Viologen-containing dendrimers (A1-A3) were prepared according to previously reported procedures.\[1\] The following donor containing dendrimers were synthesized via a similar procedure.\[2\] CB8 was prepared according to literature procedures.\[3\]

**Synthesis of 6-(4’-methoxyphenoxy)-1-hexanoic acid (1).** p-Methoxyphenol (6.21g, 50 mmol) and 6-bromo-1-hexanoic acid (9.75g, 50 mmol) were refluxed in 100 mL 10% NaOH aqueous for 3 h. The reaction mixture was poured into 600 ml water. The pH was adjusted using 2M aqueous HCl until the solution was acidic, yielding a white precipitate. The precipitate was collected by filtration and recrystallized from ethanol, affording the target product as a white solid. (Yield 6.51g, 54.7%). 1H NMR (500 MHz, CDCl3): \(\delta\) 6.83(s, 4H), 3.92(t, 2H), 3.78(s, 3H), 2.41(t, 2H), 1.75(m, 4H), 1.55(m, 2H). FAB-MS: m/z 238.

**Synthesis of dendrimer D1E (\(t\)-butyl ester termini).** In 20 mL dry DMF, 6-(4’-methoxyphenoxy)-1-hexanoic acid (524 mg, 2.20 mmol), Behera’s amine (1.00 g, 2.41 mmol), n-HATU (915 mg, 2.41 mmol) and DMAN (Proton Sponge, 943 mg, 4.40 mmol) were stirred under argon overnight. The solvent was removed under reduced pressure. The residue was diluted with 100 mL ethyl acetate, washed with 2 M HCl (2 × 50 ml), then water (2 × 50 ml). The organic layer was dried over anhydrous sodium sulfate, concentrated under vacuum and loaded into a chromatography column (SiO\(_2\), elute with CHCl\(_3\):EtOAc = 4 : 1). The appropriate fraction was dried under vacuum to afford the target compound as a white solid. (Yield: 1.34 g, 95.7%). 1H NMR (400 MHz, CDCl3): \(\delta\) 6.81 (s, 4H), 5.88 (s, 1H), 3.90 (t, \(J = 6\) Hz, 2H), 3.74 (s, 3H), 2.22 (t, \(J = 8\) Hz, 6H), 2.13 (t, \(J = 8\) Hz, 2H), 1.96 (t, \(J = 8\) Hz, 6H), 1.77 (m, 2H), 1.65 (m, 2H), 1.48 (m, 2H), 1.43 (s, 27H). 13C NMR (100 MHz, CDCl3): \(\delta\) 173.04, 172.40, 153.81, 153.31, 115.54, 114.73, 80.76, 68.41, 57.43, 55.84, 37.52, 30.13, 29.95, 29.23, 28.18, 25.89, 25.60. FAB-MS: m/z 636.

**Synthesis of dendrimers D2E and D3E.** The same procedure was used as for D1E (with the corresponding amino functionalized dendron instead of Behera’s amine). Yields: D2E, 68.3%; D3E, 42.5%.

**Dendrimer D2E.** 1H NMR (400 MHz, CDCl3): \(\delta\) 7.66 (s, 1H, -HN), 6.82 (s, 4H), 6.04 (s, 3H, -HN), 3.91 (t, \(J = 6\) Hz, 2H), 3.76 (s, 3H), 2.18 (m, 24H), 1.96 (m, 24H), 1.79 (m, 2H), 1.59 (m, 2H), 1.48 (m, 2H), 1.43 (s, 81H). 13C NMR (100 MHz, CDCl3): \(\delta\) 173.46, 173.02, 172.82, 153.78, 153.42, 115.62, 114.74, 80.72, 77.36, 68.55, 57.59, 55.86, 37.43, 32.15, 31.93, 30.01, 29.95, 29.29, 28.23, 25.97, 25.65. FAB-MS: m/z 1660.
Dendrimer D3E. $^1$H NMR (400 MHz, CDCl$_3$): $\delta$ 6.82 (s, 4H), 6.51-6.30 (b), 3.92 (t, 2H), 3.75 (s, 3H), 2.19 (m, 78H), 1.95 (m, 78H), 1.79 (m, 2H), 1.70 (m, 2H), 1.48 (m, 2H), 1.42 (s, 243H). $^{13}$C NMR (100 MHz, CDCl$_3$): $\delta$ 173.31, 173.05, 172.85, 153.79, 153.37, 115.70, 114.80, 80.52, 77.36, 68.58, 58.20, 57.58, 55.88, 32.05, 31.56, 31.18, 29.92, 29.30, 28.27, 28.06, 26.12. MALDI-TOF MS: m/z 4733.

Synthesis of dendrimers D1-D3. The corresponding t-butyl ester dendrimer D1E-D3E (200 mg) was dissolved in formic acid (10 mL) and the solution was stirred overnight at room temperature. Formic acid was removed under reduced pressure and chased with ethyl acetate. The resulting solid was dried under vacuum to afford the acid-terminated dendrimer in near quantitative yields.

Dendrimer D1. $^1$H NMR (400 MHz, D$_2$O, phosphate buffer, pD = 7): $\delta$ 6.97 (s, 4H), 4.04 (t, $J = 6$ Hz, 2H), 3.80 (s, 3H), 2.21 (m, 8H), 1.93 (m, 6H), 1.75 (m, 2H), 1.64 (m, 2H), 1.45 (m, 2H). $^{13}$C NMR (100 MHz, D$_2$O, phosphate buffer, pD = 7): $\delta$ 181.66, 176.66, 153.72, 152.84, 116.71, 115.54, 69.71, 58.38, 56.30, 36.83, 30.93, 30.48, 28.48, 25.63, 25.26. FAB-MS: m/z 469.

Dendrimer D2. $^1$H NMR (400 MHz, D$_2$O, phosphate buffer, pD = 7): $\delta$ 6.96 (s, 4H), 4.03 (t, $J = 6$ Hz, 2H), 3.79 (s, 3H), 2.21 (m, 26H), 1.95 (m, 24H), 1.77 (m, 2H), 1.65 (m, 2H), 1.47 (m, 2H). $^{13}$C NMR (100 MHz, D$_2$O, phosphate buffer, pD = 7): $\delta$ 177.87, 174.52, 173.31, 151.62, 150.81, 114.58, 113.38, 67.69, 56.06, 56.02, 54.12, 34.57, 28.86, 28.39, 27.65, 26.36, 23.38, 23.06. FAB-MS: m/z 1157.

Dendrimer D3. $^1$H NMR (400 MHz, D$_2$O, phosphate buffer, pD = 7): $\delta$ 6.90 (s, 4H), 3.97 (t, 2H), 3.75 (s, 3H), 2.20 (m, 80H), 1.96 (m, 78H), 1.74 (m, 2H), 1.65 (m, 2H), 1.45 (m, 2H). $^{13}$C NMR (100 MHz, D$_2$O, phosphate buffer, pD = 7): $\delta$ 178.79, 175.22, 153.50, 152.66, 116.24, 115.21, 69.23, 57.94, 57.81, 55.93, 31.02, 30.73, 30.25, 29.22, 28.92, 28.29, 25.14. MALDI-TOF MS: (D3$^+$Na$^-$) 3242.

References:

Figure S11. Electronic absorption spectra of a solution containing 0.5 mM A1, 0.25 mM D3, 0.25 mM CB8 and Na$_3$PO$_4$ buffered at pH 7.0 ([Na$^+$] = 0.1 M) before (dotted line) and after (solid line) reduction with Zn metal. Reduction in the absence of CB8 leads to the dashed line spectrum. Optical path = 0.1 cm.