

## SI Appendix

**Characterization.** NMR spectra were acquired on a Bruker AV-400 MHz spectrometer.  $^1\text{H}$  NMR data are reported as follows: chemical shift ( $\delta$  ppm), multiplicity (s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet, bs = broad singlet), and the peak integration. It should be mentioned that neither  $^1\text{H}$  nor  $^{13}\text{C}$  NMR data could be collected for **Eu3a**, **Eu3b**, **Gd3a**, and **Gd3b**, due to the paramagnetic nature of these materials. Mass spectra were obtained with an IonSpec ESI mass spectrometer in positive ion mode. IR spectra were measured on a Perkin Elmer Spectrum One Fourier transform infrared spectrometer as KBr pellets. Lanthanide quantification was carried out on a ICP-MS (Octapole detection, Agilent Technologies, Santa Clara, CA), which monitored Gd isotopes (157, 158 Da) and Eu isotopes (151, 153 Da). These data were used to generate a calibration curves for both  $\text{Gd}^{3+}$  and  $\text{Eu}^{3+}$ . Polymers were dissolved and diluted to the ppb range so they could be analyzed within the proper calibration curve range.

### $^1\text{H}$ and $^{13}\text{C}$ NMR Data:

#### **Poly{[ $N^2, N^3, N^4$ -tris(*tert*-butoxycarbonyl)tetraethylenetriamine]**

#### **amidodiethylenetriaminetriaacetic acid}, **2a****

$^1\text{H}$ -NMR ( $d^3$ -MeOD):  $\delta$  = 1.57 (s, 27H), 3.08 (s, 4H), 3.30 (bm, 20H), 3.43 (bs, 6H), 3.69 (s, 4h)

$^{13}\text{C}$ -NMR ( $d^3$ -MeOD):

$\delta$  = 30.21, 37.42, 40.87, 44.33, 46.03, 47.10, 49.02, 50.94, 52.63, 55.52, 57.64, 79.14, 154.95, 155.14, 169.77, 170.89, 173.02.

FTIR (KBr): See Table S1

**Poly{[ $N^2, N^3, N^4, N^5$ -tetrakis(*tert*-butoxycarbonyl)pentaethylenetetramine]amidodiethylenetriaminetriaacetic acid}, 2b**

$^1\text{H-NMR}$  ( $d^3$ -MeOD):  $\delta = 1.57$  (s, 27H), 3.10 (s, 8H), 3.28-3.53 (bm, 20H), 3.43 (bs, 6H), 3.69 (s, 4H).

$^{13}\text{C-NMR}$  ( $d^3$ -MeOD):  $\delta = 28.49, 37.27, 37.79, 40.87, 44.33, 45.04, 45.96, 46.98, 49.02, 50.94, 52.63, 55.52, 57.64, 79.14, 154.95, 155.14, 169.77, 170.89, 173.02$ .

FTIR (KBr): See Table S1

**Poly-tetraethylenepentaamineamidodiethylenetriaminetetraacetic acid, 3a.**

$^1\text{H-NMR}$  ( $\text{D}_2\text{O}$ ):  $\delta = 3.08$  (s, 4H), 3.24-3.50 (bm, 20H), 2.54 (bs, 6H), 3.77 (s, 4H)

$^{13}\text{C NMR}$  ( $\text{D}_2\text{O}$ ):  $\delta = 35.67, 43.90, 44.08, 47.56, 50.93, 53.43, 55.04, 58.72, 171.61, 173.31, 177.64$ .

FTIR (KBr): See Table S1

**Poly-pentaethylenehexamineamidodiethylenetriaminetetraacetic acid, 3b.**

$^1\text{H-NMR}$  ( $\text{D}_2\text{O}$ ):  $\delta = 3.03$  (s, 4H), 3.21-3.49 (bm, 20H), 2.54 (bs, 6H), 3.77 (s, 4H)

$^{13}\text{C NMR}$  ( $\text{D}_2\text{O}$ ):  $\delta = 35.74, 43.86, 44.28, 45.11, 47.34, 50.88, 53.39, 55.00, 58.72, 171.59, 173.29, 177.67$ .

FTIR (KBr): See Table S1