

Supporting Information

Tuning the thermotropic properties of liquid crystalline *p*-substituted aroylhydrazones

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1. General methods

Ethyl 4-aminobenzoate, 4-hydroxybenzaldehyde, 3,4-dihydroxybenzaldehyde, 3,5-dihydroxybenzaldehyde, 2,3,4-trihydroxybenzaldehyde, 3,4,5-trihydroxybenzaldehyde, hydrazine monohydrate, potassium iodide, anhydrous K_2CO_3 , bromoalkanes purchased from Aldrich Chemicals, USA were used as received. All other solvents and reagents were purchased from Merck, India. The solvents were dried using standard methods when required [S1]. Elemental analyses were performed on a CE-440 Exeter Analytical CHN analyser. IR spectra ($4000\text{--}100\text{ cm}^{-1}$) were recorded on a Varian 3100 FT-IR Excalibur series spectrophotometer. 1H and ^{13}C NMR spectra were obtained on a JEOL FT-NMR AL 300 MHz spectrometer using tetramethylsilane as the internal standard. Differential scanning calorimetry (DSC) thermograms were recorded with a Mettler Toledo TC 15 TA differential scanning calorimeter at the rate of 5.0 K min^{-1} under a nitrogen atmosphere using spec pure grade indium as standard by taking samples in close-lid aluminium pans. The transition temperatures from DSC thermograms have been determined with an accuracy of $\pm 0.1\text{ K}$. The mesophase type was identified by visual comparison with known phase standards using a Nikon Eclipse LV-100 POL polarizing optical microscope (POM) fitted with a hot stage temperature-controlled LTSE 420 heating stage (Linkam Scientific Instruments, Tadworth, Surrey, UK) with temperature controlling accuracy of 0.1 K . The GAUSSIAN-09 program [S2] package was employed to carry out DFT calculations at the Becke's three parameter functionals and Lee, Yang and Parr correlation functionals (B3LYP) [S3,4]. The internal coordinates of the system, which is used as input for GAUSSIAN-09 program was generated by the GAUSS VIEW 4.1 program [S5]. The temperature dependent Raman spectra were measured on a micro-Raman setup, Renishaw, UK equipped with a grating of $2400\text{ lines mm}^{-1}$ and CCD. The 532 nm exciting line from Nd:YAG laser was used as an excitation source. The sample was kept in a quartz sample holder which was put on a temperature-controlled THM 600 hot stage (Linkam Scientific Instruments, Tadworth, Surrey, UK) below the Lieca $50\times$ long distance microscope objective. The accuracy in the measurement of temperature was $\pm 0.1^\circ\text{C}$. The Raman scattered light was collected in the back-scattering geometry. The software used for collecting the spectra was *WiRE-4*. The recorded data were analyzed using *Spectra Calc* and *Origin* software. Variable temperature powder X-ray diffraction PXRD studies were carried out using an image plate detector (Mac Science, Japan) equipped with double mirror focusing optics, with the sample contained in a Lindmann capillary tube.

2. Synthesis and analytical data

2.1 Synthesis of *N*-(4-(*pentyl*oxy)benzylidene)-4-{4'-(*pentyl*oxy)benzylidene}amino}benzohydrazide (1, $n=5$)

The compound 1 ($n=5$) was prepared by refluxing together absolute ethanolic solution of 4-pentylxybenzaldehyde (10 mmol, 1.92 g) and 4-aminobenzohydrazide (10 mmol, 1.51 g) for $\sim 8\text{ h}$ in the presence of few drops of acetic acid and leaving solution overnight in the flask closed with guard tube. The microcrystalline white coloured solid was filtered off by suction, thoroughly washed with cold ethanol and recrystallised from a mixture of absolute ethanol and chloroform (1:2 v/v) and dried at room temperature.

All the other members of the series were prepared in a similar manner. The yield, IR, NMR and elemental data for the compounds are summarized as follows.

Yield: 80 %. IR (KBr, cm^{-1}): 3286 ν_s (N-H), 2918, 2852 (aliphatic C-H), 1649 (amide-I, C=O), 1578 (amide-II, N-H), 1628 (-C=N), 1609, 1512 (Ph), 1303, 1255 (OPh), 1029 (N-N); ^1H NMR (300 MHz, DMSO- d_6 , 60°C): δ 11.50 (s, 1H, -NH), 8.55 (s, 1H, -CH=N), 8.39 (s, 1H, -CH=N), 7.96 (dd, $J_1(\text{H,H})=8.1$ Hz, $J_2(\text{H,H})=8.7$ Hz, 4H, -C₆H₄), 7.65 (d, $J_1(\text{H,H})=8.1$ Hz, 2H, -C₆H₄), 7.31 (d, $J_1(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.08 (dd, $J_1(\text{H,H})=8.7$ Hz, $J_2(\text{H,H})=8.7$ Hz, 4H, -C₆H₄), 4.09 (m, 4H, $J_1(\text{H,H})=6.3$ Hz, -OCH₂), 1.75 (m, 4H, -OCH₂CH₂), 1.40 (m, 8H, -[CH₂]₄), 0.93 (t, $J_1(\text{H,H})=6.3$ Hz, 6H, -CH₃); ^{13}C NMR (75 MHz, DMSO- d_6 , 60°C): δ_c 176.95, 161.34, 160.22, 154.14, 147.09, 130.17, 130.08, 128.36, 127.98, 126.68, 121.42, 119.98, 115.36, 114.49, 67.53, 30.65, 28.09, 24.85, 21.36, 13.07; Elemental analyses: calculated for C₃₁H₃₇N₃O₃ (%), C, 74.52; H, 7.46; N, 8.41; Found, C, 74.44; H, 7.39; N, 8.45.

2.2 Synthesis of *N*-(4-(hexyloxy)benzylidene)-4-*l*-(4'-(hexyloxy)benzylidene)amino}benzohydrazide (*l*, *n*=6)

Yield: 82 %. IR (KBr, cm^{-1}): 3285 ν_s (N-H), 2918, 2852 (aliphatic C-H), 1650 (amide-I, C=O), 1577 (amide-II, N-H), 1629 (-C=N), 1609, 1512 (Ph), 1304, 1253 (OPh), 1028 (N-N); ^1H NMR (300 MHz, DMSO- d_6 , 60°C): δ 11.47 (s, 1H, -NH), 8.55 (s, 1H, -CH=N), 8.40 (s, 1H, -CH=N), 7.96 (dd, $J_1(\text{H,H})=8.1$ Hz, $J_2(\text{H,H})=8.7$ Hz, 4H, -C₆H₄), 7.66 (d, $J_1(\text{H,H})=8.1$ Hz, 2H, -C₆H₄), 7.31 (d, $J_1(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.08 (dd, $J_1(\text{H,H})=8.7$ Hz, $J_2(\text{H,H})=8.7$ Hz, 4H, -C₆H₄), 4.09 (m, 4H, -OCH₂), 1.76 (m, 4H, -OCH₂CH₂), 1.40 (m, 12H, -[CH₂]₆), 0.91 (t, $J_1(\text{H,H})=6.3$ Hz, 6H, -CH₃); ^{13}C NMR (75 MHz, DMSO- d_6 , 60°C): δ_c 176.95, 161.35, 160.22, 154.15, 147.09, 130.17, 130.09, 128.36, 127.98, 126.69, 121.42, 119.98, 115.35, 114.50, 67.52, 30.65, 28.10, 24.85, 21.37, 13.07; Elemental analyses: calculated for C₃₃H₄₁N₃O₃ (%), C, 75.11; H, 7.83; N, 7.96; Found, C, 75.01; H, 7.78; N, 7.92.

2.3 Synthesis of *N*-(4-(heptyloxy)benzylidene)-4-*l*-(4'-(heptyloxy)benzylidene)amino}benzohydrazide (*l*, *n*=7)

Yield: 79 %. IR (KBr, cm^{-1}): 3286 ν_s (N-H), 2920, 2851 (aliphatic C-H), 1650 (amide-I, C=O), 1576 (amide-II, N-H), 1628 (-C=N), 1610, 1513 (Ph), 1305, 1251 (OPh), 1028 (N-N); ^1H NMR (300 MHz, DMSO- d_6 , 60°C): δ 11.45 (s, 1H, -NH), 8.56 (s, 1H, -CH=N), 8.40 (s, 1H, -CH=N), 7.96 (dd, $J_1(\text{H,H})=8.1$ Hz, $J_2(\text{H,H})=8.7$ Hz, 4H, -C₆H₄), 7.65 (d, $J_1(\text{H,H})=8.1$ Hz, 2H, -C₆H₄), 7.30 (d, $J_1(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.08 (dd, $J_1(\text{H,H})=8.7$ Hz, $J_2(\text{H,H})=8.7$ Hz, 4H, -C₆H₄), 4.09 (m, 4H, $J_1(\text{H,H})=6.3$ Hz, -OCH₂), 1.76 (m, 4H, -OCH₂CH₂), 1.40 (m, 16H, -[CH₂]₈), 0.90 (t, $J_1(\text{H,H})=6.3$ Hz, 6H, -CH₃); ^{13}C NMR (75 MHz, DMSO- d_6 , 60°C): δ_c 176.94, 161.35, 160.21, 154.15, 147.10, 130.18, 130.09, 128.37, 127.98, 126.69, 121.42, 119.99, 115.35, 114.50, 67.52, 30.64, 28.10, 24.86, 21.37, 13.07; Elemental analyses: calculated for C₃₅H₄₅N₃O₃ (%), C, 75.64; H, 8.16; N, 7.56; Found, C, 75.58; H, 8.09; N, 7.49.

2.4 Synthesis of *N*-(4-(octyloxy)benzylidene)-4-*l*-(4'-(octyloxy)benzylidene)amino}benzohydrazide (*l*, *n*=8)

Yield: 83 %. IR (KBr, cm^{-1}): 3285 ν_s (N-H), 2920, 2853 (aliphatic C-H), 1648 (amide-I, C=O), 1578 (amide-II, N-H), 1630 (-C=N), 1610, 1512 (Ph), 1304, 1251 (OPh), 1029 (N-N); ^1H NMR (300 MHz, DMSO- d_6 , 60°C): δ 11.50 (s, 1H, -NH), 8.55 (s, 1H, -CH=N), 8.39 (s, 1H, -CH=N), 7.97 (dd, $J_1(\text{H,H})=8.1$ Hz, $J_2(\text{H,H})=8.7$ Hz, 4H, -C₆H₄), 7.65 (d, $J_1(\text{H,H})=8.1$ Hz, 2H, -C₆H₄), 7.30 (d, $J_1(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.10 (dd, $J_1(\text{H,H})=8.7$ Hz, $J_2(\text{H,H})=8.7$ Hz, 4H, -C₆H₄), 4.08 (m, 4H, -OCH₂), 1.76 (m, 4H, -OCH₂CH₂), 1.41 (m, 20H, -[CH₂]₁₀), 0.93 (t, $J_1(\text{H,H})=6.3$ Hz, 6H, -CH₃); ^{13}C NMR (75 MHz, DMSO- d_6 , 60°C): δ_c 176.96, 161.34, 160.21, 154.16, 147.10, 130.18, 130.10, 128.37, 127.99, 126.69, 121.43, 119.99, 115.37, 114.48, 67.52, 30.64, 28.08, 24.87, 21.37,

13.08; Elemental analyses: calculated for C₃₇H₄₉N₃O₃ (%), C, 76.12; H, 8.46; N, 7.20; Found, C, 76.01; H, 8.37; N, 7.18.

2.5 Synthesis of *N*-(4-(decyloxy)benzylidene)-4-*l*-(4'-(decyloxy)benzylidene)amino}benzohydrazide (*l*, *n*=10)

Yield: 85 %. IR (KBr, cm⁻¹): 3288 ν_s (N-H), 2918, 2851 (aliphatic C-H), 1650 (amide-I, C=O), 1578 (amide-II, N-H), 1628 (-C=N), 1609, 1510 (Ph), 1303, 1253 (OPh), 1029 (N-N); ¹H NMR (300 MHz, DMSO-d₆, 60°C): δ 11.31 (s, 1H, -NH), 8.54 (s, 1H, -CH=N), 8.40 (s, 1H, -CH=N), 7.96 (dd, J₁(H,H)=8.1 Hz, J₂(H,H)=8.7 Hz, 4H, -C₆H₄), 7.64 (d, J(H,H)=8.1 Hz, 2H, -C₆H₄), 7.29 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 7.06 (dd, J₁(H,H)=8.7 Hz, J₂(H,H)=8.7 Hz, 4H, -C₆H₄), 4.08 (m, 4H, -OCH₂), 1.78 (m, 4H, -OCH₂CH₂), 1.42-1.28 (m, 28H, -[CH₂]₁₄), 0.87 (t, J(H,H)=6.3 Hz, 6H, -CH₃); ¹³C NMR (75 MHz, DMSO-d₆, 60°C): δ_c 176.95, 161.34, 160.22, 154.14, 147.09, 130.17, 130.08, 128.36, 127.98, 126.68, 121.42, 119.98, 115.35, 114.55, 114.49, 67.52, 30.64, 28.08, 24.87, 21.37, 13.08; Elemental analyses: calculated for C₄₁H₅₇N₃O₃ (%), C, 76.95; H, 8.98; N, 6.57; Found, C, 76.88; H, 8.89; N, 6.53.

2.6 Synthesis of *N*-(4-(dodecyloxy)benzylidene)-4-*l*-(4'-(dodecyloxy)benzylidene)amino}benzohydrazide (*l*, *n*=12)

Yield: 81 %. IR (KBr, cm⁻¹): 3284 ν_s (N-H), 2917, 2850 (aliphatic C-H), 1650 (amide-I, C=O), 1576 (amide-II, N-H), 1628 (-C=N), 1611, 1512 (Ph), 1303, 1252 (OPh), 1028 (N-N); ¹H NMR (300 MHz, DMSO-d₆, 60°C): δ 11.44 (s, 1H, -NH), 8.56 (s, 1H, -CH=N), 8.40 (s, 1H, -CH=N), 7.96 (dd, J₁(H,H)=8.1 Hz, J₂(H,H)=8.7 Hz, 4H, -C₆H₄), 7.64 (d, J(H,H)=8.1 Hz, 2H, -C₆H₄), 7.30 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 7.08 (dd, J₁(H,H)=8.7 Hz, J₂(H,H)=8.7 Hz, 4H, -C₆H₄), 4.09 (m, 4H, -OCH₂), 1.78 (m, 4H, -OCH₂CH₂), 1.41-1.28 (m, 36H, -[CH₂]₁₈), 0.89 (t, J(H,H)=6.3 Hz, 6H, -CH₃); ¹³C NMR (75 MHz, DMSO-d₆, 60°C): δ_c 176.97, 161.35, 160.20, 154.16, 147.10, 130.18, 130.10, 128.37, 127.96, 126.69, 121.43, 119.97, 115.36, 114.51, 114.48, 67.52, 30.64, 28.10, 24.87, 21.37, 13.09; Elemental analyses: calculated for C₄₅H₆₅N₃O₃ (%), C, 77.65; H, 9.41; N, 6.04; Found, C, 77.57; H, 9.38; N, 5.99.

2.7 Synthesis of *N*-(4-(tetradecyloxy)benzylidene)-4-*l*-(4'-(tetradecyloxy)benzylidene)amino}benzohydrazide (*l*, *n*=14)

Yield: 78 %. IR (KBr, cm⁻¹): 3287 ν_s (N-H), 2919, 2851 (aliphatic C-H), 1648 (amide-I, C=O), 1580 (amide-II, N-H), 1630 (-C=N), 1612, 1509 (Ph), 1305, 1255 (OPh), 1027 (N-N); ¹H NMR (300 MHz, DMSO-d₆, 60°C): δ 11.46 (s, 1H, -NH), 8.55 (s, 1H, -CH=N), 8.39 (s, 1H, -CH=N), 7.97 (dd, J₁(H,H)=8.1 Hz, J₂(H,H)=8.7 Hz, 4H, -C₆H₄), 7.67 (d, J(H,H)=8.1 Hz, 2H, -C₆H₄), 7.30 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 7.08 (dd, J₁(H,H)=8.7 Hz, J₂(H,H)=8.7 Hz, 4H, -C₆H₄), 4.07 (m, 4H, -OCH₂), 1.77 (m, 4H, -OCH₂CH₂), 1.42-1.29 (m, 44H, -[CH₂]₂₂), 0.90 (t, J(H,H)=6.3 Hz, 6H, -CH₃); ¹³C NMR (75 MHz, DMSO-d₆, 60°C): δ_c 176.97, 161.36, 160.20, 154.13, 147.10, 130.18, 130.10, 128.37, 127.99, 126.69, 121.43, 119.97, 115.35, 114.47, 67.55, 30.64, 28.10, 24.87, 21.37, 13.09; Elemental analyses: calculated for C₄₉H₇₃N₃O₃ (%), C, 78.25; H, 9.78; N, 5.59; Found, C, 78.17; H, 9.69; N, 5.52.

2.8 Synthesis of *N*-(4-(hexadecyloxy)benzylidene)-4-*l*-(4'-(hexadecyloxy)benzylidene)amino}benzohydrazide (*l*, *n*=16)

Yield: 79 %. IR (KBr, cm⁻¹): 3285 ν_s (N-H), 2918, 2852 (aliphatic C-H), 1648 (amide-I, C=O), 1579 (amide-II, N-H), 1628 (-C=N), 1610, 1511 (Ph), 1304, 1254 (OPh), 1028 (N-N); ¹H NMR (300 MHz, DMSO-d₆, 60°C): δ

11.48 (s, 1H, -NH), 8.56 (s, 1H, -CH=N), 8.40 (s, 1H, -CH=N), 7.97 (dd, $J_1(\text{H,H})=8.1$ Hz, $J_2(\text{H,H})=8.7$ Hz, 4H, -C₆H₄), 7.67 (d, $J(\text{H,H})=8.1$ Hz, 2H, -C₆H₄), 7.30 (d, $J(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.10 (dd, $J_1(\text{H,H})=8.7$ Hz, $J_2(\text{H,H})=8.7$ Hz, 4H, -C₆H₄), 4.07 (m, 4H, -OCH₂), 1.78 (m, 4H, -OCH₂CH₂), 1.41-1.28 (m, 52H, -[CH₂]₂₆), 0.91 (t, $J(\text{H,H})=6.3$ Hz, 6H, -CH₃); ¹³C NMR (75 MHz, DMSO-d₆, 60°C): δ_c 176.97, 161.36, 160.21, 154.16, 147.10, 130.18, 130.10, 128.39, 127.99, 126.69, 121.45, 119.97, 115.35, 114.47, 67.55, 30.67, 28.10, 24.84, 21.37, 13.08; Elemental analyses: calculated for C₅₃H₈₁N₃O₃ (%), C, 78.76; H, 10.10; N, 5.20; Found, C, 78.73; H, 10.01; N, 5.15.

2.9 Synthesis of N-(3,4-bis(pentyloxy)benzylidene)-4-((3',4'-bis(pentyloxy)benzylidene) amino)benzohydrazide (2, n=5)

Yield: 77 %. IR (KBr, cm⁻¹): 3208 ν_s (N-H), 2920, 2850 (aliphatic C-H), 1641 (amide-I, C=O), 1576 (amide-II, N-H), 1624 (-C=N), 1599, 1513 (Ph), 1303, 1273 (OPh), 1021 (N-N); ¹H NMR (300 MHz, CDCl₃/DMSO-d₆, 30°C): δ 11.15 (s, 1H, -NH), 8.48 (s, 1H, -CH=N), 8.36 (s, 1H, -CH=N), 7.94 (d, $J_1(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.55 (s, 2H, -C₆H₃), 7.46 (d, $J_1(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.30 (dd, $J_1(\text{H,H})=9.0$ Hz, $J_2(\text{H,H})=8.1$ Hz, 2H, -C₆H₃), 7.08 (dd, $J_1(\text{H,H})=8.4$ Hz, $J_2(\text{H,H})=8.1$ Hz, 2H, -C₆H₃), 4.08 (m, 8H, $J_1(\text{H,H})=6.3$ Hz, -OCH₂), 1.78 (m, 8H, -OCH₂CH₂), 1.45-1.27 (m, 16H, -[CH₂]₈), 0.84 (t, $J_1(\text{H,H})=6.3$ Hz, 12H, -CH₃); ¹³C NMR (75 MHz, CDCl₃, 40°C): δ_c 164.70, 160.61, 154.57, 152.38, 151.85, 151.83, 149.43, 129.60, 129.09, 124.59, 120.51, 112.16, 110.98, 110.96, 105.47, 69.13, 68.70, 31.91, 29.61, 29.35, 25.98, 22.69, 14.13; Elemental analyses: calculated for C₄₁H₅₇N₃O₅ (%), C, 73.29; H, 8.55; N, 6.25; Found, C, 73.21; H, 8.51; N, 6.20.

2.10 Synthesis of N-(3,4-bis(hexyloxy)benzylidene)-4-((3',4'-bis(hexyloxy)benzylidene) amino)benzohydrazide (2, n=6)

Yield: 79 %. IR (KBr, cm⁻¹): 3206 ν_s (N-H), 2921, 2850 (aliphatic C-H), 1640 (amide-I, C=O), 1578 (amide-II, N-H), 1626 (-C=N), 1600, 1515 (Ph), 1303, 1273 (OPh), 1020 (N-N); ¹H NMR (300 MHz, CDCl₃/DMSO-d₆, 30°C): δ 11.15 (s, 1H, -NH), 8.47 (s, 1H, -CH=N), 8.36 (s, 1H, -CH=N), 7.95 (d, $J_1(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.55 (s, 2H, -C₆H₃), 7.47 (d, $J_1(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.30 (dd, $J_1(\text{H,H})=9.0$ Hz, $J_2(\text{H,H})=8.1$ Hz, 2H, -C₆H₃), 7.08 (dd, $J_1(\text{H,H})=8.4$ Hz, $J_2(\text{H,H})=8.1$ Hz, 2H, -C₆H₃), 4.09 (m, 8H, $J_1(\text{H,H})=6.3$ Hz, -OCH₂), 1.78 (m, 8H, -OCH₂CH₂), 1.46-1.27 (m, 24H, -[CH₂]₁₂), 0.84 (t, $J_1(\text{H,H})=6.3$ Hz, 12H, -CH₃); ¹³C NMR (75 MHz, CDCl₃, 40°C): δ_c 164.70, 160.62, 154.56, 152.39, 151.82, 151.83, 149.41, 129.59, 129.09, 124.59, 120.52, 112.17, 110.99, 110.94, 105.47, 69.12, 68.71, 31.91, 29.61, 29.35, 25.98, 22.69, 14.13; Elemental analyses: calculated for C₄₅H₆₅N₃O₅ (%), C, 74.24; H, 9.01; N, 5.77; Found, C, 74.13; H, 8.91; N, 5.69.

2.11 Synthesis of N-(3,4-bis(heptyloxy)benzylidene)-4-((3',4'-bis(heptyloxy)benzylidene) amino)benzohydrazide (2, n=7)

Yield: 80 %. IR (KBr, cm⁻¹): 3210 ν_s (N-H), 2920, 2852 (aliphatic C-H), 1642 (amide-I, C=O), 1580 (amide-II, N-H), 1625 (-C=N), 1598, 1515 (Ph), 1305, 1273 (OPh), 1020 (N-N); ¹H NMR (300 MHz, CDCl₃/DMSO-d₆, 30°C): δ 11.12 (s, 1H, -NH), 8.47 (s, 1H, -CH=N), 8.34 (s, 1H, -CH=N), 7.95 (d, $J_1(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.55 (s, 2H, -C₆H₃), 7.47 (d, $J_1(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.31 (dd, $J_1(\text{H,H})=9.0$ Hz, $J_2(\text{H,H})=8.1$ Hz, 2H, -C₆H₃), 7.08 (dd, $J_1(\text{H,H})=8.4$ Hz, $J_2(\text{H,H})=8.1$ Hz, 2H, -C₆H₃), 4.09 (m, 8H, $J_1(\text{H,H})=6.3$ Hz, -OCH₂), 1.78 (m, 8H, -OCH₂CH₂), 1.46-1.27 (m, 32H, -[CH₂]₁₆), 0.83 (t, $J_1(\text{H,H})=6.3$ Hz, 12H, -CH₃); ¹³C NMR (75 MHz, CDCl₃, 40°C): δ_c 164.71, 160.62, 154.57, 152.37, 151.83, 149.41, 129.60, 129.09, 124.57, 120.52, 112.16, 110.99,

110.96, 105.47, 69.12, 68.70, 31.91, 29.61, 29.35, 25.98, 22.69, 14.13; Elemental analyses: calculated for $C_{49}H_{73}N_3O_5$ (%), C, 75.06; H, 9.38; N, 5.36; Found, C, 74.96; H, 9.33; N, 5.31.

2.12 Synthesis of N-(3,4-bis(octyloxy)benzylidene)-4-((3',4'-bis(octyloxy)benzylidene)amino)benzohydrazide (2, n=8)

Yield: 76 %. IR (KBr, cm^{-1}): 3208 ν_s (N-H), 2922, 2850 (aliphatic C-H), 1644 (amide-I, C=O), 1577 (amide-II, N-H), 1626 (-C=N), 1598, 1515 (Ph), 1304, 1275 (OPh), 1020 (N-N); 1H NMR (300 MHz, $CDCl_3/DMSO-d_6$, 30°C): δ 11.13 (s, 1H, -NH), 8.50 (s, 1H, -CH=N), 8.34 (s, 1H, -CH=N), 7.93 (d, $J(H,H)=8.4$ Hz, 2H, -C₆H₄), 7.55 (s, 2H, -C₆H₃), 7.47 (d, $J(H,H)=8.4$ Hz, 2H, -C₆H₄), 7.31 (dd, $J_1(H,H)=9.0$ Hz, $J_2(H,H)=8.1$ Hz, 2H, -C₆H₃), 7.09 (dd, $J_1(H,H)=8.4$ Hz, $J_2(H,H)=8.1$ Hz, 2H, -C₆H₃), 4.09 (m, 8H, -OCH₂), 1.78 (m, 8H, -OCH₂CH₂), 1.45-1.26 (m, 40H, -[CH₂]₂₀), 0.83 (t, $J(H,H)=6.3$ Hz, 12H, -CH₃); ^{13}C NMR (75 MHz, $CDCl_3$, 40°C): δ_c 164.70, 160.63, 154.57, 152.39, 151.83, 149.41, 129.57, 129.09, 124.58, 120.53, 112.16, 110.99, 110.94, 105.47, 69.11, 68.70, 31.92, 29.61, 29.35, 25.98, 22.69, 14.13; Elemental analyses: calculated for $C_{53}H_{81}N_3O_5$ (%), C, 75.76; H, 9.72; N, 5.00; Found, C, 75.68; H, 9.68; N, 4.96.

2.13 Synthesis of N-(3,4-bis(decyloxy)benzylidene)-4-((3',4'-bis(decyloxy)benzylidene)amino)benzohydrazide (2, n=10)

Yield: 82 %. IR (KBr, cm^{-1}): 3204 ν_s (N-H), 2920, 2850 (aliphatic C-H), 1640 (amide-I, C=O), 1576 (amide-II, N-H), 1624 (-C=N), 1597, 1513 (Ph), 1303, 1273 (OPh), 1020 (N-N); 1H NMR (300 MHz, $CDCl_3/DMSO-d_6$, 30°C): δ 11.15 (s, 1H, -NH), 8.48 (s, 1H, -CH=N), 8.35 (s, 1H, -CH=N), 7.94 (d, $J(H,H)=8.4$ Hz, 2H, -C₆H₄), 7.54 (s, 2H, -C₆H₃), 7.46 (d, $J(H,H)=8.4$ Hz, 2H, -C₆H₄), 7.30 (dd, $J_1(H,H)=9.0$ Hz, $J_2(H,H)=8.1$ Hz, 2H, -C₆H₃), 7.07 (dd, $J_1(H,H)=8.4$ Hz, $J_2(H,H)=8.1$ Hz, 2H, -C₆H₃), 4.08 (m, 8H, -OCH₂), 1.77 (m, 8H, -OCH₂CH₂), 1.45-1.27 (m, 56H, -[CH₂]₂₈), 0.85 (t, $J(H,H)=6.3$ Hz, 12H, -CH₃); ^{13}C NMR (75 MHz, $CDCl_3$, 40°C): δ_c 164.70, 160.62, 154.57, 152.38, 151.84, 151.83, 149.41, 129.60, 129.09, 124.58, 120.52, 112.16, 110.99, 110.96, 105.47, 69.12, 68.70, 31.91, 29.61, 29.35, 25.98, 22.69, 14.13; Elemental analyses: calculated for $C_{61}H_{97}N_3O_5$ (%), C, 76.92; H, 10.27; N, 4.41; Found, C, 76.81; H, 10.19; N, 4.38.

2.14 Synthesis of N-(3,4-bis(dodecyloxy)benzylidene)-4-((3',4'-bis(dodecyloxy)benzylidene)amino)benzohydrazide (2, n=12)

Yield: 81 %. IR (KBr, cm^{-1}): 3212 ν_s (N-H), 2922, 2854 (aliphatic C-H), 1640 (amide-I, C=O), 1577 (amide-II, N-H), 1624 (-C=N), 1598, 1515 (Ph), 1305, 1275 (OPh), 1021 (N-N); 1H NMR (300 MHz, $CDCl_3/DMSO-d_6$, 30°C): δ 11.15 (s, 1H, -NH), 8.48 (s, 1H, -CH=N), 8.37 (s, 1H, -CH=N), 7.96 (d, $J(H,H)=8.4$ Hz, 2H, -C₆H₄), 7.52 (s, 2H, -C₆H₃), 7.46 (d, $J(H,H)=8.4$ Hz, 2H, -C₆H₄), 7.30 (dd, $J_1(H,H)=9.0$ Hz, $J_2(H,H)=8.1$ Hz, 2H, -C₆H₃), 7.07 (dd, $J_1(H,H)=8.4$ Hz, $J_2(H,H)=8.1$ Hz, 2H, -C₆H₃), 4.07 (m, 8H, -OCH₂), 1.77 (m, 8H, -OCH₂CH₂), 1.45-1.27 (m, 72H, -[CH₂]₃₆), 0.85 (t, $J(H,H)=6.3$ Hz, 12H, -CH₃); ^{13}C NMR (75 MHz, $CDCl_3$, 40°C): δ_c 164.69, 160.62, 154.58, 152.38, 151.84, 151.83, 149.41, 129.60, 129.10, 124.57, 120.52, 112.16, 110.97, 110.96, 105.48, 69.12, 68.70, 31.91, 29.61, 29.35, 25.98, 22.69, 14.13; Elemental analyses: calculated for $C_{69}H_{113}N_3O_5$ (%), C, 77.84; H, 10.70; N, 3.95; Found, C, 77.76; H, 10.61; N, 3.87.

2.15 Synthesis of N-(3,4-bis(tetradecyloxy)benzylidene)-4-((3',4'-bis(tetradecyloxy)benzylidene)amino)benzohydrazide (2, n=14)

Yield: 78 %. IR (KBr, cm^{-1}): 3206 ν_s (N-H), 2920, 2852 (aliphatic C-H), 1641 (amide-I, C=O), 1577 (amide-II, N-H), 1625 (-C=N), 1598, 1514 (Ph), 1304, 1278 (OPh), 1023 (N-N); ^1H NMR (300 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ 11.17 (s, 1H, -NH), 8.48 (s, 1H, -CH=N), 8.35 (s, 1H, -CH=N), 7.94 (d, $J(\text{H,H})=8.4$ Hz, 2H, - C_6H_4), 7.51 (s, 2H, - C_6H_3), 7.45 (d, $J(\text{H,H})=8.4$ Hz, 2H, - C_6H_4), 7.32 (dd, $J_1(\text{H,H})=9.0$ Hz, $J_2(\text{H,H})=8.1$ Hz, 2H, - C_6H_3), 7.07 (dd, $J_1(\text{H,H})=8.4$ Hz, $J_2(\text{H,H})=8.1$ Hz, 2H, - C_6H_3), 4.09 (m, 8H, - OCH_2), 1.77 (m, 8H, - OCH_2CH_2), 1.45-1.27 (m, 88H, - $[\text{CH}_2]_{44}$), 0.86 (t, $J(\text{H,H})=6.3$ Hz, 12H, - CH_3); ^{13}C NMR (75 MHz, CDCl_3 , 40°C): δ_c 164.70, 160.64, 154.57, 152.38, 151.84, 151.83, 149.41, 129.58, 129.09, 124.58, 120.52, 112.15, 110.99, 110.96, 105.47, 69.11, 68.70, 31.91, 29.61, 29.35, 25.98, 22.69, 14.14; Elemental analyses: calculated for $\text{C}_{77}\text{H}_{129}\text{N}_3\text{O}_5$ (%), C, 78.58; H, 11.05; N, 3.57; Found, C, 78.53; H, 10.97; N, 3.53.

2.16 Synthesis of N-(3,4-bis(hexadecyloxy)benzylidene)-4-((3',4'-bis(hexadecyloxy)benzylidene)amino)benzohydrazide (2, n=16)

Yield: 76 %. IR (KBr, cm^{-1}): 3207 ν_s (N-H), 2921, 2854 (aliphatic C-H), 1643 (amide-I, C=O), 1578 (amide-II, N-H), 1624 (-C=N), 1601, 1515 (Ph), 1305, 1280 (OPh), 1021 (N-N); ^1H NMR (300 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ 11.15 (s, 1H, -NH), 8.50 (s, 1H, -CH=N), 8.35 (s, 1H, -CH=N), 7.93 (d, $J(\text{H,H})=8.4$ Hz, 2H, - C_6H_4), 7.54 (s, 2H, - C_6H_3), 7.47 (d, $J(\text{H,H})=8.4$ Hz, 2H, - C_6H_4), 7.30 (dd, $J_1(\text{H,H})=9.0$ Hz, $J_2(\text{H,H})=8.1$ Hz, 2H, - C_6H_3), 7.10 (dd, $J_1(\text{H,H})=8.4$ Hz, $J_2(\text{H,H})=8.1$ Hz, 2H, - C_6H_3), 4.06 (m, 8H, - OCH_2), 1.78 (m, 8H, - OCH_2CH_2), 1.47-1.25 (m, 104H, - $[\text{CH}_2]_{52}$), 0.87 (t, $J(\text{H,H})=6.3$ Hz, 12H, - CH_3); ^{13}C NMR (75 MHz, CDCl_3 , 40°C): δ_c 164.71, 160.62, 154.57, 152.39, 151.84, 151.83, 149.41, 129.59, 129.09, 124.58, 120.53, 112.16, 110.99, 110.96, 105.47, 69.11, 68.70, 31.91, 29.62, 29.35, 25.99, 22.69, 14.13; Elemental analyses: calculated for $\text{C}_{85}\text{H}_{145}\text{N}_3\text{O}_5$ (%), C, 79.20; H, 11.34; N, 3.26; Found, C, 79.11; H, 11.28; N, 3.19.

2.17 Synthesis of N-(3,5-bis(pentyloxy)benzylidene)-4-((3',5'-bis(pentyloxy)benzylidene)amino)benzohydrazide (3, n=5)

Yield: 83 %. IR (KBr, cm^{-1}): 3220 ν_s (N-H), 2926, 2856 (aliphatic C-H), 1647 (amide-I, C=O), 1572 (amide-II, N-H), 1623 (-C=N), 1601, 1518 (Ph), 1309, 1278 (OPh), 1026 (N-N); ^1H NMR (300 MHz, CDCl_3 , 40°C): δ 9.35 (s, 1H, -NH), 8.30 (s, 1H, -CH=N), 7.94 (s, 1H, -CH=N), 7.21 (d, $J(\text{H,H})=8.4$ Hz, 2H, - C_6H_4), 6.98 (d, $J(\text{H,H})=8.4$ Hz, 2H, - C_6H_4), 6.84 (s, 4H, - C_6H_3), 6.48 (s, 2H, - C_6H_3), 4.03 (m, 8H, - OCH_2), 1.83 (m, 8H, - OCH_2CH_2), 1.45-1.27 (m, 16H, - $[\text{CH}_2]_8$), 0.90 (t, $J(\text{H,H})=6.3$ Hz, 12H, - CH_3); ^{13}C NMR (75 MHz, CDCl_3 , 40°C): δ_c 160.98, 159.12, 156.28, 147.11, 140.84, 130.33, 129.29, 128.35, 123.47, 114.57, 114.05, 107.71, 68.70, 68.06, 31.93, 30.34, 29.69, 29.39, 26.06, 22.68, 14.13; Elemental analyses: calculated for $\text{C}_{41}\text{H}_{57}\text{N}_3\text{O}_5$ (%), C, 73.29; H, 8.55; N, 6.25; Found, C, 73.18; H, 8.51; N, 6.27.

2.18 Synthesis of N-(3,5-bis(hexyloxy)benzylidene)-4-((3',5'-bis(hexyloxy)benzylidene)amino)benzohydrazide (3, n=6)

Yield: 78 %. IR (KBr, cm^{-1}): 3220 ν_s (N-H), 2924, 2856 (aliphatic C-H), 1648 (amide-I, C=O), 1572 (amide-II, N-H), 1624 (-C=N), 1601, 1518 (Ph), 1310, 1278 (OPh), 1026 (N-N); ^1H NMR (300 MHz, CDCl_3 , 40°C): δ 9.34 (s, 1H, -NH), 8.28 (s, 1H, -CH=N), 7.95 (s, 1H, -CH=N), 7.21 (d, $J(\text{H,H})=8.4$ Hz, 2H, - C_6H_4), 6.98 (d, $J(\text{H,H})=8.4$ Hz, 2H, - C_6H_4), 6.83 (s, 4H, - C_6H_3), 6.49 (s, 2H, - C_6H_3), 4.02 (m, 8H, - OCH_2), 1.83 (m, 8H, - OCH_2CH_2), 1.45-1.27 (m, 24H, - $[\text{CH}_2]_{12}$), 0.91 (t, $J(\text{H,H})=6.3$ Hz, 12H, - CH_3); ^{13}C NMR (75 MHz, CDCl_3 , 40°C): δ_c 160.95, 159.10, 156.27, 147.13, 140.84, 130.31, 129.28, 128.35, 123.49, 114.55, 114.05, 107.70,

68.72, 68.06, 31.93, 30.34, 29.68, 29.39, 26.08, 22.68, 14.10; Elemental analyses: calculated for C₄₅H₆₅N₃O₅ (%), C, 74.24; H, 9.01; N, 5.77; Found, C, 74.21; H, 9.03; N, 5.73.

2.19 Synthesis of N-(3,5-bis(heptyloxy)benzylidene)-4-((3',5'-bis(heptyloxy)benzylidene)amino)benzohydrazide (3, n=7)

Yield: 77 %. IR (KBr, cm⁻¹): 3222 v_s (N-H), 2926, 2856 (aliphatic C-H), 1644 (amide-I, C=O), 1572 (amide-II, N-H), 1623 (-C=N), 1603, 1518 (Ph), 1309, 1280 (OPh), 1026 (N-N); ¹H NMR (300 MHz, CDCl₃, 40°C): δ 9.33 (s, 1H, -NH), 8.29 (s, 1H, -CH=N), 7.94 (s, 1H, -CH=N), 7.22 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 6.98 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 6.83 (s, 4H, -C₆H₃), 6.48 (s, 2H, -C₆H₃), 4.01 (m, 8H, -OCH₂), 1.83 (m, 8H, -OCH₂CH₂), 1.45-1.27 (m, 32H, -[CH₂]₁₆), 0.90 (t, J(H,H)=6.3 Hz, 12H, -CH₃); ¹³C NMR (75 MHz, CDCl₃, 40°C): δ_c 160.96, 159.12, 156.28, 147.13, 140.81, 130.33, 129.28, 128.33, 123.49, 114.57, 114.08, 107.70, 68.70, 68.04, 31.93, 30.34, 29.68, 29.39, 26.06, 22.69, 14.10; Elemental analyses: calculated for C₄₉H₇₃N₃O₅ (%), C, 75.06; H, 9.38; N, 5.36; Found, C, 75.01; H, 9.33; N, 5.39.

2.20 Synthesis of N-(3,5-bis(octyloxy)benzylidene)-4-((3',5'-bis(octyloxy)benzylidene)amino)benzohydrazide (3, n=8)

Yield: 81 %. IR (KBr, cm⁻¹): 3220 v_s (N-H), 2928, 2858 (aliphatic C-H), 1647 (amide-I, C=O), 1572 (amide-II, N-H), 1623 (-C=N), 1601, 1520 (Ph), 1309, 1278 (OPh), 1024 (N-N); ¹H NMR (300 MHz, CDCl₃, 40°C): δ 9.34 (s, 1H, -NH), 8.28 (s, 1H, -CH=N), 7.94 (s, 1H, -CH=N), 7.21 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 6.98 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 6.82 (s, 4H, -C₆H₃), 6.47 (s, 2H, -C₆H₃), 4.02 (m, 8H, -OCH₂), 1.84 (m, 8H, -OCH₂CH₂), 1.45-1.27 (m, 40H, -[CH₂]₂₀), 0.89 (t, J(H,H)=6.3 Hz, 12H, -CH₃); ¹³C NMR (75 MHz, CDCl₃, 40°C): δ_c 160.95, 159.14, 156.28, 147.12, 140.84, 130.35, 129.28, 128.35, 123.49, 114.57, 114.05, 107.73, 68.70, 68.06, 31.94, 30.34, 29.69, 29.39, 26.06, 22.68, 14.13; Elemental analyses: calculated for C₅₃H₈₁N₃O₅ (%), C, 75.76; H, 9.72; N, 5.00; Found, C, 75.80; H, 9.74; N, 4.96.

2.21 Synthesis of N-(3,5-bis(decyloxy)benzylidene)-4-((3',5'-bis(decyloxy)benzylidene)amino)benzohydrazide (3, n=10)

Yield: 76 %. IR (KBr, cm⁻¹): 3220 v_s (N-H), 2926, 2856 (aliphatic C-H), 1646 (amide-I, C=O), 1574 (amide-II, N-H), 1623 (-C=N), 1604, 1518 (Ph), 1307, 1278 (OPh), 1026 (N-N); ¹H NMR (300 MHz, CDCl₃, 40°C): δ 9.34 (s, 1H, -NH), 8.29 (s, 1H, -CH=N), 7.94 (s, 1H, -CH=N), 7.21 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 6.98 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 6.83 (s, 4H, -C₆H₃), 6.48 (s, 2H, -C₆H₃), 4.02 (m, 8H, -OCH₂), 1.83 (m, 8H, -OCH₂CH₂), 1.45-1.27 (m, 56H, -[CH₂]₂₈), 0.90 (t, J(H,H)=6.3 Hz, 12H, -CH₃); ¹³C NMR (75 MHz, CDCl₃, 40°C): δ_c 160.95, 159.12, 156.28, 147.13, 140.84, 130.33, 129.28, 128.35, 123.49, 114.57, 114.05, 107.70, 68.70, 68.06, 31.93, 30.34, 29.68, 29.39, 26.06, 22.68, 14.10; Elemental analyses: calculated for C₆₁H₉₇N₃O₅ (%), C, 76.92; H, 10.27; N, 4.41; Found, C, 76.86; H, 10.23; N, 4.44.

2.22 Synthesis of N-(3,5-bis(dodecyloxy)benzylidene)-4-((3',5'-bis(dodecyloxy)benzylidene)amino)benzohydrazide (3, n=12)

Yield: 79 %. IR (KBr, cm⁻¹): 3218 v_s (N-H), 2926, 2856 (aliphatic C-H), 1647 (amide-I, C=O), 1572 (amide-II, N-H), 1621 (-C=N), 1601, 1519 (Ph), 1309, 1281 (OPh), 1026 (N-N); ¹H NMR (300 MHz, CDCl₃, 40°C): δ 9.35 (s, 1H, -NH), 8.29 (s, 1H, -CH=N), 7.93 (s, 1H, -CH=N), 7.21 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 6.99 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 6.83 (s, 4H, -C₆H₃), 6.48 (s, 2H, -C₆H₃), 4.03 (m, 8H, -OCH₂), 1.83 (m, 8H, -

OCH₂CH₂), 1.45-1.27 (m, 72H, -[CH₂]₃₆), 0.90 (t, J(H,H)=6.3 Hz, 12H, -CH₃); ¹³C NMR (75 MHz, CDCl₃, 40°C): δ_c 160.95, 159.12, 156.26, 147.13, 140.82, 130.33, 129.27, 128.35, 123.48, 114.57, 114.05, 107.70, 68.70, 68.07, 31.93, 30.34, 29.68, 29.37, 26.06, 22.68, 14.10; Elemental analyses: calculated for C₆₉H₁₁₃N₃O₅ (%), C, 77.84; H, 10.70; N, 3.95; Found, C, 77.76; H, 10.65; N, 3.90.

2.23 Synthesis of N-(3,5-bis(tetradecyloxy)benzylidene)-4-((3',5'-bis(tetradecyloxy)benzylidene)amino)benzohydrazide (3, n=14)

Yield: 82 %. IR (KBr, cm⁻¹): 3220 v_s (N-H), 2926, 2857 (aliphatic C-H), 1647 (amide-I, C=O), 1575 (amide-II, N-H), 1623 (-C=N), 1601, 1516 (Ph), 1309, 1278 (OPh), 1025 (N-N); ¹H NMR (300 MHz, CDCl₃, 40°C): δ 9.32 (s, 1H, -NH), 8.29 (s, 1H, -CH=N), 7.92 (s, 1H, -CH=N), 7.21 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 6.98 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 6.85 (s, 4H, -C₆H₃), 6.48 (s, 2H, -C₆H₃), 4.01 (m, 8H, -OCH₂), 1.85 (m, 8H, -OCH₂CH₂), 1.45-1.27 (m, 88H, -[CH₂]₄₄), 0.92 (t, J(H,H)=6.3 Hz, 12H, -CH₃); ¹³C NMR (75 MHz, CDCl₃, 40°C): δ_c 160.94, 159.12, 156.29, 147.13, 140.84, 130.33, 129.28, 128.34, 123.49, 114.58, 114.05, 107.70, 68.72, 68.06, 31.93, 30.34, 29.68, 29.39, 26.07, 22.68, 14.10; Elemental analyses: calculated for C₇₇H₁₂₉N₃O₅ (%), C, 78.58; H, 11.05; N, 3.57; Found, C, 78.51; H, 10.98; N, 3.53.

2.24 Synthesis of N-(3,5-bis(hexadecyloxy)benzylidene)-4-((3',5'-bis(hexadecyloxy)benzylidene)amino)benzohydrazide (3, n=16)

Yield: 80 %. IR (KBr, cm⁻¹): 3219 v_s (N-H), 2926, 2856 (aliphatic C-H), 1647 (amide-I, C=O), 1572 (amide-II, N-H), 1625 (-C=N), 1603, 1518 (Ph), 1308, 1278 (OPh), 1026 (N-N); ¹H NMR (300 MHz, CDCl₃, 40°C): δ 9.34 (s, 1H, -NH), 8.30 (s, 1H, -CH=N), 7.94 (s, 1H, -CH=N), 7.21 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 6.98 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 6.81 (s, 4H, -C₆H₃), 6.50 (s, 2H, -C₆H₃), 4.02 (m, 8H, -OCH₂), 1.83 (m, 8H, -OCH₂CH₂), 1.45-1.27 (m, 104H, -[CH₂]₅₂), 0.90 (t, J(H,H)=6.3 Hz, 12H, -CH₃); ¹³C NMR (75 MHz, CDCl₃, 40°C): δ_c 160.95, 159.13, 156.28, 147.14, 140.84, 130.32, 129.28, 128.35, 123.49, 114.57, 114.06, 107.71, 68.70, 68.06, 31.93, 30.34, 29.68, 29.39, 26.06, 22.68, 14.11; Elemental analyses: calculated for C₈₅H₁₄₅N₃O₅ (%), C, 79.20; H, 11.34; N, 3.26; Found, C, 79.12; H, 11.31; N, 3.22.

2.25 Synthesis of N-(2,3,4-tris(pentyloxy)benzylidene)-4-((2',3',4'-tris(pentyloxy)benzylidene)amino)benzohydrazide (4, n=5)

Yield: 78 %. IR (KBr, cm⁻¹): 3317 v_s (N-H), 2922, 2854 (aliphatic C-H), 1652 (amide-I, C=O), 1570 (amide-II, N-H), 1625 (-C=N), 1604, 1515 (Ph), 1303, 1285 (OPh), 1022 (N-N); ¹H NMR (300 MHz, CDCl₃/DMSO-d₆, 30°C): δ 10.70 (s, 1H, -NH), 8.78 (s, 1H, -CH=N), 8.57 (s, 1H, -CH=N), 7.96 (d, J(H,H)=7.2 Hz, 2H, -C₆H₄), 7.81 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 7.27 (d, J(H,H)=8.1 Hz, 2H, -C₆H₂), 6.78 (dd, J₁(H,H)=9.0 Hz, J₂(H,H)=9.0 Hz, 2H, -C₆H₂), 4.15 (m, 12H, -OCH₂), 1.85 (m, 12H, -OCH₂CH₂), 1.48-1.26 (m, 24H, -[CH₂]₁₂), 0.88 (t, J(H,H)=6.3 Hz, 18H, -CH₃); ¹³C NMR (75 MHz, CDCl₃/DMSO-d₆, 30°C): δ_c 163.92, 155.09, 152.80, 152.42, 151.08, 140.90, 136.21, 129.30, 122.69, 120.24, 114.25, 111.66, 108.45, 74.54, 73.39, 68.49, 31.67, 30.10, 29.36, 29.10, 25.88, 25.78, 22.45, 13.87; Elemental analyses: calculated for C₅₁H₇₇N₃O₇ (%), C, 72.56; H, 9.19; N, 4.98; Found, C, 72.50; H, 9.10; N, 4.92.

2.26 Synthesis of N-(2,3,4-tris(hexyloxy)benzylidene)-4-((2',3',4'-tris(hexyloxy)benzylidene)amino)benzohydrazide (4, n=6)

Yield: 76 %. IR (KBr, cm^{-1}): 3316 ν_s (N-H), 2921, 2856 (aliphatic C-H), 1651 (amide-I, C=O), 1571 (amide-II, N-H), 1624 (-C=N), 1606, 1515 (Ph), 1305, 1285 (OPh), 1021 (N-N); ^1H NMR (300 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ 10.69 (s, 1H, -NH), 8.78 (s, 1H, -CH=N), 8.59 (s, 1H, -CH=N), 7.96 (d, $J(\text{H,H})=7.2$ Hz, 2H, -C₆H₄), 7.82 (d, $J(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.27 (d, $J(\text{H,H})=8.1$ Hz, 2H, -C₆H₂), 6.79 (dd, $J_1(\text{H,H})=9.0$ Hz, $J_2(\text{H,H})=9.0$ Hz, 2H, -C₆H₂), 4.15 (m, 12H, -OCH₂), 1.86 (m, 12H, -OCH₂CH₂), 1.48-1.26 (m, 36H, -[CH₂]₁₈), 0.88 (t, $J(\text{H,H})=6.3$ Hz, 18H, -CH₃); ^{13}C NMR (75 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ_c 163.93, 155.07, 152.81, 152.41, 151.10, 140.89, 136.23, 129.32, 122.68, 120.25, 114.26, 111.66, 108.45, 74.56, 73.39, 68.49, 31.69, 30.09, 29.36, 29.10, 25.88, 25.79, 22.45, 13.88; Elemental analyses: calculated for C₅₇H₈₉N₃O₇ (%), C, 73.75; H, 9.66; N, 4.53; Found, C, 73.69; H, 9.68; N, 4.49.

2.27 Synthesis of N-(2,3,4-tris(heptyloxy)benzylidene)-4-[(2',3',4'-tris(heptyloxy)benzylidene)amino]benzohydrazide (4, n=7)

Yield: 77 %. IR (KBr, cm^{-1}): 3316 ν_s (N-H), 2924, 2854 (aliphatic C-H), 1653 (amide-I, C=O), 1572 (amide-II, N-H), 1624 (-C=N), 1604, 1515 (Ph), 1303, 1286 (OPh), 1021 (N-N); ^1H NMR (300 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ 10.67 (s, 1H, -NH), 8.76 (s, 1H, -CH=N), 8.59 (s, 1H, -CH=N), 7.96 (d, $J(\text{H,H})=7.2$ Hz, 2H, -C₆H₄), 7.83 (d, $J(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.27 (d, $J(\text{H,H})=8.1$ Hz, 2H, -C₆H₂), 6.77 (dd, $J_1(\text{H,H})=9.0$ Hz, $J_2(\text{H,H})=9.0$ Hz, 2H, -C₆H₂), 4.13 (m, 12H, -OCH₂), 1.85 (m, 12H, -OCH₂CH₂), 1.48-1.25 (m, 48H, -[CH₂]₂₄), 0.88 (t, $J(\text{H,H})=6.3$ Hz, 18H, -CH₃); ^{13}C NMR (75 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ_c 163.92, 155.07, 152.80, 152.42, 151.08, 140.88, 136.21, 129.31, 122.69, 120.23, 114.26, 111.67, 108.45, 74.54, 73.38, 68.49, 31.67, 30.09, 29.37, 29.10, 25.89, 25.79, 22.45, 13.87; Elemental analyses: calculated for C₆₃H₁₀₁N₃O₇ (%), C, 74.73; H, 10.05; N, 4.15; Found, C, 74.61; H, 9.96; N, 4.07.

2.28 Synthesis of N-(2,3,4-tris(octyloxy)benzylidene)-4-[(2',3',4'-tris(octyloxy)benzylidene)amino]benzohydrazide (4, n=8)

Yield: 81 %. IR (KBr, cm^{-1}): 3318 ν_s (N-H), 2920, 2852 (aliphatic C-H), 1650 (amide-I, C=O), 1574 (amide-II, N-H), 1623 (-C=N), 1606, 1518 (Ph), 1308, 1282 (OPh), 1020 (N-N); ^1H NMR (300 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ 10.65 (s, 1H, -NH), 8.75 (s, 1H, -CH=N), 8.55 (s, 1H, -CH=N), 7.95 (d, $J(\text{H,H})=7.2$ Hz, 2H, -C₆H₄), 7.85 (d, $J(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.25 (d, $J(\text{H,H})=8.1$ Hz, 2H, -C₆H₂), 6.75 (dd, $J_1(\text{H,H})=9.0$ Hz, $J_2(\text{H,H})=9.0$ Hz, 2H, -C₆H₂), 4.14 (m, 12H, -OCH₂), 1.88 (m, 12H, -OCH₂CH₂), 1.48-1.25 (m, 60H, -[CH₂]₃₀), 0.89 (t, $J(\text{H,H})=6.3$ Hz, 18H, -CH₃); ^{13}C NMR (75 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ_c 163.91, 155.08, 152.83, 152.41, 151.09, 140.89, 136.22, 129.32, 122.70, 120.24, 114.24, 111.66, 108.45, 74.54, 73.39, 68.48, 31.67, 30.09, 29.36, 29.12, 25.88, 25.80, 22.45, 13.86; Elemental analyses: calculated for C₆₉H₁₁₃N₃O₇ (%), C, 75.57; H, 10.39; N, 3.83; Found, C, 75.49; H, 10.32; N, 3.79.

2.29 Synthesis of N-(2,3,4-tris(decyloxy)benzylidene)-4-[(2',3',4'-tris(decyloxy)benzylidene)amino]benzohydrazide (4, n=10)

Yield: 80 %. IR (KBr, cm^{-1}): 3316 ν_s (N-H), 2922, 2854 (aliphatic C-H), 1651 (amide-I, C=O), 1570 (amide-II, N-H), 1624 (-C=N), 1604, 1515 (Ph), 1303, 1285 (OPh), 1021 (N-N); ^1H NMR (300 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ 10.61 (s, 1H, -NH), 8.74 (s, 1H, -CH=N), 8.58 (s, 1H, -CH=N), 7.98 (d, $J(\text{H,H})=7.2$ Hz, 2H, -C₆H₄), 7.86 (d, $J(\text{H,H})=8.4$ Hz, 2H, -C₆H₄), 7.24 (d, $J(\text{H,H})=8.1$ Hz, 2H, -C₆H₂), 6.78 (dd, $J_1(\text{H,H})=9.0$ Hz, $J_2(\text{H,H})=9.0$ Hz, 2H, -C₆H₂), 4.13 (m, 12H, -OCH₂), 1.87 (m, 12H, -OCH₂CH₂), 1.48-1.27 (m, 84H, -[CH₂]₄₂), 0.90 (t, $J(\text{H,H})=5.1$ Hz, 18H, -CH₃); ^{13}C NMR (75 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ_c 163.92, 155.07, 152.80, 152.41,

151.08, 140.89, 136.21, 129.32, 122.69, 120.24, 114.26, 111.66, 108.45, 74.54, 73.39, 68.49, 31.67, 30.09, 29.36, 29.10, 25.88, 25.79, 22.45, 13.87; Elemental analyses: calculated for C₈₁H₁₃₇N₃O₇ (%), C, 76.91; H, 10.92; N, 3.32; Found, C, 76.84; H, 10.88; N, 3.29.

2.30 Synthesis of N-(2,3,4-tris(dodecyloxy)benzylidene)-4-{(2',3',4'-tris(dodecyloxy)benzylidene)amino}benzohydrazide (4, n=12)

Yield: 82 %. IR (KBr, cm⁻¹): 3316 v_s (N-H), 2923, 2854 (aliphatic C-H), 1651 (amide-I, C=O), 1573 (amide-II, N-H), 1624 (-C=N), 1604, 1516 (Ph), 1303, 1285 (OPh), 1021 (N-N); ¹H NMR (300 MHz, CDCl₃/DMSO-d⁶, 30°C): δ 10.63 (s, 1H, -NH), 8.77 (s, 1H, -CH=N), 8.56 (s, 1H, -CH=N), 7.97 (d, J(H,H)=7.2 Hz, 2H, -C₆H₄), 7.83 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 7.24 (d, J(H,H)=8.1 Hz, 2H, -C₆H₂), 6.76 (dd, J₁(H,H)=9.0 Hz, J₂(H,H)=9.0 Hz, 2H, -C₆H₂), 4.18 (m, 12H, -OCH₂), 1.89 (m, 12H, -OCH₂CH₂), 1.49-1.27 (m, 108H, -[CH₂]₅₄), 0.86 (t, J(H,H)=6.3 Hz, 18H, -CH₃); ¹³C NMR (75 MHz, CDCl₃/DMSO-d⁶, 30°C): δ_c 163.92, 155.07, 152.79, 152.40, 151.06, 140.90, 136.21, 129.30, 122.70, 120.24, 114.26, 111.66, 108.49, 74.55, 73.39, 68.49, 31.67, 30.10, 29.36, 29.10, 25.90, 25.79, 22.46, 13.85; Elemental analyses: calculated for C₉₃H₁₆₁N₃O₇ (%), C, 77.93; H, 11.32; N, 2.93; Found, C, 77.82; H, 11.26; N, 2.88.

2.31 Synthesis of N-(2,3,4-tris(tetradecyloxy)benzylidene)-4-{(2',3',4'-tris(tetradecyloxy)benzylidene)amino}benzohydrazide (4, n=14)

Yield: 76 %. IR (KBr, cm⁻¹): 3317 v_s (N-H), 2921, 2853 (aliphatic C-H), 1652 (amide-I, C=O), 1570 (amide-II, N-H), 1626 (-C=N), 1604, 1520 (Ph), 1303, 1285 (OPh), 1023 (N-N); ¹H NMR (300 MHz, CDCl₃/DMSO-d⁶, 30°C): δ 10.60 (s, 1H, -NH), 8.72 (s, 1H, -CH=N), 8.53 (s, 1H, -CH=N), 7.99 (d, J(H,H)=7.2 Hz, 2H, -C₆H₄), 7.84 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 7.26 (d, J(H,H)=8.1 Hz, 2H, -C₆H₂), 6.74 (dd, J₁(H,H)=9.0 Hz, J₂(H,H)=9.0 Hz, 2H, -C₆H₂), 4.17 (m, 12H, -OCH₂), 1.85 (m, 12H, -OCH₂CH₂), 1.47-1.24 (m, 132H, -[CH₂]₆₆), 0.91 (t, J(H,H)=6.3 Hz, 18H, -CH₃); ¹³C NMR (75 MHz, CDCl₃/DMSO-d⁶, 30°C): δ_c 163.94, 155.10, 152.80, 152.41, 151.08, 140.89, 136.20, 129.29, 122.69, 120.22, 114.25, 111.66, 108.47, 74.54, 73.37, 68.49, 31.67, 30.09, 29.36, 29.12, 25.77, 25.79, 22.43, 13.85; Elemental analyses: calculated for C₁₀₅H₁₈₅N₃O₇ (%), C, 78.74; H, 11.64; N, 2.62; Found, C, 78.67; H, 11.57; N, 2.56.

2.32 Synthesis of N-(2,3,4-tris(hexadecyloxy)benzylidene)-4-{(2',3',4'-tris(hexadecyloxy)benzylidene)amino}benzohydrazide (4, n=16)

Yield: 78 %. IR (KBr, cm⁻¹): 3319 v_s (N-H), 2925, 2856 (aliphatic C-H), 1653 (amide-I, C=O), 1572 (amide-II, N-H), 1621 (-C=N), 1608, 1517 (Ph), 1305, 1288 (OPh), 1019 (N-N); ¹H NMR (300 MHz, CDCl₃/DMSO-d⁶, 30°C): δ 10.64 (s, 1H, -NH), 8.74 (s, 1H, -CH=N), 8.52 (s, 1H, -CH=N), 7.95 (d, J(H,H)=7.2 Hz, 2H, -C₆H₄), 7.82 (d, J(H,H)=8.4 Hz, 2H, -C₆H₄), 7.25 (d, J(H,H)=8.1 Hz, 2H, -C₆H₂), 6.73 (dd, J₁(H,H)=9.0 Hz, J₂(H,H)=9.0 Hz, 2H, -C₆H₂), 4.15 (m, 12H, -OCH₂), 1.87 (m, 12H, -OCH₂CH₂), 1.46-1.27 (m, 156H, -[CH₂]₇₈), 0.89 (t, J(H,H)=6.3 Hz, 18H, -CH₃); ¹³C NMR (75 MHz, CDCl₃/DMSO-d⁶, 30°C): δ_c 163.92, 155.08, 152.80, 152.42, 151.08, 140.90, 136.21, 129.30, 122.69, 120.24, 114.26, 111.66, 108.45, 74.54, 73.39, 68.46, 31.67, 30.09, 29.36, 29.10, 25.88, 25.79, 22.47, 13.86; Elemental analyses: calculated for C₁₁₇H₂₀₉N₃O₇ (%), C, 79.40; H, 11.90; N, 2.37; Found, C, 79.32; H, 11.80; N, 2.29.

2.33 Synthesis of N-(3,4,5-tris(pentyloxy)benzylidene)-4-{(3',4',5'-tris(pentyloxy)benzylidene)amino}benzohydrazide (5, n=5)

Yield: 73 %. IR (KBr, cm^{-1}): 3343 ν_s (N-H), 2922, 2856 (aliphatic C-H), 1640 (amide-I, C=O), 1570 (amide-II, N-H), 1628 (-C=N), 1600, 1507 (Ph), 1306, 1285 (OPh), 1016 (N-N); ^1H NMR (300 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ 9.97 (s, 1H, -NH), 8.12 (s, 2H, -CH=N), 7.73 (d, $J(\text{H,H})=5.7$ Hz, 2H, - C_6H_4), 6.92 (s, 4H, - C_6H_2), 6.68 (d, $J(\text{H,H})=7.5$ Hz, 2H, - C_6H_4), 4.01 (m, 12H, - OCH_2), 1.81 (m, 12H, - OCH_2CH_2), 1.48-1.23 (m, 24H, - $[\text{CH}_2]_{12}$), 0.89 (t, $J(\text{H,H})=6.9$ Hz, 18H, - CH_3); ^{13}C NMR (75 MHz, CDCl_3 , 40°C): δ_c 166.55, 163.74, 163.27, 162.68, 155.40, 152.29, 133.55, 132.39, 131.06, 128.39, 114.37, 113.42, 110.74, 73.08, 68.38, 31.88, 29.53, 29.34, 29.07, 25.97, 22.67, 14.09; Elemental analyses: calculated for $\text{C}_{51}\text{H}_{77}\text{N}_3\text{O}_7$ (%), C, 72.56; H, 9.19; N, 4.98; Found, C, 72.48; H, 9.13; N, 4.93.

2.34 Synthesis of N-(3,4,5-tris(hexyloxy)benzylidene)-4-((3',4',5'-tris(hexyloxy)benzylidene)amino)benzohydrazide (5, n=6)

Yield: 75 %. IR (KBr, cm^{-1}): 3340 ν_s (N-H), 2924, 2853 (aliphatic C-H), 1643 (amide-I, C=O), 1576 (amide-II, N-H), 1620 (-C=N), 1599, 1502 (Ph), 1300, 1282 (OPh), 1011 (N-N); ^1H NMR (300 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ 9.98 (s, 1H, -NH), 8.13 (s, 2H, -CH=N), 7.75 (d, $J(\text{H,H})=5.7$ Hz, 2H, - C_6H_4), 6.97 (s, 4H, - C_6H_2), 6.71 (d, $J(\text{H,H})=7.5$ Hz, 2H, - C_6H_4), 4.05 (m, 12H, - OCH_2), 1.80 (m, 12H, - OCH_2CH_2), 1.47-1.28 (m, 36H, - $[\text{CH}_2]_{18}$), 0.88 (t, $J(\text{H,H})=6.9$ Hz, 18H, - CH_3); ^{13}C NMR (75 MHz, CDCl_3 , 40°C): δ_c 166.55, 163.74, 163.28, 162.69, 155.42, 152.30, 133.55, 132.38, 131.06, 128.36, 114.37, 113.43, 110.75, 73.08, 68.37, 31.86, 29.54, 29.33, 29.07, 25.97, 22.67, 14.12; Elemental analyses: calculated for $\text{C}_{57}\text{H}_{89}\text{N}_3\text{O}_7$ (%), C, 73.75; H, 9.66; N, 4.53; Found, C, 73.63; H, 9.62; N, 4.49.

2.35 Synthesis of N-(3,4,5-tris(heptyloxy)benzylidene)-4-((3',4',5'-tris(heptyloxy)benzylidene)amino)benzohydrazide (5, n=7)

Yield: 76 %. IR (KBr, cm^{-1}): 3347 ν_s (N-H), 2925, 2851 (aliphatic C-H), 1641 (amide-I, C=O), 1571 (amide-II, N-H), 1627 (-C=N), 1602, 1506 (Ph), 1303, 1284 (OPh), 1016 (N-N); ^1H NMR (300 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ 10.03 (s, 1H, -NH), 8.10 (s, 2H, -CH=N), 7.76 (d, $J(\text{H,H})=5.7$ Hz, 2H, - C_6H_4), 6.98 (s, 4H, - C_6H_2), 6.69 (d, $J(\text{H,H})=7.5$ Hz, 2H, - C_6H_4), 4.02 (m, 12H, - OCH_2), 1.80 (m, 12H, - OCH_2CH_2), 1.47-1.26 (m, 48H, - $[\text{CH}_2]_{24}$), 0.88 (t, $J(\text{H,H})=6.9$ Hz, 18H, - CH_3); ^{13}C NMR (75 MHz, CDCl_3 , 40°C): δ_c 166.55, 163.74, 163.27, 162.68, 155.42, 152.28, 133.55, 132.38, 131.07, 128.37, 114.37, 113.43, 110.75, 73.08, 68.38, 31.88, 29.54, 29.35, 29.07, 25.97, 22.68, 14.10; Elemental analyses: calculated for $\text{C}_{63}\text{H}_{101}\text{N}_3\text{O}_7$ (%), C, 74.73; H, 10.05; N, 4.15; Found, C, 74.63; H, 9.97; N, 4.09.

2.36 Synthesis of N-(3,4,5-tris(octyloxy)benzylidene)-4-((3',4',5'-tris(octyloxy)benzylidene)amino)benzohydrazide (5, n=8)

Yield: 77 %. IR (KBr, cm^{-1}): 3346 ν_s (N-H), 2927, 2850 (aliphatic C-H), 1645 (amide-I, C=O), 1574 (amide-II, N-H), 1626 (-C=N), 1597, 1501 (Ph), 1304, 1284 (OPh), 1010 (N-N); ^1H NMR (300 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ 10.02 (s, 1H, -NH), 8.17 (s, 2H, -CH=N), 7.78 (d, $J(\text{H,H})=5.7$ Hz, 2H, - C_6H_4), 6.94 (s, 4H, - C_6H_2), 6.68 (d, $J(\text{H,H})=7.5$ Hz, 2H, - C_6H_4), 4.04 (m, 12H, - OCH_2), 1.80 (m, 12H, - OCH_2CH_2), 1.46-1.27 (m, 60H, - $[\text{CH}_2]_{60}$), 0.89 (t, $J(\text{H,H})=6.9$ Hz, 18H, - CH_3); ^{13}C NMR (75 MHz, CDCl_3 , 40°C): δ_c 166.55, 163.74, 163.28, 162.67, 155.42, 152.29, 133.55, 132.39, 131.06, 128.37, 114.37, 113.40, 110.75, 73.08, 68.37, 31.89, 29.54, 29.34, 29.07, 25.98, 22.67, 14.11; Elemental analyses: calculated for $\text{C}_{69}\text{H}_{113}\text{N}_3\text{O}_7$ (%), C, 75.57; H, 10.39; N, 3.83; Found, C, 75.50; H, 10.31; N, 3.85.

2.37 Synthesis of *N*-(3,4,5-tris(decyloxy)benzylidene)-4-*-(3',4',5'-tris(decyloxy)benzylidene)amino*benzohydrazide (5, *n*=10)

Yield: 79 %. IR (KBr, cm^{-1}): 3340 ν_s (N-H), 2924, 2853 (aliphatic C-H), 1643 (amide-I, C=O), 1576 (amide-II, N-H), 1620 (-C=N), 1599, 1502 (Ph), 1300, 1282 (OPh), 1011 (N-N); ^1H NMR (300 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ 10.00 (s, 1H, -NH), 8.15 (s, 2H, -CH=N), 7.77 (d, $J(\text{H,H})=5.7$ Hz, 2H, -C₆H₄), 6.95 (s, 4H, -C₆H₂), 6.69 (d, $J(\text{H,H})=7.5$ Hz, 2H, -C₆H₄), 4.02 (m, 12H, -OCH₂), 1.79 (m, 12H, -OCH₂CH₂), 1.46-1.27 (m, 84H, -[CH₂]₄₂), 0.88 (t, $J(\text{H,H})=6.9$ Hz, 18H, -CH₃); ^{13}C NMR (75 MHz, CDCl_3 , 40°C): δ_c 166.55, 163.74, 163.28, 162.68, 155.42, 152.29, 133.55, 132.38, 131.06, 128.37, 114.37, 113.42, 110.75, 73.08, 68.37, 31.88, 29.54, 29.34, 29.07, 25.97, 22.67, 14.10; Elemental analyses: calculated for C₈₁H₁₃₇N₃O₇ (%), C, 76.91; H, 10.92; N, 3.32; Found, C, 76.85; H, 10.85; N, 3.28.

2.38 Synthesis of *N*-(3,4,5-tris(dodecyloxy)benzylidene)-4-*-(3',4',5'-tris(dodecyloxy)benzylidene)amino*benzohydrazide (5, *n*=12)

Yield: 76 %. IR (KBr, cm^{-1}): 3342 ν_s (N-H), 2926, 2849 (aliphatic C-H), 1640 (amide-I, C=O), 1573 (amide-II, N-H), 1623 (-C=N), 1596, 1503 (Ph), 1305, 1281 (OPh), 1013 (N-N); ^1H NMR (300 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ 10.04 (s, 1H, -NH), 8.12 (s, 2H, -CH=N), 7.74 (d, $J(\text{H,H})=5.7$ Hz, 2H, -C₆H₄), 6.95 (s, 4H, -C₆H₂), 6.70 (d, $J(\text{H,H})=7.5$ Hz, 2H, -C₆H₄), 4.03 (m, 12H, -OCH₂), 1.78 (m, 12H, -OCH₂CH₂), 1.46-1.26 (m, 108H, -[CH₂]₅₄), 0.90 (t, $J(\text{H,H})=6.9$ Hz, 18H, -CH₃); ^{13}C NMR (75 MHz, CDCl_3 , 40°C): δ_c 166.54, 163.74, 163.28, 162.69, 155.42, 152.29, 133.54, 132.38, 131.06, 128.36, 114.37, 113.42, 110.77, 73.08, 68.37, 31.87, 29.54, 29.35, 29.07, 25.97, 22.67, 14.10; Elemental analyses: calculated for C₉₃H₁₆₁N₃O₇ (%), C, 77.93; H, 11.32; N, 2.93; Found, C, 77.82; H, 11.27; N, 2.88.

2.39 Synthesis of *N*-(3,4,5-tris(tetradecyloxy)benzylidene)-4-*-(3',4',5'-tris(tetradecyloxy)benzylidene)amino*benzohydrazide (5, *n*=14)

Yield: 74 %. IR (KBr, cm^{-1}): 3341 ν_s (N-H), 2923, 2853 (aliphatic C-H), 1641 (amide-I, C=O), 1575 (amide-II, N-H), 1621 (-C=N), 1599, 1501 (Ph), 1303, 1282 (OPh), 1015 (N-N); ^1H NMR (300 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ 10.01 (s, 1H, -NH), 8.14 (s, 2H, -CH=N), 7.76 (d, $J(\text{H,H})=5.7$ Hz, 2H, -C₆H₄), 6.93 (s, 4H, -C₆H₂), 6.67 (d, $J(\text{H,H})=7.5$ Hz, 2H, -C₆H₄), 4.01 (m, 12H, -OCH₂), 1.77 (m, 12H, -OCH₂CH₂), 1.46-1.27 (m, 132H, -[CH₂]₆₆), 0.89 (t, $J(\text{H,H})=6.9$ Hz, 18H, -CH₃); ^{13}C NMR (75 MHz, CDCl_3 , 40°C): δ_c 166.55, 163.73, 163.28, 162.67, 155.42, 152.28, 133.55, 132.37, 131.06, 128.37, 114.35, 113.42, 110.74, 73.08, 68.36, 31.88, 29.54, 29.34, 29.07, 25.97, 22.67, 14.10; Elemental analyses: calculated for C₁₀₅H₁₈₅N₃O₇ (%), C, 78.74; H, 11.64; N, 2.62; Found, C, 78.66; H, 11.59; N, 2.59.

2.40 Synthesis of *N*-(3,4,5-tris(hexadecyloxy)benzylidene)-4-*-(3',4',5'-tris(hexadecyloxy)benzylidene)amino*benzohydrazide (5, *n*=16)

Yield: 78 %. IR (KBr, cm^{-1}): 3344 ν_s (N-H), 2924, 2851 (aliphatic C-H), 1644 (amide-I, C=O), 1574 (amide-II, N-H), 1620 (-C=N), 1595, 1504 (Ph), 1300, 1283 (OPh), 1009 (N-N); ^1H NMR (300 MHz, $\text{CDCl}_3/\text{DMSO-d}_6$, 30°C): δ 9.99 (s, 1H, -NH), 8.11 (s, 2H, -CH=N), 7.73 (d, $J(\text{H,H})=5.7$ Hz, 2H, -C₆H₄), 6.98 (s, 4H, -C₆H₂), 6.65 (d, $J(\text{H,H})=7.5$ Hz, 2H, -C₆H₄), 4.05 (m, 12H, -OCH₂), 1.81 (m, 12H, -OCH₂CH₂), 1.45-1.26 (m, 156H, -[CH₂]₇₈), 0.90 (t, $J(\text{H,H})=6.9$ Hz, 18H, -CH₃); ^{13}C NMR (75 MHz, CDCl_3 , 40°C): δ_c 166.53, 163.74, 163.29, 162.68, 155.43, 152.29, 133.56, 132.38, 131.07, 128.39, 114.37, 113.43, 110.75, 73.09, 68.37, 31.88, 29.54,

29.34, 29.07, 25.97, 22.67, 14.10; Elemental analyses: calculated for C₁₁₇H₂₀₉N₃O₇ (%), C, 79.40; H, 11.90; N, 2.37; Found, C, 79.31; H, 11.87; N, 2.32.

References:

- S1** A. Weisberger and F. S. Praskver, in *Organic Solvents*, International publishers Inc., New York, 1956, pp. 1263.
- S2** M. J. Frisch, G. W. Trucks, H. B. Schlegel, G. E. Scuseria, M. A. Robb, J. R. Cheeseman, G. Scalmani, V. Barone, B. Mennucci, G. A. Petersson, H. Nakatsuji, M. Caricato, X. Li, H. P. Hratchian, A. F. Izmaylov, J. Bloino, G. Zheng, J. L. Sonnenberg, M. Hada, M. Ehara, K. Toyota, R. Fukuda, J. Hasegawa, M. Ishida, T. Nakajima, Y. Honda, O. Kitao, H. Nakai, T. Vreven, J. A. Montgomery, Jr., J. E. Peralta, F. Ogliaro, M. Bearpark, J. J. Heyd, E. Brothers, K. N. Kudin, V. N. Staroverov, R. Kobayashi, J. Normand, K. Raghavachari, A. Rendell, J. C. Burant, S. S. Iyengar, J. Tomasi, M. Cossi, N. Rega, J. M. Millam, M. Klene, J. E. Knox, J. B. Cross, V. Bakken, C. Adamo, J. Jaramillo, R. Gomperts, R. E. Stratmann, O. Yazyev, A. J. Austin, R. Cammi, C. Pomelli, J. W. Ochterski, R. L. Martin, K. Morokuma, V. G. Zakrzewski, G. A. Voth, P. Salvador, J. J. Dannenberg, S. Dapprich, A. D. Daniels, Ö. Farkas, J. B. Foresman, J. V. Ortiz, J. Cioslowski, and D. J. Fox, *Gaussian 09 (Revision A.02)*, Gaussian, Inc., Wallingford CT, 2009.
- S3** A.D. Becke, *J. Chem. Phys.*, 1993, 98, 5648.
- S4** C. Lee, W. Yang and R.G. Parr, *Phys. Rev. B*, 1988, 37, 785.
- S5** R. Dennington II, T. Keith, J. Millam, K. Eppinnett, W. L. Hovell and R. Gilliland, *Gauss View 03*, Semichem, Inc., Shawnee Mission, KS. 2003.

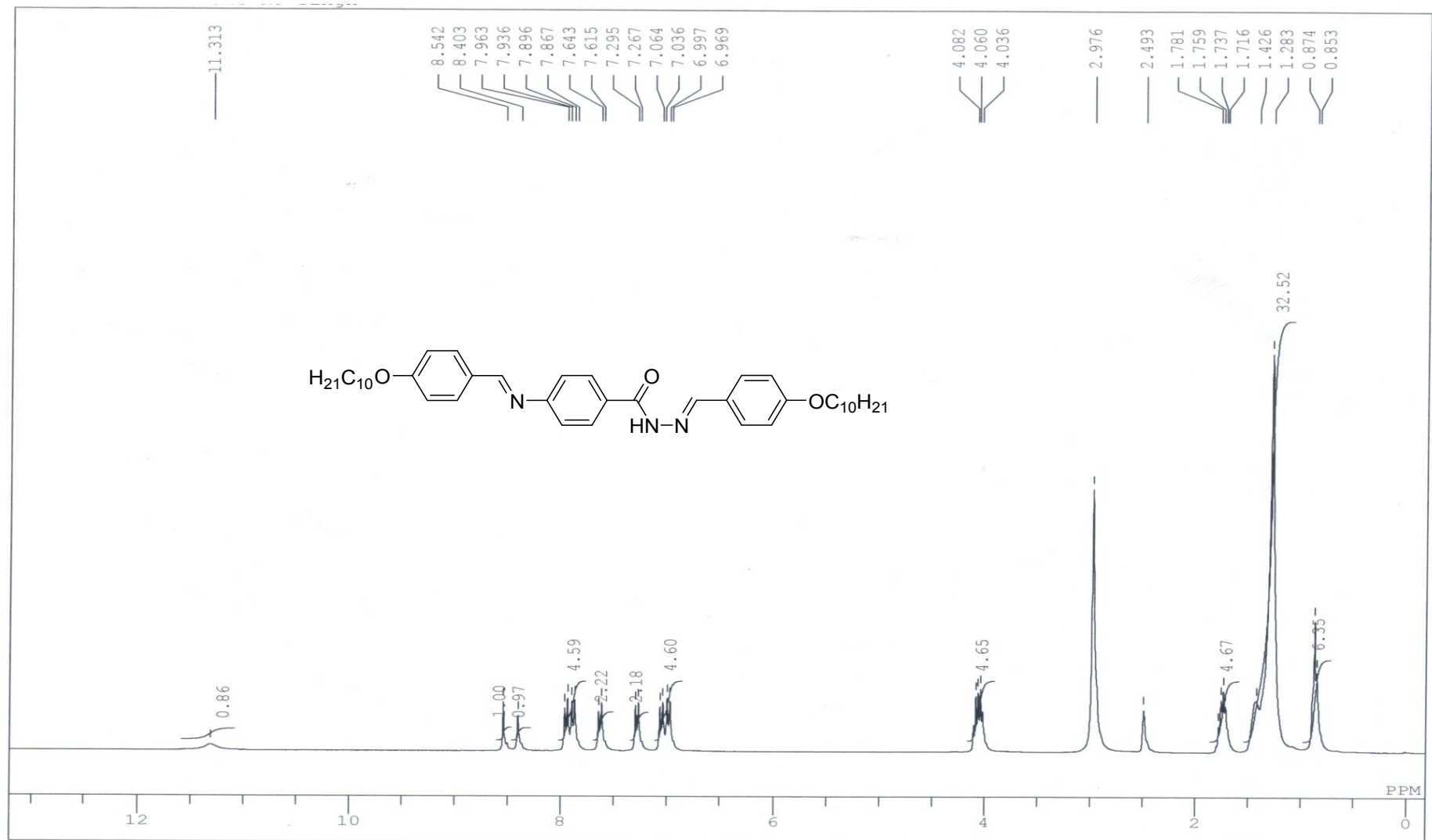


Fig. SI 1 ¹H NMR spectrum of N-(4-(decyloxy)benzylidene)-4-((4'-(decyloxy)benzylidene)amino)benzohydrazide, **1** (n=10)

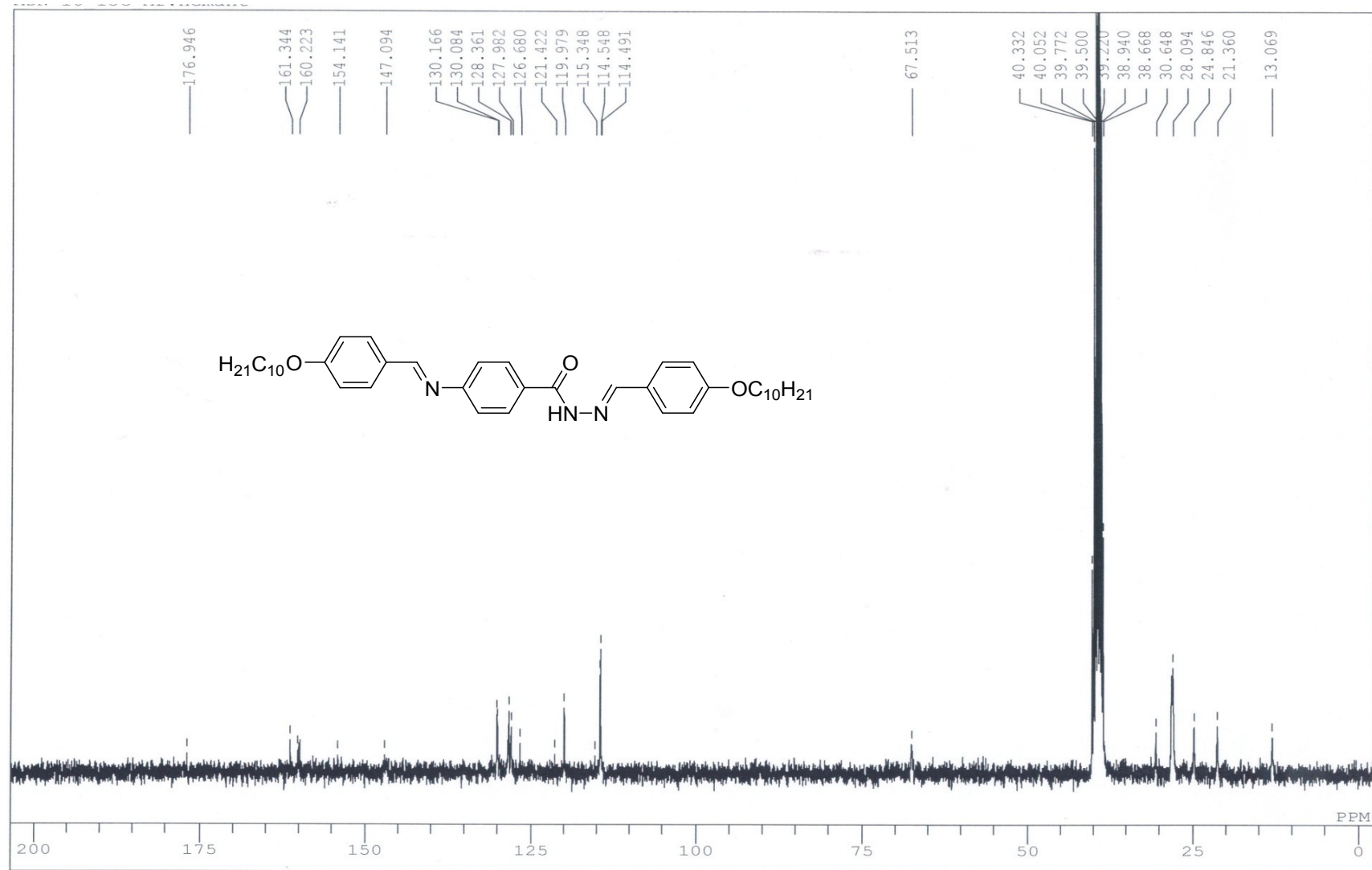


Fig. SI 2 ¹³C NMR spectrum of N-(4-(decyloxy)benzylidene)-4-((4'-(decyloxy)benzylidene)amino)benzohydrazide, **1** (n=10)

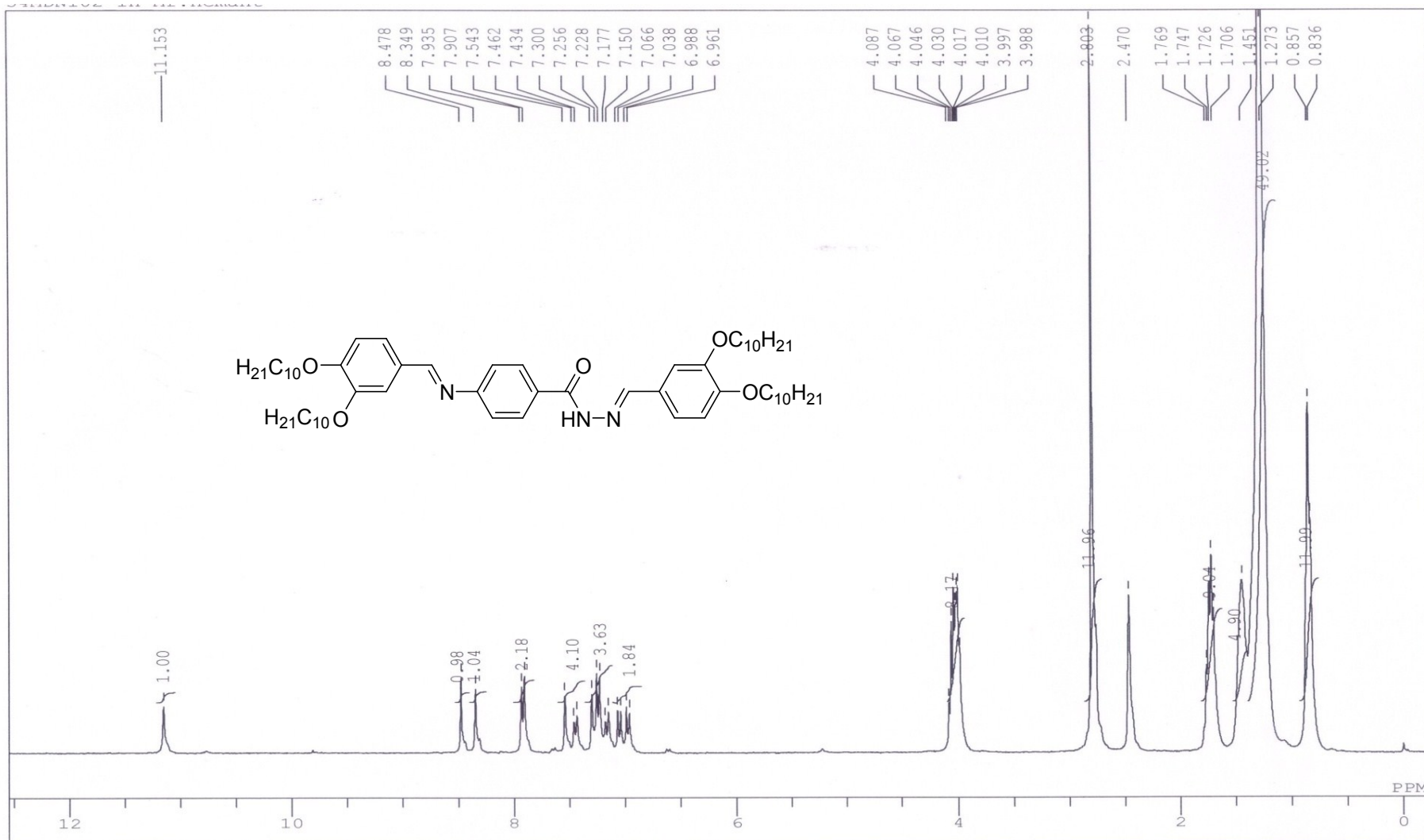


Fig. SI 3 ¹H NMR spectrum of N-(3,4-bis(decyloxy)benzylidene)-4-((3',4'-bis(decyloxy)benzylidene)amino)benzohydrazide, 2 (n=10)

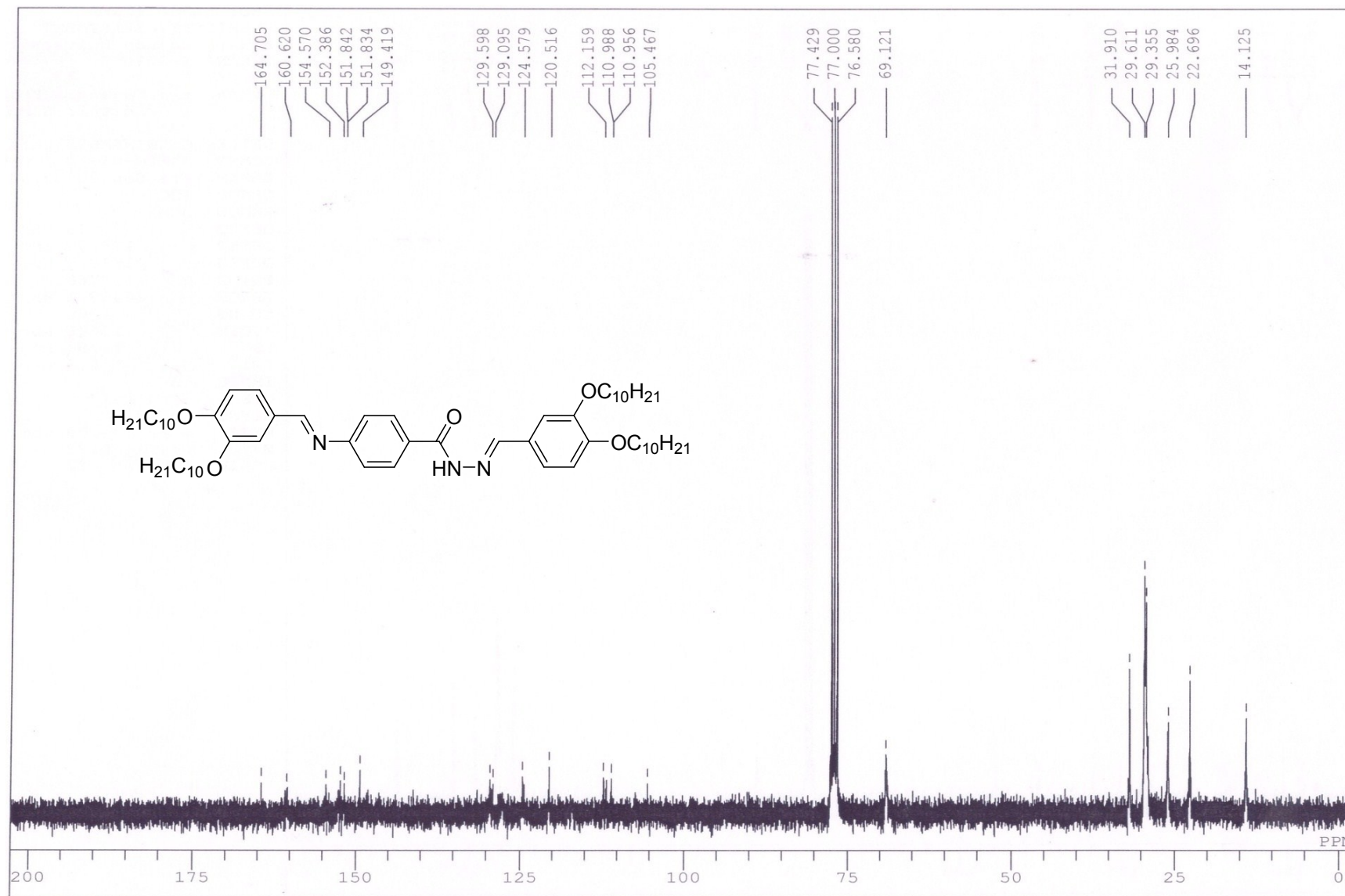


Fig. SI 4 ¹³C NMR spectrum of N-(3,4-bis(decyloxy)benzylidene)-4-((3',4'-bis(decyloxy)benzylidene)amino)benzohydrazide, **2** (n=10)

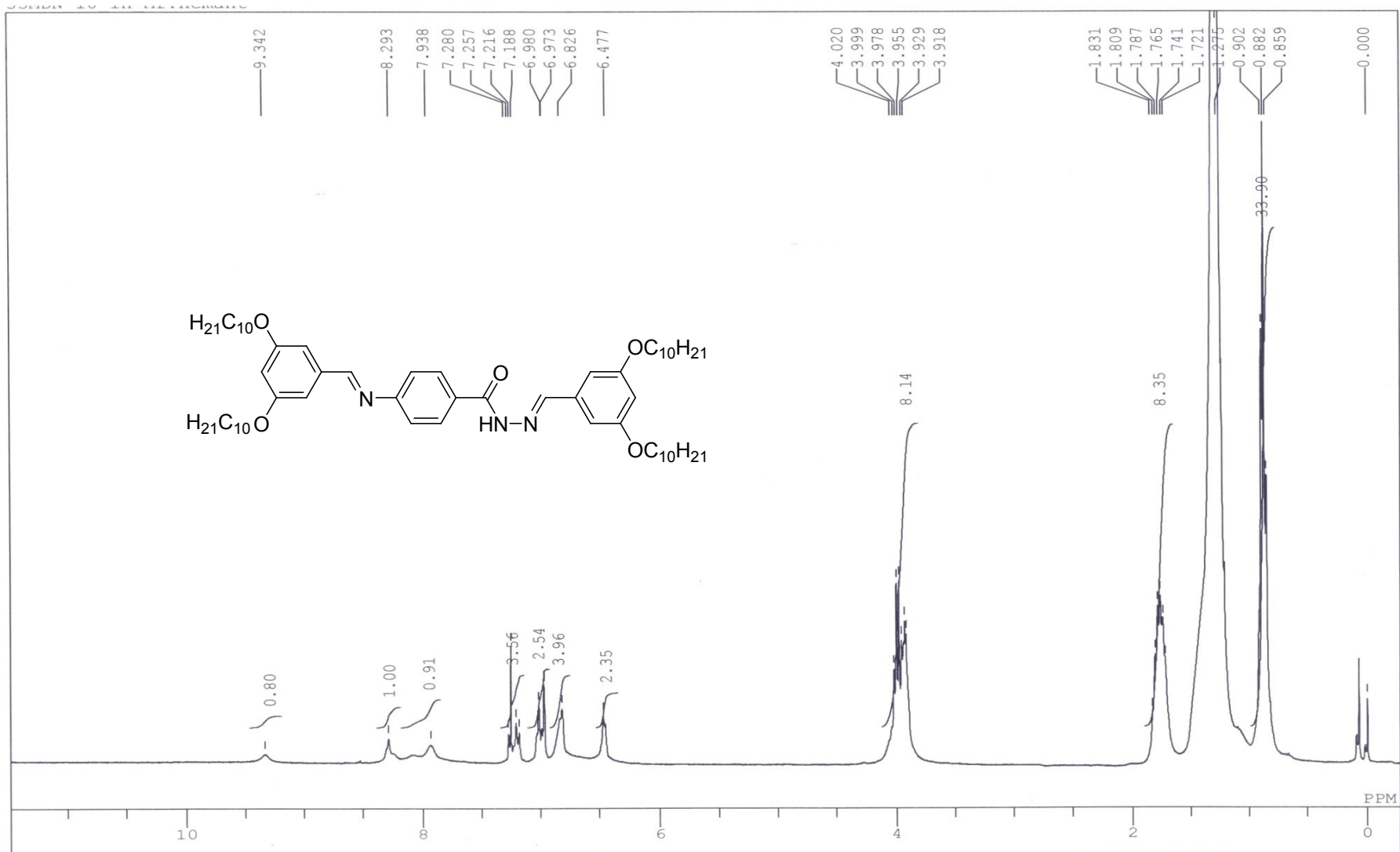


Fig. SI 5 ¹H NMR spectrum of N-(3,5-bis(decyloxy)benzylidene)-4-((3',5'-bis(decyloxy)benzylidene)amino)benzohydrazide, **3** (n=10)

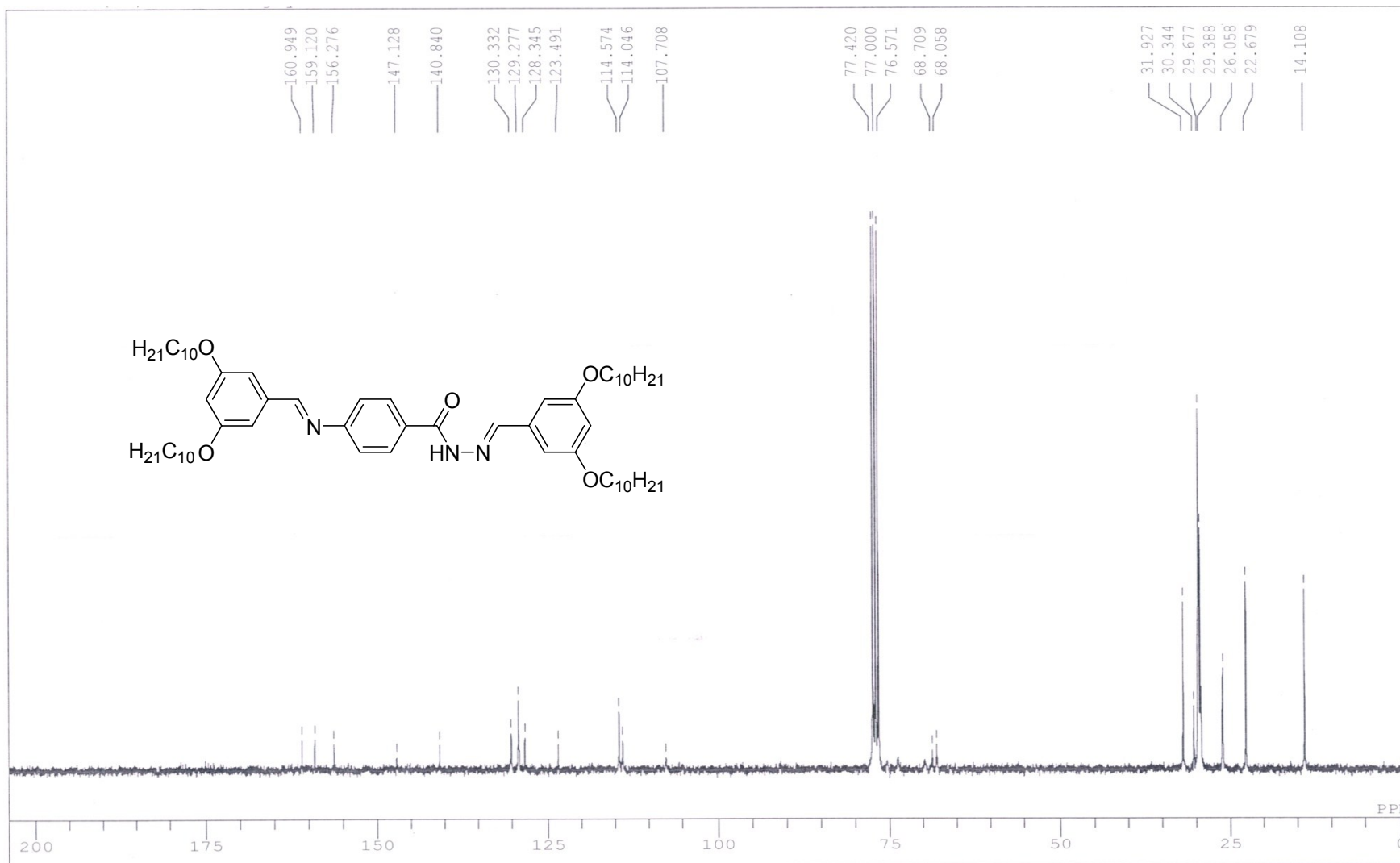


Fig. SI 6 ¹³C NMR spectrum of N-(3,5-bis(decyloxy)benzylidene)-4-((3',5'-bis(decyloxy)benzylidene)amino)benzohydrazide, **3** (n=10)

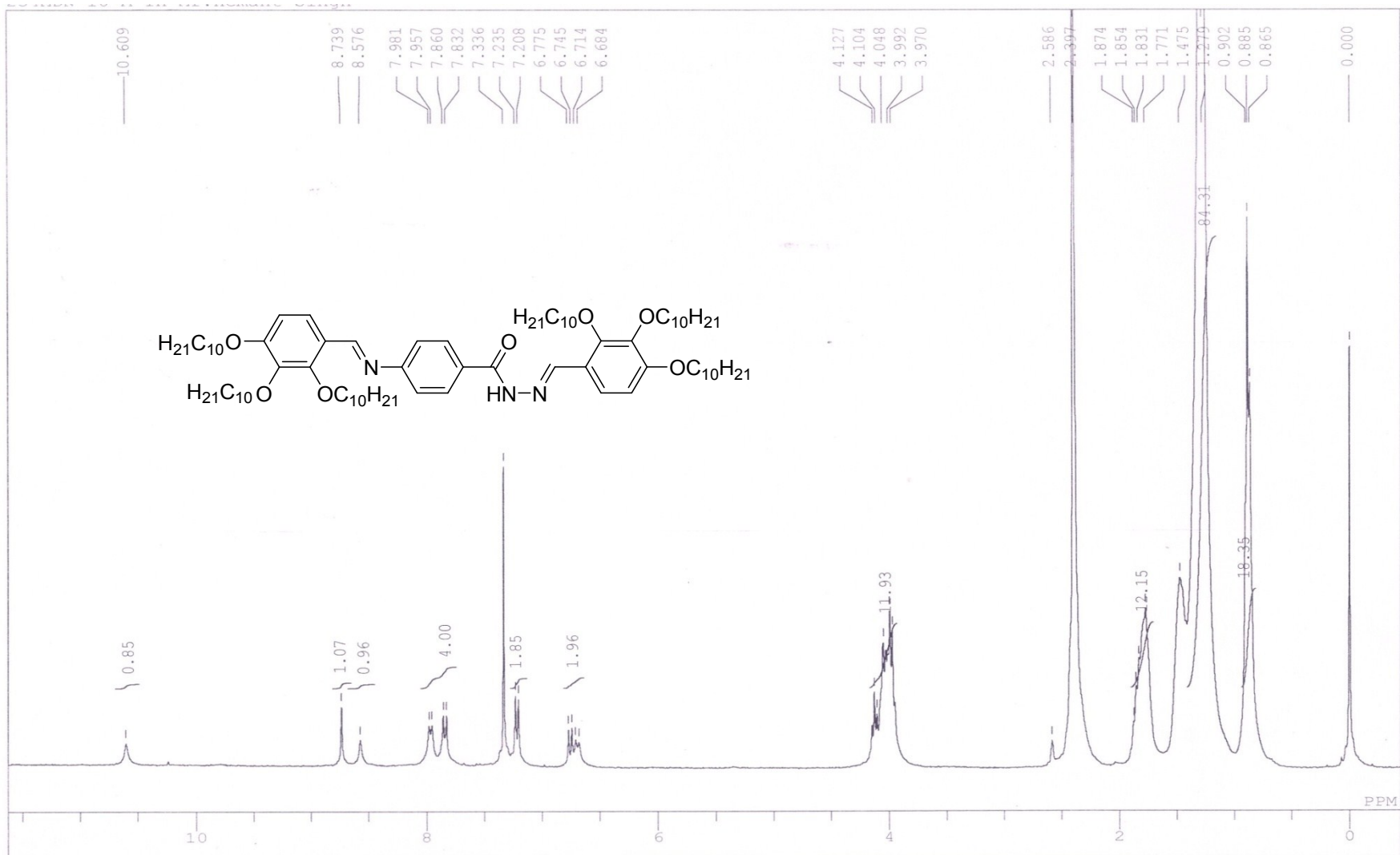


Fig. SI 7 ¹H NMR spectrum of N-(2,3,4-tris(decyloxy)benzylidene)-4-((2',3',4'-tris(decyloxy)benzylidene)amino)benzohydrazide, **4** (n=10)

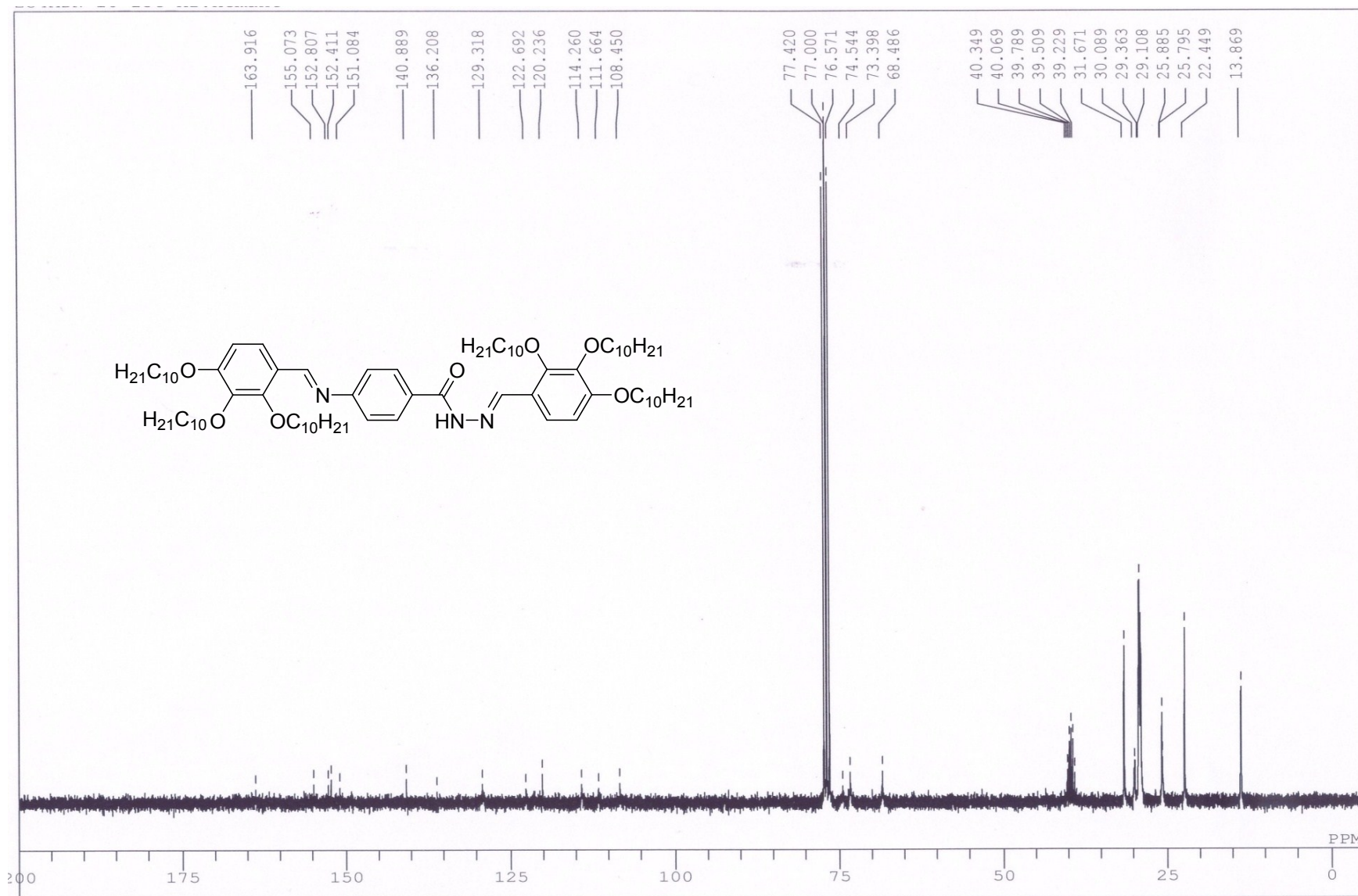


Fig. SI 8 ^{13}C NMR spectrum of N-(2,3,4-tris(decyloxy)benzylidene)-4-((2',3',4'-tris(decyloxy)benzylidene)amino)benzohydrazide, **4** (n=10)

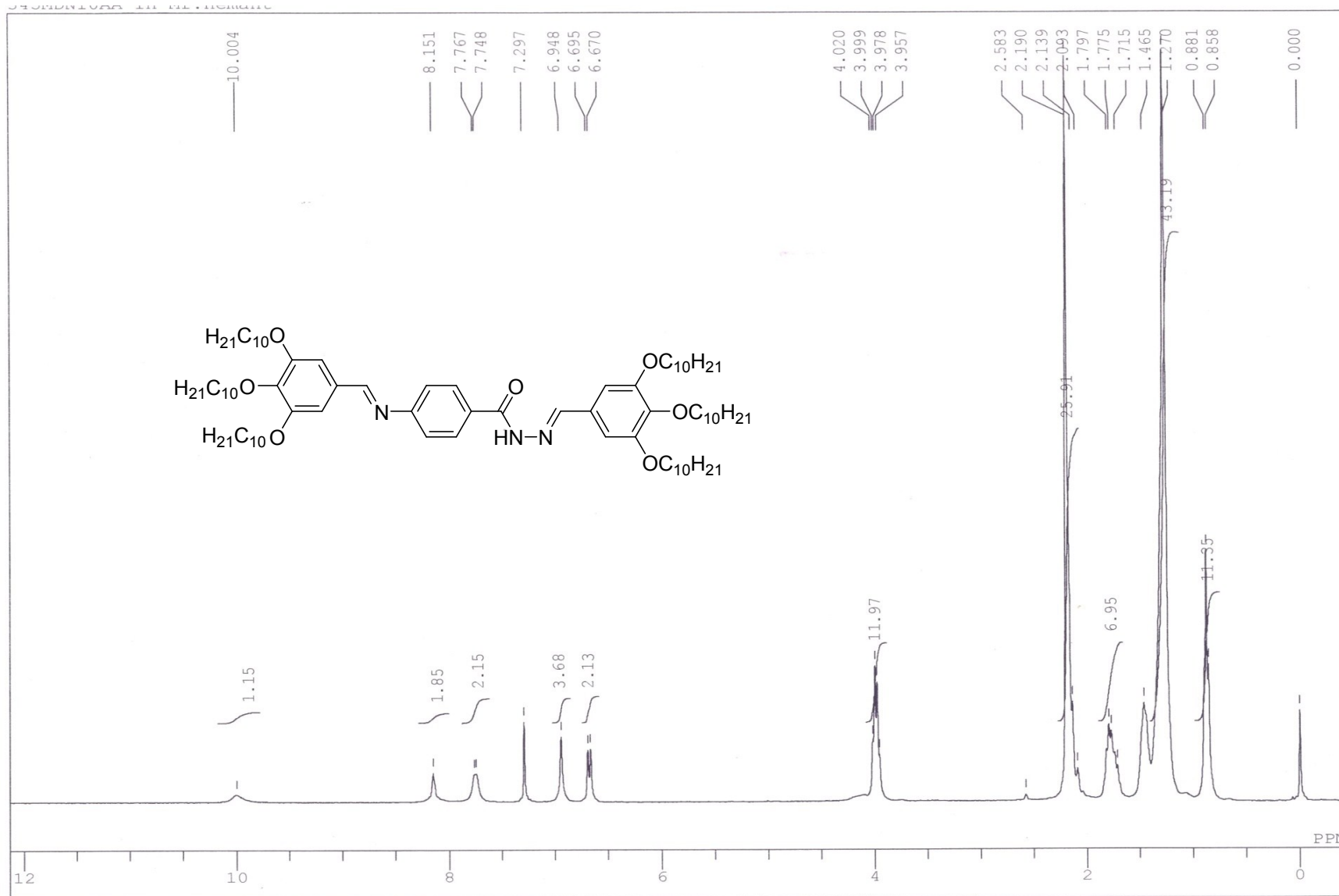


Fig. SI 9 ¹H NMR spectrum of N-(3,4,5-tris(decyloxy)benzylidene)-4-((3',4',5'-tris(decyloxy)benzylidene)amino)benzohydrazide, **5** ($n=10$)

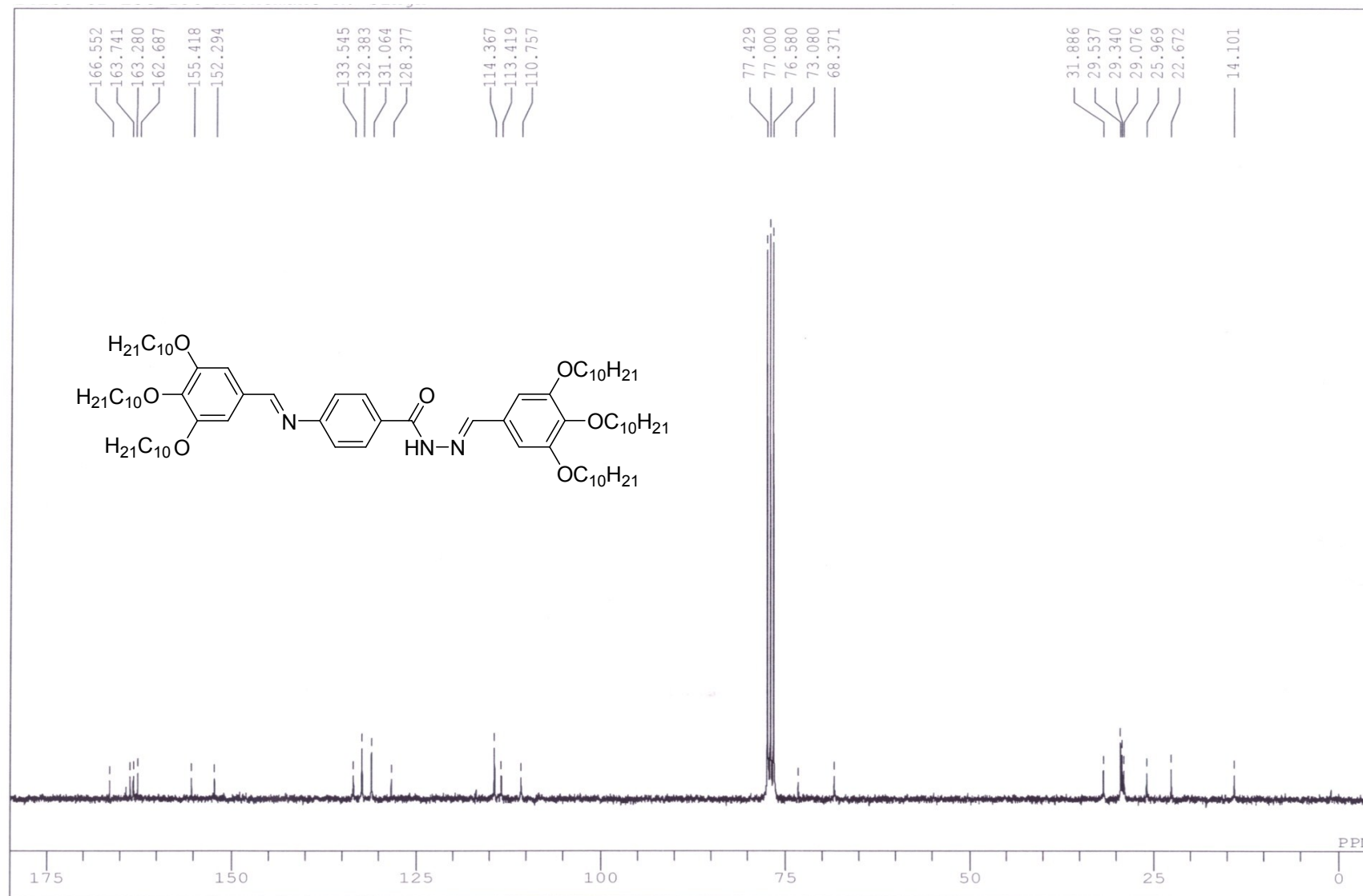
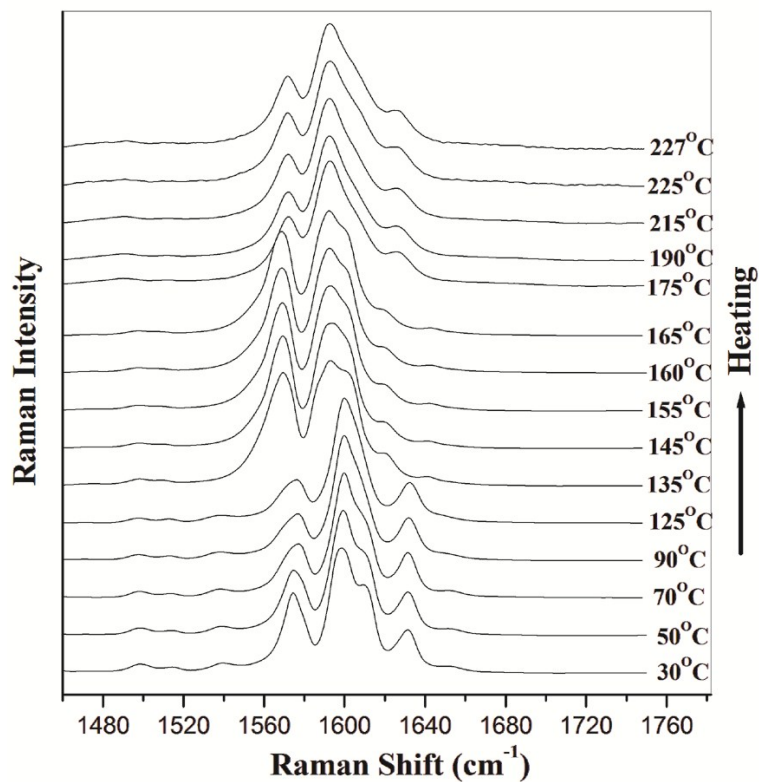
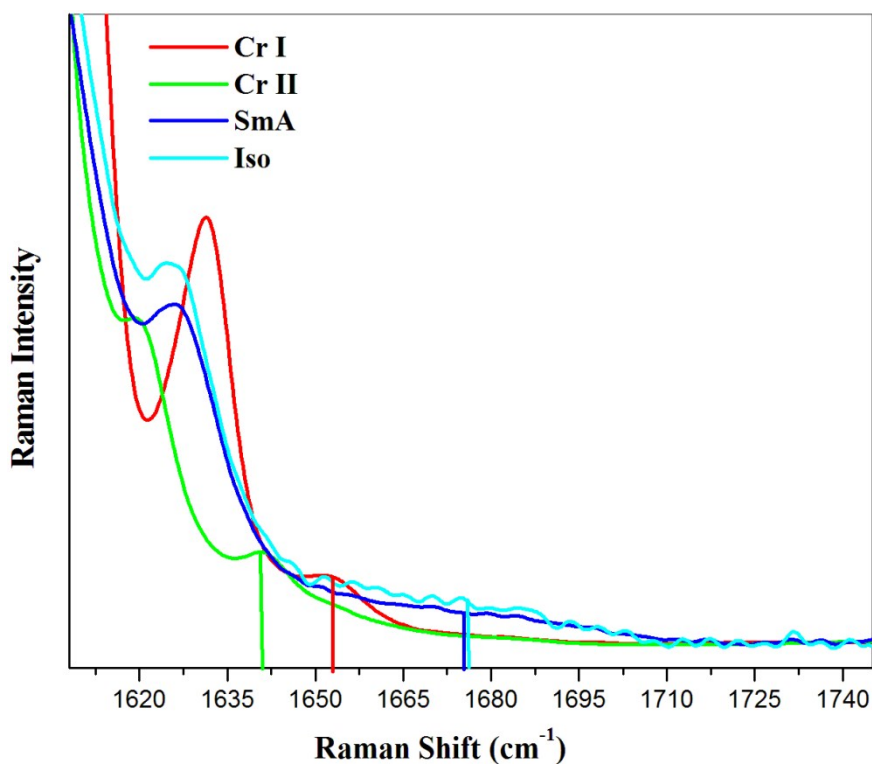


Fig. SI 10 ¹³C NMR spectrum of N-(3,4,5-tris(decyloxy)benzylidene)-4-((3',4',5'-tris(decyloxy)benzylidene)amino)benzohydrazide, **5** (n=10)



(a)



(b)

Fig. SI 11 Raman Spectra of compound N-(4-(dodecyloxy)benzylidene)-4-((4'-(dodecyloxy)benzylidene)amino)benzohydrazide, **1** ($n=12$) at different phases (Cr \rightarrow Cr' 171.6 $^{\circ}$ C \rightarrow SmA 221.8 $^{\circ}$ C \rightarrow i) (a) 1460-1740 cm^{-1} and (b) 1610-1740 cm^{-1} .

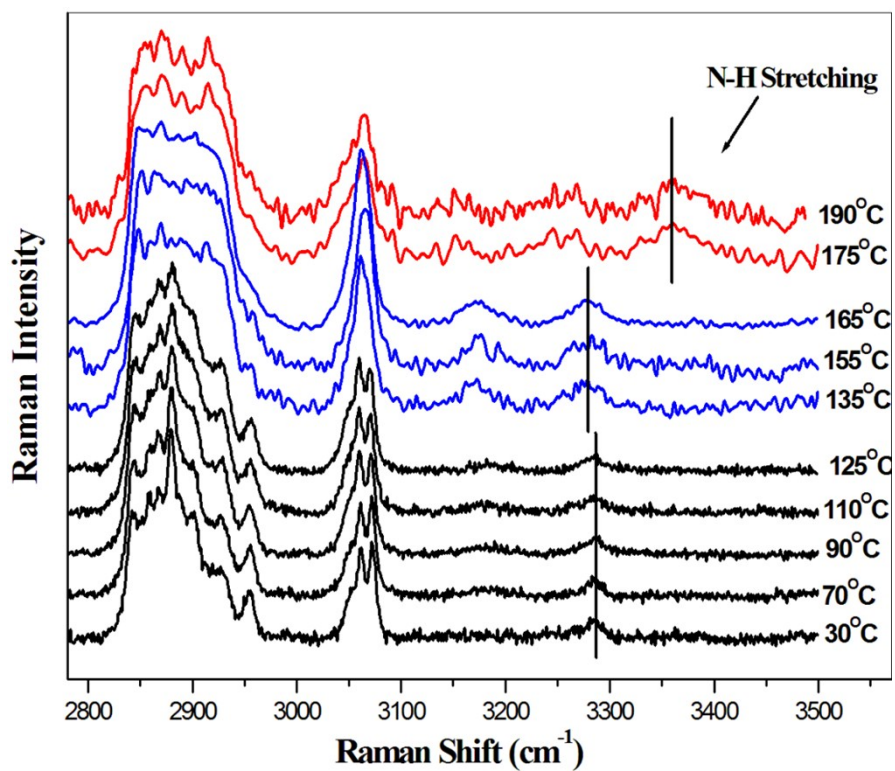


Fig. SI 12 Raman Spectra of compound N-(4-(dodecyloxy)benzylidene)-4-{{4'-(dodecyloxy)benzylidene)amino} benzohydrazide, **1**(n=12) at different phases, 2770-3500 cm^{-1} .

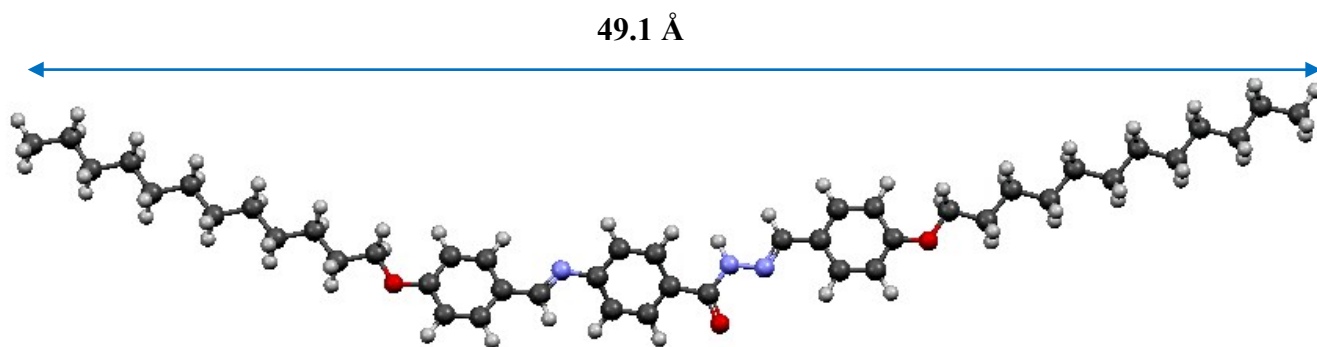


Fig. SI 13 The optimized structure of N-(4-(dodecyloxy)benzylidene)-4-{{4'-(dodecyloxy)benzylidene)amino} benzohydrazide, **1**(n=12) by DFT method at B3LYP/6-311++G(d,p) level.

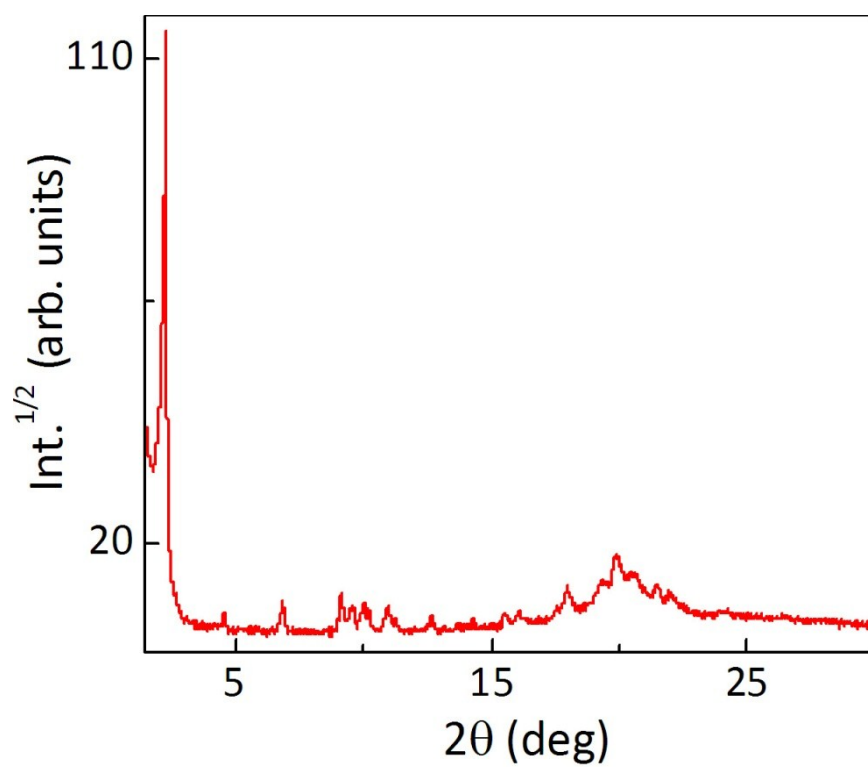


Fig. SI 14 XRD pattern of compound 2 (n=12) at 125°C.

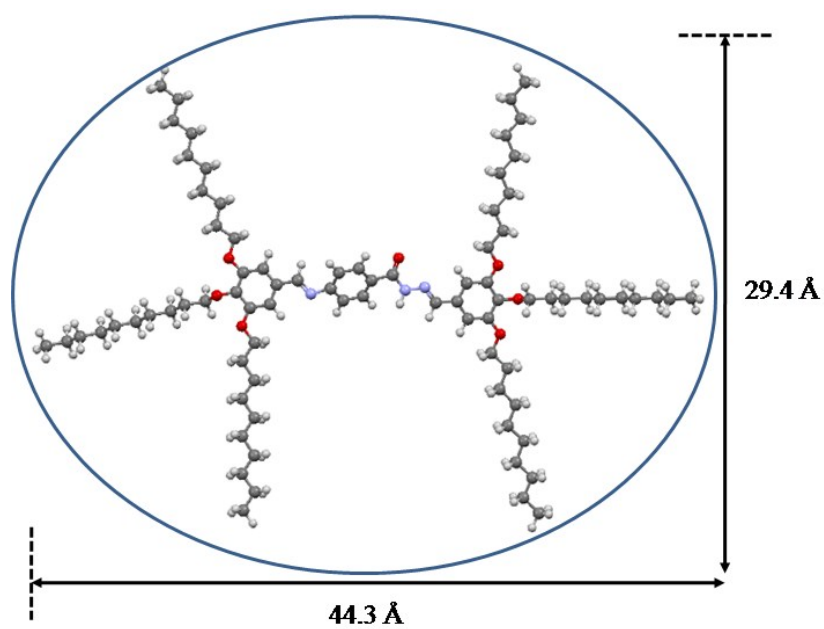


Fig. SI 15 The optimized structure of N-(3,4,5-tris(decyloxy)benzylidene)-4-((3',4',5'-tris(decyloxy)benzylidene)amino)benzohydrazide (**5**, n=10).

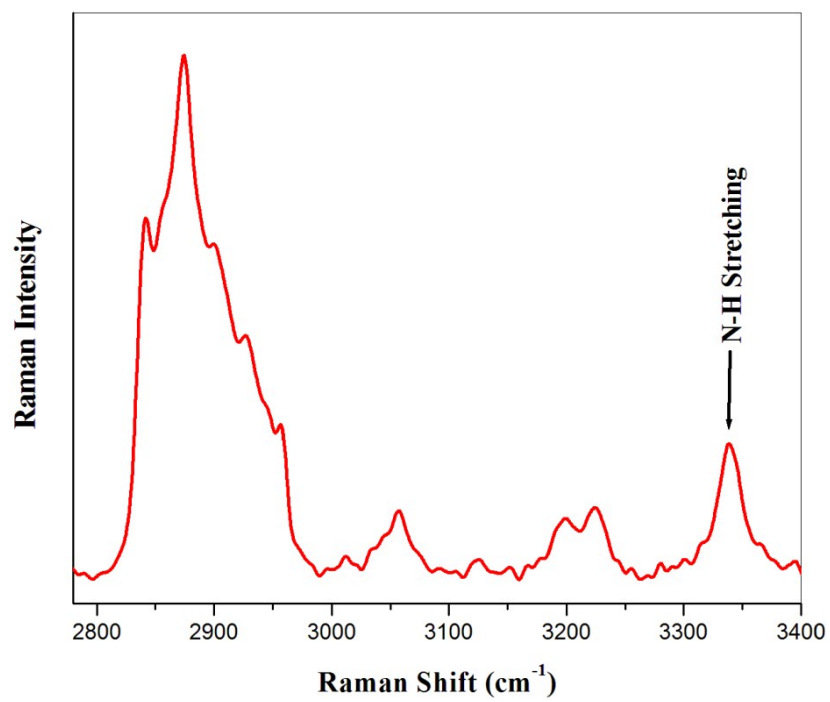
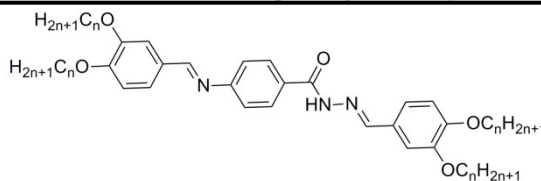
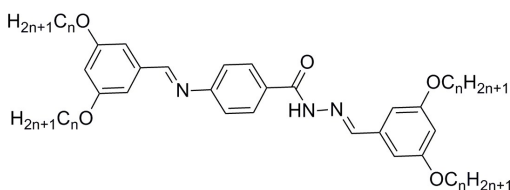


Fig. SI 16 Room temperature Raman spectrum of compound **5**(n=10).

Table SI 1. Thermal Transitions and Corresponding Thermodynamic Parameters

Compound	T, °C [ΔH , kJ/mol; ΔS , J/(mol K)]	
	Heating	Cooling
2 (n=5)	Cr 139.8 (16.2; 39.1) Cr' 197.6 (32.5; 69.1) i	i 193.2 (-38.8; -83.2) Cr
2 (n=6)	Cr 173.5 (23.2; 51.9) Cr' 192.3 (28.8; 61.8) i	i 184.7 (-36.8; -80.4) Cr
2 (n=7)	Cr 188.5 (41.7; 90.4) i	i 183.8 (-29.7; -65.0) Cr' 113.8 (-20.1; -52.0) Cr
2 (n=8)	Cr 182.9 (49.4; 108.3) i	i 171.5 (-46.3; -104.1) Cr
2 (n=10)	Cr 141.9 (57.5; 138.3) Cr' 180.3 (12.9; 28.4) i	i 175.0 (-7.2; -16.1) Cr
2 (n=12)	Cr 169.7 (42.4; 95.7) i	i 163.9 (-23.6; -53.9) Cr' 136.3 (-12.5; -30.6) Cr
2 (n=14)	Cr 163.7 (72.2; 165.4) i	i 161.2 (-54.2; -124.9) Cr
2 (n=16)	Cr 157.5 (60.2; 139.8) i	i 148.9 (-36.5; -86.6) Cr

Abbreviations: Cr & Cr' = crystalline state, i = isotropic liquid.

Table SI 2. Thermal Transitions and Corresponding Thermodynamic Parameters

Compound	T, °C [ΔH , kJ/mol; ΔS , J/(mol K)]	
	Heating	Cooling
3 (n=5)	Cr 172.2 (55.1; 123.8) i	i 160.2 (-42.1; -97.2) Cr' 146.7 (-9.3; -22.1) Cr
3 (n=6)	Cr 130.9 (32.4; 80.2) Cr' 163.8 (24.6; 56.5) i	i 157.3 (-57.0; -132.4) Cr
3 (n=7)	Cr 161.6 (45.1; 103.8) i	i 160.8 (-36.6; -84.4) Cr' 153.5 (-15.7; -36.7) Cr
3 (n=8)	Cr 115.8 (26.6; 68.4) Cr' 154.8 (67.8; 158.4) i	i 144.2 (-46.6; -111.4) Cr
3 (n=10)	Cr 150.9 (84.8; 199.9) i	i 135.6 (-82.1; -200.9) Cr
3 (n=12)	Cr 134.5 (13.4; 32.8) Cr' 148.1 (29.2; 69.2) i	i 142.5 (-37.1; -89.2) Cr
3 (n=14)	Cr 143.1 (49.4; 118.6) i	i 138.5 (-48.0; -116.7) Cr
3 (n=16)	Cr 140.8 (26.2; 63.2) i	i 136.9 (-42.4; -103.4) Cr

Abbreviations: Cr & Cr' = crystalline state, i = isotropic liquid.