

Supporting Information

Degradable Sugar-based Magnetic Hybrid Nanoparticles for Recovery of Crude Oil from Aqueous Environments

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Materials

The bicyclic glucose-based carbonate monomer, methyl-2,3-*O*-ethyloxycarbonyl-4,6-*O*-carbonyl- α -D-glucopyranoside (GC) and Grubbs' G3 catalyst were synthesized according to previously-published procedures.^{1,2} α -Methoxy- ω -amino polyethylene glycol (PEG) was purchased from Rapp Polymere GmbH (Tübingen, Germany). 3-(3-Dimethylaminopropyl)-1-ethyl-carbodiimide hydrochloride (EDC hydrochloride) and 1,5,7-triazabicyclo[4.4.0]dec-5-ene (TBD) were purchased from TCI America (Portland, OR), and TBD was degassed and stored in a glovebox under Ar atmosphere. Other chemicals and reagents were purchased from Sigma-Aldrich, Co. (St. Louis, MO) and were used as received, unless otherwise noted. Tetrahydrofuran (THF), dichloromethane (DCM), and *N,N*-dimethylformamide (DMF) were purified by passage through a solvent purification system (J. C. Meyer Solvent Systems, Inc., Laguna Beach, CA). Nanopure water (18 M Ω -cm) was obtained from a Milli-Q water filtration system (Millipore Corp, USA). Dialysis membrane tubing with a molar mass cut off (MWCO) of 6-8 kDa was purchased from Spectrum Laboratories, Inc. (Rancho Dominguez, CA) and soaked for 5 min in nanopure water at room temperature (rt) before use. Column chromatography was performed on a CombiFlash Rf4x (Teledyne ISCO) with RediSep Rf columns (Teledyne ISCO).

Instrumentation

¹H NMR and ¹³C NMR spectra were recorded on Varian Inova 500 spectrometer (Varian, Inc., Palo Alto, CA) interfaced to a UNIX computer using the VnmrJ software. Chemical shifts for ¹H NMR and ¹³C NMR signals were referenced to the solvent resonance frequencies. Fourier transform infrared (FT-IR) spectra were recorded on an IR Prestige 21 system, equipped with a diamond attenuated total reflection (ATR) lens (Shimadzu Corp., Japan), and analyzed using IRsolution v. 1.40 software.

Size exclusion chromatography (SEC) eluting with tetrahydrofuran (THF) was performed on a Waters Chromatography, Inc. (Milford, MA) system equipped with an isocratic pump (model 1515), a differential refractometer (model 2414), and column set comprised of a PLgel 5 μ m guard column (50 \times 7.5 mm), a PLgel 5 μ m Mixed C column (300 \times 7.5 mm, Agilent Technologies) and two Styragel[®] columns (500 Å and 104 Å, 300 \times 7.5 mm, Waters Chromatography, Inc.). The system was operated at 40 °C with a flow rate of 1 mL/min. Data were analyzed using Breeze software from Waters Chromatography, Inc. (Milford, MA). Molar masses were determined relative to polystyrene standards (580–3,250,000 Da) purchased from Polymer Laboratories, Inc.

(Amherst, MA). Polymer solutions were prepared at a concentration of ca. 3 mg/mL with 0.05 vol% toluene as the flow rate marker; an injection volume of 200 μ L was used.

Glass transition temperatures (T_g) were measured by differential scanning calorimetry (DSC) on a Mettler-Toledo DSC3/700/1190 (Mettler-Toledo, Inc., Columbus, OH) under a nitrogen gas atmosphere. Measurements were performed with a heating and cooling rate of 10 $^{\circ}$ C/min, and three heating and cooling cycles were conducted. Measurements were analyzed using Mettler-Toledo STAR^e v. 15.00a software. The T_g was taken as the midpoint of the inflection tangent of the third heating scan.

Thermogravimetric analysis (TGA) was performed under N₂ atmosphere using a Mettler-Toledo TGA2/1100/464, with a heating rate of 10 $^{\circ}$ C/min. Data were analyzed using Mettler-Toledo STAR^e v. 15.00a software.

Electrospray ionization mass spectrometry (ESI-MS) was performed using an Applied Biosystems PE SCIEX QSTAR instrument.

Magnetic measurements were carried out on a Quantum Design MPMS XL superconducting quantum interference device (SQUID) magnetometer at room temperature.

Transmission electron microscopy (TEM) images were collected on a JEOL 1200EX operated at 100 kV, and micrographs were recorded using an SIA-15C CCD camera. Samples for TEM were prepared as follows: 20 μ L of polymer solution in nanopure water (1 mg/mL) was deposited onto a carbon-coated copper grid, and after 1 min, excess solution was quickly wicked away by a piece of filter paper. The samples were then negatively stained with a 1 wt% phosphotungstic acid (PTA) aqueous solution (20 μ L). After 30 s, excess staining solution was quickly wicked away by a piece of filter paper and the samples were left to dry under ambient conditions prior to imaging. High-resolution scanning transmission electron microscopy (STEM) and elemental mapping were conducted on a FEI TECNAI G2 F20 ST FE-TEM – MATERIALS coupled with Oxford instruments energy-dispersive X-ray spectroscopy (EDS) detector, operating at a voltage of 200 kV with a Gatan 2k x 2k CCD camera.

Atomic force microscopy (AFM) was performed using the Multimode 8 system (Bruker) in PeakForce™ tapping mode using a ScanAsyst-Air Silicon Nitride probe ($k = 0.4$ N/m, $f_0 = 70$ kHz, Bruker). AFM samples were prepared by depositing nanocomposites (1 mg/mL in nanopure water, 20 μ L) on freshly cleaved mica surface. The mica surface was allowed to dry in air. AFM images were analyzed using Nanoscope Analysis software from Bruker.

Dynamic light scattering (DLS) measurements were conducted using a Zetasizer Nano ZS instrument (Malvern Panalytical Ltd., Malvern, United Kingdom) equipped with a laser diode operating at 633 nm. Scattered light was detected at 175° and analyzed using a log correlator for a 0.5 mL of sample in a disposable cell (capacity = 0.9 mL). The photomultiplier aperture and attenuator were adjusted automatically. The particle size distribution and distribution averages were calculated using particle size distribution analysis routines in Zetasizer 7.13 software. The number of accumulations and measurement duration were adjusted automatically. All measurements were repeated 3 times. The average diameter of the particles is reported as the intensity-, volume- and number-average particle diameter from three measurements.

Synthetic Procedures

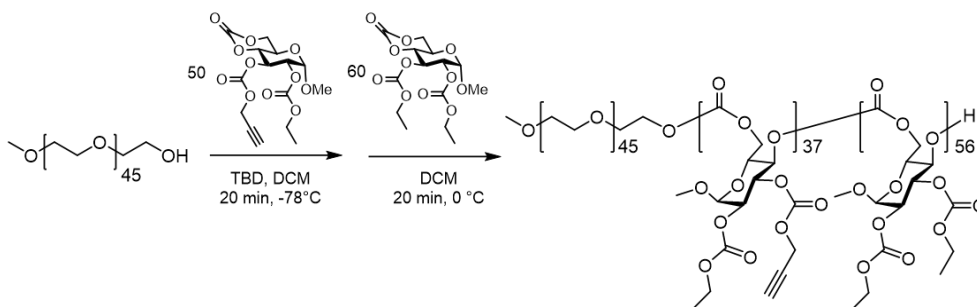
Synthesis of Fe₃O₄ nanoparticles *via* thermal decomposition method³

A flame-dried, 50 mL three-neck flask equipped with a magnetic stir bar and condenser was charged with iron(III) acetylacetonate, Fe(acac)₃ (1 equiv, 720.1 mg, 2.039 mmol), oleic acid (3 equiv, 2.3 g, 6.0 mmol) and oleyl amine (3 equiv, 2.0 g, 6.0 mmol). After the addition of solvent benzyl ether (20 mL) and 1,2-hexadecanediol (5 equiv, 2.6 g, 10 mmol), the reaction mixture was deoxygenated by a flow of nitrogen for 30 min. The reaction temperature was then increased to 140 °C for 1 h. After an hour, the reaction temperature was taken to 200 °C for one hour and then, under a blanket of nitrogen, heated to reflux at 250 - 300 °C for an additional hour. Once the reaction mixture was cooled to room temperature, it was transferred into a centrifuge tube and centrifugation (8000 rpm, 10 min) was applied to remove any undispersed residue. The black-colored mixture was then precipitated with ethanol, centrifuged (8000 rpm, 10 min) to remove the solvent, and resuspended into THF for more than three cycles, until the supernatant became clear (Yield: 81%).

Synthesis of PEG-*b*-PGC(EPC)-*b*-PGC(EC) triblock terpolymers 1 (Scheme S1)

Monomers GC(EPC) and GC(EC) were dried under vacuum over P₂O₅ for 3 days before transferring to a glovebox for storage under an inert atmosphere. All stock solutions were prepared in the glovebox under Ar atmosphere and the reaction conducted in a fume hood. To a solution of GC(EPC) (52 equiv., 48.0 mg, 128 μmol) and α-methoxy-ω-hydroxy poly(ethylene glycol)₄₅ (1 equiv., 4.9 mg, 2.5 μmol) in anhydrous DCM (3.0 mL), organocatalyst TBD (0.38 mg, 2.7 μmol) in DCM (200 μL) was added under Ar atmosphere at -78 °C. After stirring for 20 min, the DCM solution (500 μL) of GC(EC) (58 equiv., 52.8 mg, 145 μmol) was added and allowed to stir for 20 min at 0 °C, then quenched by addition of acetic acid. The polymer was purified by

precipitation from THF into diethyl ether (3×) and dried under vacuum to give a white powder. Yield = 90%. ¹H NMR (500 MHz, CDCl₃) δ ppm 5.45 – 5.26 (m, C³H), 5.02 (m, C¹H), 4.95 – 4.79 (m, C⁴H), 4.77 – 4.64 (m, C²H, OCH₂CCH), 4.36 – 4.23 (m, C⁶H₂), 4.19 (m, OCH₂CH₃), 4.02 (m, C⁵H), 3.64 (s, OCH₂CH₂ in PEG unit), 3.45 – 3.34 (s, OCH₃ in glucose unit), 2.59 (s, OCH₂CCH), 1.35 – 1.24 (m, OCH₂CH₃). ¹³C NMR (126 MHz, CDCl₃) δ ppm 154.08, 154.02, 153.73, 153.62, 153.53, 96.40, 76.18, 74.11, 73.65, 73.39, 72.70, 72.18, 70.52, 66.68, 65.99, 65.69, 64.77, 64.67, 64.58, 55.87, 55.74, 55.60, 46.88, 37.71, 25.58, 20.84, 15.23, 14.08. FT-IR (ATR, cm⁻¹) 3707–3356, 3335–3145, 2889, 1767, 1464, 1450, 1373, 1344, 1238, 1142, 1099, 1016, 993, 961, 869, 841, 781. *M_n* (NMR) = 36200 g/mol. *M_n* (SEC) = 30400 g/mol. *D* = 1.08. *T_g* = 70 °C. TGA in N₂, 172-189 °C, 8% mass loss; 230-275 °C, 50% mass loss; 363–407 °C, 16% mass loss.

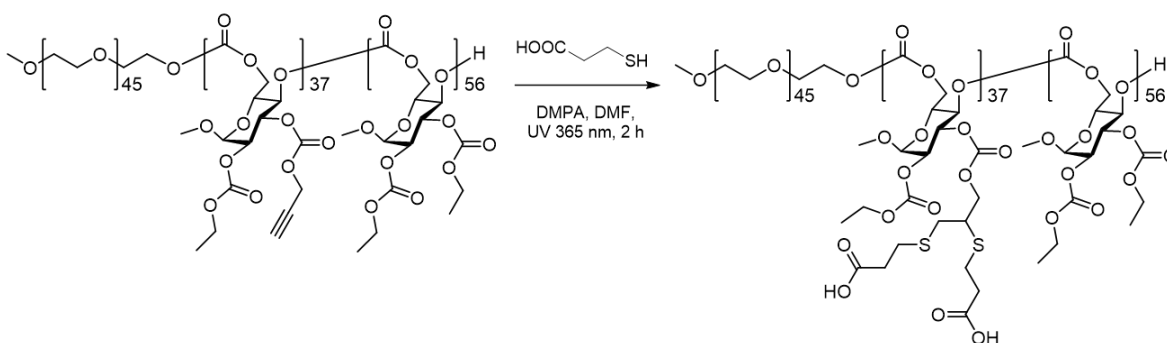


Scheme S1. Synthetic procedure for **1**.

Synthesis of PEG-*b*-PGC(EPC-MPA)-*b*-PGC(EC) **2** by post-polymerization modification of **1** with 3-mercaptopropionic acid (Scheme S2)

Terpolymer **1** (1 equiv., 80.3 mg, 2.21 μmol), 3-mercaptopropionic acid (742 equiv., 174 mg, 143 μL, 1.64 mmol), and 2,2-dimethoxy-2-phenylacetophenone (DMPA) (371 equiv., 210 mg, 0.818 mmol) were dissolved in anhydrous DMF (10 mL), deoxygenated under N₂ for 30 min, and irradiated under UV light (365 nm) for 2 h. The resulting mixture was transferred to dialysis tubing and dialyzed against sodium bicarbonate solution followed by nanopure water for 3 days to remove excess thiol and solvent residue. The solution was lyophilized and further purified by precipitation from THF into diethyl ether (3×) to remove excess impurities, then dried under vacuum to give the polymer PEG-*b*-PGC(EPC-MPA)-*b*-PGC(EC) as a white powder. Yield = 92%. ¹H NMR (500 MHz, DMSO-*d*₆) δ ppm 5.12 (m, C¹H), 4.99 (m, C⁴H), 4.95 – 4.71 (m, C²H), 4.44 – 4.22 (m, C⁵H), 4.13 (m, OCH₂CHS, OCH₂CH₃), 3.99 (m, C⁶H), 3.50 (s, OCH₂CH₂ in PEG unit), 3.31 (s, OCH₃), 2.72 (m, SCH₂CH₂COOH), 2.49 (m, OCH₂CHCH₂S), 1.19 (m, OCH₂CH₃). ¹³C NMR (126 MHz, DMSO-*d*₆) δ ppm 173.40, 154.30, 153.96, 76.14, 70.23, 67.47, 64.90, 62.63, 60.73, 55.36, 49.04, 35.26, 34.92, 31.14, 25.57, 14.33. FT-IR (ATR, cm⁻¹) 3696–3206,

3055–2733, 1753, 1466, 1450, 1395, 1371, 1342, 1244, 1144, 1101, 1032, 1018, 964, 872, 781.
 $T_g = 5\text{ }^\circ\text{C}, 60\text{ }^\circ\text{C}$. TGA in N_2 , 298–329 $^\circ\text{C}$, 38% mass loss; 379–402 $^\circ\text{C}$, 24% mass loss.

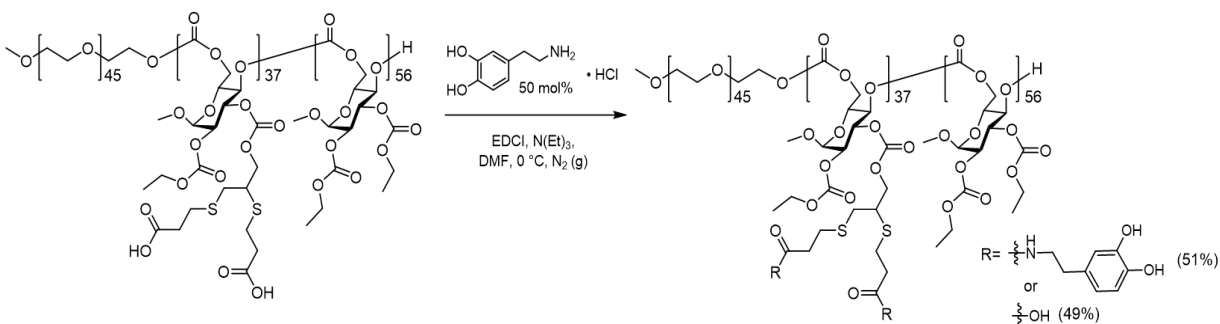


Scheme S2. Synthetic procedure for **2**.

Synthesis of PEG-*b*-PGC[(EPC-MPA)-*co*-(EPC-DOPA)]-*b*-PGC(EG) **3** by conjugation of dopamine hydrochloride with **2** (Scheme S3)

PEG-*b*-PGC(EPC-MPA)-*b*-PGC(EG) (1 equiv., 15 mg, 0.34 μmol), dopamine hydrochloride (0.7 equiv. relative to carboxylic acid groups, 3.5 mg, 19 μmol), and EDC hydrochloride (2 equiv. relative to carboxylic acid groups, 9.5 mg, 50 μmol) were dissolved in 10 mL dry DMF and added with triethyl amine (TEA, 1 equiv. relative to carboxylic acid groups, 2.55 mg, 3.51 μL , 25 μmol) solution afterwards in the glovebox under Ar atmosphere. The mixture was transferred to the fume hood and allowed to react at 0 $^\circ\text{C}$ for 20 min, then slowly turn back to room temperature and react for another 10 h under nitrogen flow and aluminum foil to avoid polymerization of dopamine. The resulting mixture was transferred to a 6 – 8 kDa dialysis tubing and dialyzed against nanopure water to remove solvent residues for 3 days. The solution was lyophilized and further purified by precipitation from THF into diethyl ether (3 \times) to remove impurities insoluble in water, then dried under vacuum to give the polymer PEG-*b*-PGC[(EPC-MPA)-*co*-(EPC-DOPA)]-*b*-PGC(EG) as a white powder. Yield = 87%. Dopamine conjugation percentage = 51%. ^1H NMR (400 MHz, $\text{DMSO-}d_6$) δ ppm 11.42 (s, $\text{SCH}_2\text{CH}_2\text{COOH}$), 8.95 (m, protons from dopamine diol), 7.94 (m, CONHCH_2), 6.94 – 6.01 (m, *Ar*), 5.35 – 4.59 (m, $\text{C}^1\text{H}, \text{C}^4\text{H}, \text{C}^2\text{H}$), 4.49 – 3.88 (m, $\text{C}^5\text{H}, \text{OCH}_2\text{CHS}, \text{OCH}_2\text{CH}_3, \text{C}^6\text{H}$), 3.50 (s, OCH_2CH_2 in PEG unit), 1.09 (m, OCH_2CH_3). ^{13}C NMR (126 MHz, CDCl_3) δ ppm 195.17, 154.10, 136.85, 134.23, 132.86, 129.99, 128.91, 128.52, 128.13, 126.93, 103.58, 96.33, 73.69, 72.70, 70.55, 64.69, 55.63, 50.05, 28.30, 23.58, 19.32, 14.11. FT-IR (ATR, cm^{-1})

3688-3186, 3071-2823, 2754-2167, 1929, 1759, 1659, 1620, 1366, 1242, 995, 879, 833, 779, 702. $T_g = 93\text{ }^\circ\text{C}$. TGA in N_2 , 213–260 $^\circ\text{C}$, 38% mass loss; 393-401 $^\circ\text{C}$, 12% mass loss.



Scheme S3. Synthetic procedure for **3**.

Preparation of nanocomposites **4** by co-assembly of triblock polymer **3** and iron oxide nanoparticles

PEG-*b*-PGC(MPA)-*b*-PGC(EC) was dissolved in DMF (4 mg/mL, 5 mL, 1 vol%), and mixed with 0.2 mg/mL iron oxide nanoparticle solution in THF (5 mL, 1 vol%). The mixture was sonicated for 10 min. Nanopure water (10 mL, 2 vol%) was added into the sonicated mixture dropwise by a syringe pump at 2 mL/h. Additional nanopure water (10 mL, 2 vol%) was added afterwards within 15 min. The resulting solution mixture was transferred to 6 – 8 kDa molecular weight cut-off dialysis tubing immediately and dialyzed against nanopure water for 1 day. The solution was lyophilized and resuspended in nanopure water directly to afford a 1 mg/mL solution.

Representative Procedure for Oil Sequestration Experiments

Weathered crude oil from the Texas–Oklahoma pipeline (light sweet crude oil, density = *ca.* 0.76 g/mL) was added to a 4 mL shell vial containing nanopure water (2 mL). Powdered glucose-based magnetic nanoparticles (2 mg) were added to the oil-contaminated water and allowed to rest with minimal movement. After *ca.* 10 h, the loaded material was attracted by an external magnetic field for *ca.* 10 min, followed by decanting of the contaminated water for oil extraction.

Oil Recovery Experiments

The crude oil trapped within the nanocomposites was extracted with diethyl ether (1 mL) twice. The nanocomposites were vortexed and sonicated for *ca.* 1 min to allow complete recovery of crude oil. The composite materials were recovered using magnet (neodymium magnet, 90 lb pull) and the crude oil-containing organic phase was transferred to a pre-weighed clean vial. After the evaporation of diethyl ether, the mass of the extracted crude oil was recorded. Experiments were conducted in triplicate.

Recyclability Tests

The composite materials with 15 mg initial amount were reused for another two cycles of oil sequestration after the extraction of captured crude oil. For each cycle, 150 μ L of crude oil was applied. The recovered composite materials were redeployed into contaminated water (2 mL) and exposed to the external magnetic field followed by the same procedure as previously described. The experiment was repeated for three times, and the loading capacity of the reused materials [sequestered crude oil/nanocomposite (mg/mg)] was recorded.

Degradation Study

Degradation study in organic media: PEG-*b*-PGC[(EPC-MPA)-*co*-(EPC-DOPA)]-*b*-PGC(EC) (5.0 mg) were added in DMSO-*d*₆ to an NMR tube containing 0.2 mL D₂O, and 6 mg TEA to yield a 5 mg/mL solution. The solution was incubated at 37 °C, and the degradation process was monitored by ¹H NMR spectroscopy. There was no obvious change in NMR spectra over 100 days, 100 μ L of the reaction mixture was removed and directly analyzed by ESI-MS.

Hydrolytic degradation study in aqueous media: PEG-*b*-PGC[(EPC-MPA)-*co*-(EPC-DOPA)]-*b*-PGC(EC) (5.0 mg) was added to D₂O in an NMR tube containing NaOD to yield a 5 mg/mL solution (pH = 10). The solution was incubated at 37 °C, and the degradation process was monitored by ¹H NMR spectroscopy. No obvious change in NMR spectra over 60 d, reaction mixture (100 μ L) was removed and directly analyzed by ESI-MS.

Degradation study in an environmental chamber: Nanocomposites (5 mg) were applied into crude oil (20 μ L)-contaminated water (2 mL), which was adjusted to pH 8 with NaOH to mimic sea water. Crude oil sequestration experiments by nanocomposites were conducted as previously stated. The mixture of uncovered crude oil, alkaline water, and nanocomposites remaining in water was transferred to an environmental chamber that was set to 22 °C and 70% relative humidity. An aliquot of the reaction mixture (100 μ L) was removed and directly analyzed by ESI-MS immediately and after 20, 50, and 100 d.

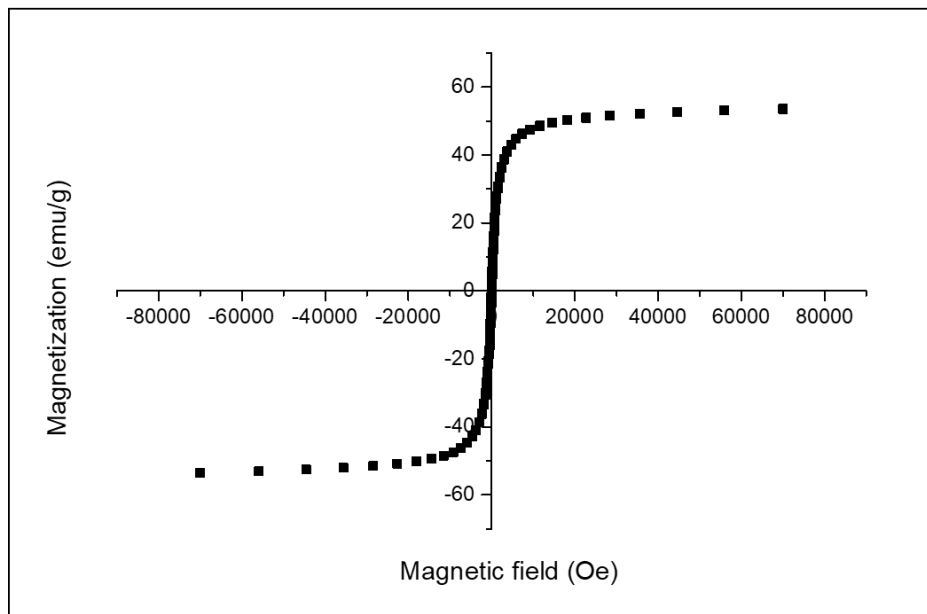


Figure S1. SQUID magnetometry analysis showing magnetization vs. applied magnetic field for iron oxide nanoparticles.

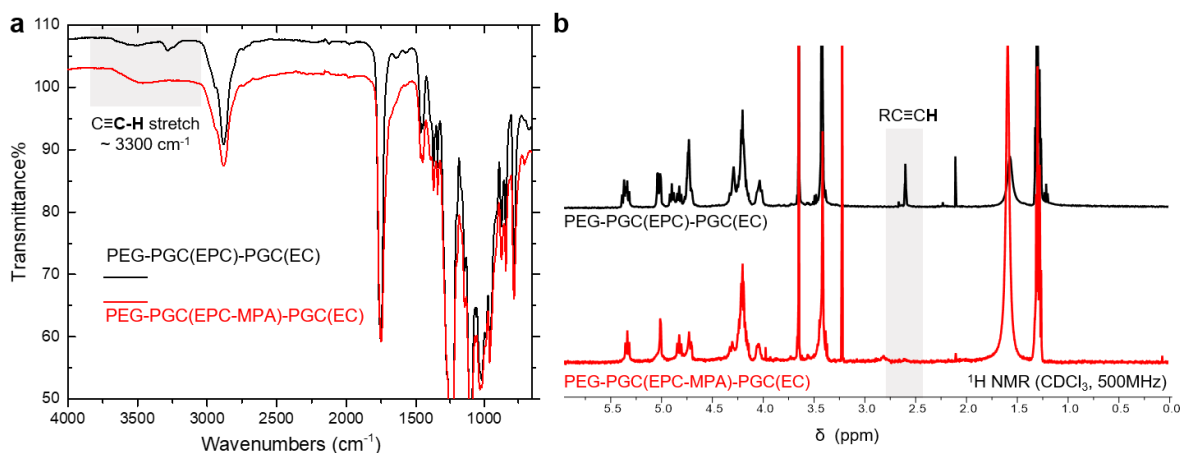


Figure S2. (a) FT-IR spectra of triblock polymers **1** (black line) and **2** (red line) before and after the thiol-yne click reaction with MPA, showing the disappearance of the alkynyl C-H stretch at 3300 cm^{-1} . (b) ^1H NMR spectra (500 Hz, CDCl_3) of polymers **1** and **2**, showing the disappearance of the proton resonance from the alkyne group at 2.60 ppm.

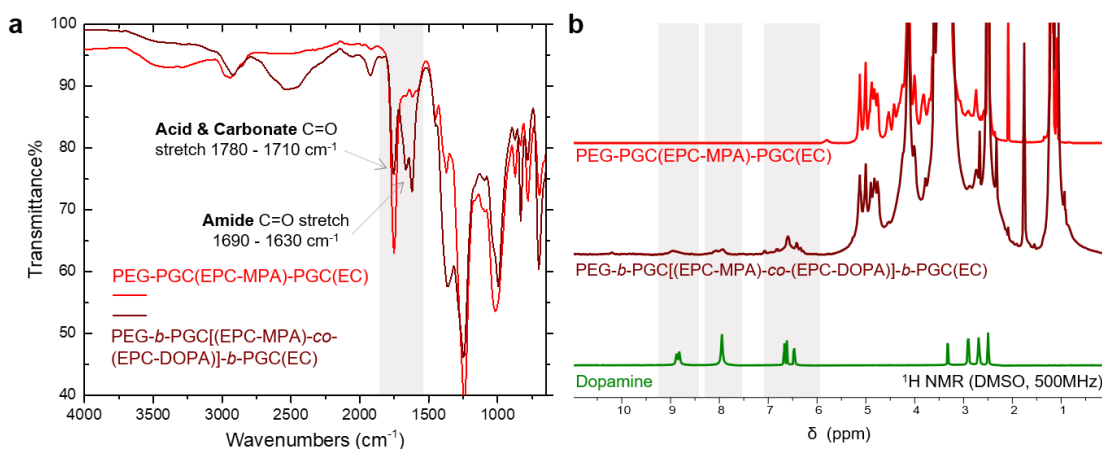


Figure S3. (a) FT-IR spectra of triblock polymer **2** (red line) and **3** (maroon line) before and after the amidation reaction with dopamine, showing the shift of the C=O stretch from the carboxylic acid to amide carbonyl groups. (b) ^1H NMR spectra (500 Hz, $\text{DMSO}-d_6$) of triblock polymers **2**, **3**, and dopamine hydrochloride (green line), showing the appearance of proton resonance from dopamine at 6.25 – 6.75 ppm, 7.75 - 8.25 ppm, and 8.75 – 9.25 ppm.

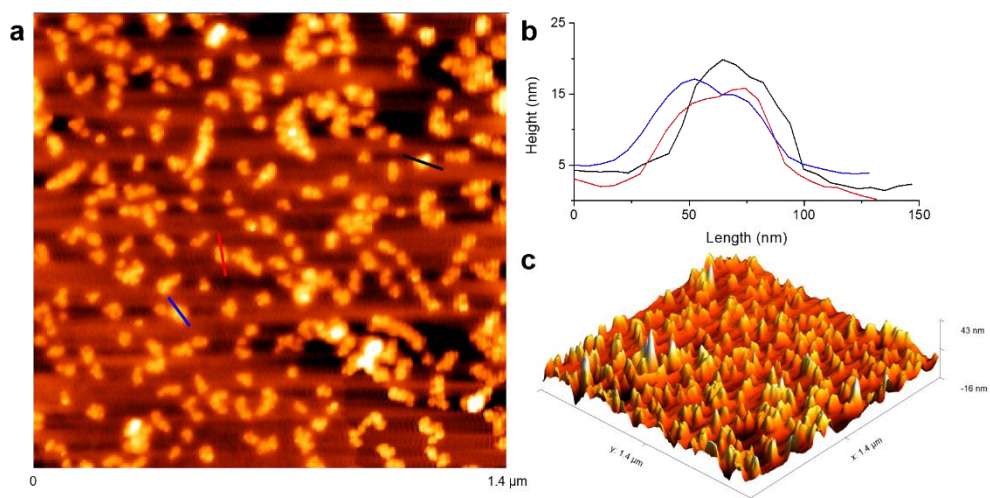


Figure S4. (a) AFM micrographs, (b) height profile, (c) 3D image of nanocomposites **4**. Sample was drop cast onto the freshly cleaved mica, and then left to dry under the ambient condition. The height profiles were measured along the colored lines shown on the micrographs.

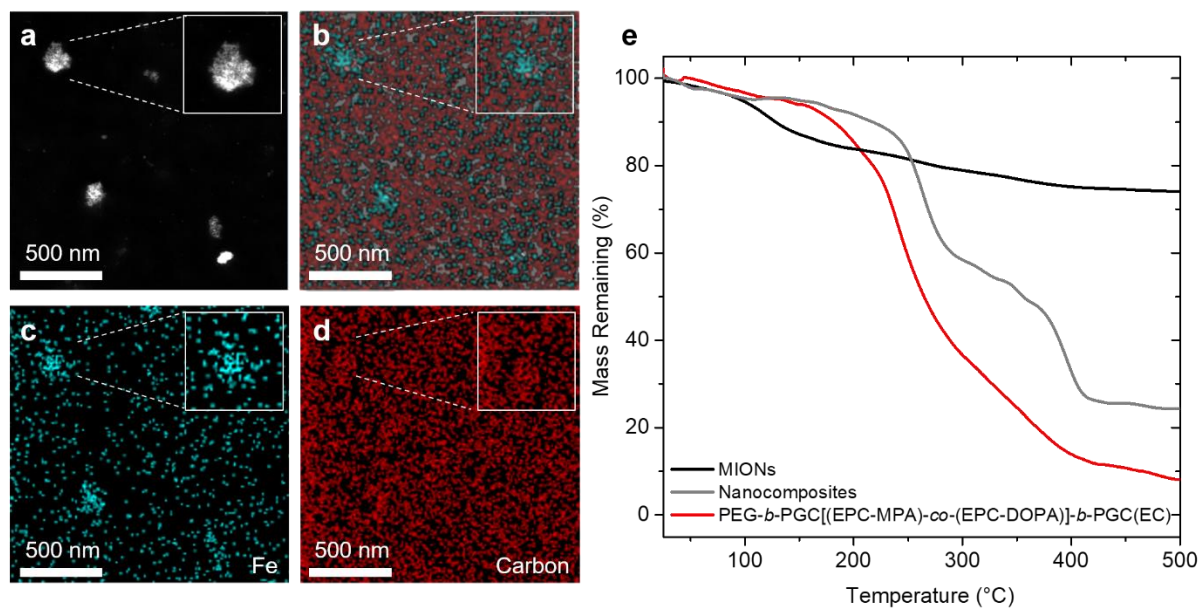


Figure S5. Dark-field STEM image and elemental mapping of nanocomposite **4** by EDS detector in STEM mode (a) dark-field STEM image, (b) merged elemental mapping images of iron (Fe) and carbon, (c) elemental mapping images of iron and (d) carbon. (e) Thermogravimetric analysis of MIONs (black line), polymer **3** (red line), and nanocomposites (grey line).

Table S1. Quantitative evaluation of crude oil loading capacity by nanocomposites

	1:5 ^a	1:10	1:15	1:20	1:25	1:30
1 st experiment	4.4 ^b mg	8.9 mg	13.9 mg	14.5 mg	15.3 mg	18.9 mg
2 nd experiment	3.8 mg	7.6 mg	10.4 mg	16.8 mg	15.5 mg	14.8 mg
3 rd experiment	4.2 mg	8.8 mg	12.2 mg	20.6 mg	15.3 mg	16.7 mg
Average (mg per mg of nanocomposites).	2.1 ± 0.1	4.2 ± 0.3	6.1 ± 0.7	8.7 ± 1.3	7.7 ± 0.1	8.4 ± 1.7
$\frac{\text{Sequestered Oil (mg)}}{\text{Initial Oil (mg)}}$	0.54	0.56	0.53	0.57	0.40	0.37

^a The actual amount of nanocomposites and initial volume of crude oil used for 1:5, 1:10, 1:15, 1:20, 1:25, 1:30 are 2 mg nanocomposites and 10, 20, 30, 40, 50, 60 µL crude oil;

^b The amount of crude oil (mg) recovered from 2 mL water by diethyl ether wash.

Table S2. Quantitative evaluation of nanocomposite recyclability

	1 st cycle ^a			2 nd cycle			3 rd cycle		
	M ^b (mg)	N ^c (mg)	M/N ^d	M (mg)	N (mg)	M/N	M (mg)	N (mg)	M/N
1 st experiment	78.9	15.0	5.3	43.0	9.5	4.5	29.0	7.4	3.9
2 nd experiment	73.0	15.0	4.9	34.5	8.0	4.3	27.1	6.0	4.5
3 rd experiment	70.3	15.0	4.7	33.4	7.4	4.5	26.2	5.4	4.9
Average	74.1 ± 3.6	15.0 ± 0	4.9 ± 0.2	37.0 ± 4.3	8.3 ± 0.9	4.5 ± 0.1	27.4 ±1.2	6.3 ± 0.8	4.4 ± 0.4

^a The amount of nanocomposites and initial volume of crude oil for each cycle are 15 mg and 150 µL.

^b The amount of crude oil (mg) recovered by diethyl ether wash;

^c The actual amount of nanocomposite **4** (mg) retrieved from last cycle and used to recover oil for next cycle;

^d The amount of recovered crude oil (mg) per unit mass of nanocomposite **4** (mg).

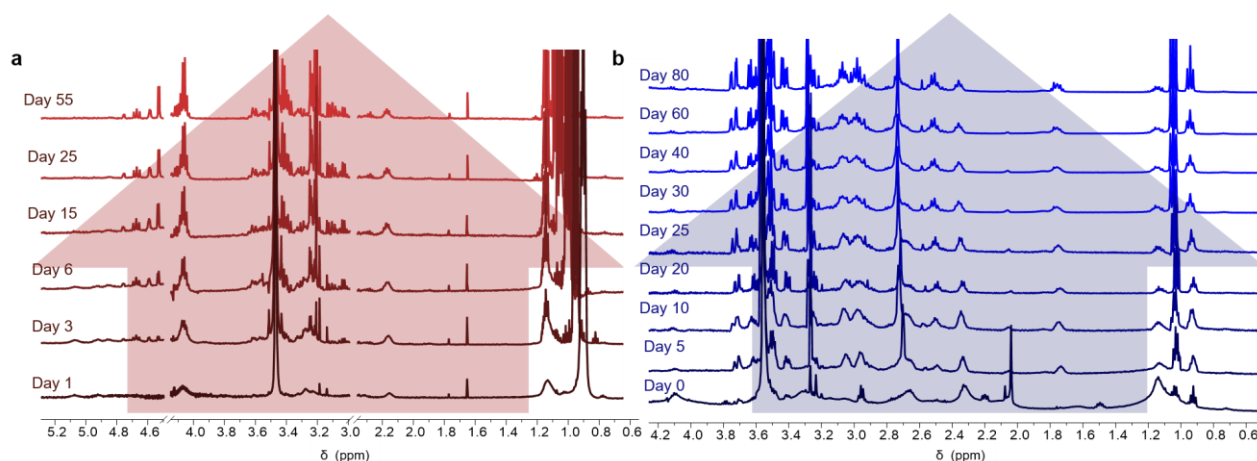


Figure S6. ^1H NMR (500 MHz) spectra of triblock polymers **3** acquired in (a) $\text{DMSO-}d_6$ (5 mg/mL) with addition of D_2O (20 vol %) and TEA (6 mg/mL) during incubation at 37°C over 55 days and (b) in D_2O (5 mg/mL) with addition of NaOD adjusted to pH 10 during incubation at 37°C over 80 days. It is noted that resonance of TEA was cut in spectrum (a).



Figure S7. By clicking the .mp4 file above, or on the next page, see a video of the addition of lyophilized magnetic nanocomposites to a weathered mixture of crude oil and water in a petri dish.



Figure S8. By clicking the .mp4 file above, or on the next page, see a video of the magnetic action of lyophilized magnetic nanocomposites after absorption of crude oil from a weathered mixture of crude oil and water in a petri dish.

References

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