Supplementary Information

A Hydrogen Bonding Motif for Forming Extended Assemblies

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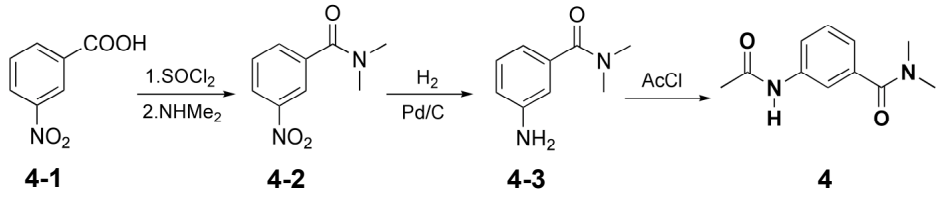
I. Synthetic Procedures

General. Chemicals were purchased from commercial sources and used as received. Unless otherwise specified, all solvents were removed with a rotary evaporator. Silica gel for analytical thin layer chromatography (TLC) and column chromatography (200~300 mesh) were purchased from Qingdao Haiyang Chemical Co., Ltd & Spegial Silica Gel Factory. The ^{1}H NMR spectra were recorded at 400 MHz and ^{13}C NMR spectra were measured at 100 MHz on a Bruker AV400 spectrometer at ambient temperature or 75℃ using $CDCl_3$ or DMSO-d_6 as solvent (purchased from Beijing Chongxi High-Tech Incubator Co.,Ltd). Chemical shifts are reported in parts per million downfield from TMS (tetramethylsilane). Coupling constant in ^{1}H NMR are expressed in Hertz. NOESY spectra (Bruker 600MHz) were performed at room temperature. All the Mass (ESI) Spectra were determined on a mass spectrometer (LCMS-2010, Shimadzu) at the Center of Mass Spectrum in the Institute of Chemistry, Chinese Academy of Science. Melting points were measured on a microscope hot stage melting point apparatus and are uncorrected. Elemental Analysis was performed on a Perkin Elmer 240 CHN Analyzer.

I-1. Synthesis of 4

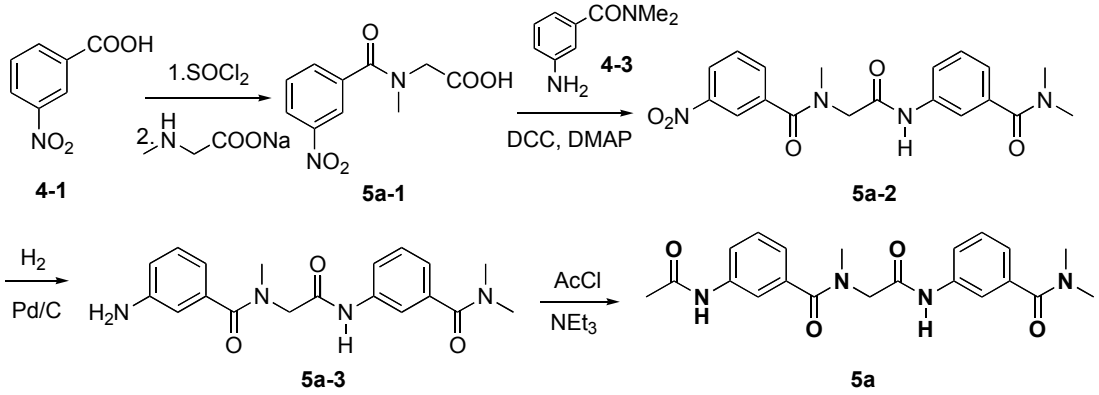


***N,N*-dimethyl-3-nitrobenzamide (4-2).** To a solution of **4-1** (30 g, 180 mmol) in thionyl chloride (40 mL) was added two drops of DMF. The mixture was stirred and refluxed for 8 hours. After removal of unreacted thionyl chloride under reduced pressure, the acid chloride was directly used for the next step. To a solution of dimethylamine hydrochloride (15.1 g, 185 mmol) and triethylamine (55 mL, 396 mmol) in dry dichloromethane (200 mL) cooled with an ice-bath was added dropwise the above acid chloride dissolved in 50 mL of CH_2Cl_2. The reaction mixture was stirred at room temperature overnight, then washed with brine, and dried over anhydrous Na_2SO_4. After removing the solvent and other volatile fractions, the remaining residue was recrystallized in methanol. Compound **4-2** (29.3 g, 83.8%) was obtained as slight yellow solid. (R_f=0.64, Petroleum ether/Acetone 1:1), m.p. 84.4-85.0 ℃. ^{1}H NMR (400 MHz, $CDCl_3$) δ3.02 (s, 3H), 3.16 (s, 3H), 7.63 (t, J = 7.8 Hz, 1H), 7.78 (d, J = 7.6 Hz, 1H), 8.28 (d, J = 8.6 Hz, 1H), 8.30 (s, 1H)

3-Acetamido-*N,N*-dimethylbenzamide (4). To a solution of **4-2** (3.88 g, 20 mmoL) in THF (30 mL) and methanol (20 mL) was added 10% palladium on carbon (200 mg). The mixture was hydrogenated under 4 atmosphere pressure at room temperature until the reaction had completed as indicated by TLC (about 18 hours). The catalyst was removed by filtration, which was followed by evaporation of solvent. The remaining residue was directly used for the next step. To the obtained residue and triethylamine (4.2 mL, 30 mmol) in dry CH_2Cl_2 (20 mL) cooled in an

ice-bath was added dropwise acetyl chloride (1.5 mL, 20 mmol) dissolved in dry CH_2Cl_2 (10 mL). The mixture was stirred at room temperature overnight and was then washed with brine, dried over anhydrous Na_2SO_4. After removing the solvent, the remaining residue was purified by flash column chromatography (R_f =0.20, petroleum ether/acetone = 1/1) to afford **4** (3.18 g, 77.1%) as bright white solid. m.p.129.8-131.4℃. ^{1}H NMR (400 MHz, $CDCl_3$) δ2.15 (s, 3H), 2.99 (s, 3H), 3.11 (s, 3H), 7.08 (d, J = 7.6 Hz, 1H), 7.31(t, J = 7.8Hz, 1H), 7.46 (s, 1H), 7.64 (d, J = 8.0 Hz, 1H), 8.09 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ24.5, 35.4, 39.6, 118.6, 121.2, 122.2, 129.0, 136.6, 138.4, 168.7, 171.4. EI-MS(m/z): 206 (M^+, 46%), 162 (100%), 147 (34%), 134 (48%), 120 (47%), 92 (42%), 72 (11%), 65 (32%), 43 (54%). Anal. Calcd. for $C_{11}H_{14}N_2O_2$(**4**): C, 64.06; H, 6.84; N, 13.58. Found: C, 63.99; H, 6.48; N,13.57

I-2. Synthesis of 5a

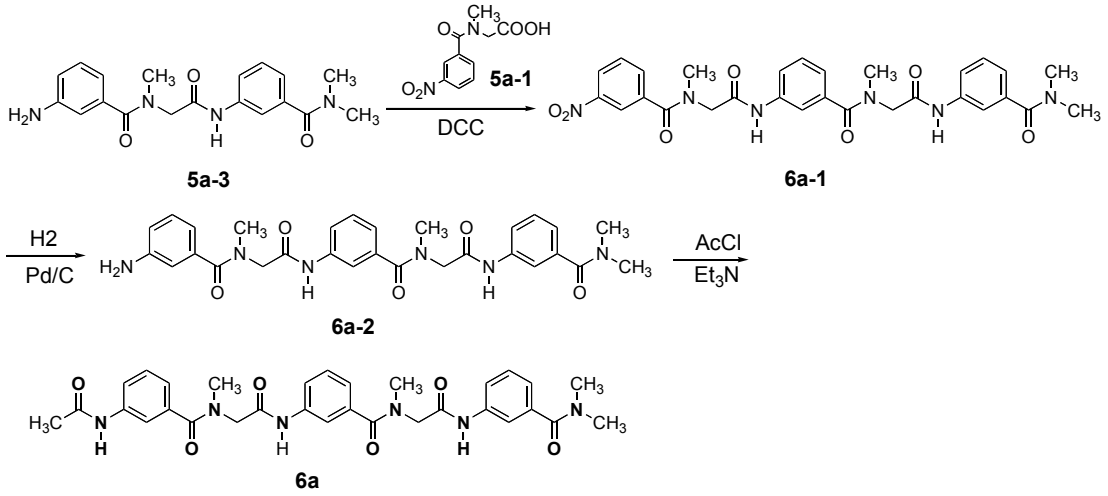


2-(*N*-Methyl-3-nitrobenzamido) acetic acid (5a-1). To a mixture of 3-nitrobenzoic acid (30 g, 180 mmol) and thionyl chloride (40 mL) was added two drops of DMF. The mixture was stirred and refluxed for 8 hours. After removal of the unreacted thionyl chloride under reduced pressure, the acid chloride, which was a liquid, was directly used for the next step without being dissolved in any solvent. To sarcosine (32 g, 360 mmol) and sodium hydroxide (20.8 g, 540 mmol) in water (150 mL) in an ice-bath was added dropwise the above acid chloride. The reaction mixture was stirred at room temperature overnight, to which 10g of NaCl was then added. The product precipiatated out of the solution. After filtration, the obtained solid was washed with cold water and was then stirred in dilute H_2SO_4 (pH ≈1). The volume of the mixture should be kept under 100 mL. The solid was collected by filration. Acid **5a-1** was obtained as a yellowish solid (29.6 g, 69%). m.p.144.0-145.0℃. (Note: Acid **5a-1** exists as a mixture of two conformers due to limited rotation around the tertiary amide C-N bond (*trans*: 75%; *cis*: 25% in $CDCl_3$) at room temperature, and as a single conformer at an elevated temperature, which is revealed by its ^{1}H-NMR spectra recorded at 25 ℃ and 75 ℃ in DMSO-d_6 respectively). ^{1}H NMR (400 MHz, $CDCl_3$, 25℃) Major conformer: δ3.10 (s, 3H), 4.35 (s, 3H), 7.66 (t, J = 8.0 Hz, 1H) , 7.76 (d, J = 7.3Hz, 1H), 8.33 (d, J = 8.5 Hz, 1H), 8.36 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$, 25℃) Major conformer: δ38.9, 49.3, 122.4, 125.0, 129.9, 133.3, 136.5, 148.0, 169.9, 172.8. ESI-MS: 237.0 ($M-H^+$). Anal. Calcd. for $C_{10}H_{10}N_2O_5$: C, 50.42; H, 4.23; N, 11.76. Found: C, 50.22; H, 4.14; N, 11.85.

***N,N*-Dimethyl-3-(2-(N-methyl-3-nitrobenzamido)acetamido)benzamide (5a-2).** To compound **5a-1** (6.00 g, 25 mmol), DCC (6.19 g, 30 mmol) and DMAP (0.61 g, 5 mmol) in dry THF (60 mL) cooled in an ice-bath was added dropwise amine **4-3**（4.11 g, 25 mmol) dissolved in dry THF (40 mL). The mixture was stirred at room temperature for 24 hours and was then filtrated to remove insoluble fractions. THF was then removed from the filtrate, which left a residue that was purified by flash column chromatography to afford **5a-2** (6.18 g, 64.3%) as a white solid. (R_f =0.18，petroleum ether/acetone 1:1) M.p=179.6-182.8 ℃. ^{1}H NMR (400 MHz, $CDCl_3$) Major conformer: δ2.98 (s, 3H), 3.11 (s, 3H), 3.13 (s, 3H), 4.31 (s, 3H), 7.03 -8.38(m, 8H), 9.36 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) Major conformer: δ35.4, 39.3, 39.6, 52.4, 118.6, 121.2, 122.3, 122.6, 125.0, 129.0, 129.8, 133.4, 136.6, 136.7, 138.2, 148.0, 166.4, 170.0, 171.4. ESI-MS: 385.2 ($M+H^+$)，407.1 ($M+Na^+$), 423.1 ($M+K^+$). ESI-HRMS：Cacld. for $C_{19}H_{20}N_4O_5$, ($M+H^+$): 385.1512; found 385.1511.

***N,N*-dimethyl-3-(2-(N-methyl-3-acetamidobenzamido)acetamido)benzamide (5a)**. To a solution of **5a-2** (1.47 g, 3 mmoL) in THF (10 mL) and methanol (20 mL) was added 10% palladium on carbon (100 mg). The mixture was hydrogenated under 4 atmosphere pressure at room temperature until the starting mafterial was shown by TLC analysis to have completely reacted (~24 hours). Removal of the catalyst by filtration, followed by the evaporation of solvent, led to a residue (petroleum ether/acetone 1:2, R_f =0.21) that was used for the next step directly. To the residue and triethylamine (0.7 mL, 5 mmol) dissolved in dry CH_2Cl_2 (10 mL) placed in an ice-bath was added dropwise acetyl chloride (0.4 mL, 5 mmol) dissolved in dry dichloromethane (10 mL). The mixture was stirred at room temperature for overnight. After removing the solvent, the remaining residue was purified by flash column chromatography (R_f =0.16, petroleum ether/acetone = 1/2) to afford **5a** (0.87 g, 73.0%) as a white solid. M.p = 178.7-179.6 ℃. ^{1}H NMR (400 MHz, DMSO-d_6, 75℃) δ2.04 (s, 3H), 2.94 (s, 6H), 3.01 (s, 3H), 4.20 (s, 2H), 7.06 (d, J = 7.6 Hz, 2H), 7.31-7.37(m, 2H)，7.56-7.62 (m, 2H)，7.67 (s, 2H), 9.83 (s, 1H)，9.94 (s, 1H). ^{13}C NMR (100 MHz, DMSO-d_6, 75℃) δ24.3, 37.1, 38.8, 51.4, 54.3, 118.1, 118.5, 120.5, 120.6, 121.6, 122.2, 129.0, 129.1, 137.2, 137.7, 139.2, 139.8,167.5, 168.8, 170.4, 171.3. ESI-MS: 397.2 ($M+H^+$)，419.2 ($M+Na^+$), 435.2 ($M+K^+$). Anal. Calcd. for $C_{21}H_{24}N_4O_4$(**5a**): C, 63.62; H, 6.10; N, 14.13. Found: C, 63.77; H, 6.14; N, 14.53.

I-3. Synthesis of 6a

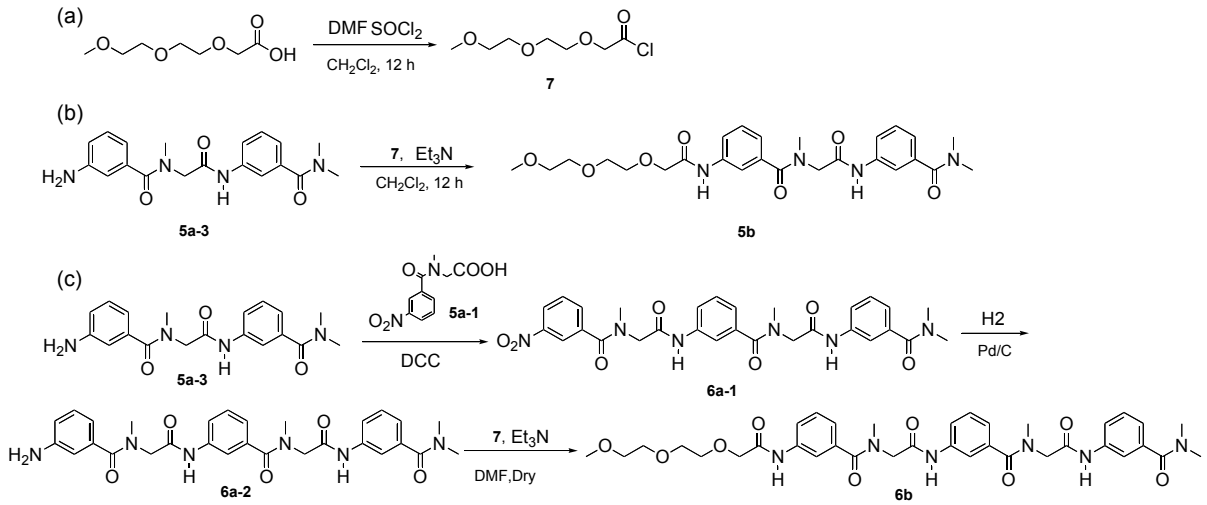


***N,N*-dimethyl-3-(2-(N-methyl-3-(2-(N-methyl-3-nitrobenzamido)acetamido) benzamido)acetamido)benzamide (6a-1).** To the compound **5a-1** (1.66 g, 7 mmol), DCC (1.86 g, 9 mmol) and DMAP (0.17 g, 1.4 mmol) in dry THF (40 mL) with ice-bath was added dropwise the amine **5a-3**（2.48 g, 7 mmol) in dry THF (10 mL). The mixture was stirred at room temperature for 24 hours. And was then filtrated, after evaporation of the THF of the filtrate, the residue was purified by flash column chromatography (R_f =0.13, Petroleum ether/Acetone 1:2) to afford **6a-1** (1.58 g, 39.2 %) as white solid. ^{1}H NMR (400 MHz, DMSO-d_6, 75℃) δ2.96 (s, 6H), 3.03 (s, 3H), 3.04 (s, 3H), 4.23 (s, 4H), 7.08 (d, J = 7.6 Hz, 1H), 7.13(d, J = 7.1 Hz, 1H), 7.35-7.41 (m, 2H), 7.58-7.65 (m, 2H), 7.68 (s, 1H), 7.74-7.78 (m, 2H), 7.88 (s, 1H), 8.24 (s, 1H), 8.29 (d, J = 7.6 Hz, 1H), 9.98 (s, 2H). ^{13}C NMR (100 MHz, DMSO-d_6, 75℃) δ34.3, 37.2, 38.7, 41.8, 51.5, 56.2, 118.3, 118.4, 120.5, 120.6, 120.8, 120.9, 122.0, 122.2, 124.6, 129.1, 129.2, 130.7, 133.6, 137.3, 137.6, 138.2, 139.0, 148.3, 167.2, 167.4, 169.4, 170.4, 171.2. ESI-MS: 575.2 ($M+H^+$)，597.2 ($M+Na^+$), 613.3 ($M+K^+$). Anal. Calcd. for $C_{29}H_{30}N_6O_7$(12): C, 60.62; H, 5.26; N, 14.63. Found: C, 60.59; H, 5.10; N, 14.99.

***N,N*-dimethyl-3-(2-(N-methyl-3-(2-(N-methyl-3-acetamidobenzamido)acetamido)benzamido)acetamido)benza mide (6a).** To a solution of **6a-1** (1.09 g, 2 mmoL) in THF (10 mL) and methanol (20 mL) was added 10% palladium on carbon (80 mg). The mixture was hydrogenolysed under 4 atmosphere pressure at room temperature until all of the starting mafterial was shown by TLC analysis to have reacted (about 24 hours). Removal of the catalyst by filtration, after the evaporation of the solvent, the residue (Petroleum ether/Acetone 1:2, R_f =0.09) was used for the next step directly. To the residue (**6a-2**) above and triethylamine (0.7 mL, 5 mmol) in dry dichloromethane (10 mL) with ice-bath was added dropwise acetyl chloride (0.4 mL, 5 mmol) in dry dichloromethane (10 mL). The mixture was stirred at room temperature overnight and after the removal the solvent, the residue was purified by flash column chromatography (Petroleum ether/Acetone 1:2)(TLC, $CHCl_3$:MeOH=10:1, R_f=0.25) to afford **6a** (0.73 g, 62.2%) as white solid. m.p=243.7-245.9℃. ^{1}H NMR (400 MHz, DMSO-d_6, 75 ℃) δ2.06 (s, 3H), 2.96 (s, 6H), 3.02 (s, 3H), 3.04 (s, 3H), 4.22 (s, 4H), 7.07-7.13 (m, 3H), 7.33-7.41 (m, 3H), 7.58-7.73 (m, 6H), 9.85 (s, 1H), 9.98 (s, 2H). ^{13}C NMR (400 MHz, DMSO-d_6, 75 ℃) δ24.3, 34.7, 37.2, 39.0, 51.2,

54.0, 54.6, 118.0, 118.3, 120.4, 120.5, 120.7, 121.6, 122.0, 122.2, 129.0, 129.1, 129.2, 137.1, 137.3, 137.7, 139.0, 139.1, 139.7, 139.8, 167.4, 167.5, 168.8, 168.9, 170.5, 171.3. ESI-MS: 587.4 ($M+H^+$)，609.3 ($M+Na^+$). ESI-HRMS: Calcd. for $C_{31}H_{34}N_6O_6$(T5), ($M+H^+$): 587.2618; Found: 587.2614.

I-4. Synthesis of compound 5b and 6b



***N,N*-Dimethyl-3-(2-(N-methyl-3-(2-(2-(2-methoxyethoxy)ethoxy)acetamido)benzamido)acetamido) benzamide (5b)**. To a mixture of of 2-(2-(2-methoxyethoxy)ethoxy)acetic acid (3.56 g, 20 mmol) and thionyl chloride (2.2 mL, 30 mmol) in CH_2Cl_2 was added two drops of DMF. The mixture was stirred and refluxed for 12 hours at room temperature. After removal of CH_2Cl_2 and unreacted thionyl chloride under reduced pressure, the generated acid chloride **7** was used for the next step directly. To a solution of **5a-3** (1.77 g, 5 mmol) and Et_3N (0.8 mL, 6 mmol) in dry CH_2Cl_2 (10 mL) placed in an ice-bath was added dropwise the acid chloride **7** (1.18 g, 6 mmol) in dry CH_2Cl_2 (10 mL). The reaction mixture was stirred at room temperature overnight. After removal of the solvent, the residue was purified by flash column chromatography (R_f =0.10, $CHCl_3$:MeOH=20:1) to afford **5b** (1.74 g, 67.8%) as a white solid. M.p=91.2-92.7℃. ^{1}H NMR (400 MHz, $CDCl_3$) δ2.99 (s, 3H), 3.09 (s, 3H), 3.16 (s, 3H), 3.32 (s, 3H), 3.56 (m, 2H), 3.71 (m, 4H), 3.77 (d, *J* = 4.3 Hz, 2H), 4.12 (s, 2H), 4.25 (s, 2H), 7.12 (d, *J* = 7.5 Hz, 1H), 7.25 (m, 1H), 7.33 (t, *J* = 8.0 Hz, 1H), 7.38 (m, 1H), 7.65-7.57 (s, 3H), 7.88 (s, 1H), 8.90 (s, 1H), 9.10 (s, 1H). ^{13}C NMR (100 MHz, $CDCl_3$) δ35.4, 39.2, 39.6, 53.2, 58.9, 70.2, 70.5, 70.7, 71.3, 71.8, 118.6, 119.1, 121.0, 121.8, 122.5, 123.3, 129.0, 129.2, 135.6, 136.9, 137.7, 138.2, 167.0, 168.6, 171.3, 172.3. ESI-MS: 515.3 ($M+H^+$), 537.3 ($M+Na^+$). ESI-HRMS: Calcd. for $C_{26}H_{34}N_4O_7Na$, ($M+Na^+$): 537.2325; found: 537.2314.

N,N-dimethyl-3-(2-(N-methyl-3-(2-(N-methyl-3-nitrobenzamido)acetamido)benzamido)acetamido) benzamide (6a-1). To a solution of compound **5a-1** (1.66 g, 7 mmol), DCC (1.86 g, 9 mmol) and DMAP (0.17 g, 1.4 mmol) dissolved in dry THF (40 mL) placed in an ice-bath was added dropwise amine **5a-3** (2.48 g, 7 mmol) in dry THF (10 mL). The reaction mixture was stirred at room temperature for 24 hrs and was then filtrated, followed by

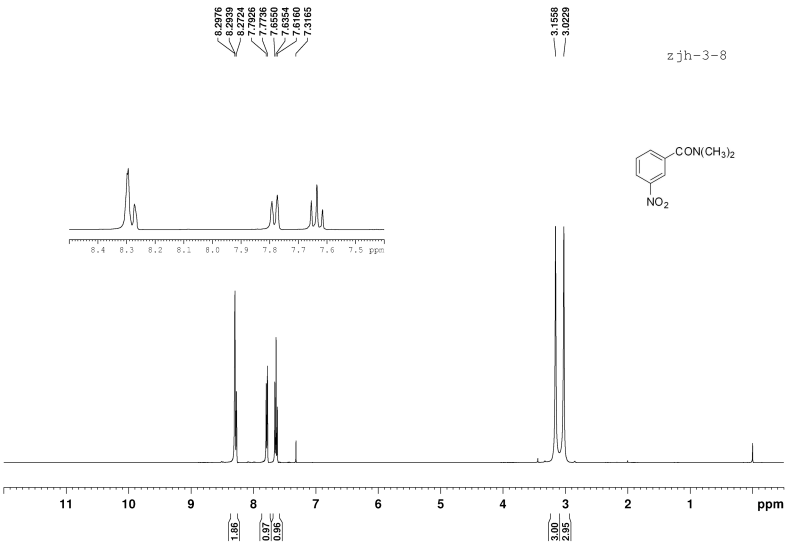
evaporation of THF. The remaining residue was purified by flash column chromatography (R_f =0.13, petroleum ether/acetone 1:2) to afford **6a-1** (1.58 g, 39.2 %) as a white solid. ^{1}H NMR (400 MHz, DMSO-d_6, 75℃) δ2.96 (s, 6H), 3.03 (s, 3H), 3.04 (s, 3H), 4.23 (s, 4H), 7.08 (d, J = 7.6 Hz, 1H), 7.13 (d, J = 7.1 Hz, 1H), 7.35-7.41 (m, 2H), 7.58-7.65 (m, 2H), 7.68 (s, 1H), 7.74-7.78 (m, 2H), 7.88 (s, 1H), 8.24 (s, 1H), 8.29 (d, J = 7.6 Hz, 1H), 9.98 (s, 2H). ^{13}C NMR (100 MHz, DMSO-d_6, 75 ℃) δ34.3, 37.2, 38.7, 41.8, 51.5, 56.2, 118.3, 118.4, 120.8, 120.5, 120.6, 120.9, 122.0, 122.2, 124.6, 129.1, 129.2, 130.7, 133.6, 137.3, 137.6, 138.2, 139.0, 148.3, 167.2, 167.4, 169.4, 170.4, 171.2. ESI-MS: 575.2 ($M+H^+$), 597.2 ($M+Na^+$), 613.3 ($M+K^+$). Anal. Calcd. for $C_{29}H_{30}N_6O_7$: C, 60.62; H, 5.26; N, 14.63. Found: C, 60.59; H, 5.10; N, 14.99.

N,N-dimethyl-3-(2-(N-methyl-3-(2-(N-methyl-3-(2-(2-(2-methoxyethoxy)ethoxy)acetamido)benzamido)acetamido)benzamido)acetamido)benzamide (6b). To a solution of **6a-1** (2.87g, 5 mmoL) in THF (25 mL) and methanol (50 mL) was added 10% palladium on carbon (200 mg). The mixture was hydrogenolysed under 4 atmosphere pressure at room temperature until all of the starting mafterial was shown by TLC analysis to have reacted (~24 hours). Removal of the catalyst by filtration, followed by the evaporation of solvent, led to **6a-2** (petroleum ether/acetone 1:2, R_f =0.09) which was used in the next step directly. To a solution of **6a-2** (5 mmol) and Et_3N (0.8 mL, 6 mmol) in dry DMF (10 mL) placed in an ice-bath was added dropwise acid chloride **7** (1.18 g, 6 mmol) in dry DMF (10 mL). The mixture was stirred at room temperature for overnight. Removing the solvent under reduced pressure led to a solid residue that was purified by flash column chromatography (R_f =0.35, $CHCl_3$:MeOH=10:1). Compound **6b** was obtained as a white solid (2.26 g, 64.3%). M.p=170.6-172.5℃. ^{1}H NMR (400 MHz, DMSO-d_6, 75 ºC): δ3.01-2.94 (m, 12H), 3.24 (s, 3H), 3.46 (t, J = 5.0 Hz, 2H), 3.57 (t, J = 4.4 Hz, 2H), 3.62 (d, J = 4.5 Hz, 2H), 3.68 (d, J = 4.2 Hz, 2H), 4.07 (s, 2H), 4.02 (s, 4H), 7.07 (m, 3H), 7.35 (m, 3H), 7.73-7.56 (m, 6H), 9.53 (s, 1H), 9.96 (s, 2H). ^{13}C NMR (100 MHz, DMSO-d_6, 75 ºC): δ34.4, 36.3, 37.6, 39.1, 51.6, 54.5, 58.5, 70.1, 70.2, 70.9, 71.0, 71.8, 118.3, 118.4, 118.7, 120.6, 120.7, 121.1, 122.0, 122.2, 122.3, 129.0, 129.1, 129.2, 137.2, 137.3, 137.7, 138.8, 139.1, 139.2, 167.5, 168.9, 170.4, 171.2. ESI-MS: 705.5 ($M+H^+$), 727.5 ($M+Na^+$). ESI-HRMS: Calcd. for $C_{36}H_{44}N_6O_9Na$, ($M+Na^+$): 727.3067; Found: 727.3039.

II. NMR Spectra

II-1. 1D ^{1}H and ^{13}C NMR spectra

4-2



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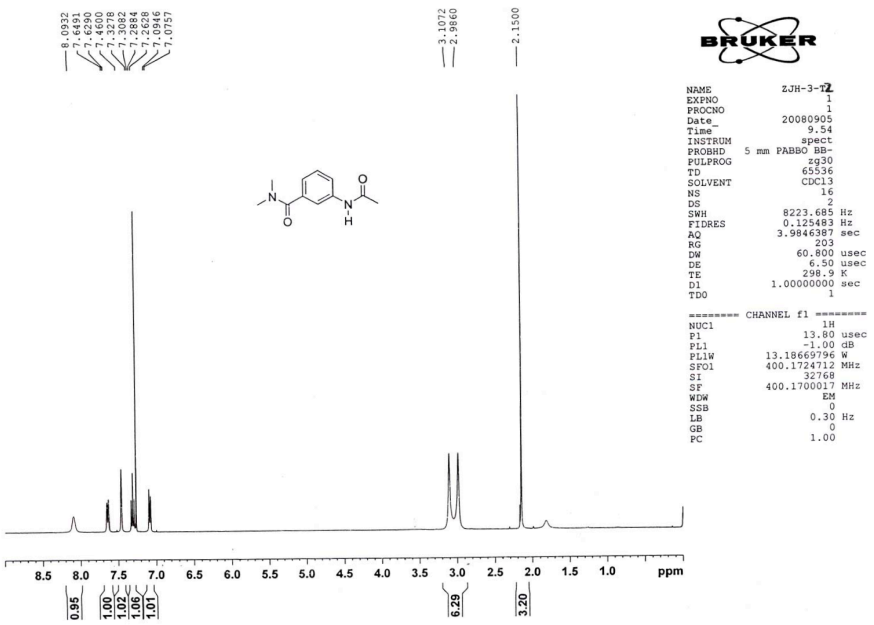

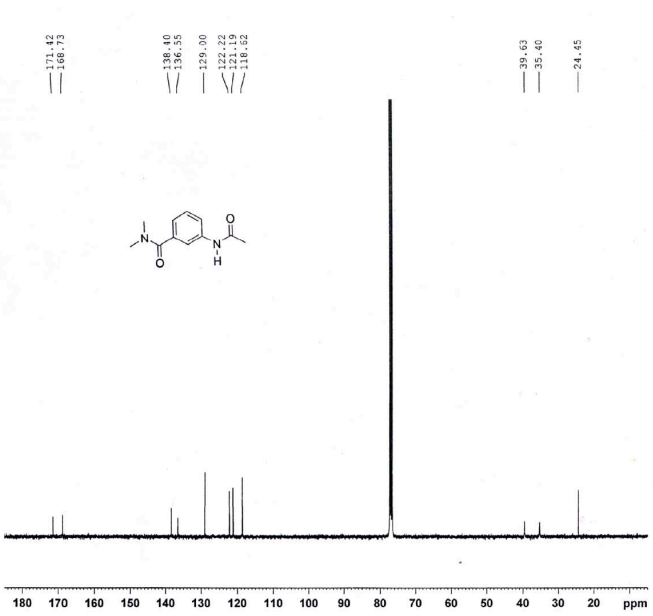

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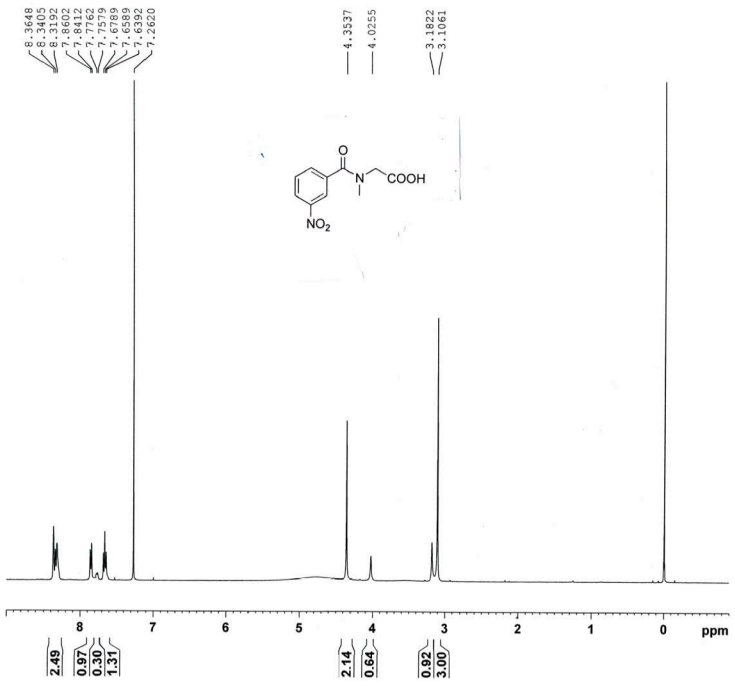

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PULPROG           zg30
TD              65536
SOLVENT         CDCl3
NS                 16
DS                  2
SWH          8223.685 Hz
FIDRES      0.125483 Hz
AQ          3.9846387 sec
RG                203
DW             60.800 usec
DE              6.50 usec
TE             297.7 K
D1          1.00000000 sec
TD0                 1

========== CHANNEL f1 ========
NUC1               1H
P1              13.80 usec
PL1             -1.00 dB
PL1W       13.18669796 W
SFO1      400.1724712 MHz
SI             32768
SF        400.1700022 MHz
WDW               EM
SSB                0
LB              0.30 Hz
GB                 0
PC              1.00
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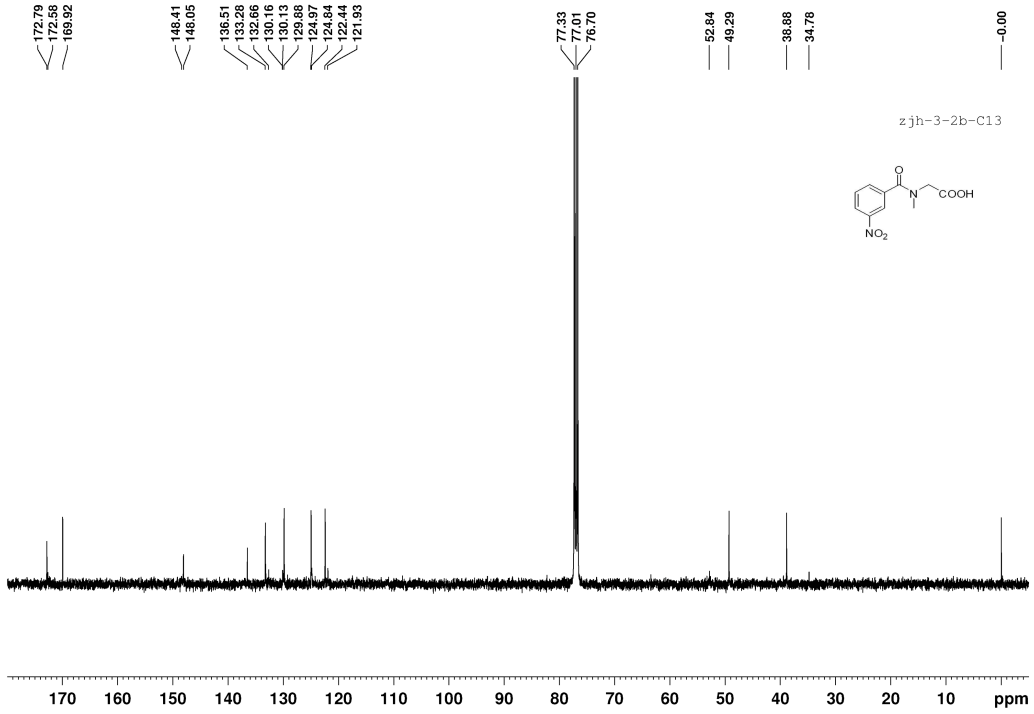
5a-1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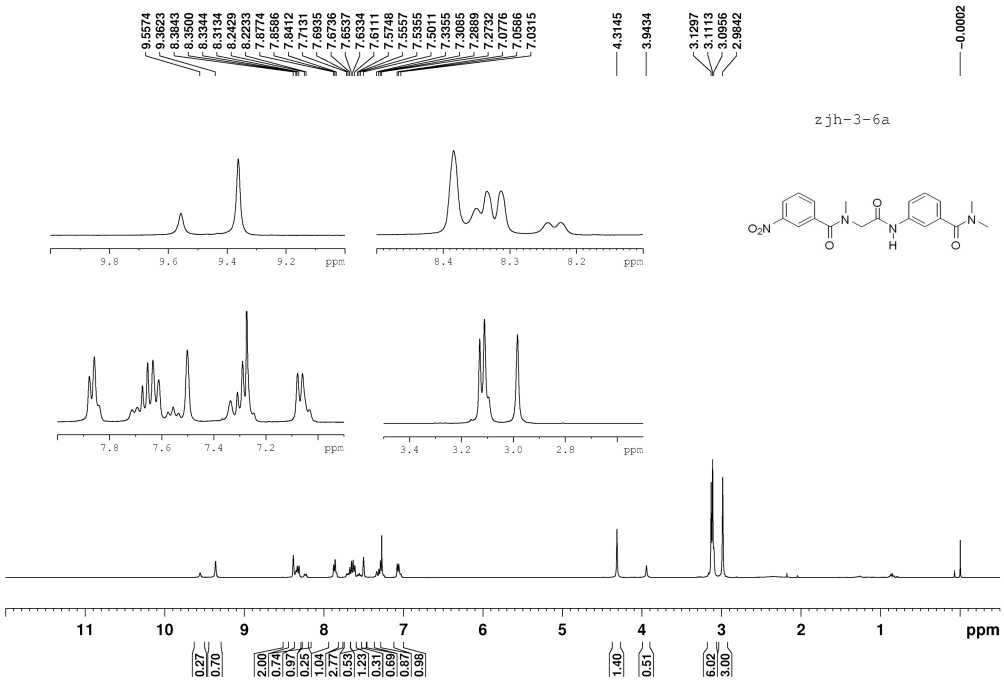
172.56
169.92
148.41
148.05
136.51
133.28
132.66
130.16
130.13
129.88
124.97
124.84
122.44
121.93
77.33
77.01
76.70
52.84
49.29
38.88
34.78
-0.00
zjh-3-2b-C13
COOH
NO2
170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 ppm

5a-1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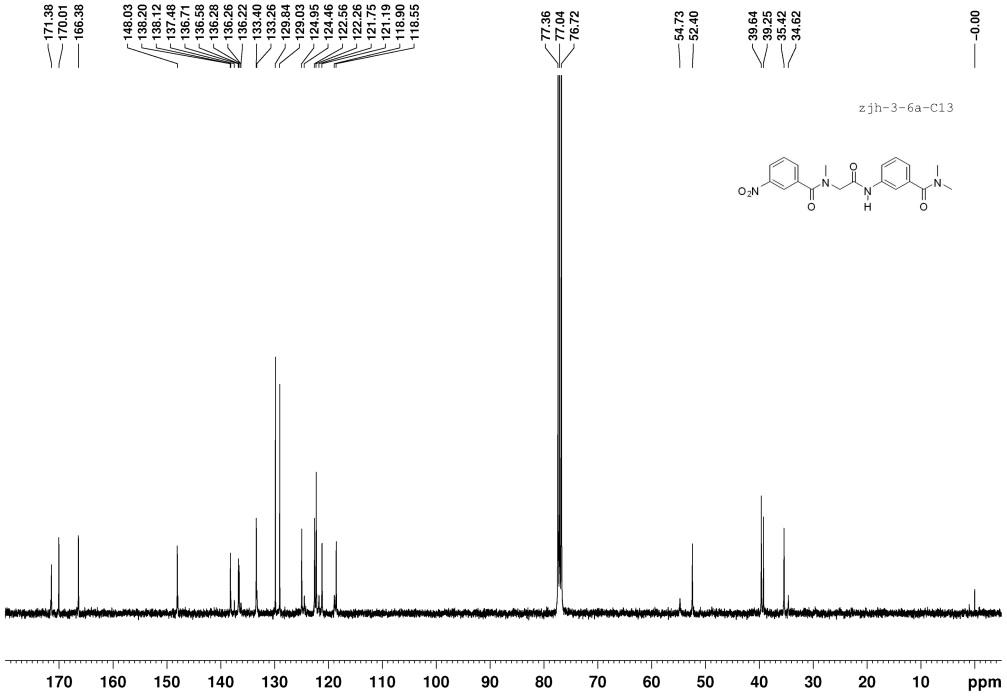
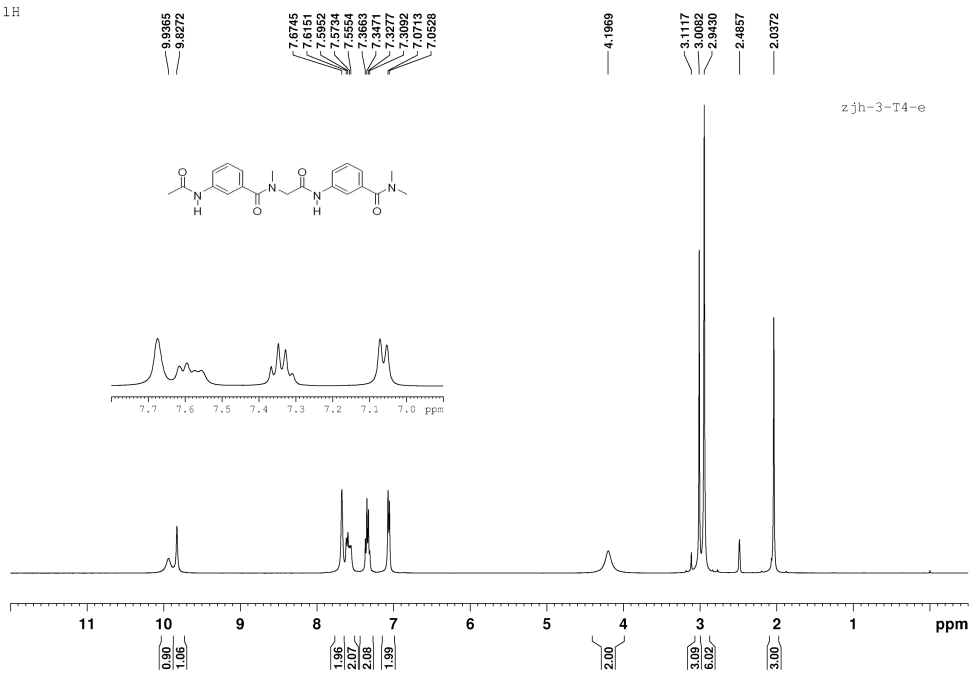
9.3623
8.3843
8.3500
8.3344
8.3134
8.2429
8.2233
7.8774
7.8586
7.8412
7.7131
7.6935
7.6736
7.6537
7.6334
7.6111
7.5748
7.5557
7.5355
7.5011
7.3355
7.3085
7.2889
7.2732
7.0776
7.0586
7.0315
4.3145
3.9434
3.1297
3.1113
3.0956
2.9842
-0.0002
zjh-3-6a
9.8 9.6 9.4 9.2 ppm
8.4 8.3 8.2 ppm
7.8 7.6 7.4 7.2 ppm
3.4 3.2 3.0 2.8 ppm
11 10 9 8 7 6 5 4 3 2 1 ppm
0.27
0.70
2.00
0.74
0.97
0.25
1.04
2.77
0.53
1.23
0.31
0.69
0.87
0.98
1.40
0.51
6.02
3.00

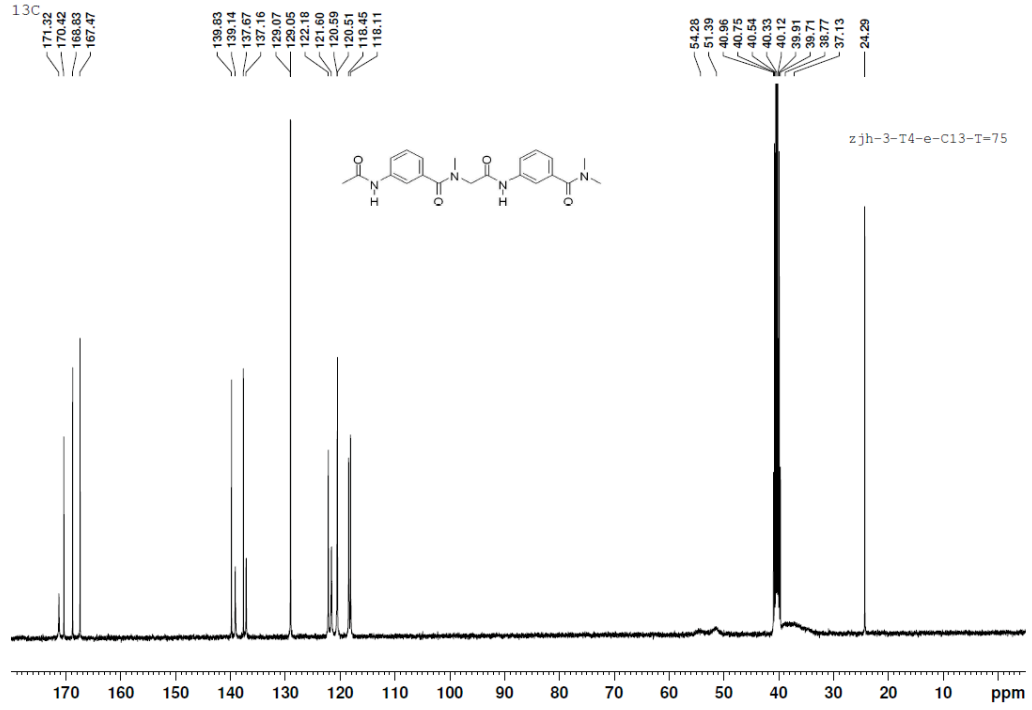
5a-2


171.38
170.01
166.38
148.03
138.20
138.12
137.48
136.71
136.58
136.28
136.26
136.22
133.40
133.26
129.84
129.03
124.95
124.46
122.56
122.26
121.75
121.19
118.90
118.55
77.36
77.04
76.72
54.73
52.40
39.64
39.25
35.42
34.62
0.00
zjh-3-6a-C13
170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 ppm

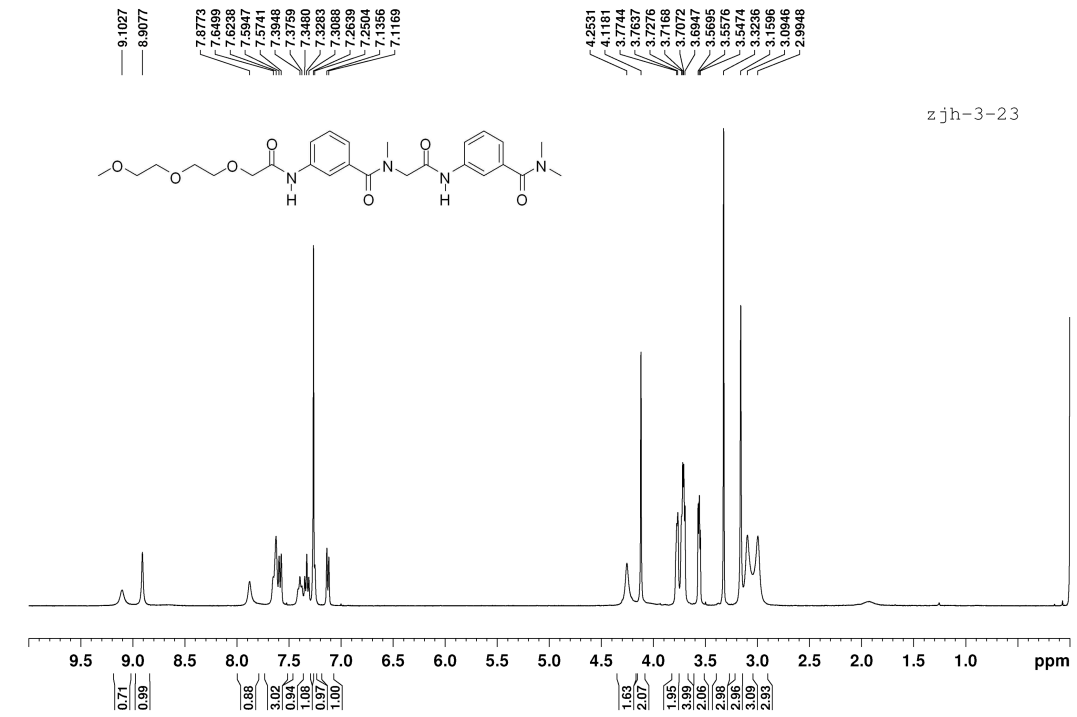
5a-2


1H
9.9365
8.8272
7.6745
7.6151
7.5952
7.5734
7.5554
7.3663
7.3471
7.3277
7.3092
7.0713
7.0528
4.1969
3.1117
3.0082
2.9430
2.4857
2.0372
zjh-3-T4-e
7.7 7.6 7.5 7.4 7.3 7.2 7.1 7.0 ppm
11 10 9 8 7 6 5 4 3 2 1 ppm
0.90
1.06
1.96
2.07
2.08
1.99
2.00
3.09
6.02
3.00

5a


13C
171.32
170.42
168.83
167.47
139.83
139.14
137.67
137.16
129.07
129.05
122.18
121.60
120.59
120.51
118.45
118.11
54.28
51.39
40.96
40.75
40.54
40.33
40.12
39.91
39.71
38.77
37.13
24.29
zjh-3-T4-e-C13-T=75
170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 ppm

5a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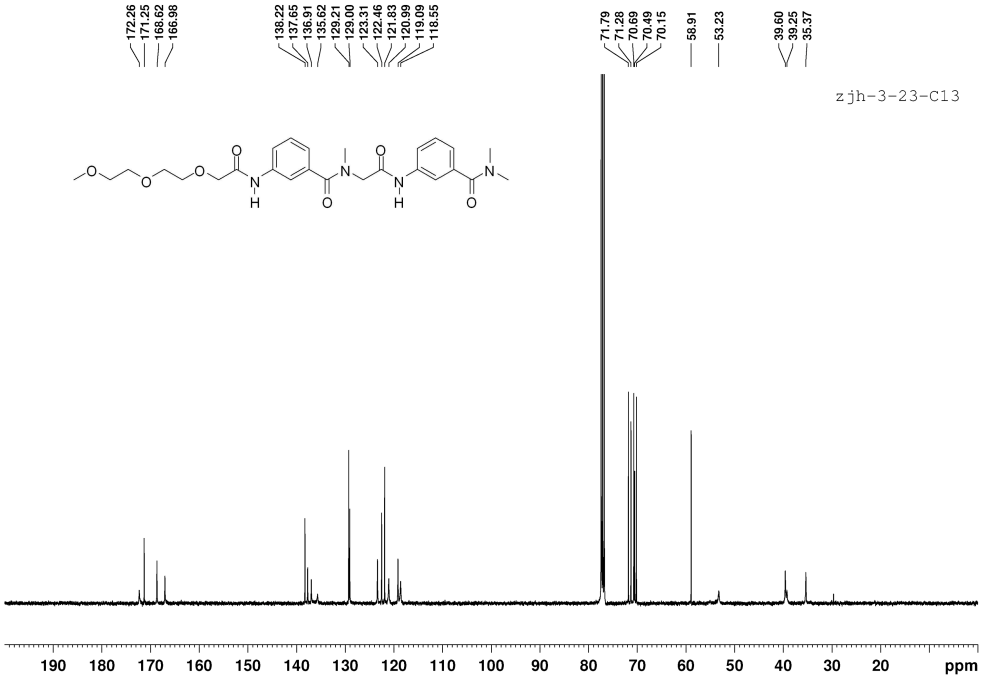
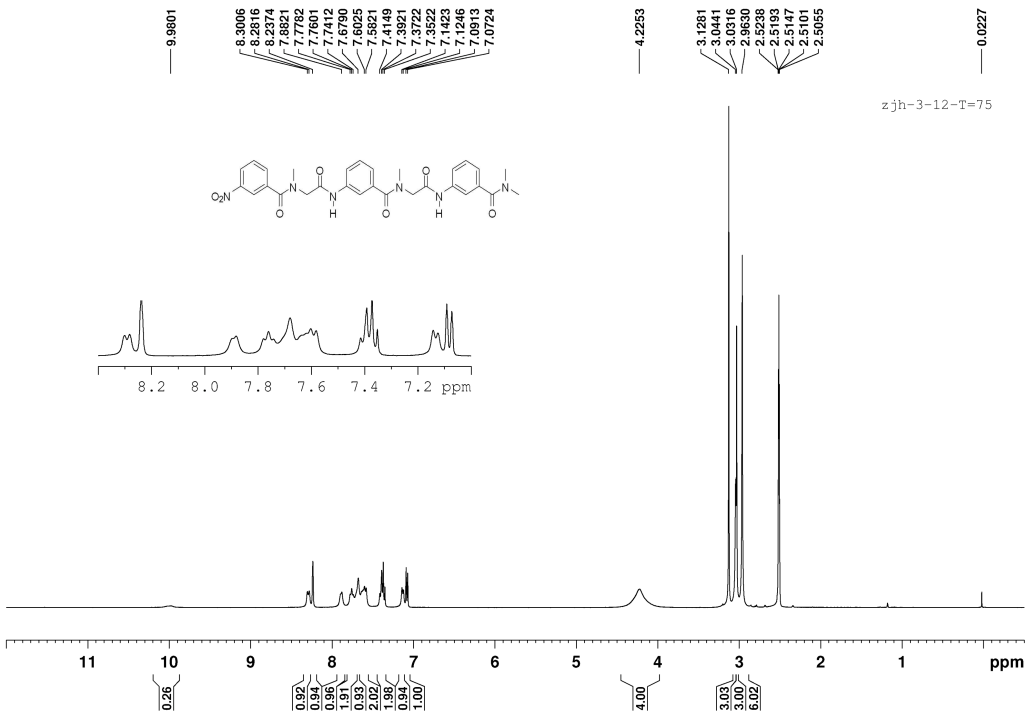
8.9077
7.8773
7.6499
7.6238
7.5947
7.5741
7.3948
7.3759
7.3480
7.3283
7.3088
7.2639
7.2504
7.1356
7.1169
4.2531
4.1181
3.7744
3.7637
3.7276
3.7168
3.7072
3.6947
3.5695
3.5576
3.5474
3.3236
3.1596
3.0946
2.9948
zjh-3-23
9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 ppm
0.71
0.99
0.88
3.02
0.94
1.08
0.97
1.00
1.63
2.07
1.95
3.99
2.06
2.98
2.96
3.09
2.93

5b

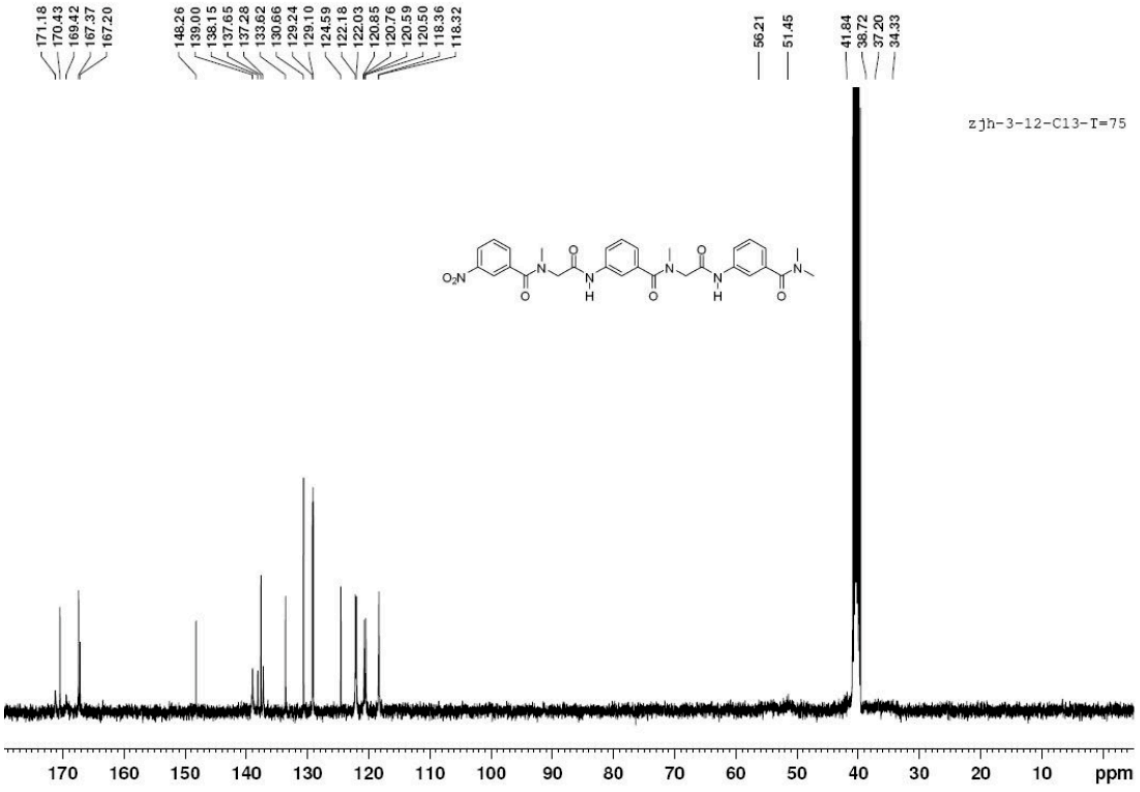

172.26
171.25
168.62
166.98
138.22
137.65
136.91
135.62
129.21
129.00
123.31
122.46
121.83
120.99
119.09
118.55
71.79
71.28
70.69
70.49
70.15
58.91
53.23
39.60
39.25
35.37
zjh-3-23-C13
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 ppm

5b

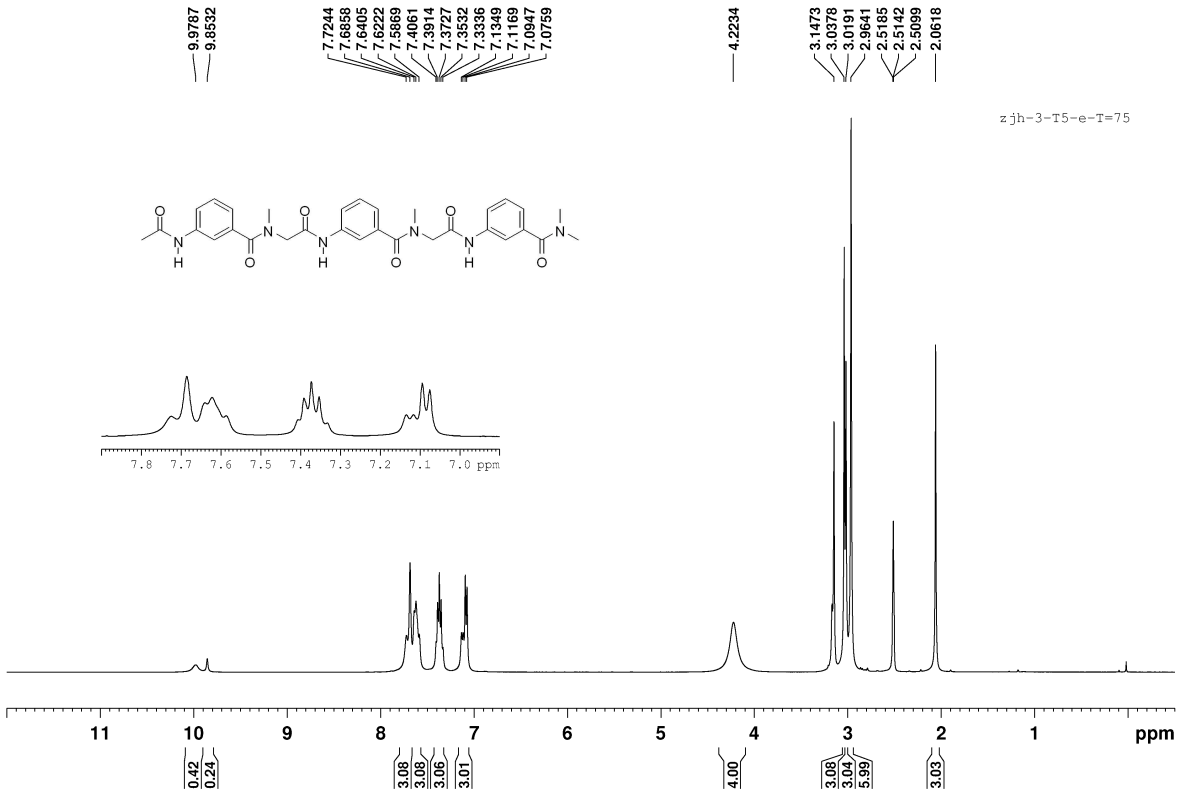

9.9801
8.3006
8.2816
8.2374
7.8821
7.7782
7.7601
7.7412
7.6790
7.6025
7.5821
7.4149
7.3921
7.3722
7.3522
7.1423
7.1246
7.0913
7.0724
4.2253
3.1261
3.0441
3.0316
2.9630
2.5238
2.5193
2.5147
2.5101
2.5055
0.0227
zjh-3-12-T=75
8.2 8.0 7.8 7.6 7.4 7.2 ppm
11 10 9 8 7 6 5 4 3 2 1 ppm
0.26
0.92
0.94
0.96
1.91
0.93
2.02
1.98
0.94
1.00
4.00
3.03
3.00
6.02

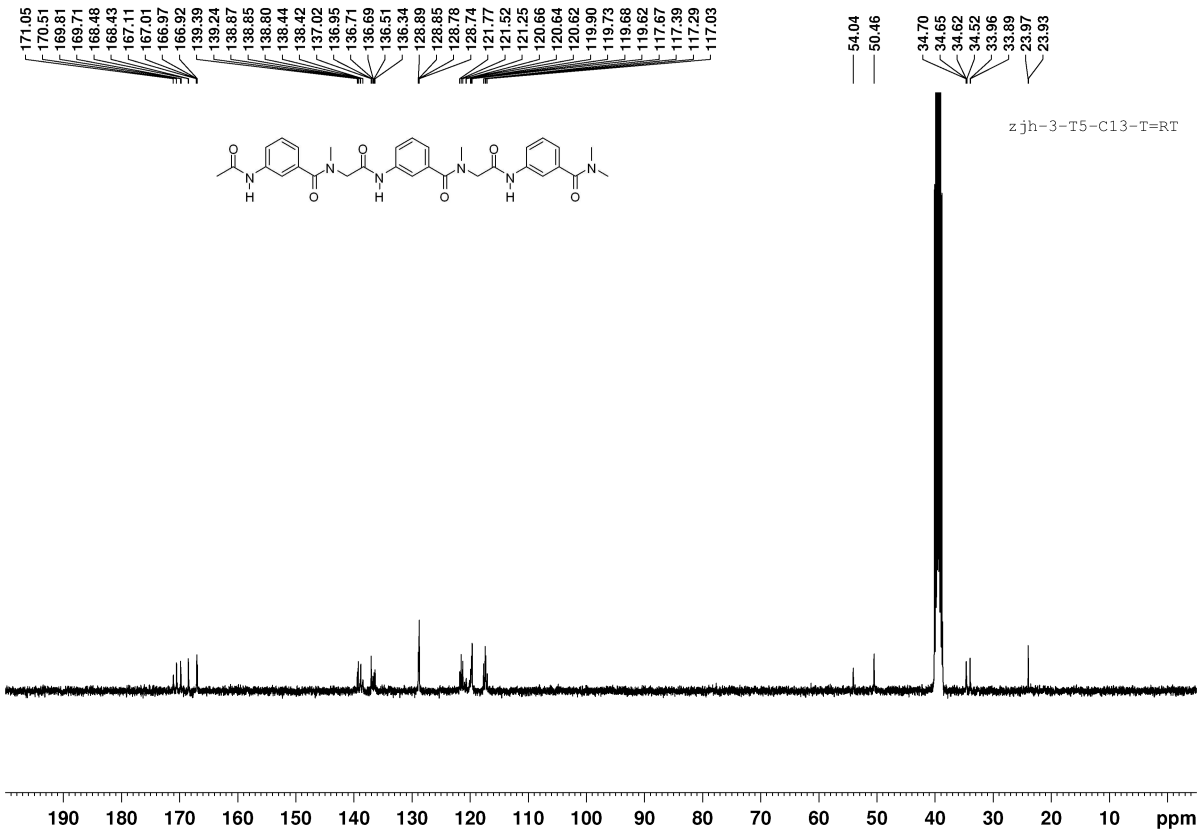
6a-1

6a-1

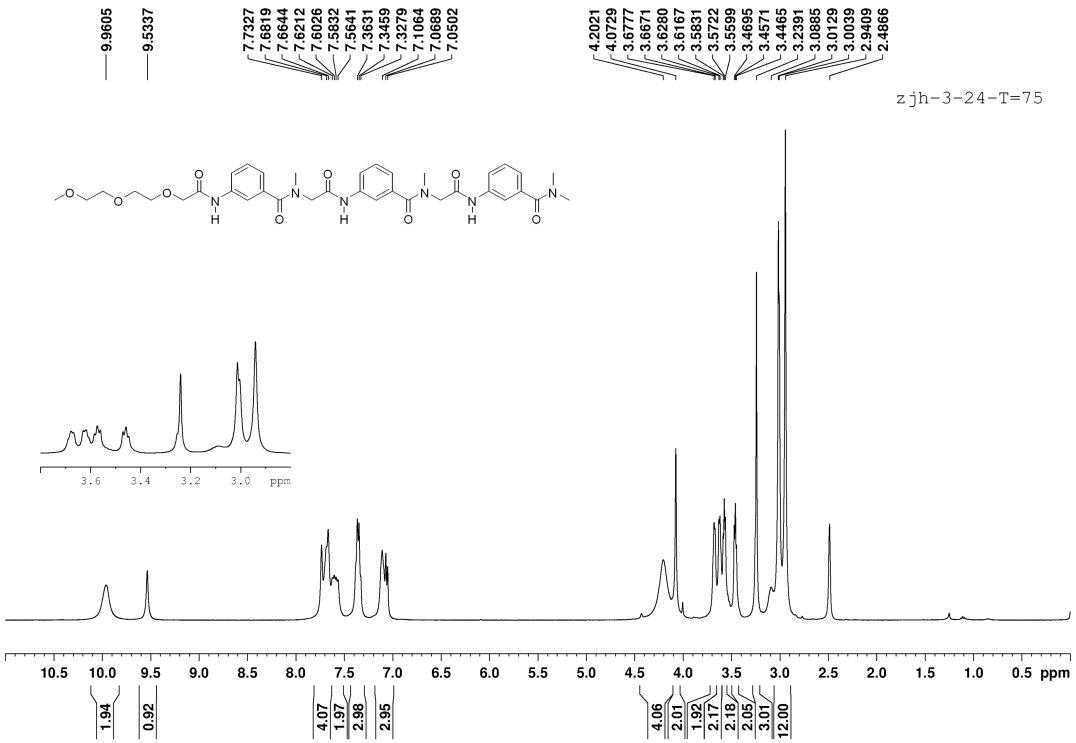

171.18
170.43
169.42
167.37
167.20
148.26
139.00
138.15
137.65
137.28
133.62
130.66
129.24
129.10
124.59
122.18
122.03
120.85
120.76
120.59
120.50
118.36
118.32
56.21
51.45
41.84
38.72
37.20
34.33
zjh-3-12-C13-T=75
170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 ppm

6a


9.9787
9.8532
7.7244
7.6858
7.6405
7.6222
7.5869
7.4061
7.3914
7.3727
7.3532
7.3336
7.1349
7.1169
7.0947
7.0759
4.2234
3.1473
3.0378
3.0191
2.9641
2.5185
2.5142
2.5099
2.0618
zjh-3-T5-e-T=75
7.8 7.7 7.6 7.5 7.4 7.3 7.2 7.1 7.0 ppm
11 10 9 8 7 6 5 4 3 2 1 ppm
0.42
0.24
3.08
3.08
3.06
3.01
4.00
3.08
3.04
5.99
3.03


171.05
170.51
169.81
169.71
168.48
168.43
167.11
167.01
166.97
166.92
139.39
139.24
138.87
138.85
138.80
138.44
138.42
137.02
136.95
136.71
136.69
136.51
136.34
128.89
128.85
128.78
128.74
121.77
121.52
121.25
120.66
120.64
120.62
119.90
119.73
119.68
119.62
117.67
117.39
117.29
117.03
54.04
50.46
34.70
34.65
34.62
34.52
33.96
33.89
23.97
23.93
zjh-3-T5-C13-T=RT
190
180
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10
ppm

6a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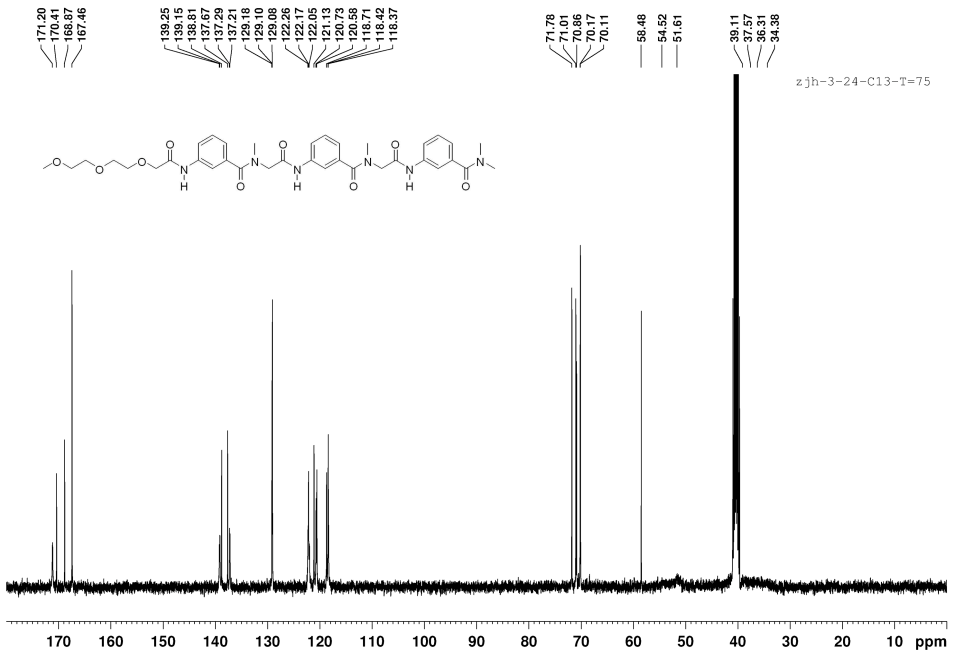
7.7327
7.6819
7.6644
7.6212
7.6026
7.5832
7.5641
7.3631
7.3459
7.3279
7.1064
7.0689
7.0502
4.2021
4.0729
3.6777
3.6671
3.6280
3.6167
3.5831
3.5722
3.5599
3.4695
3.4571
3.4465
3.2391
3.0885
3.0129
3.0039
2.9409
2.4866
zjh-3-24-T=75
3.6
3.4
3.2
3.0
ppm
10.5
10.0
9.5
9.0
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
1.5
1.0
0.5 ppm
1.94
0.92
4.07
1.97
2.98
2.95
4.06
2.01
1.92
2.17
2.18
2.05
3.01
12.00

6b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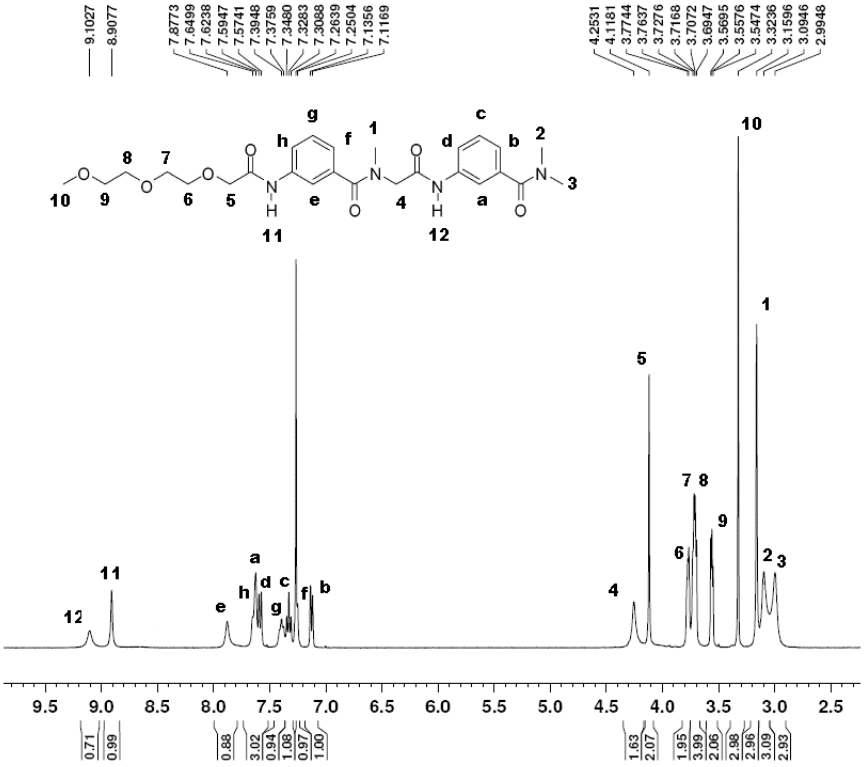
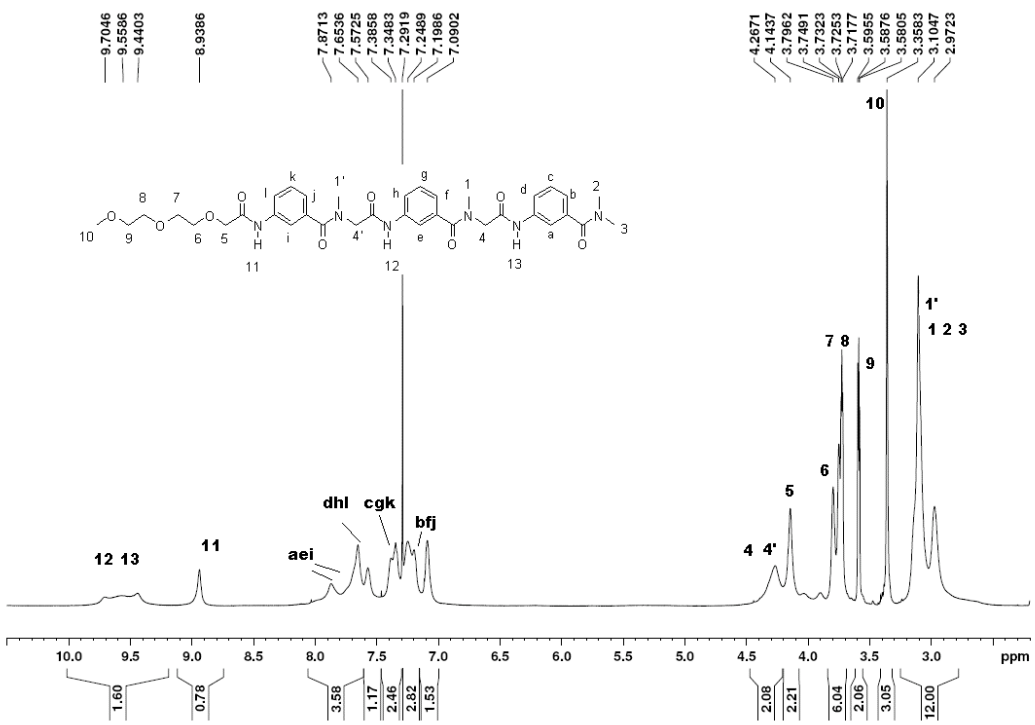
170.41
169.87
167.46
139.25
139.15
138.81
137.67
137.29
137.21
129.18
129.10
129.08
122.26
122.17
122.05
121.13
120.73
120.58
118.71
118.42
118.37
71.78
71.01
70.86
70.17
70.11
58.48
54.52
51.61
39.11
37.57
36.31
34.38
zjh-3-24-C13-T=75
170
160
150
140
130
120
110
100
90
80
70
60
50
40
30
20
10 ppm

6b

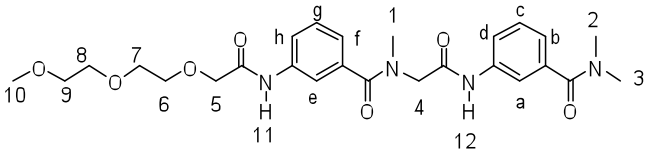
II-2. 1D and 2D ^{1}H NMR spectra of 5b and 6b

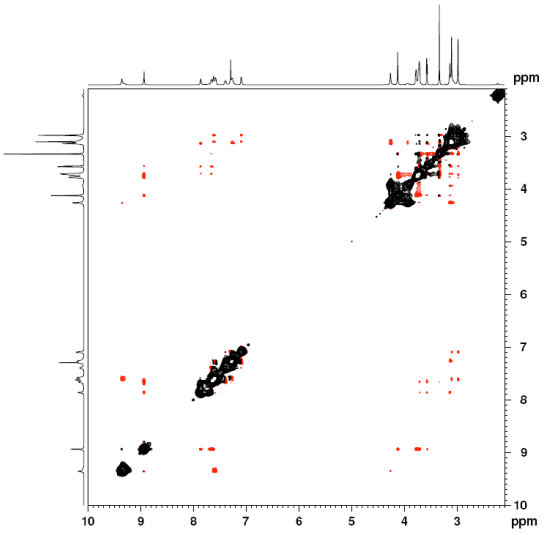
(Recorded on a Bruker Avance 600 MHz NMR Spectrometer)

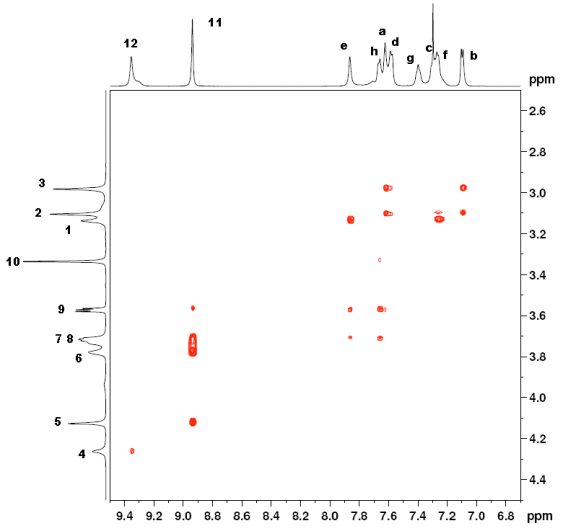


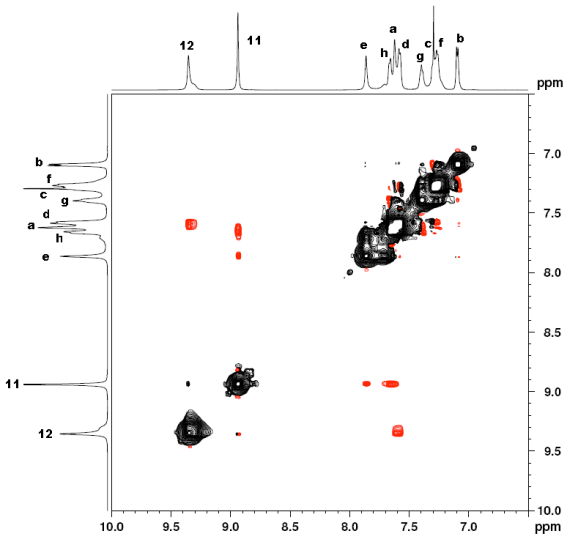


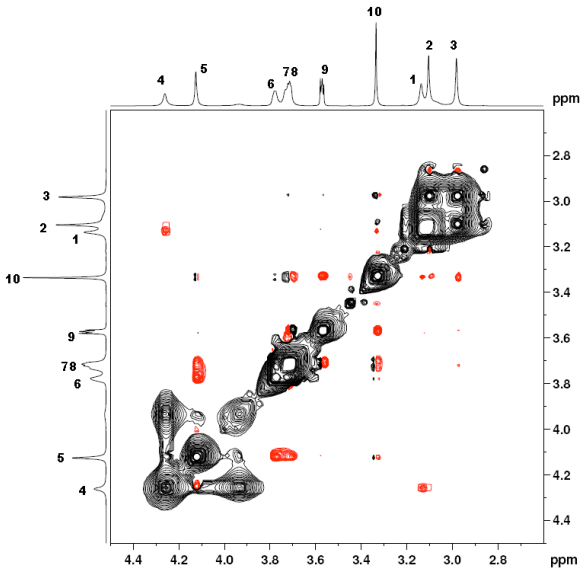
NOESY spectra of **5b** (~46 mM in $CDCl_3$, room temp., mixing time = 0.8s)



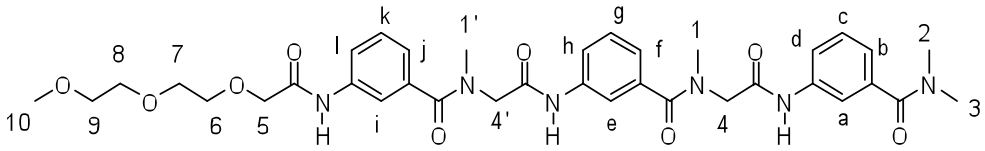


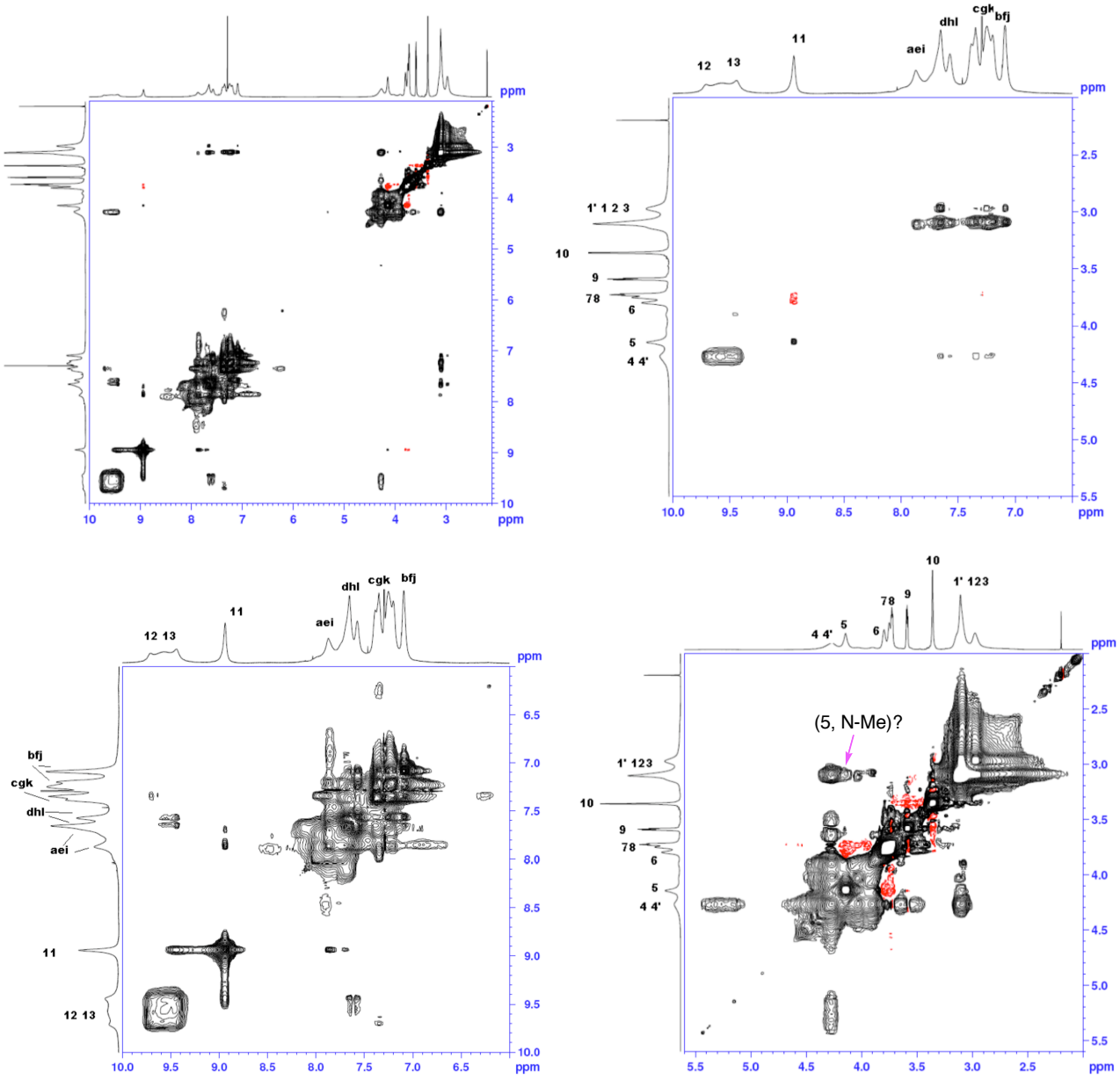






The NOESY spectra of **6b** (~15 mM in $CDCl_3$, room temp., mixing time = 0.8s）



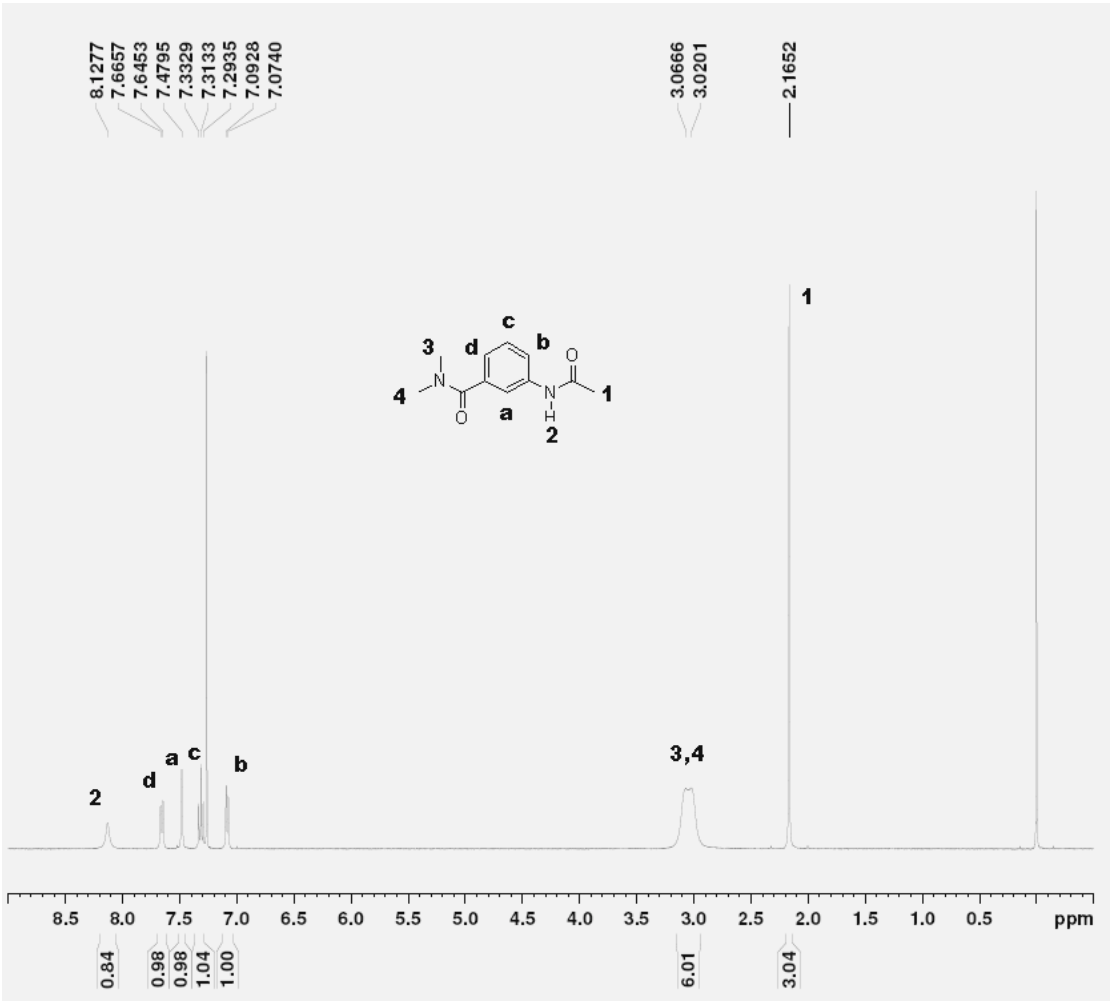


(The NOE between proton 5 and those of one of the N-methyls may be the result of an antiparallel of the molecules in solution.)

II-3. 1D ^{1}H NMR spectra recorded in $CDCl_3$ vs those recorded in DMSO-d_6

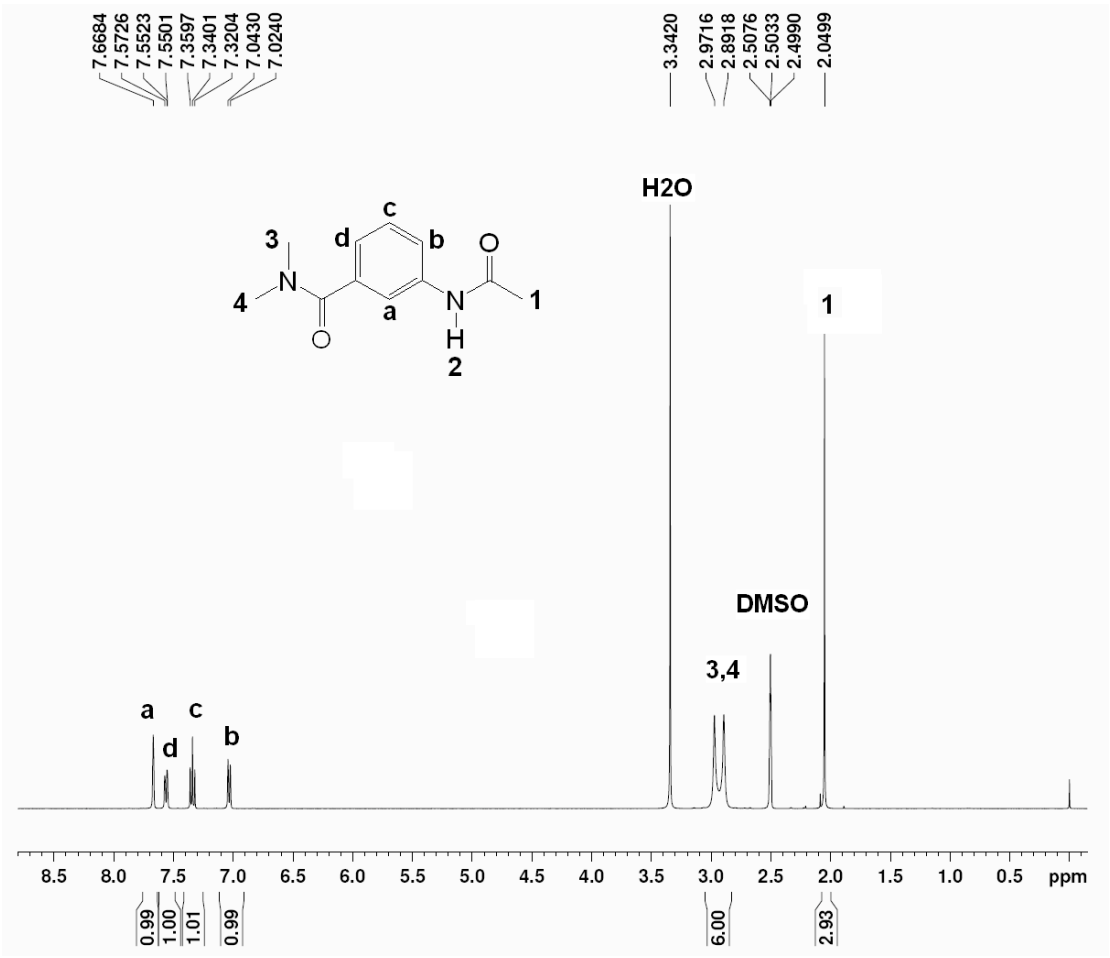
^{1}H-NMR(Bruker 400MHz) of Compound 4：

Temperature: 298K; Solvent: $CDCl_3$; Concentration: 17 mM



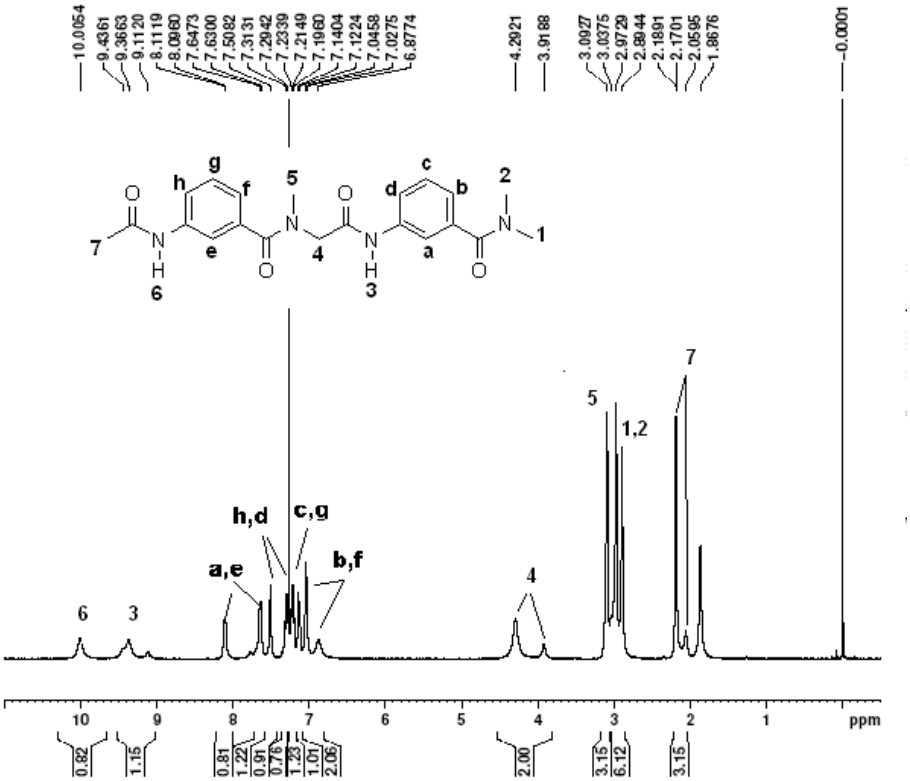
^{1}H-NMR(Bruker 400MHz) of Compound 4：

Temperature: 298K; Solvent: DMSO-d_6; Concentration: 109 mM



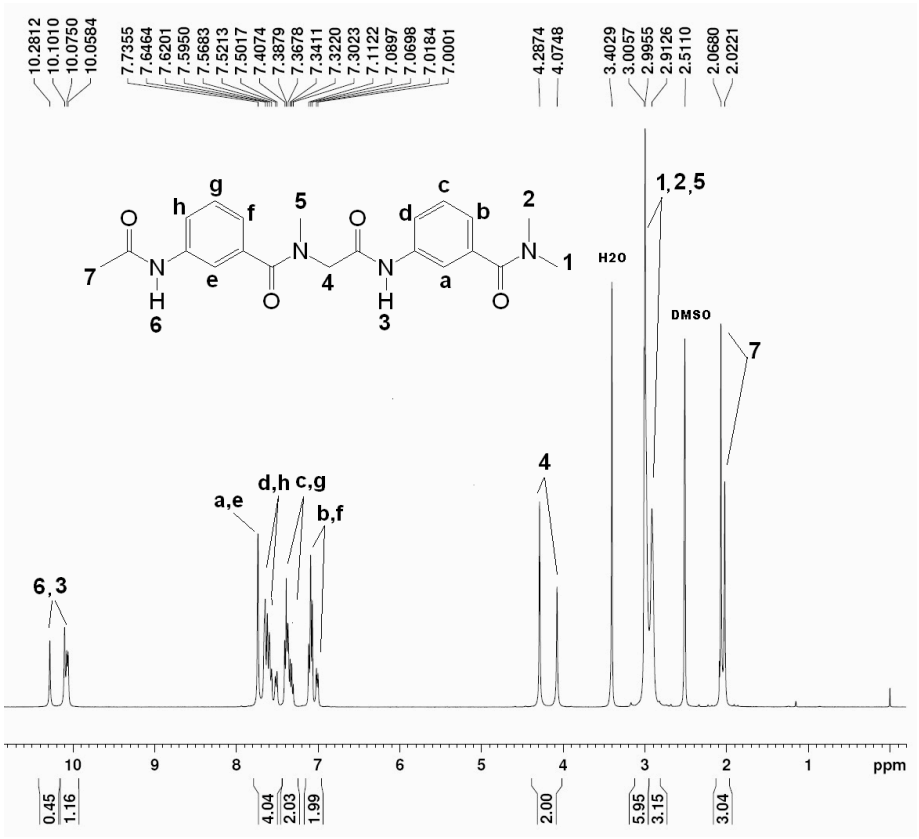
^{1}H-NMR(Bruker 400MHz) of Compound 5a:

Temperature: 298K; Solvent: $CDCl_3$; Concentration: 21mM



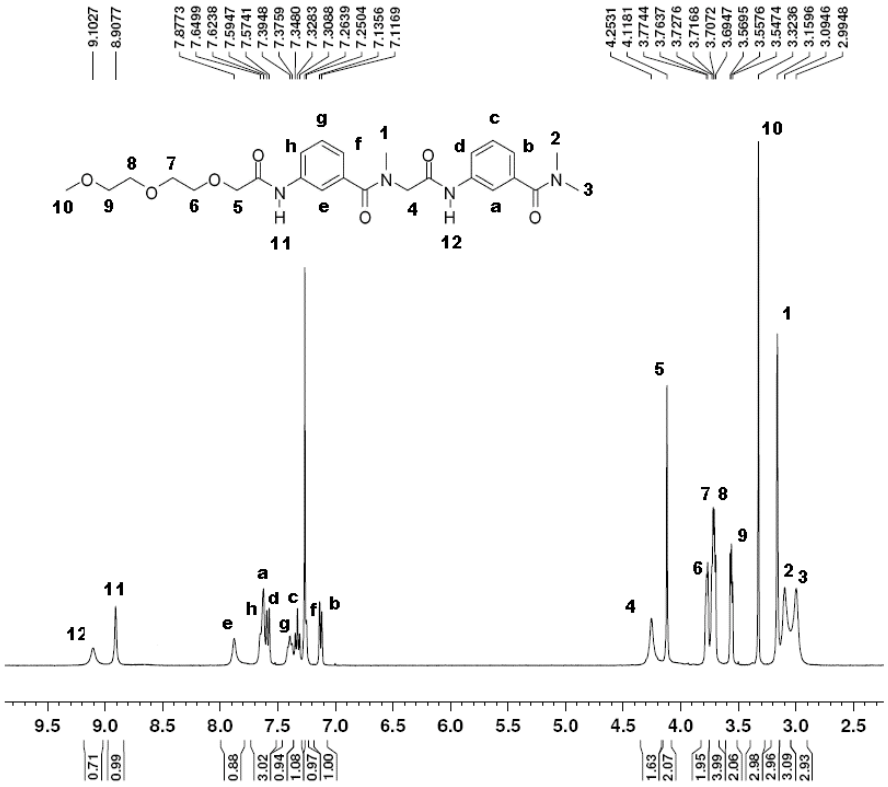
^{1}H-NMR(Bruker 400MHz) of Compound 5a:

Temperature: 298K; Solvent: DMSO-d_6; Concentration: 78 mM



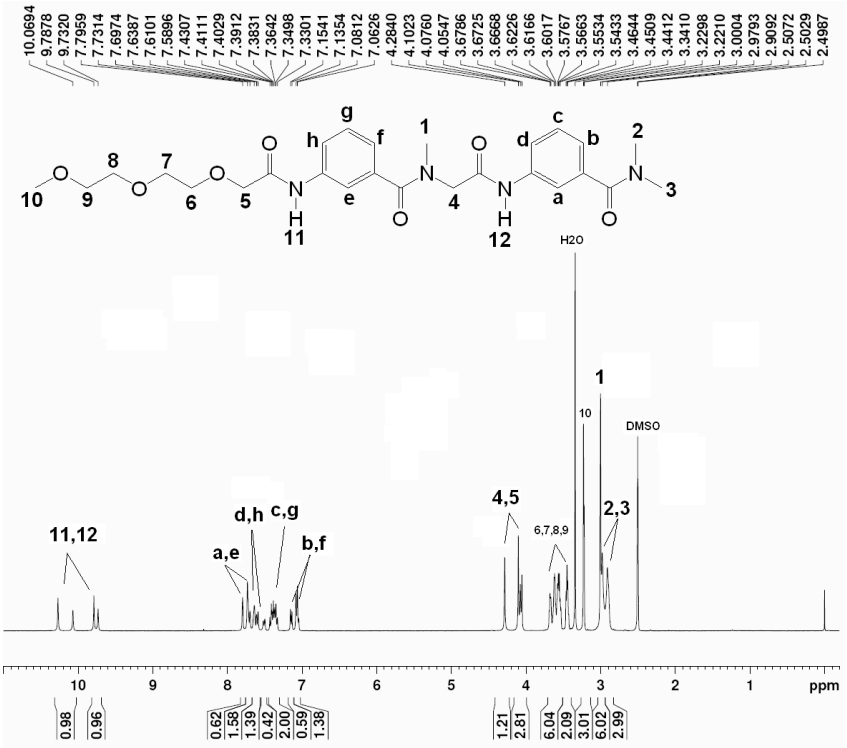
^{1}H-NMR(Bruker 400MHz) of Compound 5b:

Temperature: 298K; Solvent: $CDCl_3$; Concentration: 46 mM



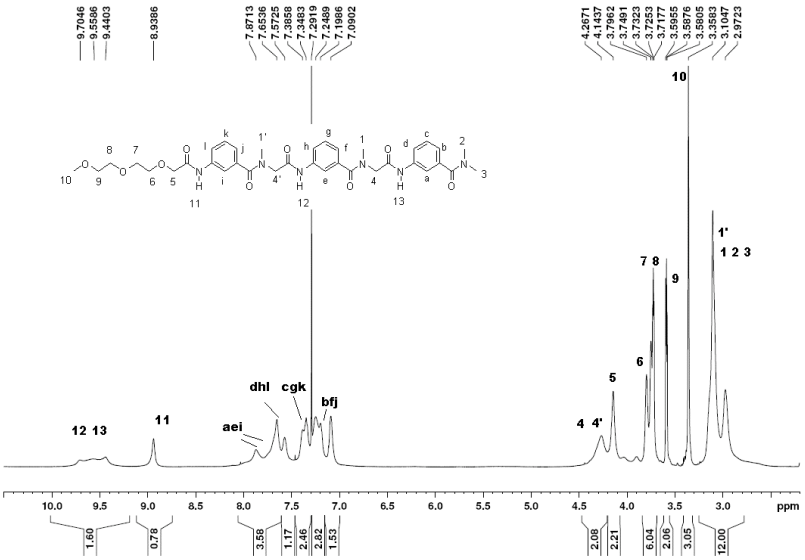
^{1}H-NMR(Bruker 400MHz) of Compound T4b:

Temperature: 298K; Solvent: DMSO-d_6; Concentration: 58 mM



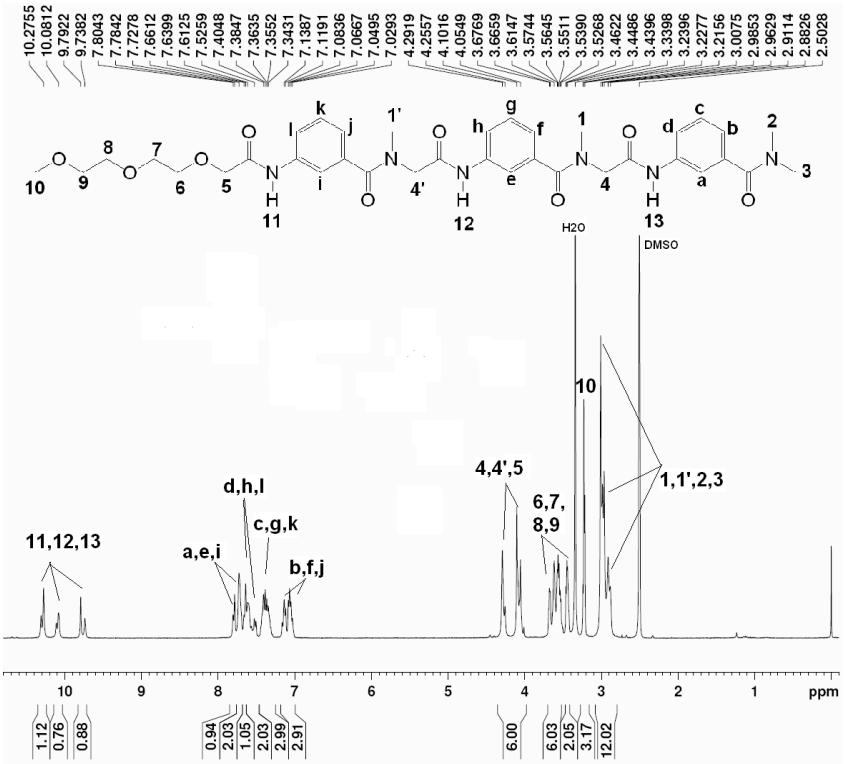
^{1}H-NMR(Bruker 400MHz) of Compound 6b:

Temperature: 298K; Solvent: $CDCl_3$; Concentration: 0.015mol • L^{-1}



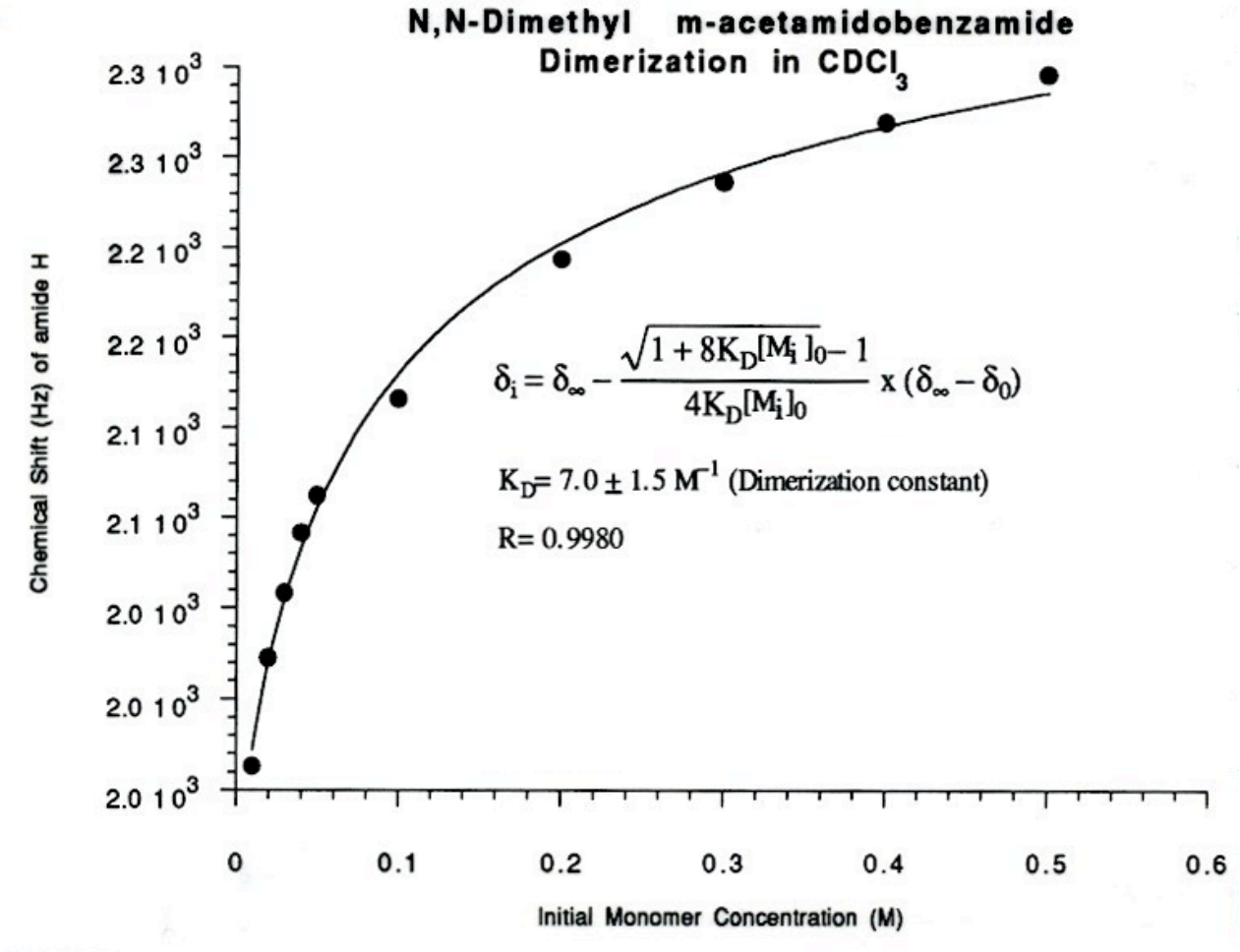
^{1}H-NMR(Bruker 400MHz) of Compound T5b:

Temperature: 298K; Solvent: DMSO-d_6; Concentration: 36 mM

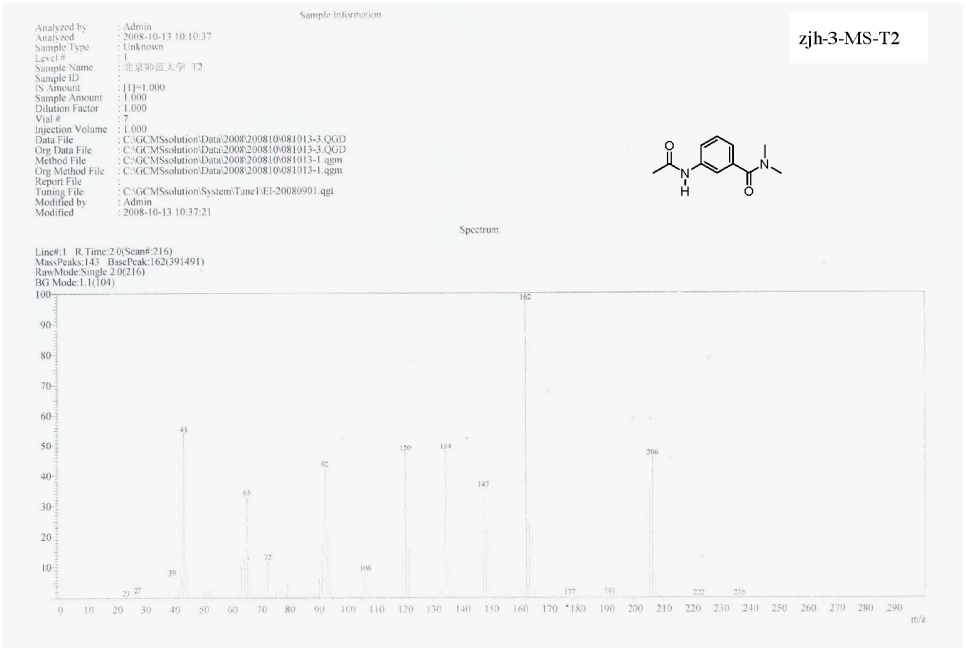


II-4. ^{1}H NMR dilution experiments on compound 4

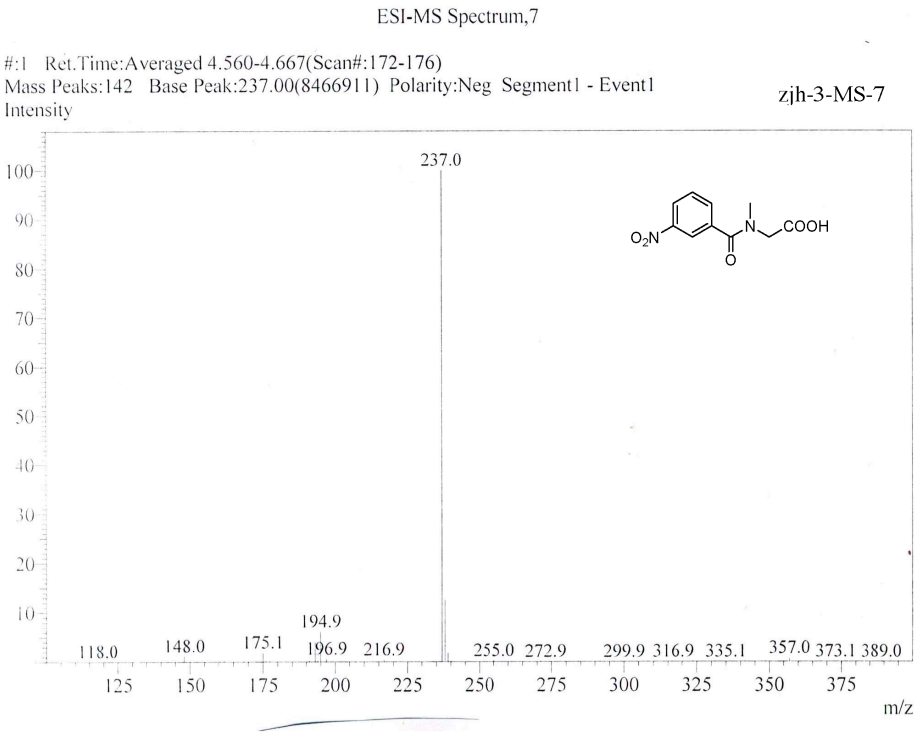
	Added monomer concentration (M)	Chem. shift (Hz) of amide H
1	0.0100000	1963.590
2	0.020000	2022.940
3	0.030000	2058.710
4	0.040000	2091.620
5	0.050000	2112.190
6	0.10000	2166.230
7	0.20000	2243.860
8	0.30000	2286.600
9	0.40000	2319.710
10	0.50000	2346.260



III. Mass (ESI) Spectra



4

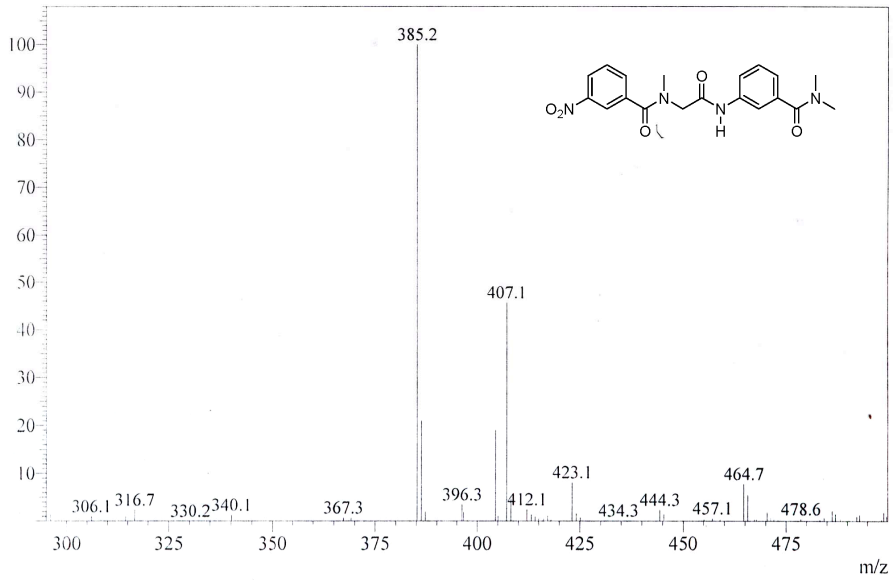


2b-1

ESI-MS Spectrum,9

#:1 Ret.Time:Averaged 3.413-3.493(Scan#:129-132)
Mass Peaks:209 Base Peak:385.15(5935823) Polarity:Pos Segment1 - Event1
Intensity

zjh-3-MS-9

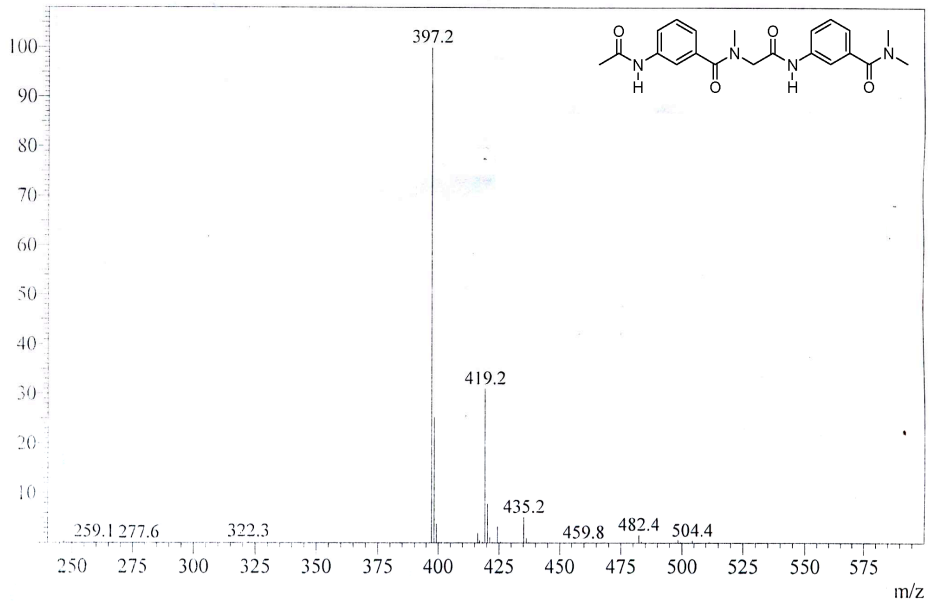


2b-2

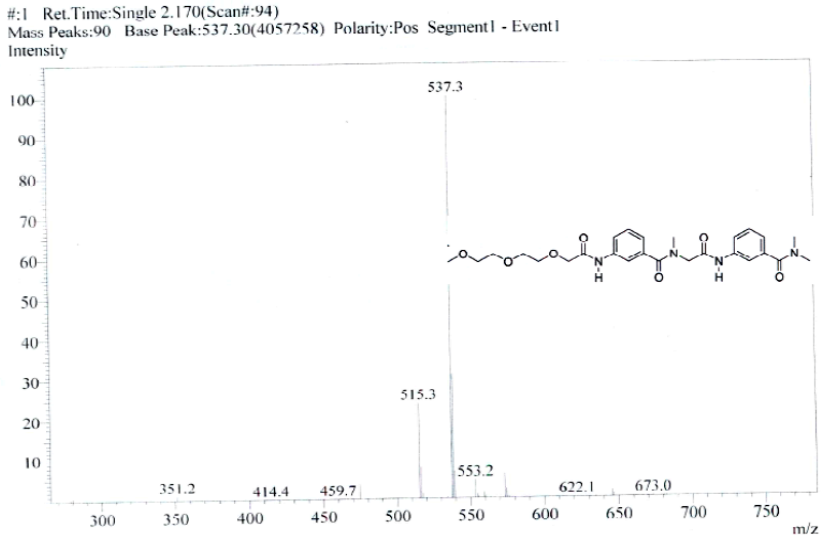
ESI-MS Spectrum,T4

#:1 Ret.Time:Averaged 1.280-1.307(Scan#:49-50)
Mass Peaks:90 Base Peak:397.20(7290817) Polarity:Pos Segment1 - Event1
Intensity

zjh-3-MS-T4



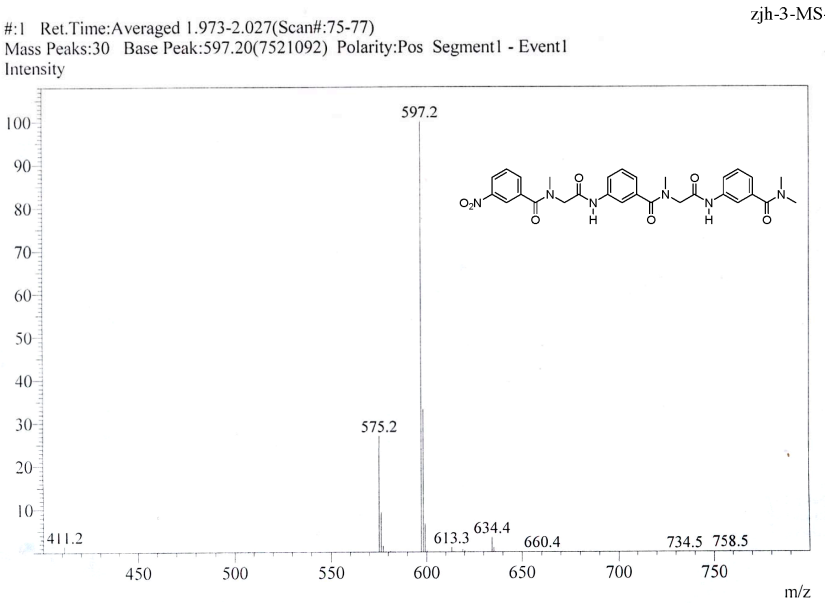
2b



5b

ESI-MS Spectrum, 12

zjh-3-MS-12

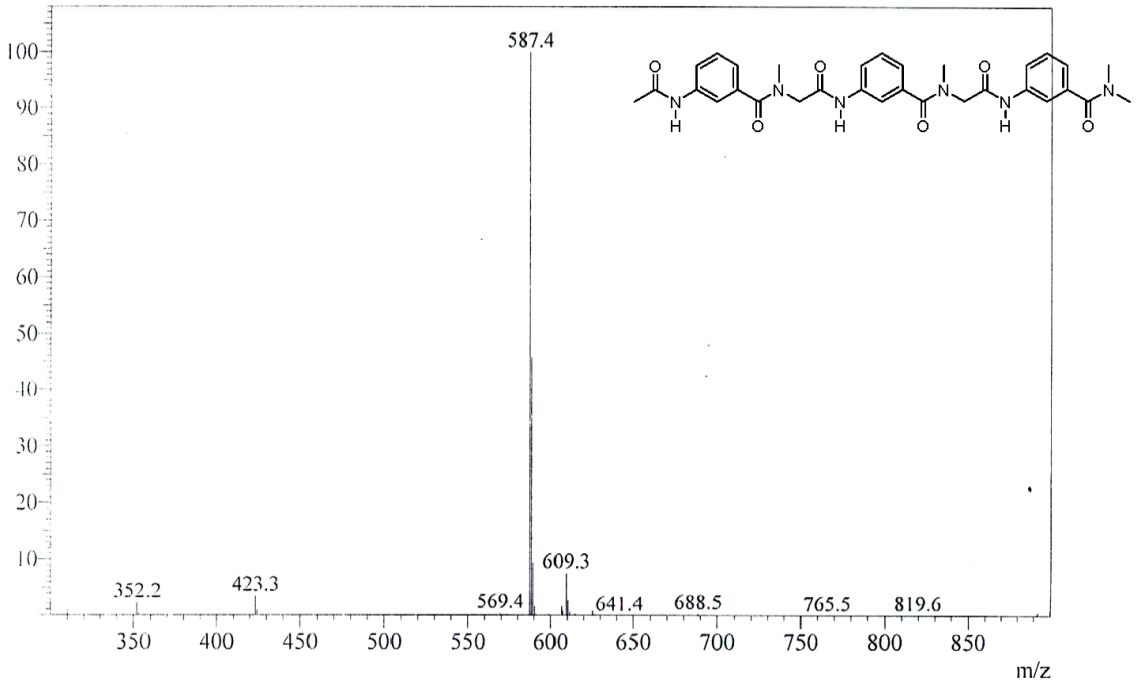


6b-1

zjh-3-MS-T5

ESI-MS Spectrum,T5

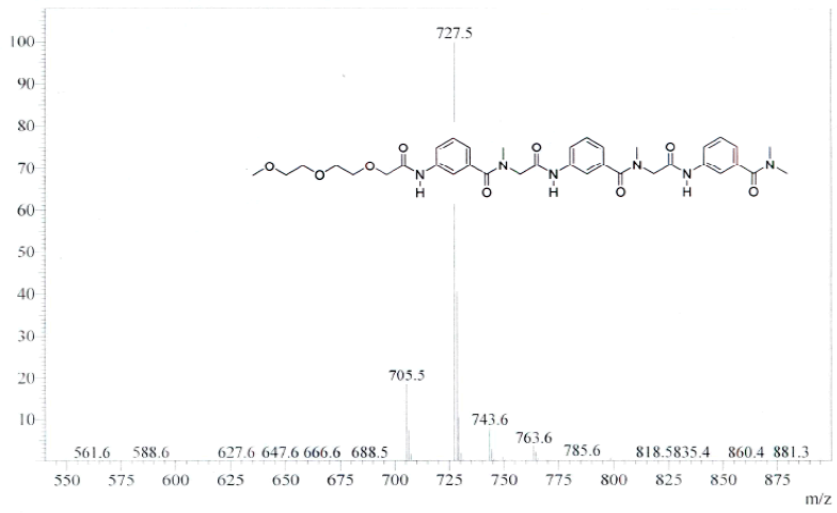
#:1 Ret.Time:Averaged 2.907-2.960(Scan#:110-112)
Mass Peaks:79 Base Peak:587.35(10744318) Polarity:Pos Segment1 - Event1
Intensity



6a

ESI-MS Spectrum, zjh-24

#:1 Ret.Time:Single 5.273(Scan#:227)
Mass Peaks:131 Base Peak:727.50(2767689) Polarity:Pos Segment1 - Event1
Intensity



6b

IV. Procedures of DLS Experiment

Instruments and Materials: The average sizes of the aggregates formed by **5b** and **6b** were measured on a Zetasizer Nano ZS, purchased from Malvern Instruments Co., Ltd (UK), which served as the laser particle size analyzer. Syringe Filter (Φ13mm, 0.45□m, organic solvent) was purchased from Tianjin Jinteng Experiment Equipment Co., Ltd. Chloroform, purchased from Beijing Chemical Reagents Company, was distilled before use.

Preparation of solution: A solution of **5b** (5.15 mg, 0.01 mmol) in ~9 mL of chloroform was added to a 10.0 mL volumetric flask at room temperature. After the solid had dissolved, additional chloroform was added to make the total volume of the solution to be 10.0 mL, and thus the concentration of **5b** to be 1 mM. The volumetric flask was then placed in ultrasonic cleaner for 2 minutes to ensure the homogeneity of the solution. A fraction of the solution (~1.2 mL) was injected into a quartz cell a the syringe filter which was attached to a glass syringe. Measurement was performed after the filtered solution was stabilized for 1 mintinue. The solution of **6b** in chloroform was similarly prepared.

Measurment: At room temperature, the following parameters set before measurements started: Chloroform: refractive index 1.4467, dielectric constant 4.81, viscosity 0.57 mPa·s. The quartz cell was placed into the Zetasizer Nano ZS to start the measurement.

V. Procedures of VPO Experiment

VPO experiment was performed on a WESCOR VAPRO 5520. Polystyrene (Mw 1500) was used as standard. All sample and standard solutions were determined minimum 5 times. Calibration curve of instrument reading values versus concentrations were obtained by least square fit of 7 data points (2-160 mM) of standard solution. Aggregation numbers are obtained by converting experimental data into effective concentration based on the calibration curve.