Supplementary Information for

Fusion of Capsules to Produce Liquid-Filled Monoliths for Carbon Capture

Chia-Min Hsieh,[a] Luma Al-Mahbobi,[b] Smita S. Dasari,[c] Mohd Avais,[b] Huaixuan Cao,[c] Peiran Wei,[d] Yifei Wang,[b] Micah J. Green,[c] and Emily B. Pentzer*[ab]

[a]Department of Chemistry, Texas A&M University, College Station, TX 77843, USA

[b]Department of Materials Science and Engineering, Texas A&M University, College Station, TX 77840, USA

[c]Artie McFerrin Department of Chemical Engineering, Texas A&M University, College Station, TX 77843, USA

[d]Soft Matter Facility, Texas A&M University, 1313 Research Pkwy, College Station, TX 77845, USA

*Email: emilypentzer@tamu.edu

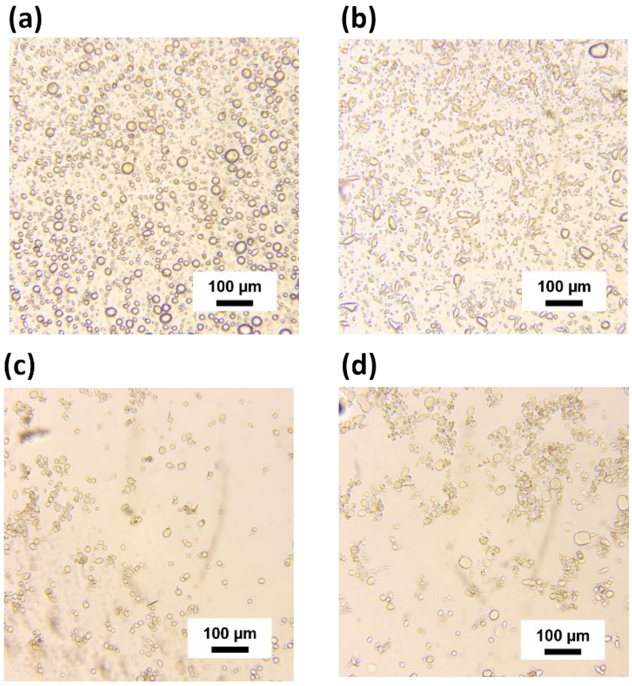

Fig. S1 Optical microscope images of emulsions: (a) IPDI+PAO-in-water, (b) BZDI+PAO-in-water, (c) Cys+IL-in-octane, and (d) EDA+IL-in-octane.

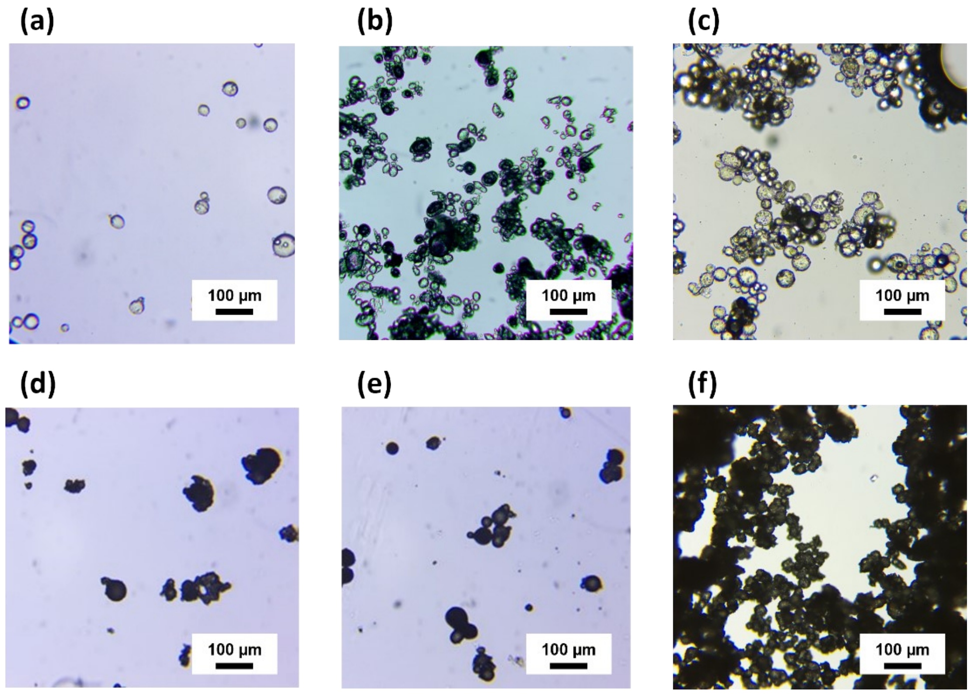

Fig. S2 Optical microscopy images of capsules: (a) IPDIxCys_PAO, (b) BZDIxCys_PAO, and (c) IPDIxEDA_PAO redispersed in water. (d) IPDIxCys_IL, (e) BZDIxCys_IL, and (f) IPDIxEDA_IL redispersed in hexanes.

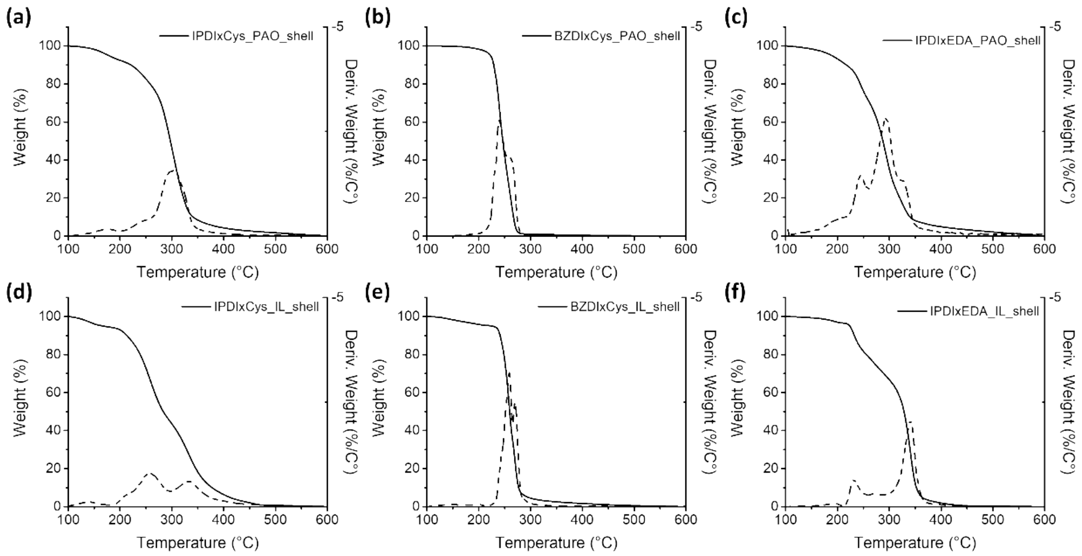

Fig. S3 TGA weight loss profiles of capsule shells: (a) IPDIxCys_PAO, (b) BZDIxCys_PAO, (c) IPDIxEDA_PAO, (d) IPDIxCys_IL, (e) BZDIxCys_IL, and (f) IPDIxEDA_IL.

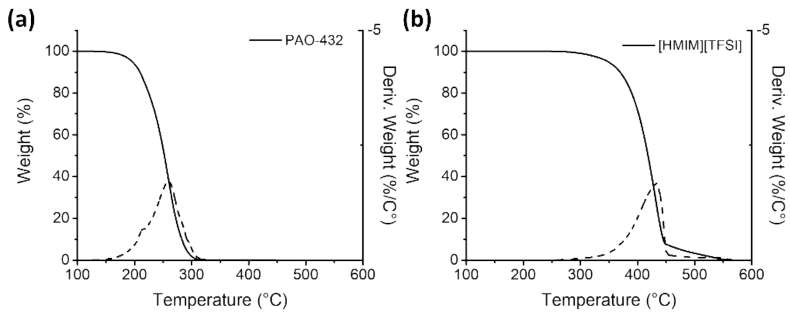

Fig. S4 TGA weight loss profiles of (a) PAO-432 and (b) [HMIM][TFSI].

Table S1 Summary of DTG peaks (unit: °C).

	IPDIxCys_PAO	BZDIxCys_PAO	IPDIxEDA_PAO	IPDIxCys_IL	BZDIxCys_IL	IPDIxEDA_IL
Capsule	245 339	264	259 350	269, 398, 430	223, 272, 434, 464	311, 450, 471
Shell	171, 246, 304	242, 261	200, 245, 293, 329	142, 219, 258, 332	259, 270	193, 232, 341
Core	260			432		

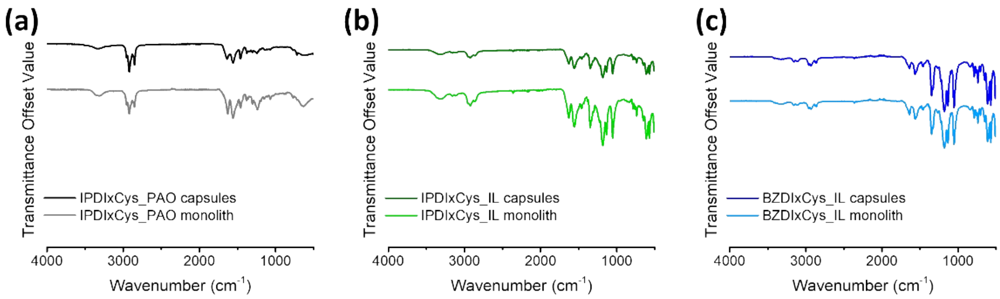

Fig. S5 Offset of IR spectra of (a) IPDIxCys_PAO, (b) IPDIxCys_IL, and (c) BZDIxCys_IL capsules and monoliths.

Solid-state Electron Paramagnetic Resonance (EPR) Characterization:

The basic formula for calculating the g-value in EPR spectroscopy is $g = \frac{hv}{\beta B}$ (**Eq. S1**), where h represents Planck's constant (6.626×10^{-34} J·Hz^{-1}), v is the microwave frequency (9.357×10^{9} Hz), β is the Bohr magneton (9.274×10^{-24} J·T^{-1}), and B is the magnetic field strength (in Tesla).

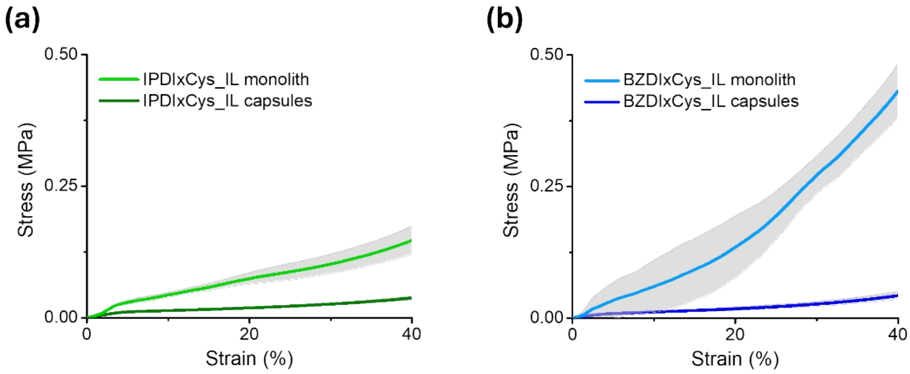

Fig. S6 Stress-strain curves from compression tests of (a) IPDIxCys_IL and (b) BZDIxCys_IL compact capsules and fused monoliths. The Shaded area indicates the standard deviation (n=3).

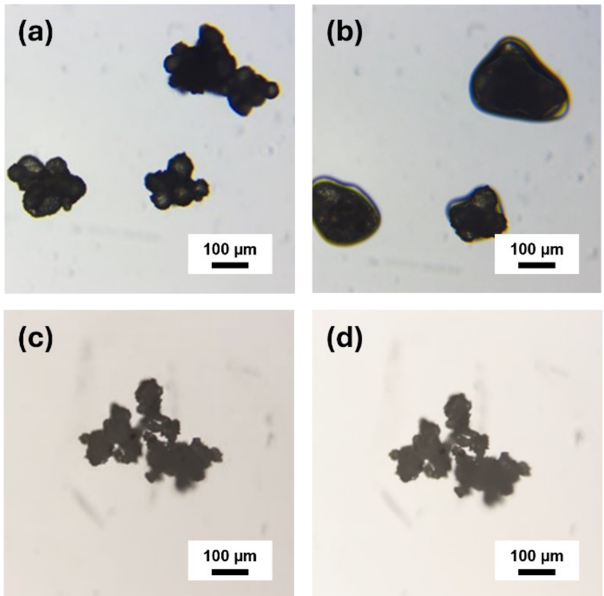

Fig. S7 Optical microscopy images of IPDIxCys_IL capsules at (a) 22 and (b) 100 °C; IPDIxEDA_IL capsules at (a) 22 °C and (b) 100 °C for 10 minutes.

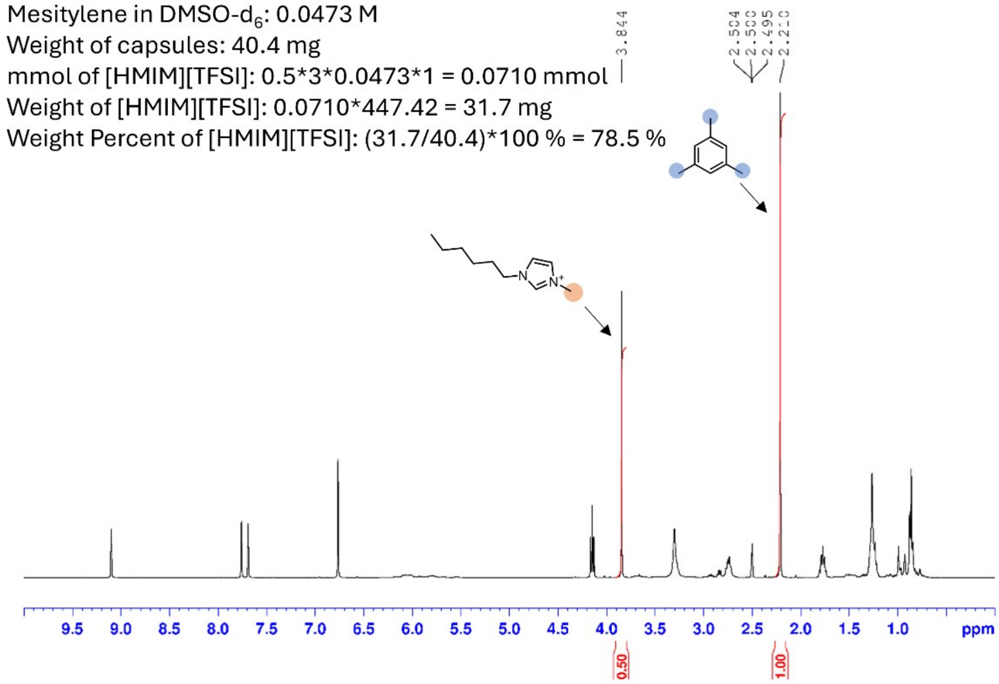

Fig. S8 A representative ^{1}H NMR spectrum of the extractant from IPDIxCys_IL monolith using DMSO-d_6 and mesitylene as the internal standard. Integration was used to calculate wt% of the IL.

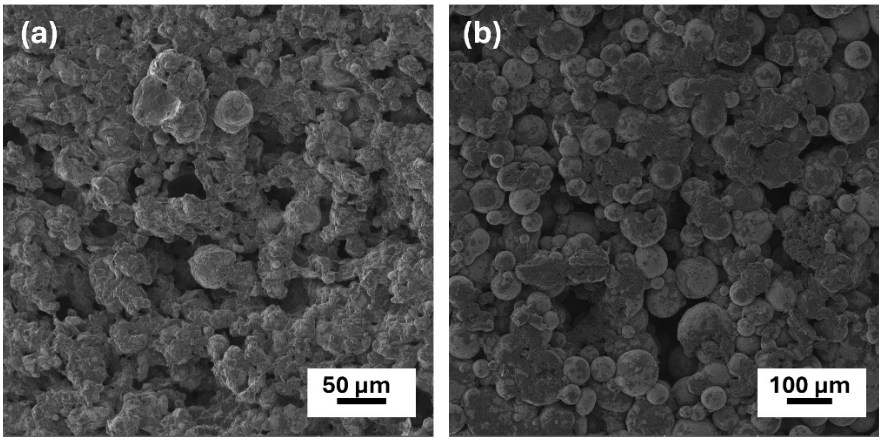

Fig. S9 Monoliths washed with acetone: (a) IPDIxCys_IL and (b) BZDIxCys_IL.

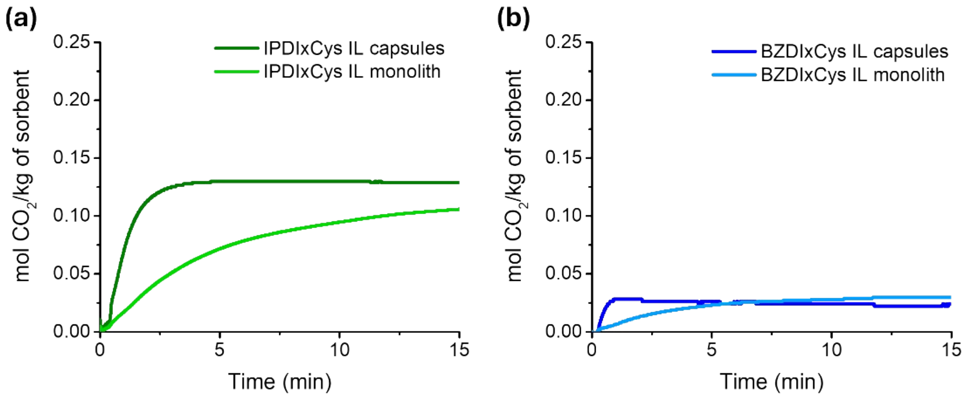

Fig. S10 Preliminary CO_2 absorption tests of (a) IPDIxCys_IL and (b) BZDIxCys_IL.

Table S2 Comparison of CO2 capacity of analogues systems containing polymeric capsules with ionic liquid cores.

Shell	Core	Conditions	CO_2 Capacity (mol/kg)	Reference
Responsive disulfide Polyurea	[HMIM][TFSI]	1 bar, 25 ˚C	0.12	This work
Polyurea	[HMIM][TFSI]	1 bar, 20 ˚C	0.068	*Ind. Eng. Chem. Res.*, 2019, **58**, 10503–10509.
	[BMIM][PF_6]	1 bar, 20 ˚C	0.025-0.065	*J. Polym. Sci.*, 2021, **59**, 2980–2989.
	[EMIM][2-CNpyr] + 1,3-propandiol	1 bar, 25 ˚C	1.40	*ACS Sustain. Chem. Eng.*, 2024, **12**, 7882–7893.
	[EMIM][2-CNpyr] + diethylene glycol		1.03	
	[BMIM][BF_4]	1 bar, 25 ˚C	0.2	*ACS Appl. Eng. Mater.*, 2024, **2**, 1298–1305.
	[BMIM][BF_4] + piperazine		0.4	
	[EMIM][2-CNpyr]	1 bar, 25 ˚C	3.0	*ACS Appl. Mater. Interfaces*, 2020, **12**, 19184–19193.
	[EMIM][2-CNpyr]0.5[TFSI]0.5		0.9	
	[EMIM][2-CNpyr]0.5[TCM]0.5		1.3	
Polysulfone	[EMIM][TFSI]	1 bar, 25 ˚C	0.90	*Environ. Chall.*, 2021, **4**, 100109.

	[BMIM][TFSI]		0.84	
	[HMIM][TFSI]		0.79	
	[BMIM][TFSI] + Fe_2O_3	4 bar, 45 ˚C	1.3	*J. Environ. Chem. Eng.*, 2021, **9**(1), 104781.
	[EMIM][TFSI]	4.3 bar, 45 ˚C	1.22	*Heliyon*, 2023, **9**(2), e13298.
Poly(vinylidene fluoride-co-hexafluoropropylene)	[EMIM][TFSI]	5 bar, 23 ˚C	0.465	*J. Chem. Eng.*, 2018, **354**, 753–757.
	[HMIM][TFSI]		0.565	
Polystyrene	[BMIM][PF_6]	1 bar, 25˚C	0.45	*Mater. Chem. Phys.*, 2020, **251**, 122982.
Acrylic-based Polymer	[EMIM][TFO]	4 bar, 45 ˚C	1.2	*J. Mol. Liq.*, 2023, **385**, 122394.
Acrylic-based Polymer	NDIL0231	Not reported	~1.8	*Energy Procedia*, 2017, **114**, 860–865.
Poly(3-[Tris(trimethylsiloxy)silyl]propyl methacrylate)	NDIL0231	0.19 bar, 25˚C	~0.5	*Faraday Discuss.*, 2016, **192**, 271.

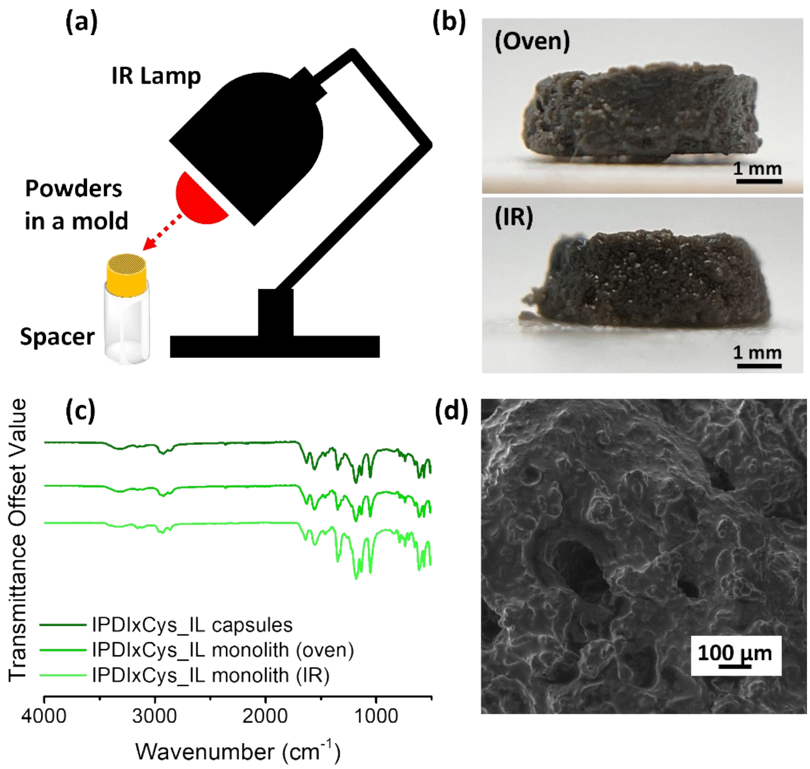

Fig. S11 (a) Schematic of the IR fusion experiment setup. (b) IPDIxCys_IL monoliths prepared using an oven (top) and IR lamp (bottom). (c) Offset of IR spectra of IPDIxCys_IL capsules and monoliths prepared using an oven and IR lamp. (d) SEM image of the IPDIxCys_IL monolith cross-section fused using an IR lamp.

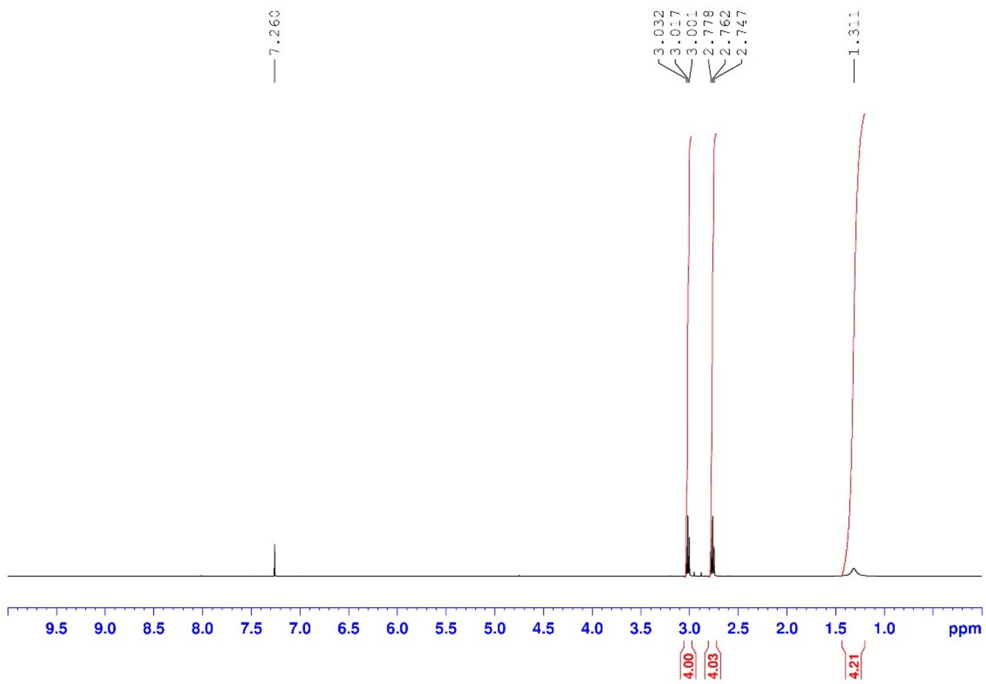

Fig. S12 ^{1}H NMR spectrum of cystamine.

Aqueous Emulsion

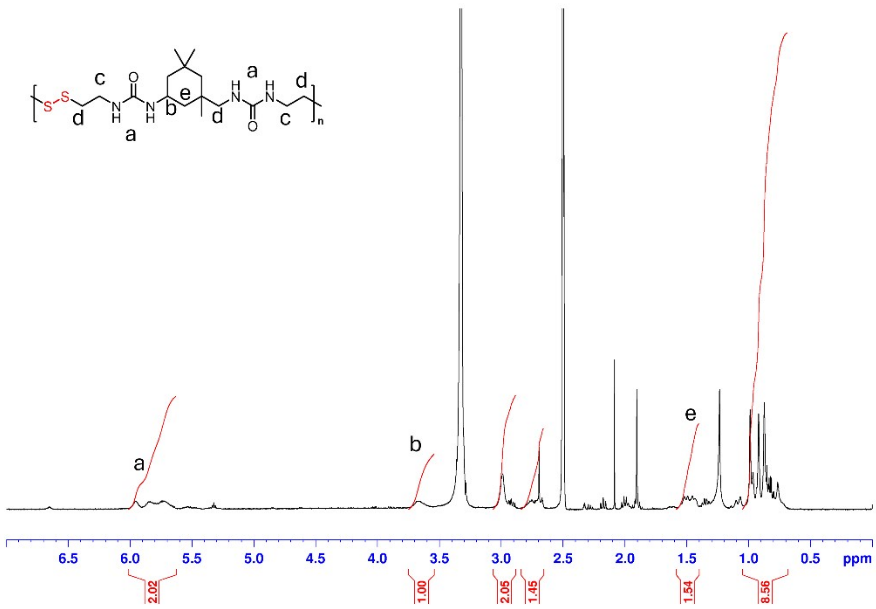

Fig. S13 ^{1}H NMR spectrum of the IPDIxCys_PAO shell in DMSO-d_6.

Non-Aqueous Emulsion

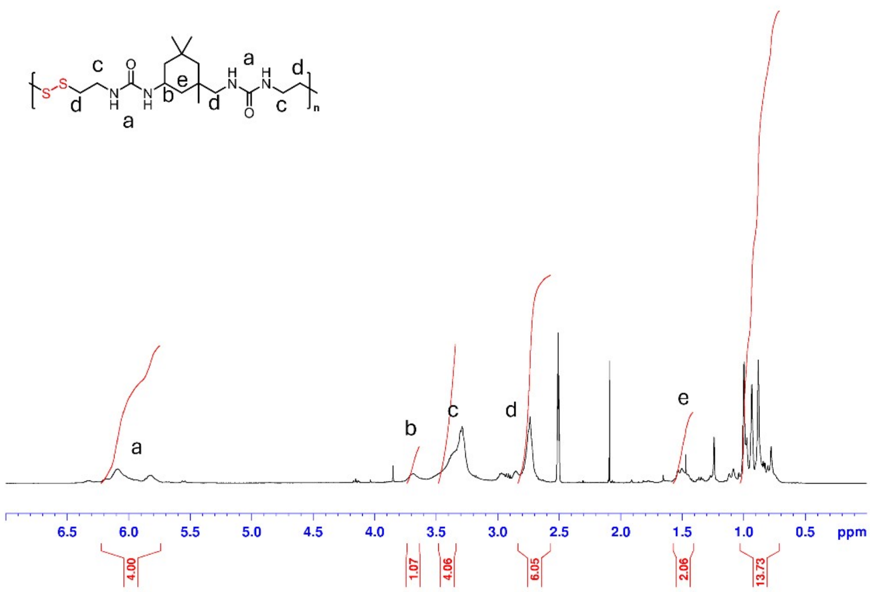

Fig. S14 ^{1}H NMR spectrum of the IPDIxCys_IL shell in DMSO-d_6.

Aqueous Emulsion

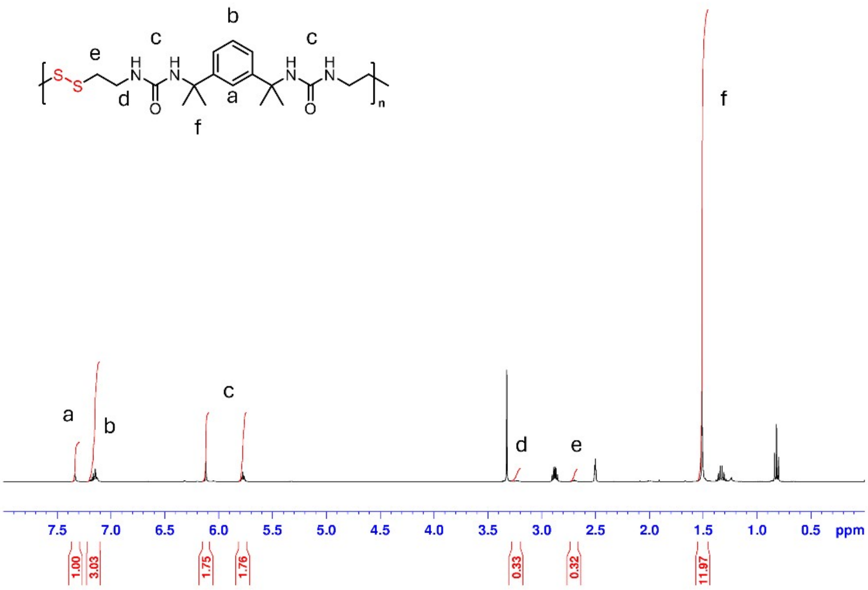

Fig. S15 ^{1}H NMR spectrum of the BZDIxCys_PAO shell in DMSO-d_6.

Non-Aqueous Emulsion

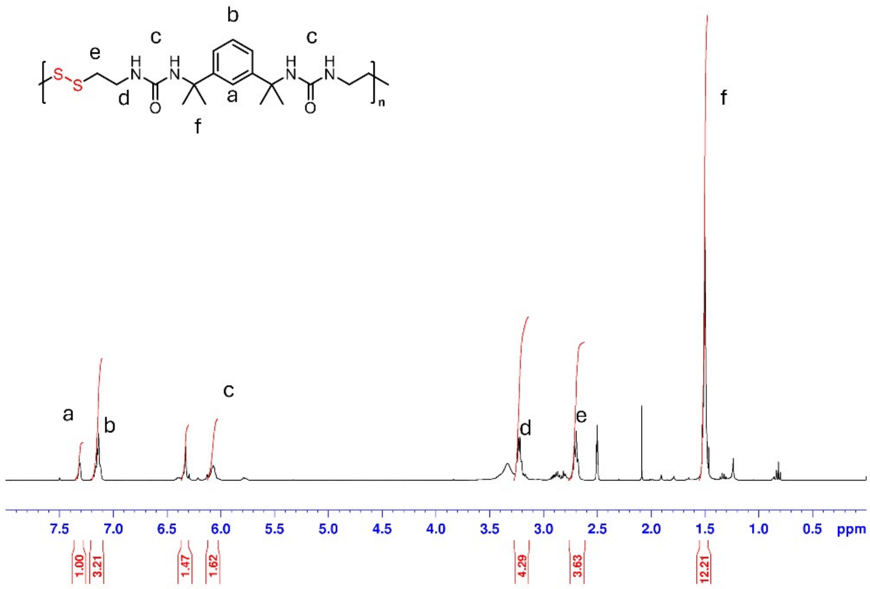

Fig. S16 ^{1}H NMR spectrum of the BZDIxCys_IL shell in DMSO-d_6.

Aqueous Emulsion

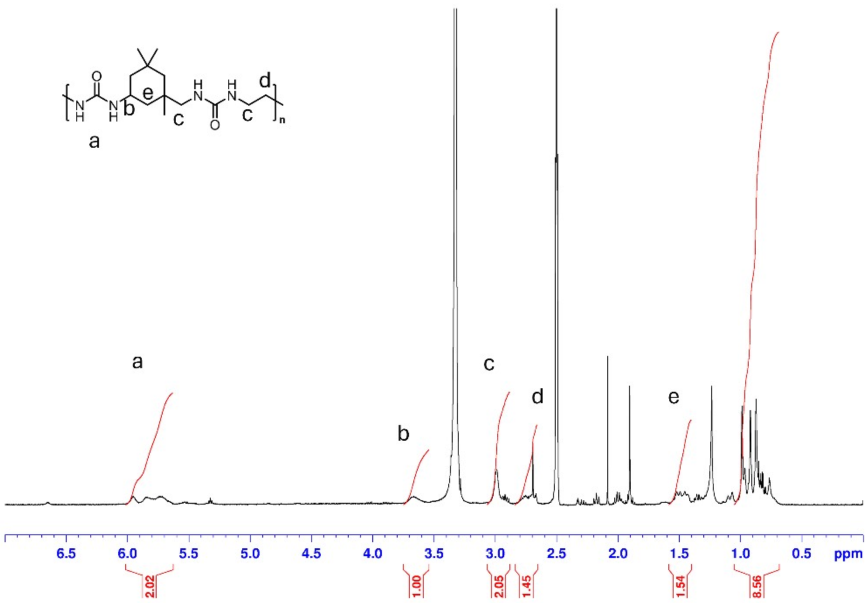

Fig. S17 ^{1}H NMR spectrum of the IPDIxEDA_PAO shell in DMSO-d_6.

Non-Aqueous Emulsion

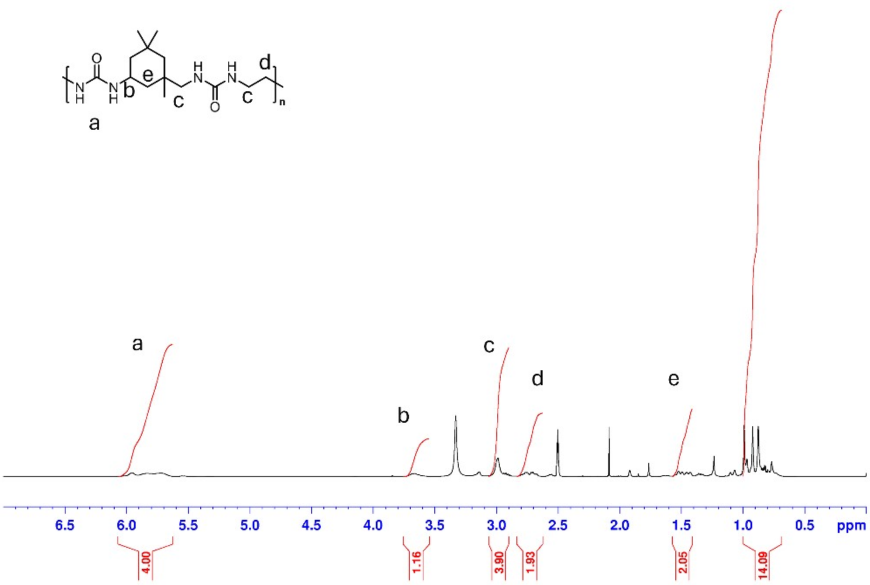

Fig. S18 ^{1}H NMR spectrum of the IPDIxEDA_IL shell in DMSO-d_6.