Supplementary Information (SI) for Polymer Chemistry.

Fenton–RAFT Polymerization in Organic Media

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- **Characterization date of the Fe(II)-MOF particles**

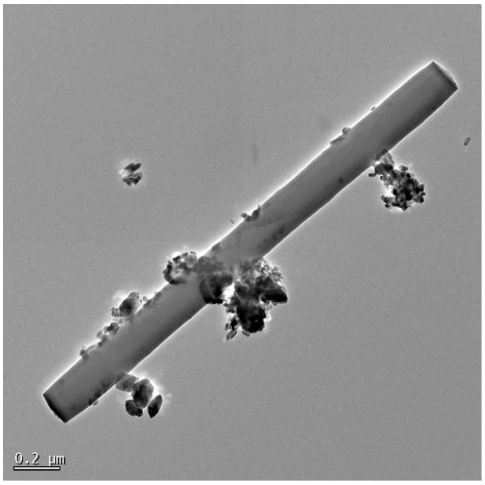

Figure S1. HR-TEM image of the employed Fe(II)–MOF particles in this study.

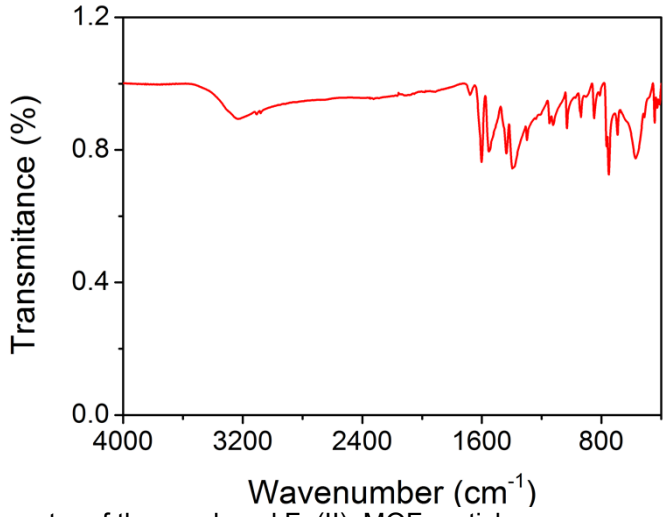

Figure S2. FTIR spectra of the employed Fe(II)–MOF particles.

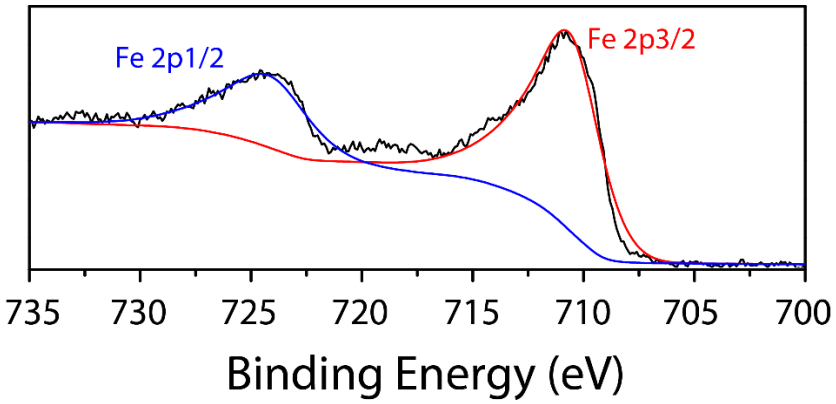

Figure S3. XPS analysis of the Fe(II)–MOF particles before the polymerization.[1]

- **Non-deoxygenated organic Fenton-RAFT polymerization**

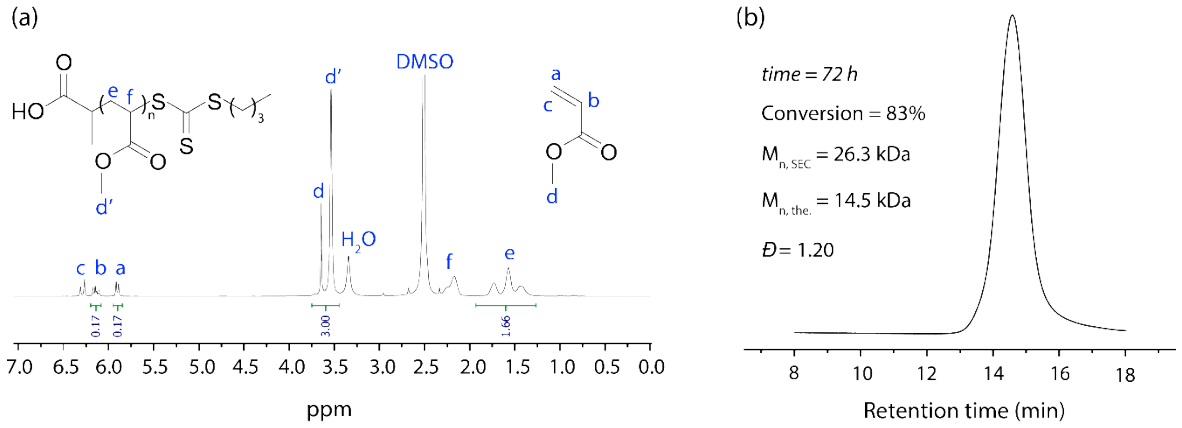

Figure S4. ^{1}H NMR and SEC peaks of the synthesized PMA with TTC-1 *via* the non-deoxygenated Fenton-RAFT in DMSO. [Fe(II)-MOF] = 1.6 mg mL^{-1}, and $[H_2O_2]_0$ = 35 mM.

- **Deoxygenated organic Fenton-RAFT polymerization**

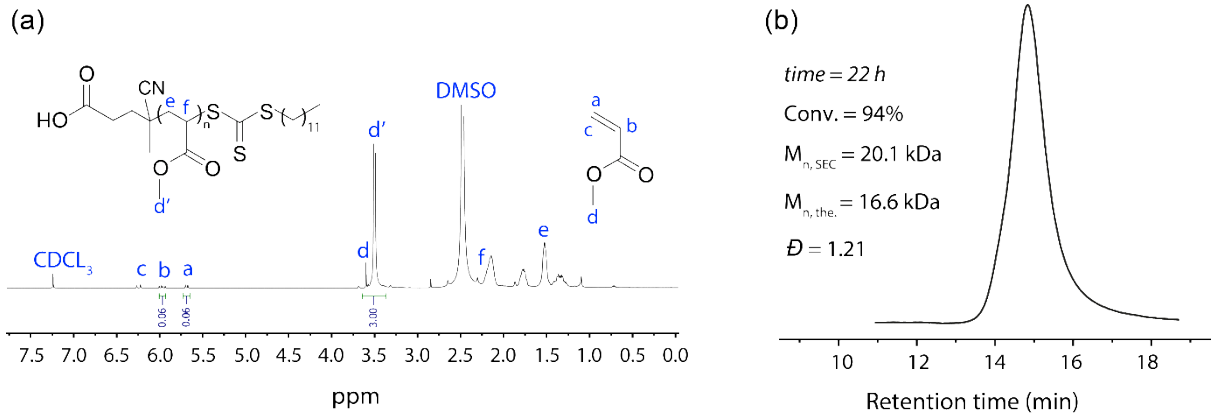

Figure S5. ^{1}H NMR and SEC peaks of the synthesized PMA with TTC-2 *via* the deoxygenated Fenton-RAFT in DMSO. [Fe(II)-MOF] = 3.2 mg mL^{-1}, and $[H_2O_2]_0$ = 70 mM.

- Kinetic study of the organic Fenton-RAFT polymerization

Table S1. Characterization data of the synthesized PMA via the kinetic study when [Fe(II)-MOF] = 9.6 mg mL^{-1} and $[H_2O_2]_0$ = 70 mM.

time (h)	Conversion (%)	$M_{n,\ the.}$ (kDa)	$M_{n,\ SEC}$ (kDa)	*Đ* (-)
0.5	2	-	-	-
1	15	-	-	-
3	34	6.1	3.2	1.34
5	46	8.2	4.3	1.35
7	56	9.9	7.4	1.26
9	71	12.5	10.0	1.19
11	80	14.1	12.8	1.15

- **Fenton reaction-mediated bulk polymerization**

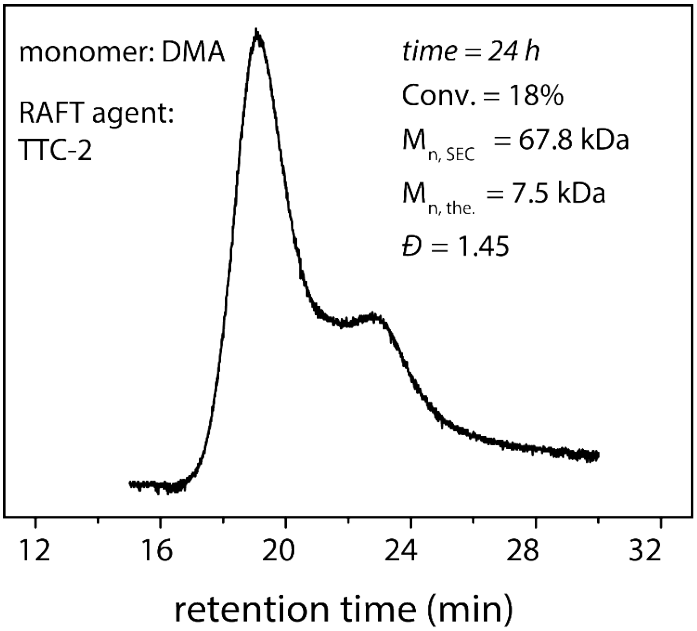

Figure S6. SEC chromatograms of PDMA synthesized via a bulk RAFT polymerization. [Fe(II)-MOF] = 9.6 mg mL^{-1}, and $[H_2O_2]_0$ = 70 mM.
Note: $M_{n, SEC}$ and *Đ* values of this polymer were obtained by using a light scattering detector coupled with SEC through an aqueous system.

- **Effect of MA/DMSO ratio on the polymerization rate**

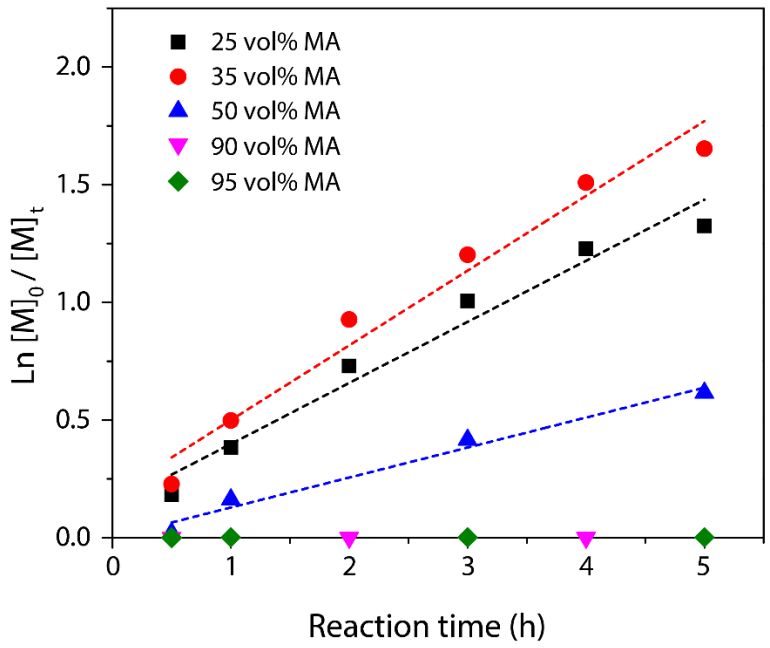

Figure S7. Rate of organic Fenton-RAFT polymerization at different volume percentages of MA in DMSO when targeted DP = 200, $[Fe(II)\text{-}MOF]_0$ = 9.6 mg mL^{-1} and $[H_2O_2]$ = 70 mM.

Table S2. The experimental data obtained from the reactions comparing polymerization rates at different MA/DMSO volume ratios.

MA/DMSO (vol%)	Time (h)	Conversion (%)	Ln $[M]_0/[M]_t$	$M_{n,\,SEC}$ (kDa)	*Đ* (-)
25	0.5	16.7	0.183	-	-
	1	31.9	0.384	3.0	1.19
	2	51.8	0.730	6.6	1.15
	3	63.4	1.006	9.1	1.13
	4	70.7	1.228	10.7	1.11
	5	73.4	1.325	11.8	1.10
35	0.5	20.4	0.228	-	-
	1	39.2	0.498	4.1	1.20
	2	60.4	0.927	8.4	1.14
	3	69.9	1.202	11.2	1.15
	4	77.9	1.509	13.0	1.12
	5	80.9	1.653	13.9	1.11
50	0.5	2	0.020	-	-
	1	15	0.163	-	-
	2	Not taken	Not taken	-	-
	3	34	0.416	3.2	1.34
	4	Not taken	Not taken	-	-
	5	46	0.616	4.3	1.35
90 and 95	0.5	0	0	-	-
	1	0	0	-	-
	2	0	0	-	-
	3	0	0	-	-
	4	0	0	-	-
	5	0	0	-	-

- **Recycling experiments**

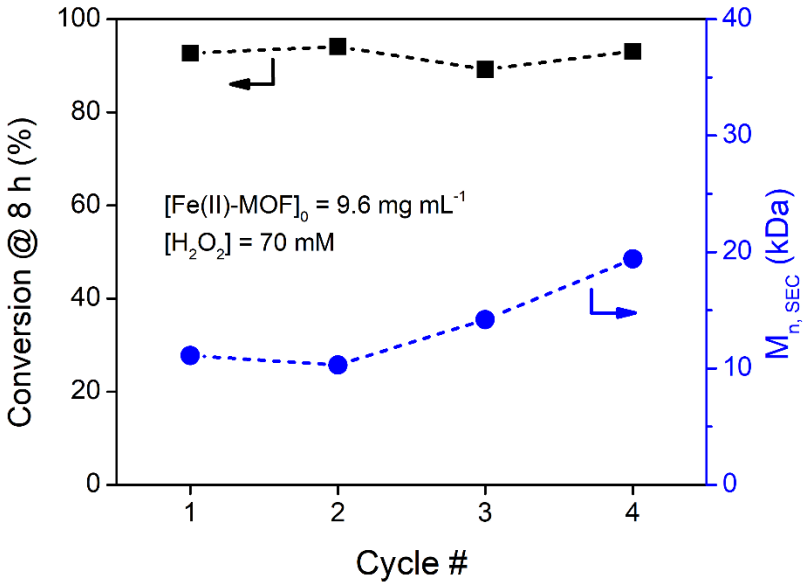

Figure S8. Recyclability of the Fe(II)-MOF particles: monomer conversion after 8 h via recycling runs of Fe(II)-MOF particles in the organic Fenton-RAFT polymerization when CTA was TTC-3 and targeted DP was 200.

Table S3. Characterization data of the synthesized PMA after 8 h *via* the experiments with recovered MOFs when targeted DP = 200, [Fe(II)-MOF]$_0$ = 9.6 mg mL^{-1} and [H_2O_2] = 70 mM.

Cycle #	Conversion (%)	$M_{n, SEC}$ (kDa)	*Đ* (-)
1	92.7	11.1	1.15
2	94.1	10.3	1.11
3	89.2	14.2	1.10
4	93.1	19.4	1.12

- **Temperature effect on the polymerization rate**

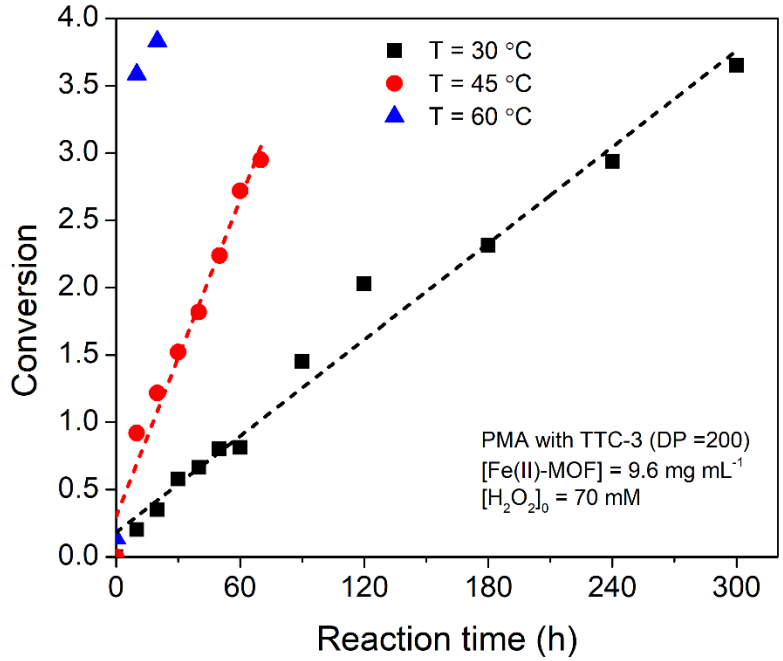

Figure S9. Rate of organic Fenton-RAFT polymerization at different temperatures (T = 30, 45, and 60 °C) when targeted DP = 200, $[Fe(II)\text{-}MOF]_0$ = 9.6 mg mL^{-1} and $[H_2O_2]$ = 70 mM.

Table S4. Characterization data of the synthesized PMA in the experiments with different Ts when targeted CTA is TTC-3, DP = 200, $[Fe(II)\text{-}MOF]_0$ = 9.6 mg mL^{-1}, and $[H_2O_2]$ = 70 mM.

T (°)	Time (min)	Conversion (%)	$M_{n,\,the.}$ (kDa)	$M_{n,\,SEC}$ (kDa)	*Đ* (-)
30	0	0.0	-	-	-
	10	18.3	-	-	-
	20	29.6	5.4	3.0	1.19
	30	43.9	7.8	4.6	1.22
	40	48.5	8.6	6.0	1.21
	50	55.2	9.8	7.5	1.19
	60	55.6	9.9	8.5	1.16
	90	76.6	13.5	11.1	1.18
	120	86.8	15.2	12.8	1.19
	180	90.1	15.8	14.8	1.21
	240	94.7	16.6	16.0	1.22
	300	97.4	17.0	16.7	1.23
45	0	0.0	-	-	-
	10	60.2	10.7	6.7	1.19
	20	70.4	12.4	8.8	1.15
	30	78.2	13.8	10.2	1.15
	40	83.8	14.8	11.0	1.15
	50	89.3	15.7	12.2	1.16
	60	93.4	16.3	13.1	1.21
	70	94.8	16.6	13.5	1.17
60	0	12.7	-	-	-
	10	97.2	17022.122	10802	1.30
	20	97.8	17126.096	10684	1.31

References

1. A. Reyhani, H. Ranji-Burachaloo, T. G. McKenzie, Q. Fu and G. G. Qiao, *Macromolecules*, 2019, **52**, 3278-3287.