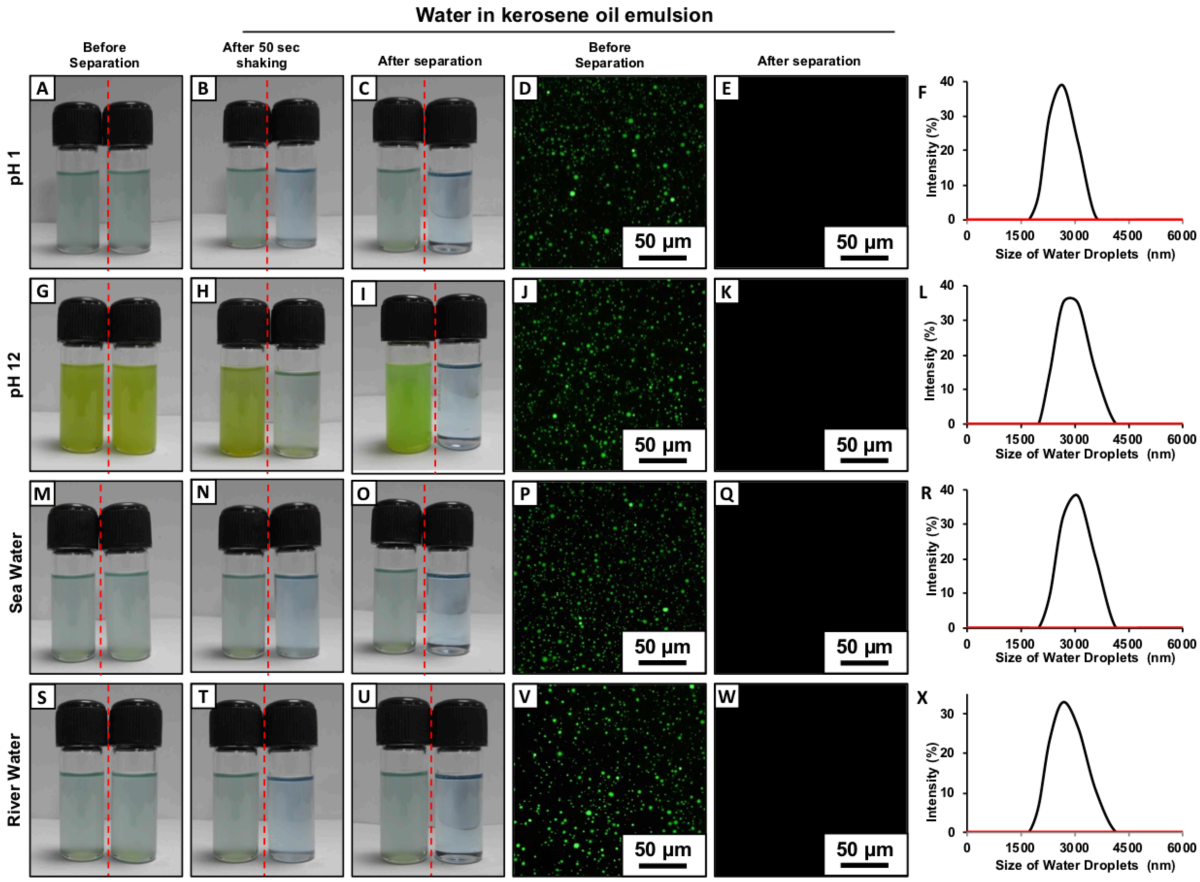

Figure S22. A-C, G-I, M-O, S-U) Digital images of water-in-kerosene oil emulsions before (A,G,M,S), during (after 50 sec; B,H,N,T) and after (C,I,O,U) oil/water separation by MASHGO in presence of different aqueous environment (pH 1 (A-C), pH 12 (G-I), sea water (M-O), river water(S-U)). There are two separate vials of the same water-in-kerosene oil emulsion solution in presence (right-side vial) or absence (left-side vial) of added MASHGO. D-E, J-K, P-Q, V-W) Fluorescence microscope images of different water-in-kerosene oil emulsions before (D,J,P,V) and after (E,K,Q,W) oil/water separation by MASHGO. F,L,R,X) DLS plots of different water-in-kerosene oil emulsions before (black) and after (red) oil/water separation by MASHGO.

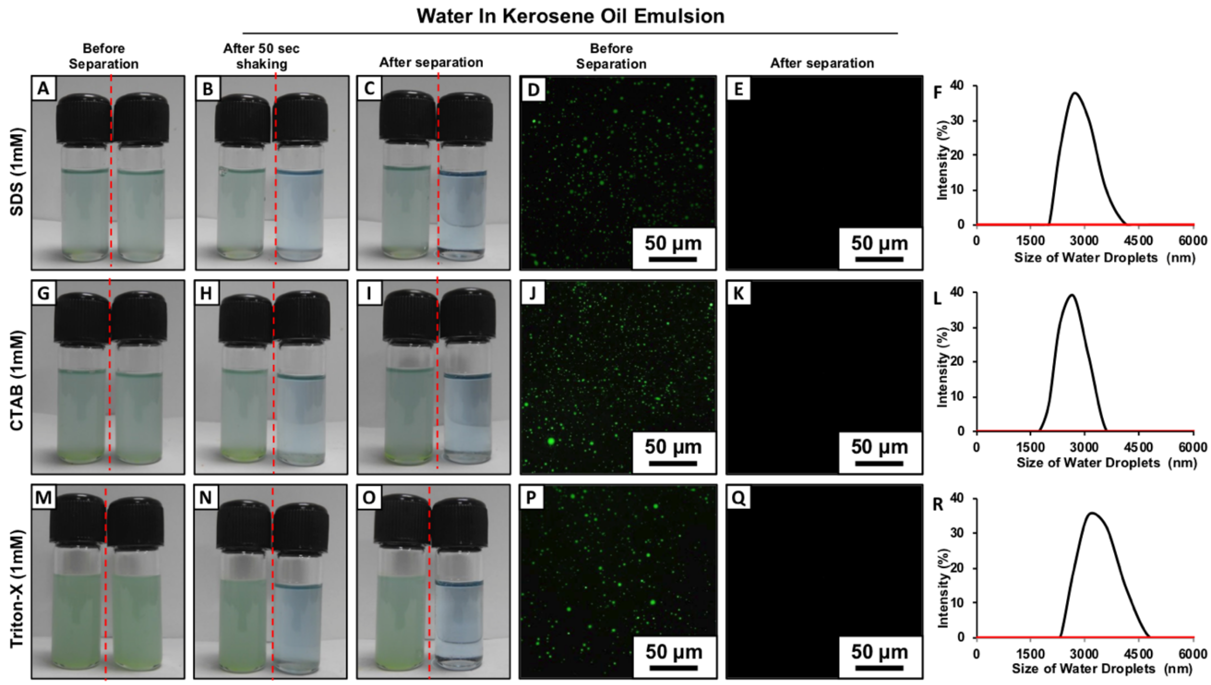

Figure S23. A-C, G-I, M-O) Digital images of water-in-kerosene oil emulsion before (A,G,M), during (after 50 sec, B,H,N,) and after (C,I,O) oil/water separation by MASHGO in presence of different surfactants (SDS; A-F, CTAB; G-L, Triton-X; M-R). The left-sided and right-sided vials of the dotted red lines denoted water-in-kerosene oil emulsion without and with MASHGO. D-E, J-K, P-Q) Fluorescence microscope images of water-in-kerosene oil emulsion before (D,J,P) and after (E,K,Q) oil/water separation by MASHGO in presence of different surfactants (SDS; D-E, CTAB; J-K, Triton-X; P-Q). F,L,R) DLS plots of water-in-kerosene oil emulsion before (black) and after (red) oil/water separation by MASHGO in presence of different surfactant.(SDS; F, CTAB; L, Triton-X; R).