

Manuscript title: Highly Shifted LIPOCEST Agents Based on the Encapsulation of Neutral Polynuclear Paramagnetic Shift Reagents

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Supplementary information

- *Characterization of ligand 1*:

^1H NMR (14.1 T, solvent D_2O , pD 7, 298 K): δ 3.89 (b, 6H), 3.56 (b, 8H), 3.26 (b, 16H), 3.04 (b, 22H), 1.35 (s, 4H), 1.17 (s, 4H).

^{13}C NMR (14.1 T, solvent D_2O , pD 7, 298 K, $\{^1\text{H}\}$): δ 175.0 (b), 171.0 (b), 167.1 (b), 56.7, 54.9 (b), 52.6 (b), 49.8 (b), 41.1, 29.7, 27.2.

ESI-MS: (m+1)/z calculated: 889.4916, found: 889.42

- *Characterization of Tm-1 complex*:

ESI-MS (m+1)/z calculated: 1221.8292, found: 1221.67

- *Characterization of ligand 2*:

^1H NMR (14.1 T, solvent D_2O , pD 7, 298 K): δ 3.90 (b, 2H), 3.85 (b, 1H), 3.78 (b, 2H), 3.74 (b, 9H), 3.68 (d, $J = 10.0$ Hz, 4H), 3.65 (d, $J = 8.6$ Hz, 2H), 3.63 (b, 2H), 3.57 (b, 2H), 3.47 (b, 4H), 3.45-3.38 (b, 5H), 3.30 (b, 20H), 3.21 (b, 5H), 3.12-2.85 (b, 16H).

^{13}C NMR (14.1 T, solvent D_2O , pD 7, 298 K, $\{^1\text{H}\}$) δ 174.0 (b), 173.1 (b), 171.2 (b), 169.8 (b), 166.7, 66.2, 56.8, 56.2, 56.0, 55.0, 54.1 (b), 53.0, 51.5 (b), 50.4, 49.8 (b), 39.3, 39.0.

ESI-MS (m+1)/z calc'd.: 1250.6150, found: 1250.62.

- *Characterization of Tm-2 complex*:

ESI-MS (m+1)/z calculated: 1749.0490, found: 1749.56