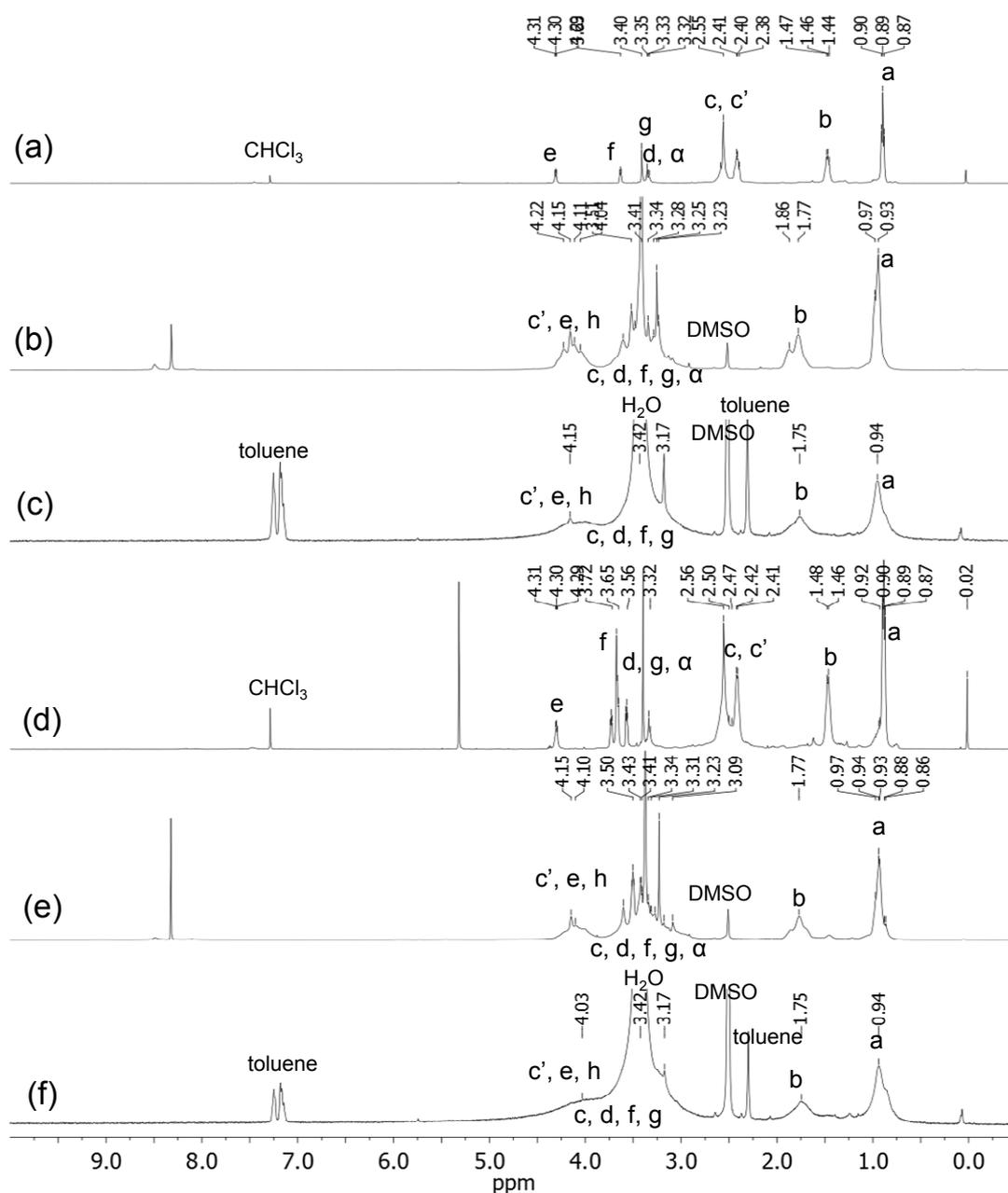


Supporting Information

Figure S1. $^1\text{H-NMR}$ spectra of (a) **7** in CDCl_3 ; (b) **9** in $\text{DMSO-}d_6$; (c) **1** in $\text{DMSO-}d_6$ -toluene- d_8 ; (d) **8** in CDCl_3 ; (e) **10** in $\text{DMSO-}d_6$; and (f) **2** in $\text{DMSO-}d_6$ -toluene- d_8 .



All proton peaks of compounds **7**, **9**, **1** (Figure S1) were assigned with the corresponding proton numbers indicated on the compound structures shown in Scheme 1. Clear chemical shift assignment of all protons in the structure methoxyethyleneglycol- $[N,N',N,N,N,N$ -hexapropyl-hexa(aminoethyl)amino]-malonamide ester, $\text{ME}_1\text{N}_6\text{C}_3$ (**7**) was used to correlate the chemical shift changes of $\text{ME}_1\text{N}_6^+\text{C}_3$ (**9**) and $\text{C}_{60}>(\text{ME}_1\text{N}_6^+\text{C}_3)$ (**1**) upon $\text{CH}_3\text{-I}$ quaternization and C_{60} reaction. Accordingly, we assigned the peak at δ 0.89 (H_a), 1.44 (H_b), 2.38–2.53 (H_c and $\text{H}_{c'}$) of **7** to the chemical shift of methyl protons $\text{CH}_3\text{-CH}_2\text{-CH}_2\text{-N-}$, methylene protons $\text{CH}_3\text{-CH}_2\text{-CH}_2\text{-N-}$, and N -attached ethyleneamino $\text{-CH}_2\text{-}$ protons; two peaks at δ 3.32 (H_d) and 4.29 (H_e) to the chemical shift of methylene protons of $\text{-CH}_2\text{-NH-C=O}$ and

$-\text{CH}_2\text{-O-C=O}$, respectively; and two peaks at δ 3.61 (H_f) and 3.39 (H_g) to the methylene and methyl protons of $-\text{CH}_2\text{-O-}$ and $\text{CH}_3\text{-O-}$, respectively. The chemical shift of malonate α -proton was found to be δ 3.33 as a singlet. Quaternization reaction of $\text{ME}_1\text{N}_6\text{C}_3$ (**7**) with iodomethane to $\text{ME}_1\text{N}_6^+\text{C}_3$ (**9**) resulted in the introduction of methylated quaternary amine arm in the structure that caused a large down-field shift of chemical shifts (more than 1.0 ppm) of all *N*-attached ethyleneamino $-\text{CH}_2-$ protons at δ 2.38–2.53 to δ 3.22–4.25 (H_c , $\text{H}_{c'}$, and H_h) for quaternary ammonium methylene ($-\text{N}^+-\text{CH}_2-$), and additional methyl protons ($-\text{N}^+-\text{CH}_3$), with the overlap of H_d , H_e , H_f , H_g , and H_a , as a broad band. Chemical shifts of all methyl or methylene proton peaks of $\text{CH}_3\text{-CH}_2\text{-CH}_2\text{-N}^+$ and $\text{CH}_3\text{-CH}_2\text{-CH}_2\text{-N}^+$ were also down-field shifted to δ 0.95 (H_a) and δ 1.50–1.85 (H_b), respectively.

Similar chemical shifts were made for assignments of all proton peaks of $\text{ME}_3\text{N}_6\text{C}_3$ (**8**) to those of $\text{ME}_1\text{N}_6\text{C}_3$ (**7**), as shown in Figure S1, except more complicated peaks at δ 3.55–3.71 for the methylene protons of $-\text{CH}_2\text{-O-}$ (H_f). Other peaks are almost identical, those of all methyl or methylene proton peaks in the moieties of $\text{CH}_3\text{-O-}$, $\text{CH}_3\text{-CH}_2\text{-CH}_2\text{-N-}$, $\text{CH}_3\text{-CH}_2\text{-CH}_2\text{-N-}$ and $-\text{CH}_2\text{-N-}$ to δ 3.38 (H_g), 0.90 (H_a), δ 1.47 (H_b), and 2.40–2.54 (H_c and $\text{H}_{c'}$) respectively. After methylated quaternization and the reaction with C_{60} , ^1H NMR spectrum of $\text{C}_{60}(>\text{ME}_3\text{N}_6^+\text{C}_3)$ (**2**) showed nearly identical chemical shifts as those of $\text{C}_{60}(>\text{ME}_1\text{N}_6^+\text{C}_3)$ (**1**), except a slight shift of chemical shifts of all methyl or methylene proton peaks in the moieties of $\text{CH}_3\text{-CH}_2\text{-CH}_2\text{-N-}$, $\text{CH}_3\text{-CH}_2\text{-CH}_2\text{-N-}$, $-\text{CH}_2\text{-N-}$ ($\text{CH}_3\text{-N}^+$) to δ 0.94 (H_a), δ 1.75 (H_b), and δ 2.90–4.20 (H_c , $\text{H}_{c'}$, H_h), respectively.

Figure S2. MALDI-TOF mass spectra of $\text{C}_{60}(>\text{ME}_1\text{N}_6^+\text{C}_3)$ (**1**) using sinapic acid as the matrix.

