Supplementary Material for *Organic & Biomolecular Chemistry*

Four-component assembly in the crystalline state driven by amidinium-carboxylate salt bridge formation from an aqueous solution

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Outline

1. Supplementary figures and tables

1.1 ^{1}H NMR spectra of complexaion with 1 and terephthalic acid disodium salt 2a.

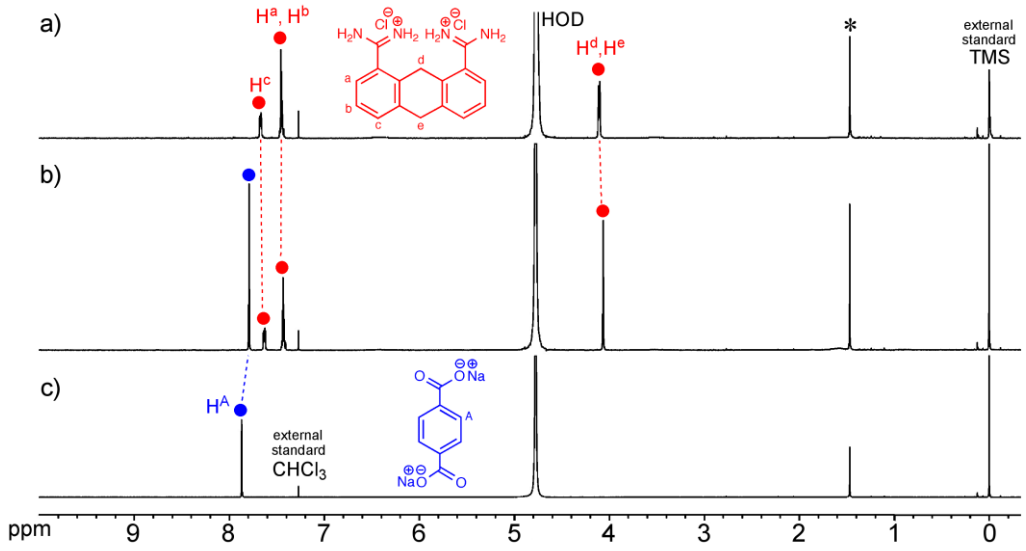


Fig. S1. ^{1}H NMR spectra (1.89 mM in D_2O, 298 K) of a) diamidine dihydrochloride **1a**, b) 1:1 mixture of **1a** and **2a**, and c) terephthalic acid disodium salt **2a**. Asterisks indicate signals from the external standard (TMS/$CDCl_3$ in glass capillary).

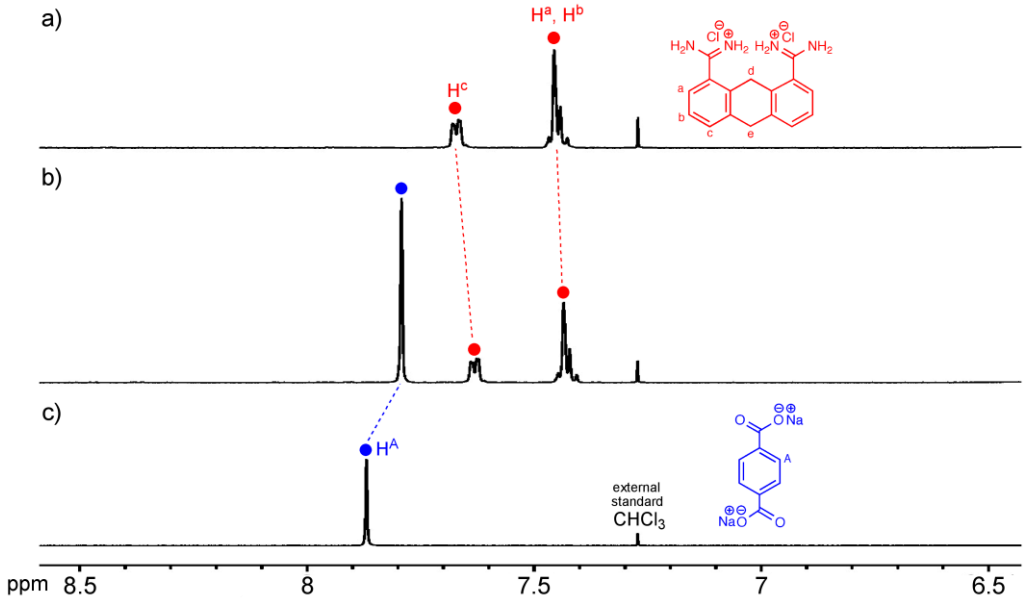


Fig. S1 (expanded). ^{1}H NMR spectra (1.89 mM in D_2O, 298 K) of a) diamidine dihydrochloride **1a**, b) 1:1 mixture of **1a** and **2a**, and c) terephthalic acid disodium salt **2a**.

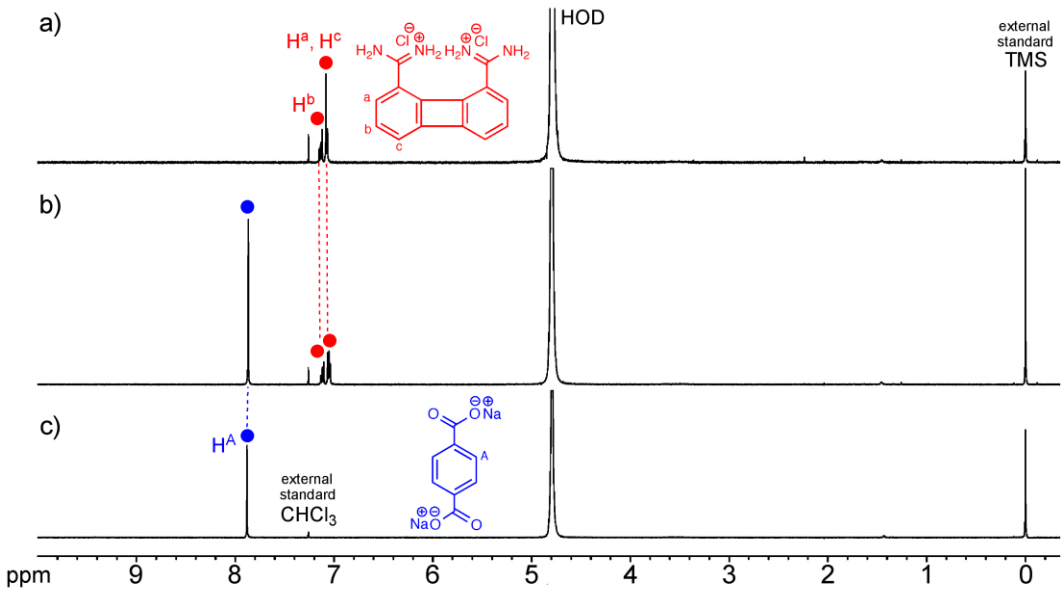


Fig. S2. ^{1}H NMR spectra (1.89 mM in D_2O, 298 K) of a) diamidine dihydrochloride **1b**, b) 1:1 mixture of **1b** and **2a**, and c) terephthalic acid disodium salt **2a**.

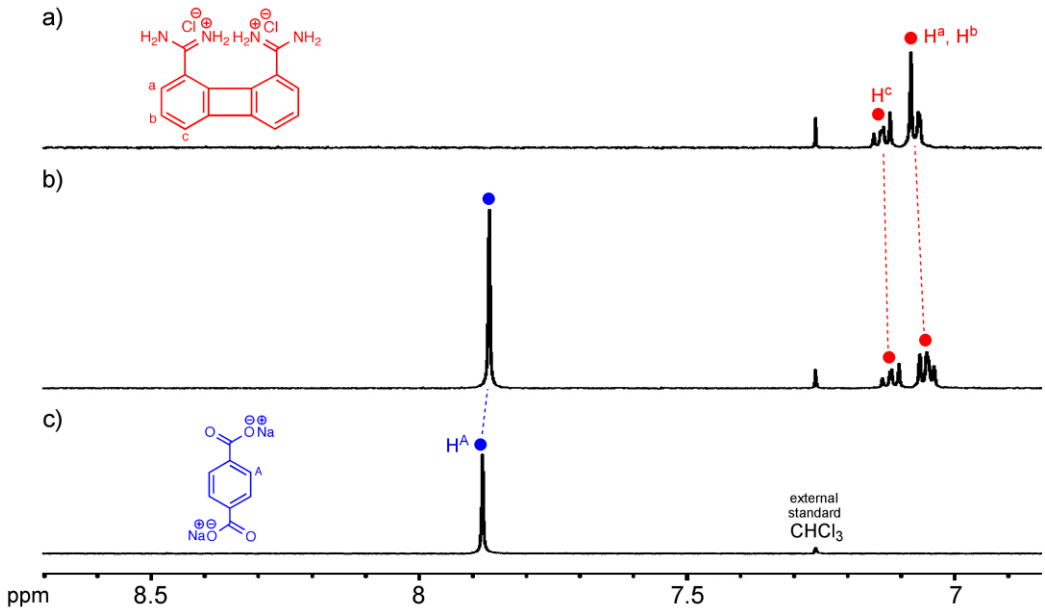


Fig. S2 (expanded). ^{1}H NMR spectra (1.89 mM in D_2O, 298 K) of a) diamidine dihydrochloride **1b**, b) 1:1 mixture of **1b** and **2a**, and c) terephthalic acid disodium salt **2a**.

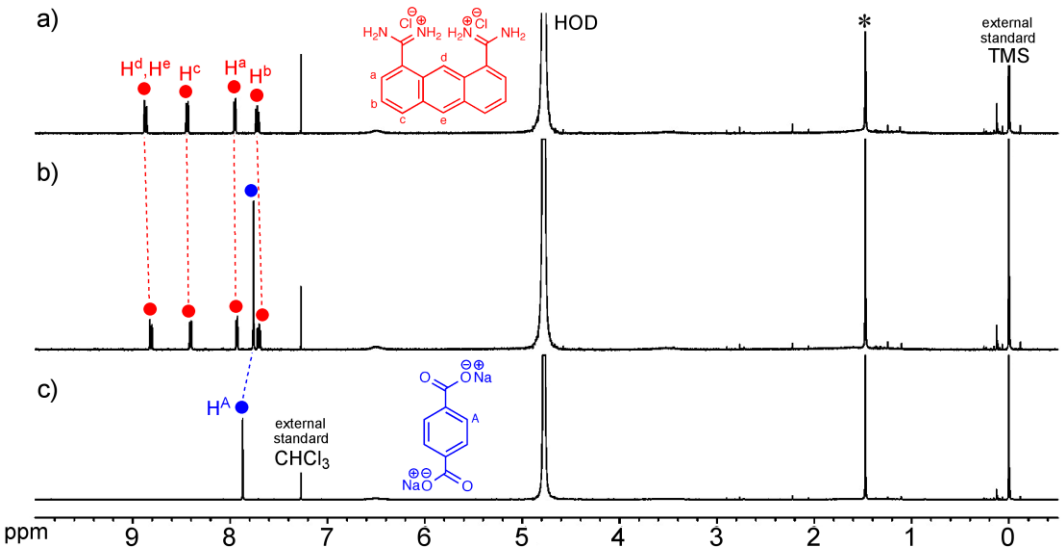


Fig. S3. ^{1}H NMR spectra (0.63 mM in D_2O, 298 K) of a) diamidine dihydrochloride **1c**, b) 1:1 mixture of **1c** and **2a**, and c) terephthalic acid disodium salt **2a**. Asterisks indicate signals from the external standard (TMS/$CDCl_3$ in glass capillary).

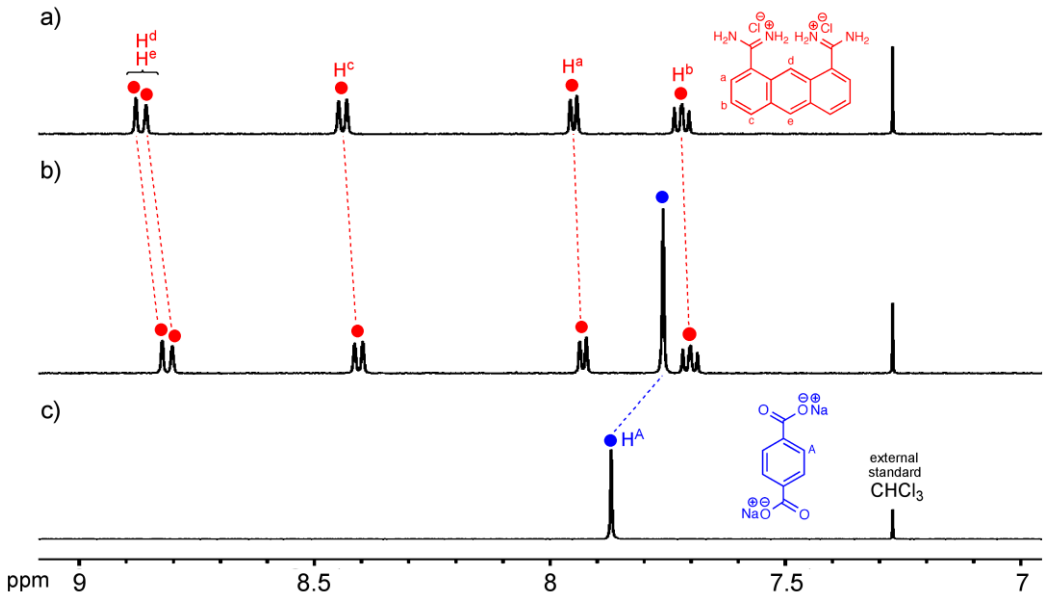


Fig. S3 (expanded). ^{1}H NMR spectra (0.63 mM in D_2O, 298 K) of a) diamidine dihydrochloride **1c**, b) 1:1 mixture of **1c** and **2a**, and c) terephthalic acid disodium salt **2a**.

2. DOSY spectra

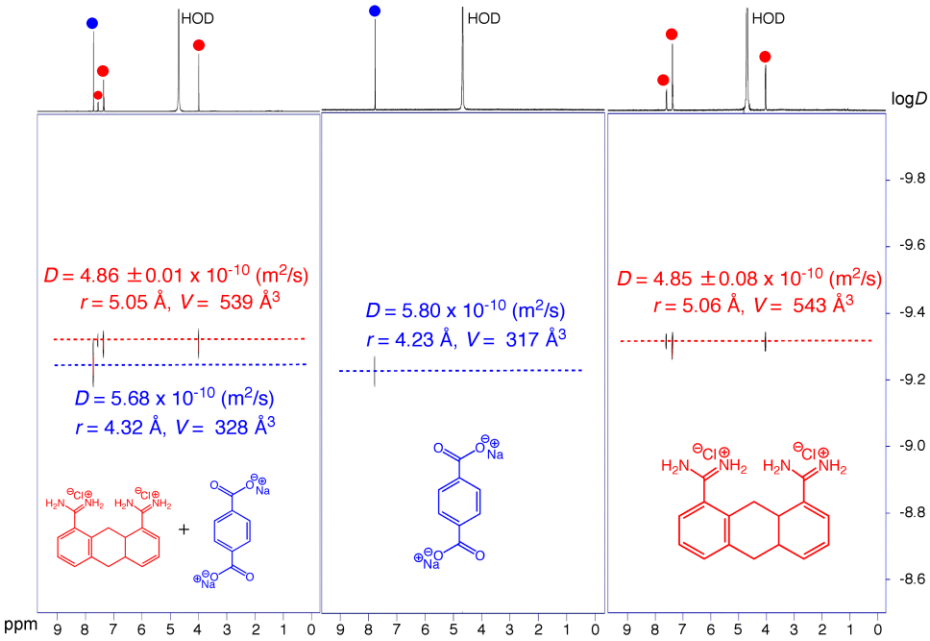


Fig. S4. ^{1}H DOSY spectra (1.89 mM in D_2O, 25 °C, diffusion time Δ = 200 ms) of a) complex with **1a** and **2a**, b) terephthalic acid disodium salt **2a**, c) diamidine **1a**.

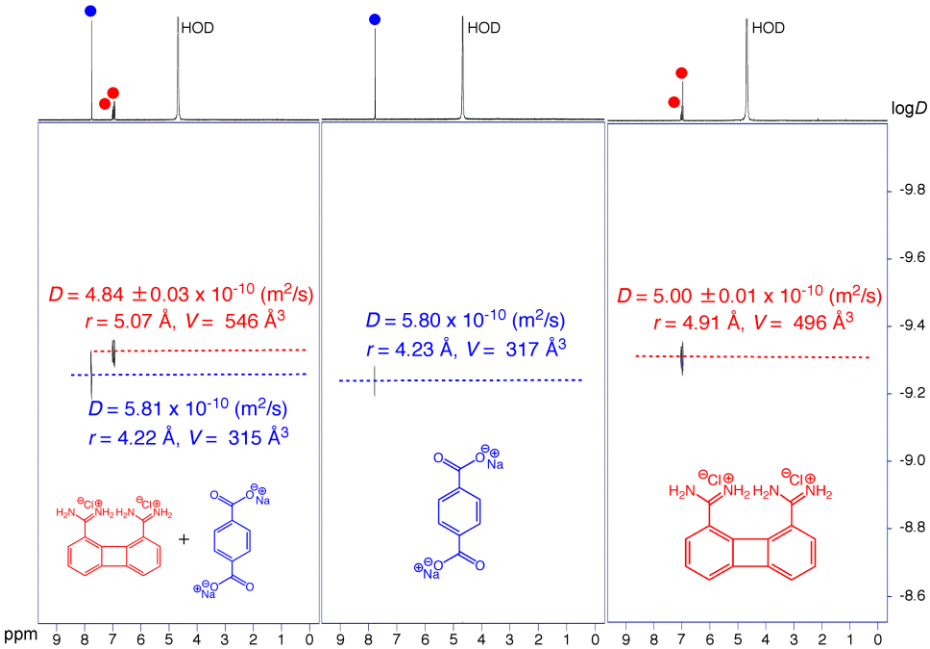


Fig. S5. ^{1}H DOSY spectra (1.89 mM in D_2O, 25 °C, diffusion time Δ = 200 ms) of a) complex with **1b** and **2a**, b) terephthalic acid disodium salt **2a**, c) diamidine **1b**.

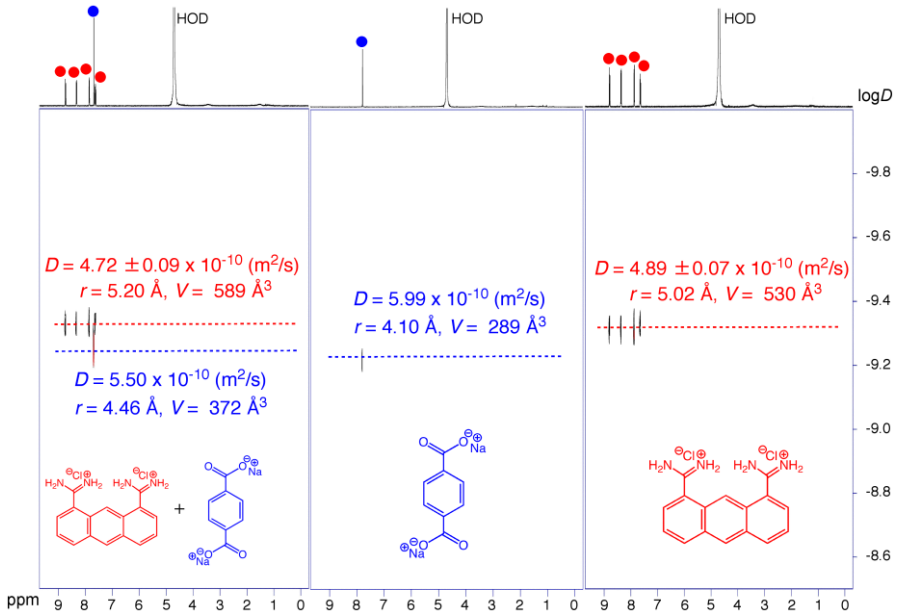


Fig. S6. ^{1}H DOSY spectra (0.63 mM in D_2O, 25 °C, diffusion time Δ = 200 ms) of a) complex with **1c** and **2a**, b) terephthalic acid disodium salt **2a**, c) diamidine **1c**.

Table S1. Comparison of diffusion coefficients, molecular radii and volumes[a]

sample	conc. (mM)	D (m^2 s^{-1})	r_H (Å)	V (Å^3)
1a	1.89	$4.85 \pm 0.08 \times 10^{-10}$	5.06	543
1b	1.89	$5.00 \pm 0.01 \times 10^{-10}$	4.91	496
1c	0.63	$4.89 \pm 0.07 \times 10^{-10}$	5.02	530
2a	1.89	5.80×10^{-10}	4.23	317
2a	0.63	5.99×10^{-10}	4.10	289
1a + **2a**	1.89	$4.86 \pm 0.01 \times 10^{-10}$ (A)[b] 5.68×10^{-10} (C)[c]	5.05 4.32	539 328
1b + **2a**	1.89	$4.84 \pm 0.03 \times 10^{-10}$ (A)[b] 5.81×10^{-10} (C)[c]	5.07 4.22	546 315
1c + **2a**	0.63	$4.72 \pm 0.09 \times 10^{-10}$ (A)[b] 5.50×10^{-10} (C)[c]	5.20 4.46	589 372

[a] Diffusion coefficients were recorded at 25 °C (diffusion time Δ = 200 ms), and molecular radii were calculated using the Stokes-Einstein equation.

[b] Derived from diamidine dihydrochloride **1**.

[c] Derived from terephthalic acid disodium salt **2a**.

3. Crystallographic data

Table S2. Crystallographic data for **1**

Compound	**1a**	**1b'**	**1c**
Chemical formula	$C_{16}H_{18}Cl_2N_4$•H_2O	$C_{14}H_{14}N_4Br_2$	$C_{16}H_{16}Cl_2N_4$•$2H_2O$
Temperature /K	213	213	213
Crystal system	monoclinic	monoclinic	monoclinic
Space group	*P* 2_1/a	*C* 2/c	*P* 2_1/n
a /Å	10.8452(19)	9.3274(13)	22.853(5)
b /Å	20.440(3)	21.826(3)	7.5764(14)
c /Å	8.5123(18)	7.1188(9)	10.434(2)
β /°	106.969(15)	94.249(11)	96.5625(17)
V /Å^3	1804.8(6)	1445.3(3)	1794.6(6)
Z	4	4	4
Radiation type	CuK_α	CuK_α	CuK_α
$D_{calcd.}$ /Mgm^{-3}	1.307	1.829	1.374
Collected reflections	4345	1789	4309
Unique reflections	3177	1297	3183
R_{int}	0.069	0.037	0.039
R_1 ($I > 2\sigma(I)$)	0.0509	0.0367	0.0423
R_1 (all data)	0.1092	0.0389	0.0481
R_w (all data)	0.1532	0.0997	0.1547
Goodness-of fit	1.011	1.108	1.025

Table S3. Crystallographic data for **1•2**.

Compound	**1a•2a**	**1a•2b**	**1b•2a**	**1c•2a**
Chemical formula	$C_{24}H_{22}N_4O_4$•$4H_2O$	$C_{24}H_{18}F_4N_4O_4$•$3H_2O$	$C_{22}H_{18}N_4O_4$•$5H_2O$	$C_{24}H_{20}N_4O_4$•$4H_2O$
Temperature /K	213	213	213	213
Crystal system	triclinic	triclinic	triclinic	triclinic
Space group	*P*-1	*P*-1	*P*-1	*P*-1
a /Å	11.677(3)	11.4606(7)	11.197(5)	11.6126(14)
b /Å	11.8984(14)	12.5728(12)	11.525(6)	12.017(2)
c /Å	9.9909(12)	10.4779(8)	10.753(5)	10.144(2)
α /°	103.393(9)	108.785(6)	116.02(3)	108.959(14)
β /°	108.395(12)	102.511(6)	90.97(3)	94.679(15)
γ /°	89.725(13)	63.700(6)	73.52(4)	114.816(11)
V /Å^3	1277.9(3)	1276.52(17)	1186.3(9)	1175.4(3)
Z	2	2	2	2
Radiation type	CuK_α	CuK_α	CuK_α	CuK_α
$D_{calcd.}$ /Mgm^{-3}	1.306	1.448	1.379	1.414
Collected reflections	6152	6221	5774	5778
Unique reflections	4465	4537	4217	4211
R_{int}	0.043	0.046	0.045	0.041
R_1 ($I > 2\sigma(I)$)	0.0631	0.0527	0.0913	0.0758
R_1 (all data)	0.0979	0.0602	0.1859	0.1472
R_w (all data)	0.2094	0.1536	0.3154	0.2518
Goodness-of fit	1.065	1.045	1.046	1.073

Table S4. Crystallographic data for **1•2**.

Compound	**1c•2b**	**1c•3a**	**1c•3b**	**1c•3c**
Chemical formula	$C_{24}H_{16}F_4N_2O_4$•$2H_2O$	$C_{22}H_{24}N_4O_4$•$2H_2O$	$C_{24}H_{28}N_4O_4$•$2H_2O$	$C_{26}H_{32}N_4O_4$
Temperature /K	213	213	213	213
Crystal system	triclinic	triclinic	triclinic	triclinic
Space group	*P*-1	*P*-1	*P*-1	*P*-1
a /Å	11.480(5)	11.5243(13)	12.054(3)	9.7411(15)
b /Å	11.934(5)	11.7284(16)	12.563(4)	16.079(3)
c /Å	10.976(4)	10.0810(14)	9.714(3)	7.9526(19)
α /°	109.92(3)	108.915(11)	105.63(2)	98.725(16)
β /°	102.60(3)	105.206(10)	109.560(20)	103.439(15)
γ /°	111.76(3)	105.742(10)	102.83(2)	79.976(12)
V /Å^3	1206.6(8)	1145.2(3)	1253.3(6)	1184.9(4)
Z	2	2	2	2
Radiation type	CuK_α	CuK_α	CuK_α	CuK_α
$D_{calcd.}$ /Mgm^{-3}	1.476	1.289	1.252	1.302
Collected reflections	5843	5563	6054	5824
Unique reflections	4266	4057	4398	4209
R_{int}	0.070	0.015	0.128	0.048
R_1 ($I > 2\sigma(I)$)	0.0566	0.0423	0.0771	0.0439
R_1 (all data)	0.0973	0.0590	0.0891	0.1805
R_w (all data)	0.1920	0.1569	0.2256	0.1458
Goodness-of fit	1.025	1.186	1.071	0.929

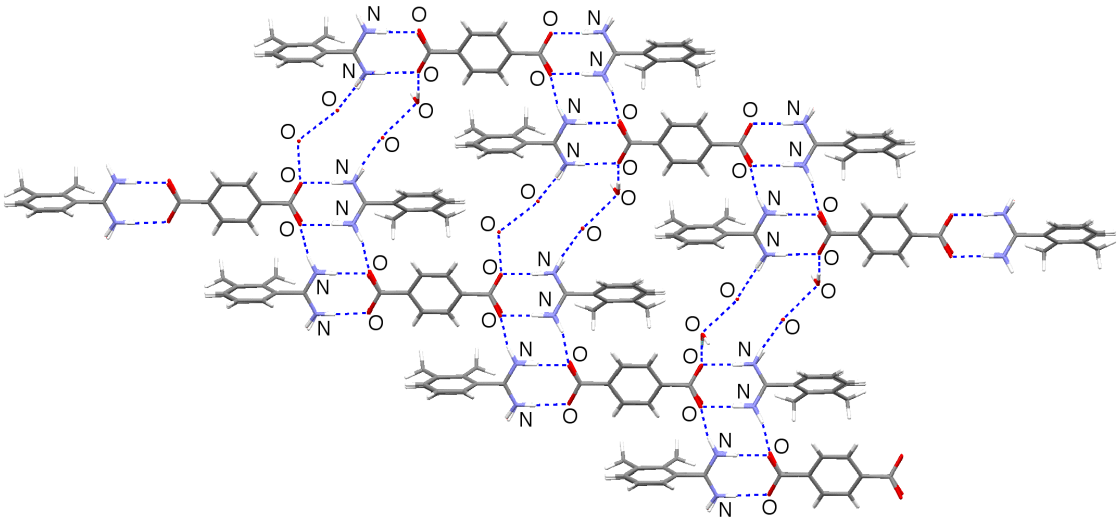


Fig. S7. Crystal structure of **1a$_2$•2a$_2$** showing the formation of the two-dimensional hydrogen-bonding network.

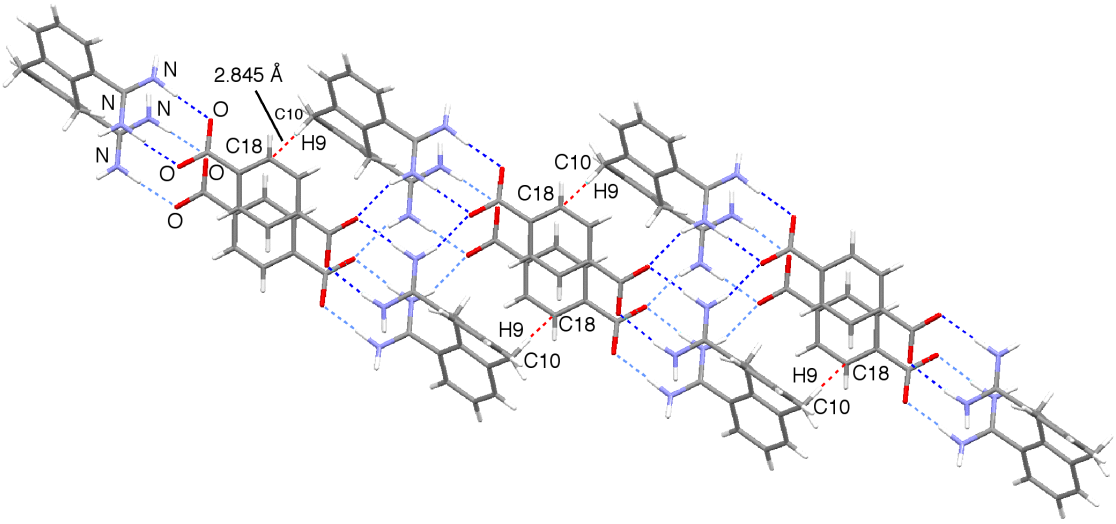


Fig. S8. Crystal structure of **1a**$_2$•**2a**$_2$ showing the formation of the CH-π interaction between **1a** and **2a**. Selected distance (Å): H9-C18: 2.845.

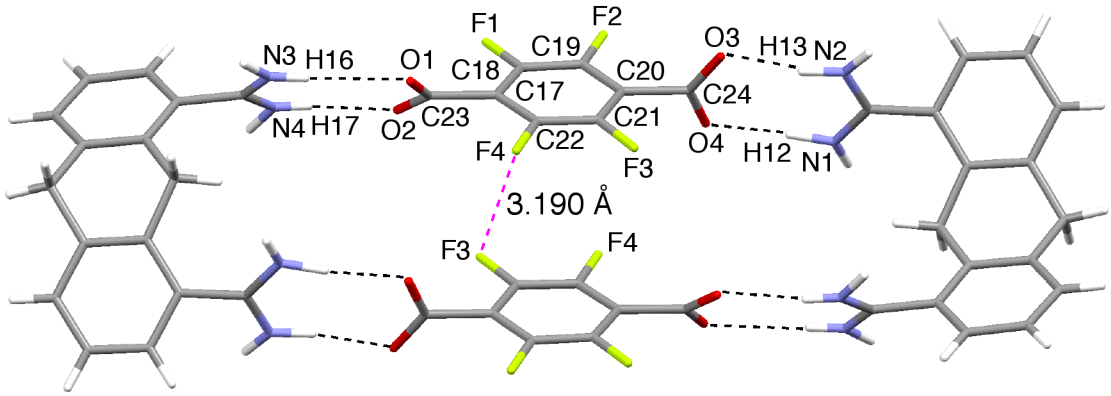


Fig. S9. Crystal structure of **1a**$_2$•**2b**$_2$ showing the formation of the four-component assembly. Selected distances (Å): N1-O4: 2.913(2); N2-O3: 2.794(3); N3-O1: 3.192(3); N4-O2: 2.781(2).

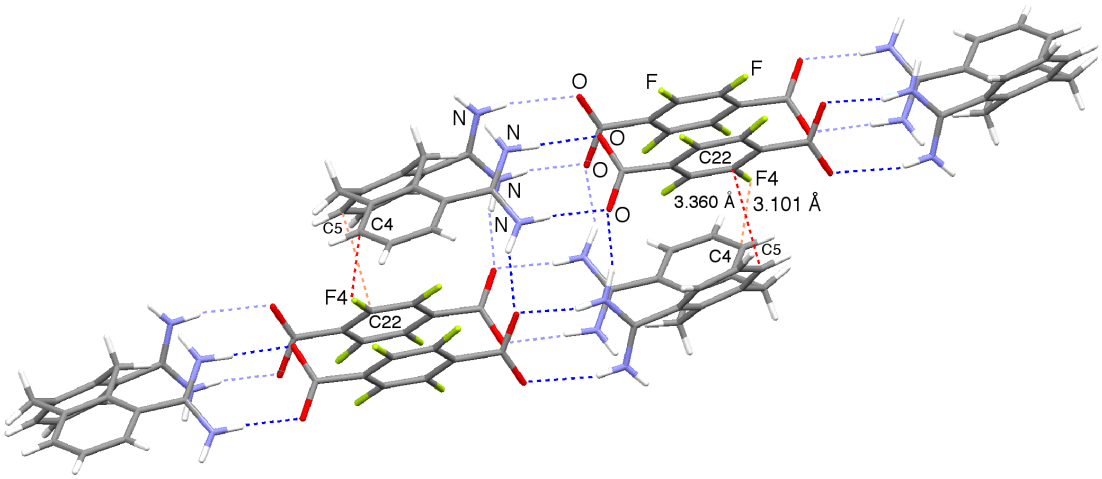


Fig. S10. Crystal structure of **1a**$_2$•**2b**$_2$ showing the formation of the π-π interaction between **1a** and **2b**. Selected distances (Å): C4-F4: 3.101; C5-C22: 3.360.

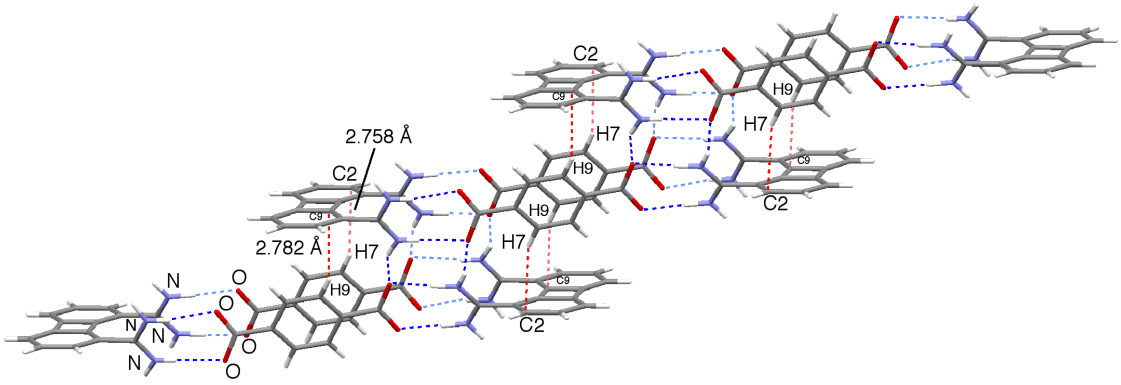


Fig. S11. Crystal structure of **1b**$_2$•**2a**$_2$ showing the formation of the CH-π interaction between **1b** and **2a**. Selected distances (Å): C2-H7: 2.758; C9-H9: 2.782.

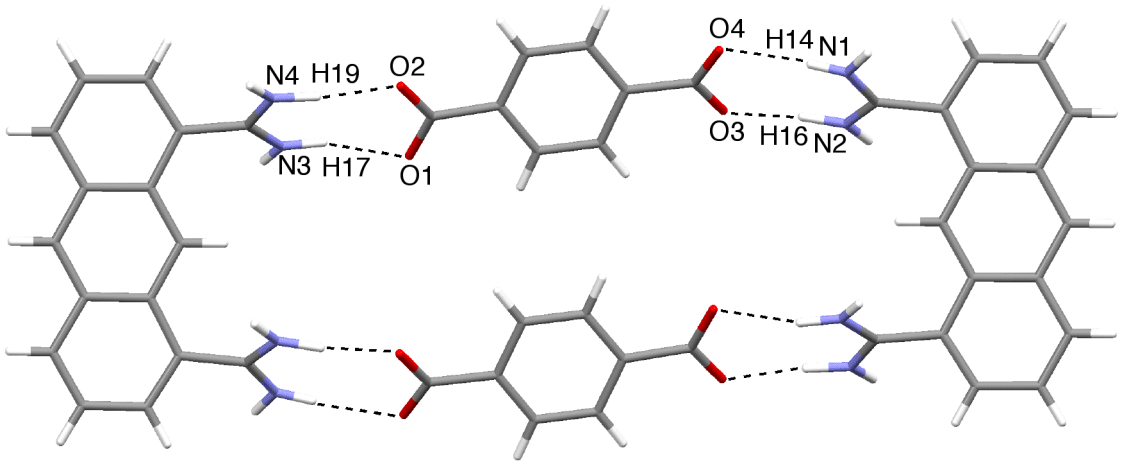


Fig. S12. Crystal structure of **1c$_2$•2a$_2$** showing the formation of the four-component assembly. Selected distances (Å): N1-O4: 2.895(5); N2-O3: 2.730(5); N3-O1: 2.849(5); N4-O2: 2.849(5).

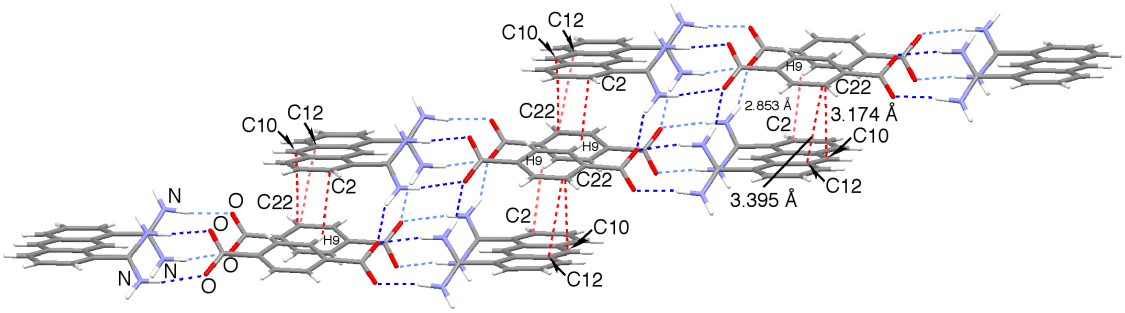


Fig. S13. Crystal structure of **1c$_2$•2a$_2$** showing the formation of the π-π interaction between **1c** and **2a**. Selected distances (Å): C10-C22: 3.174; C12-C22: 3.395.

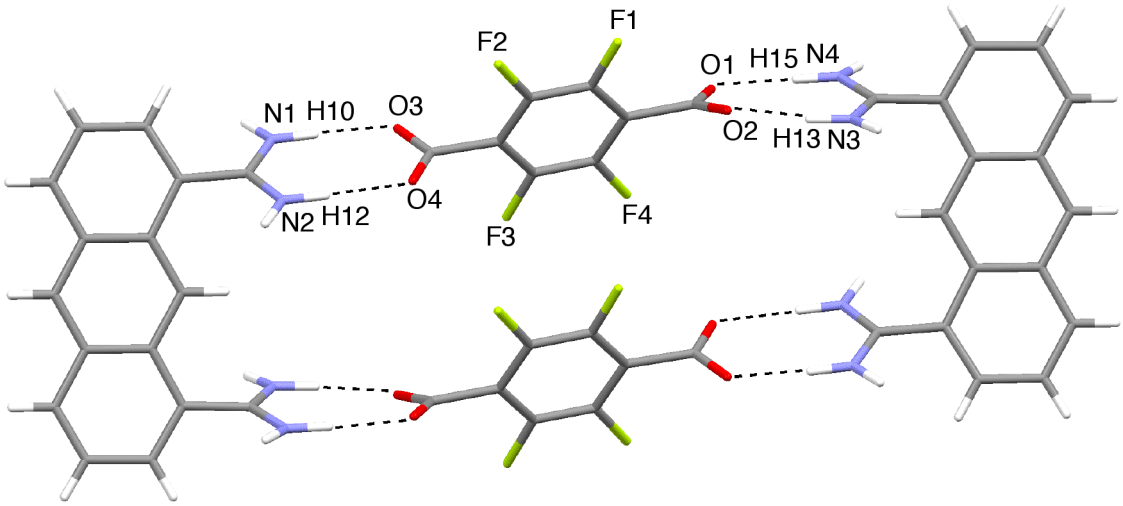


Fig. S14. Crystal structure of **1c**$_2$•**2b**$_2$ showing the formation of the four-component assembly. Selected distances (Å): N1-O3: 2.769(5); N2-O4: 2.888(5); N3-O2: 2.779(5); N4-O1: 2.845(5).

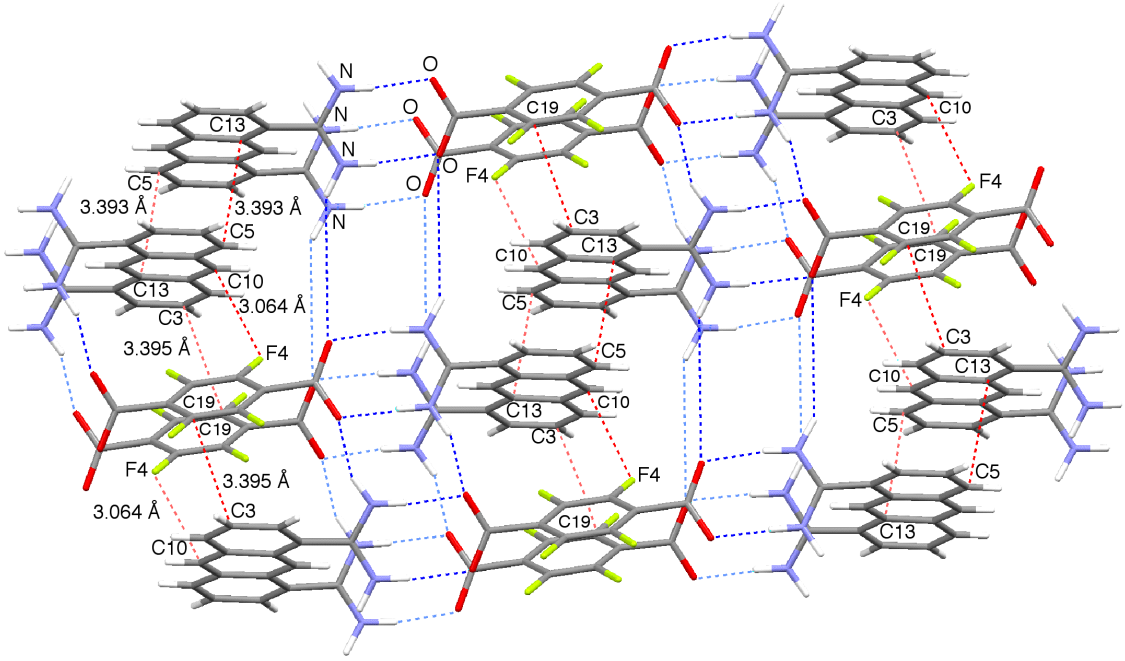


Fig. S15. Crystal structure of **1c**$_2$•**2b**$_2$ showing the formation of the π-π interaction between **1c** and **2b**. Selected distances (Å): C5-C13: 3.393; C3-C19: 3.395; C10-F4: 3.064.

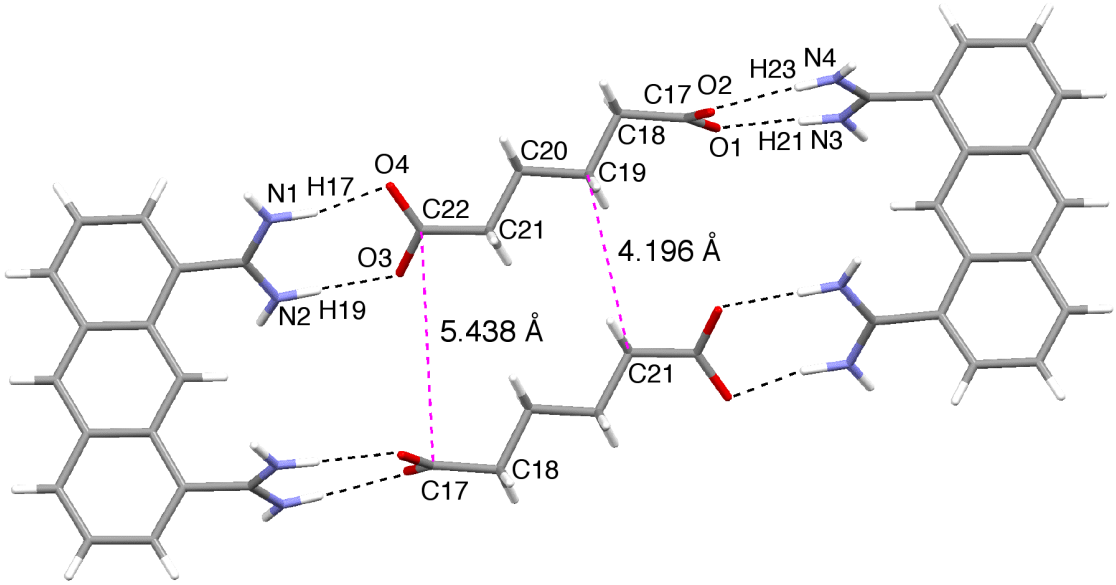


Fig. S16. Crystal structure of **1c$_2$•3a$_2$** showing the formation of the four-component assembly. Selected distances (Å): N1-O4: 2.799(2); N2-O3: 2.792(2); N3-O1: 2.809(2); N4-O2: 2.897(2).

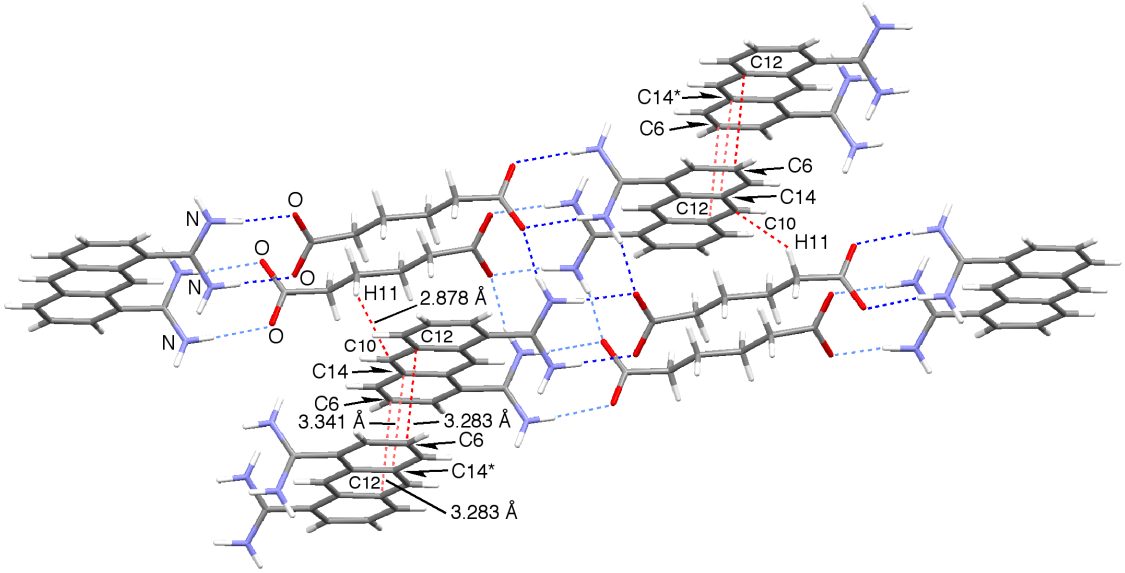


Fig. S17. Crystal structure of **1c$_2$•3a$_2$** showing the formation of the π-π and CH-π interaction. Selected distances (Å): C6-C12: 3.283; C14-C14*: 3.341; C10-H11: 2.877.

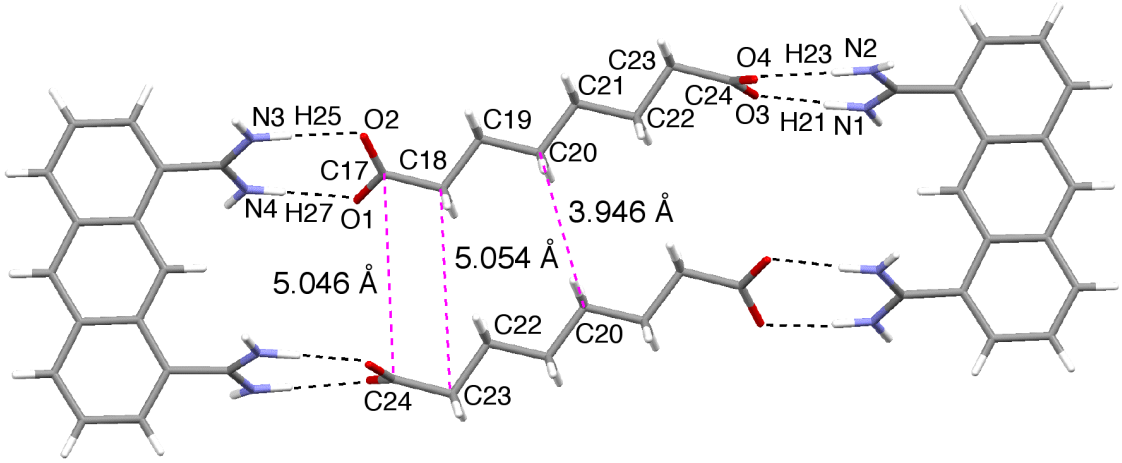


Fig. S18. Crystal structure of **1c$_2$•3b$_2$** showing the formation of the four-component assembly. Selected distances (Å): N1-O3: 2.746(3); N2-O4: 2.893(3); N3-O2: 2.777(3); N4-O1: 2.747(3).

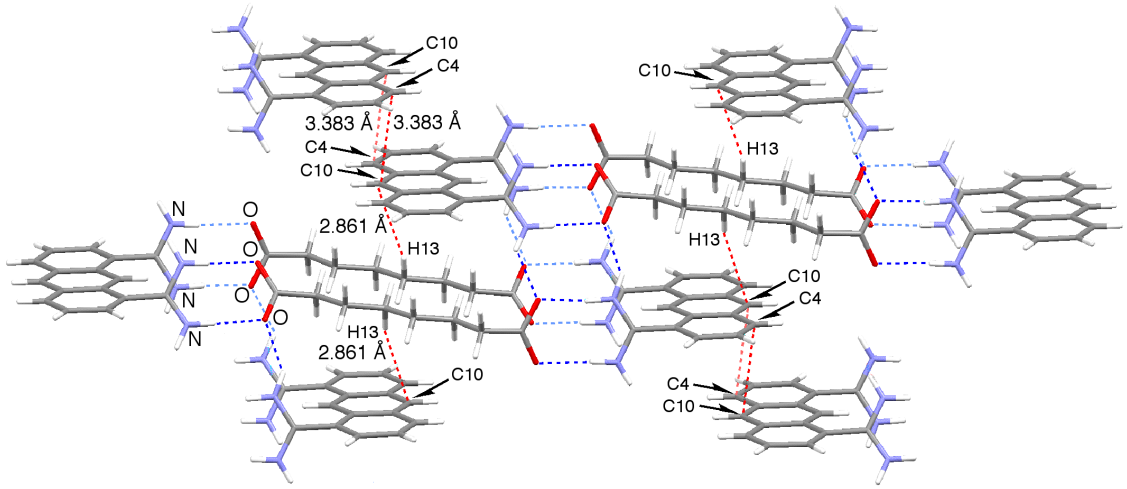


Fig. S19. Crystal structure of **1c$_2$•3b$_2$** showing the formation of the π-π and CH-π interaction. Selected distances (Å): C4-C10: 3.383; C10-H13: 2.861.

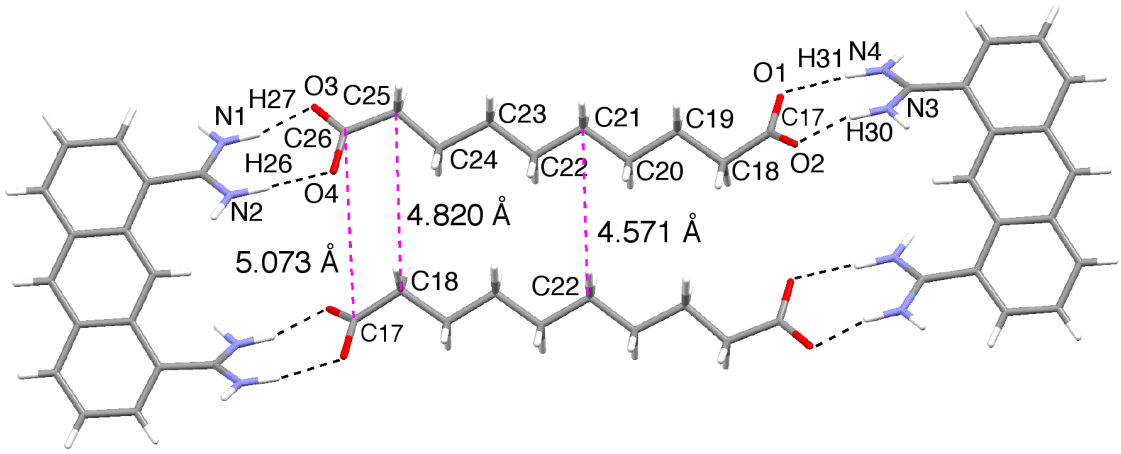


Fig. S20. Crystal structure of **1c**$_2$•**3c**$_2$ showing the formation of the four-component assembly. Selected distances (Å): N1-O3: 2.728(3); N2-O4: 2.873(3); N3-O2: 2.727(3); N4-O1: 2.894(3).

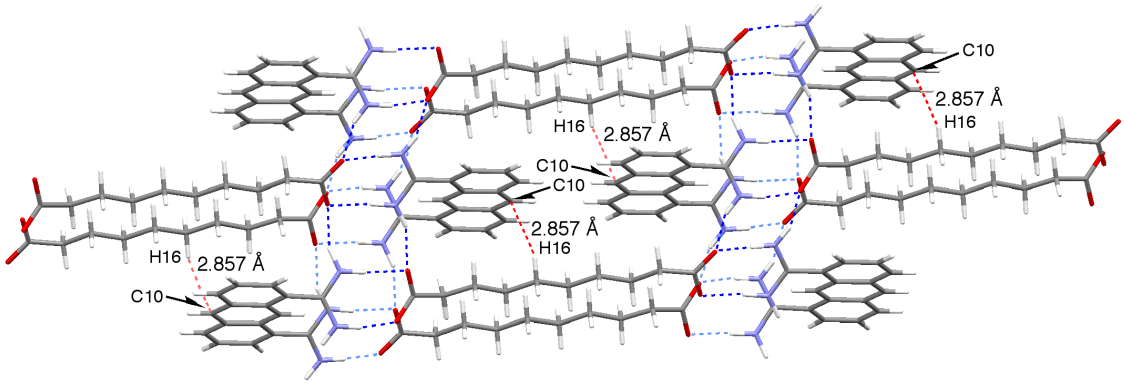


Fig. S21. Crystal structure of **1c**$_2$•**3c**$_2$ showing the formation of the CH-π interaction between **1c** and **3c**. Selected distance (Å): C10-H16: 2.857.

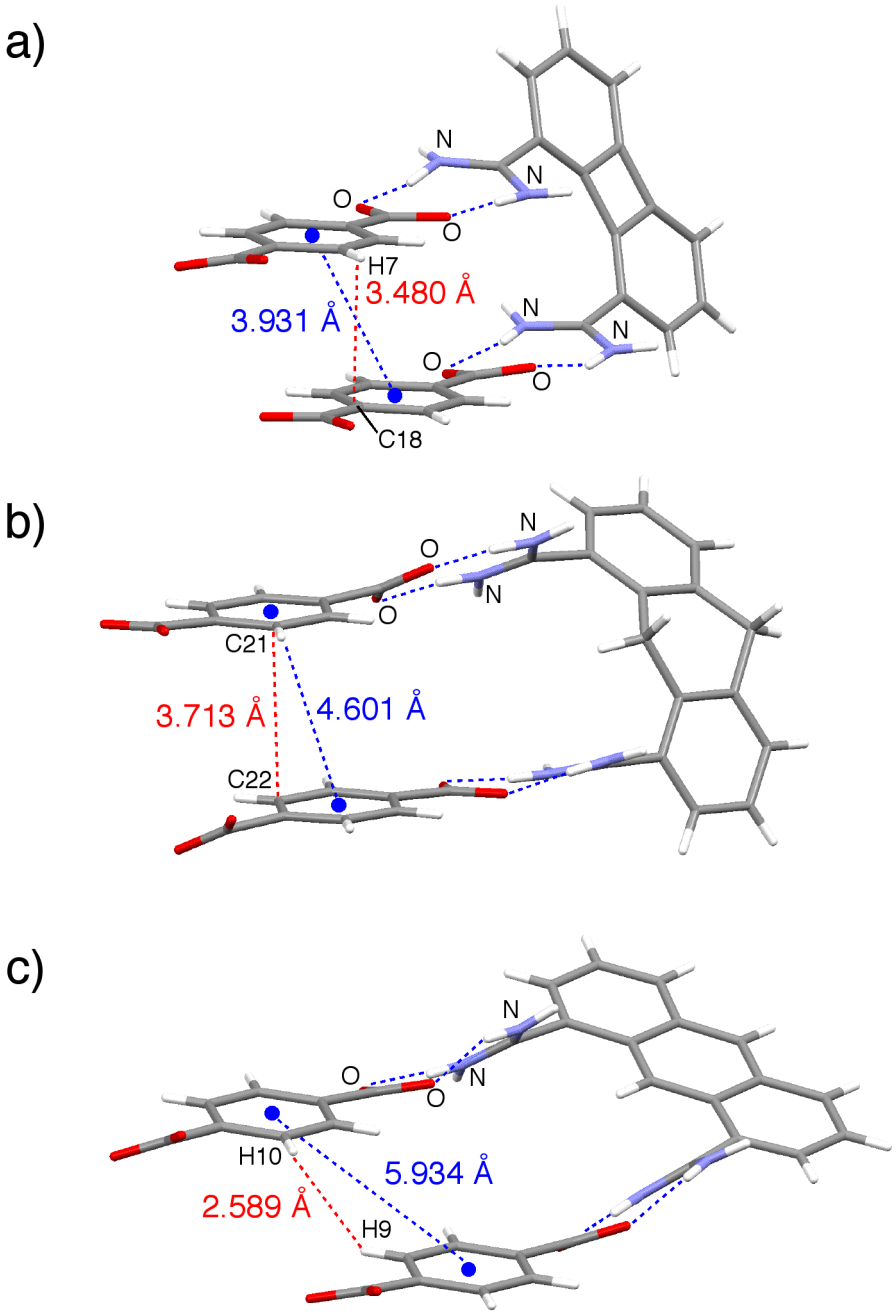


Fig. S22. Comparison of the orientation of terephthalic acid **2a** in the four-component assemblies, a) crystal structure of **1b**$_2$•**2a**$_2$, b) **1a**$_2$•**2a**$_2$, c) **1c**$_2$•**2a**$_2$, one diamidine units **1** are omitted for clarity.

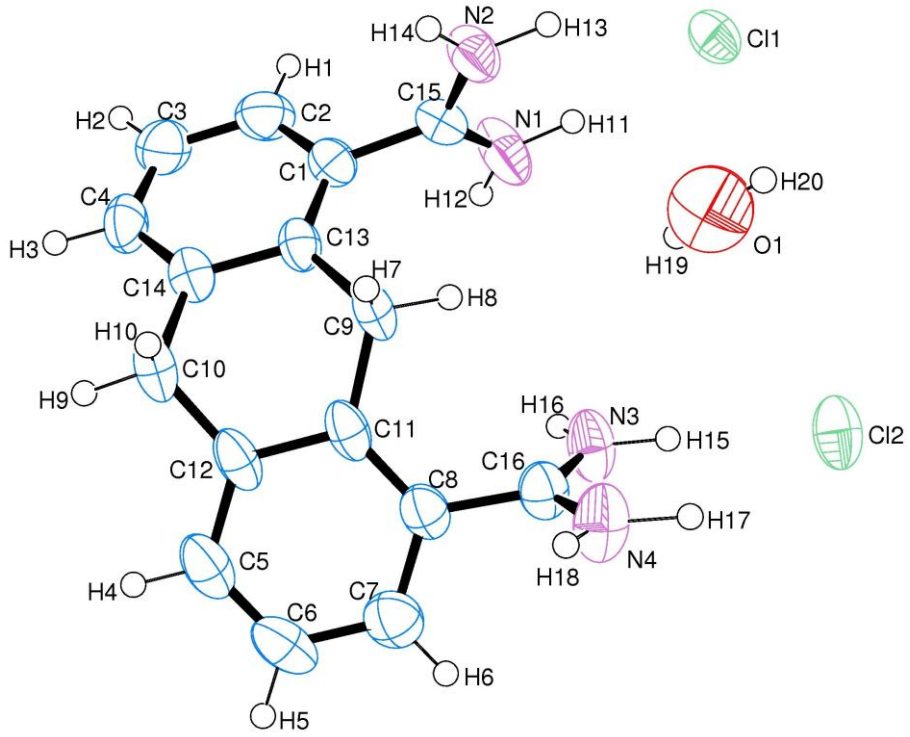


Fig. S23. ORTEP drawing of **1a**.

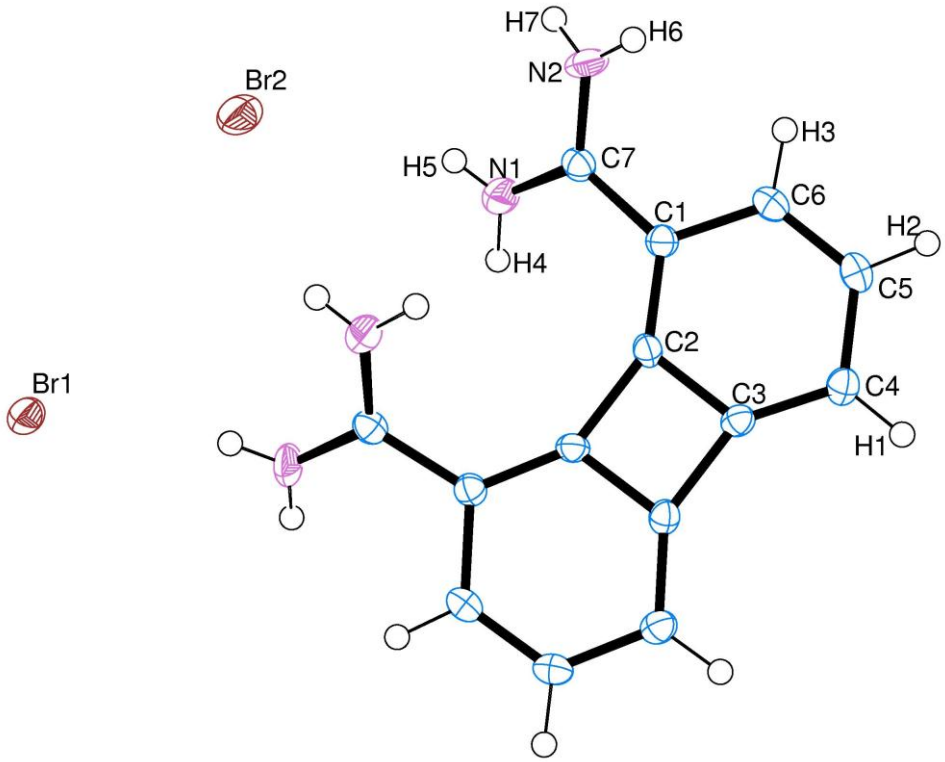


Fig. S24. ORTEP drawing of **1b'**.

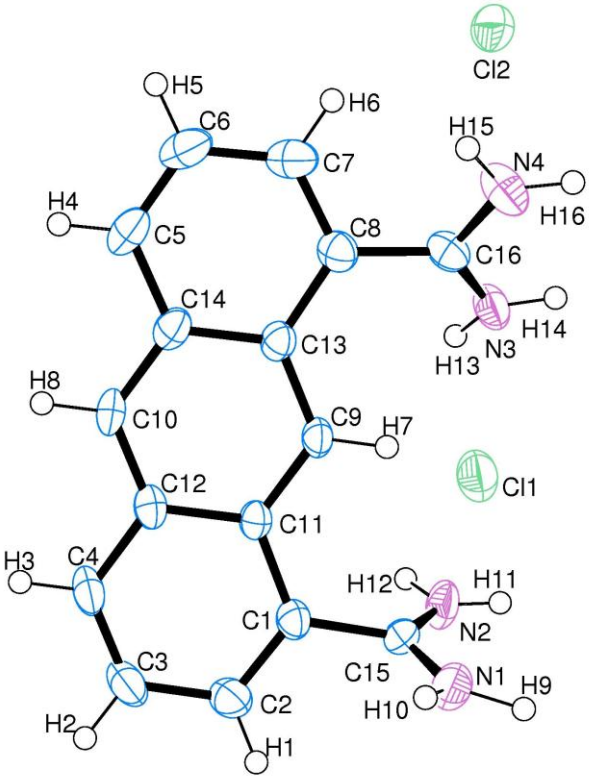


Fig. S25. ORTEP drawing of **1c**. Water molecules are omitted for clarity.

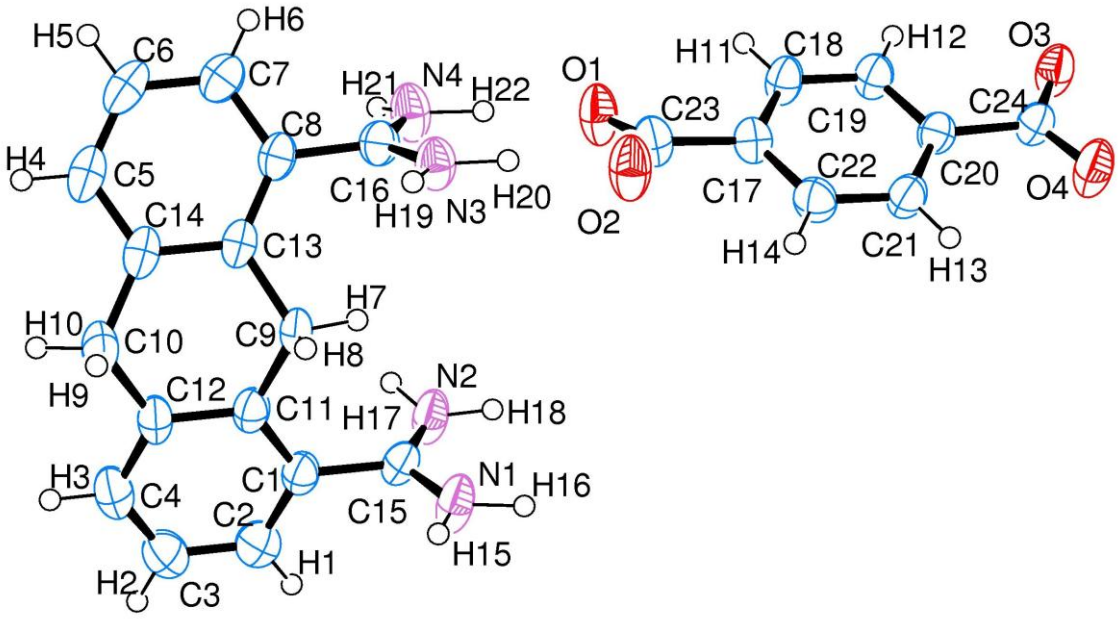


Fig. S26. ORTEP drawing of **1a•2a**. Water molecules are omitted for clarity.

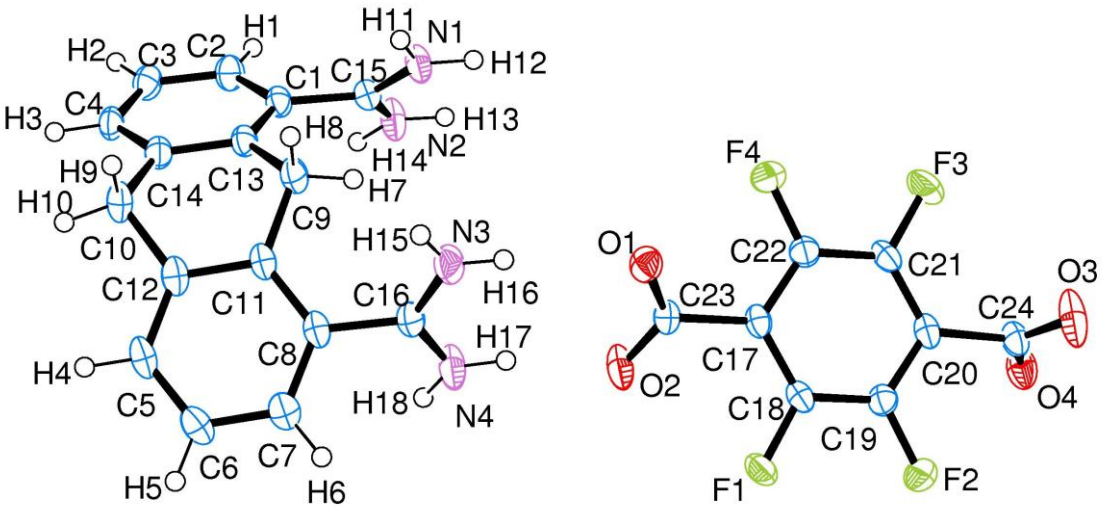


Fig. S27. ORTEP drawing of **1a•2b**. Water molecules are omitted for clarity.

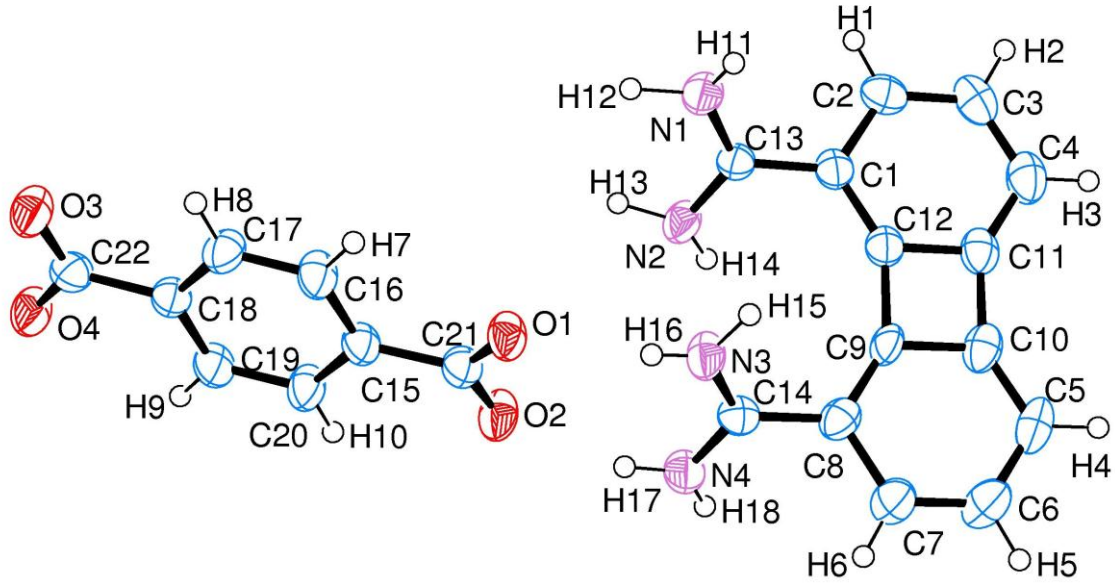


Fig. S28. ORTEP drawing of **1b•2a**. Water molecules are omitted for clarity.

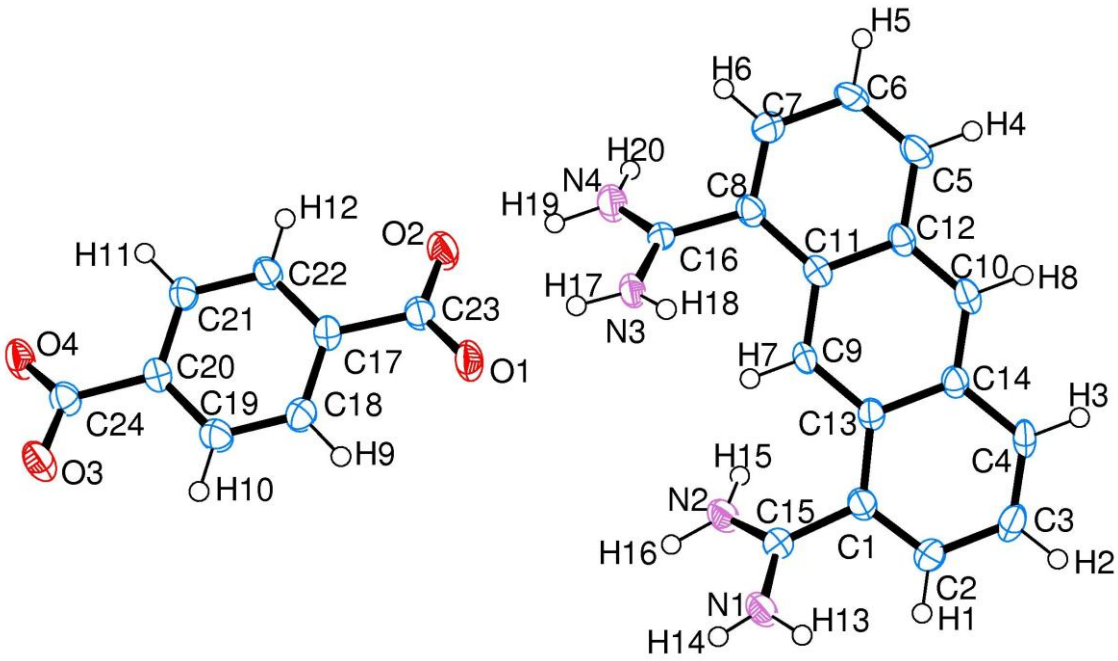


Fig. S29. ORTEP drawing of **1c•2a**. Water molecules are omitted for clarity.

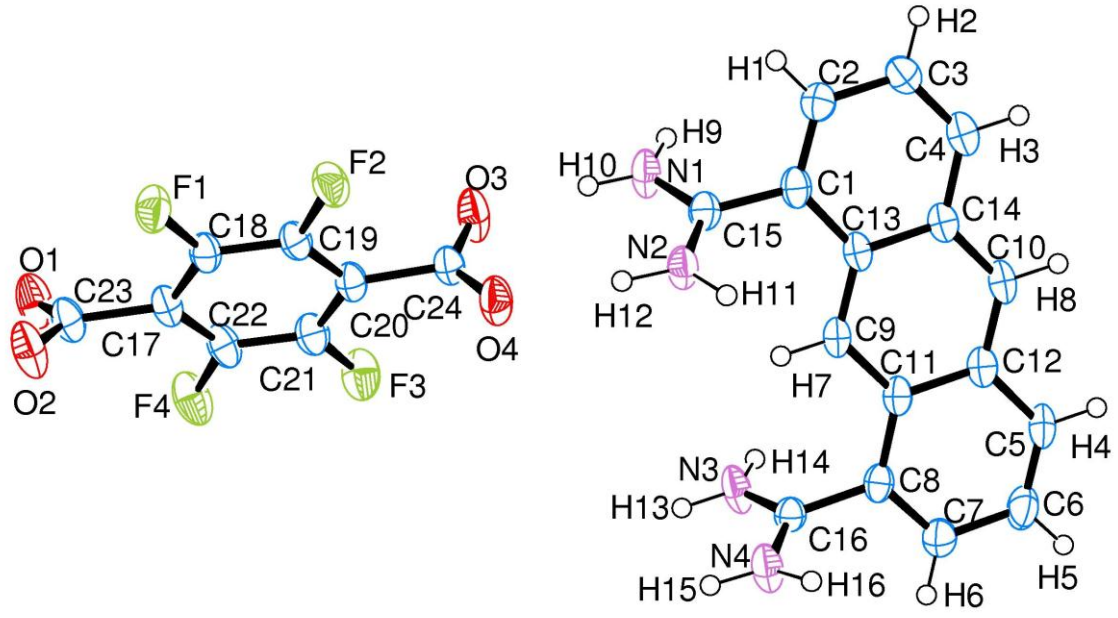


Fig. S30. ORTEP drawing of **1c•2b**. Water molecules are omitted for clarity.

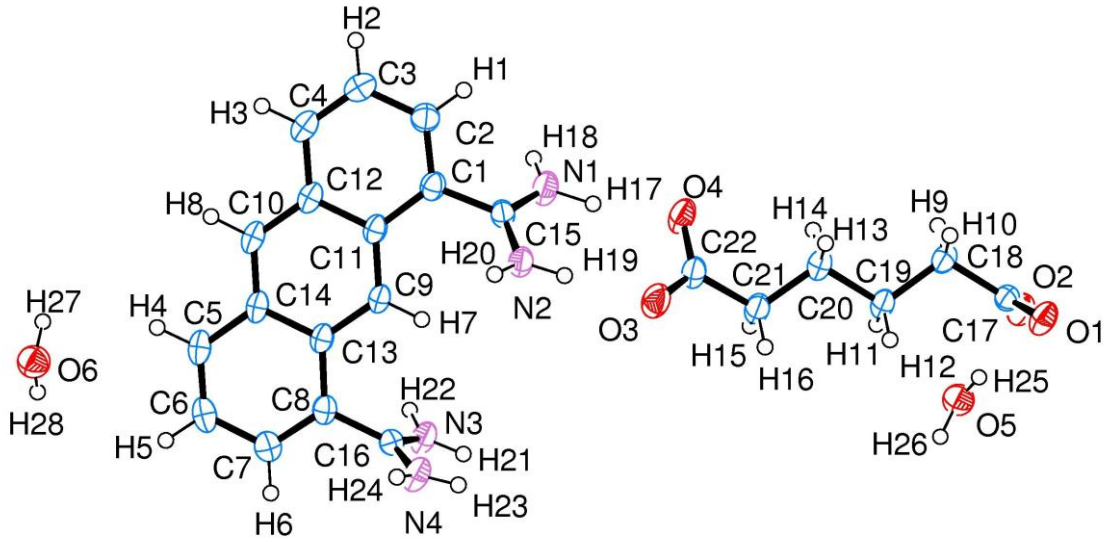


Fig. S31. ORTEP drawing of **1c•3a**.

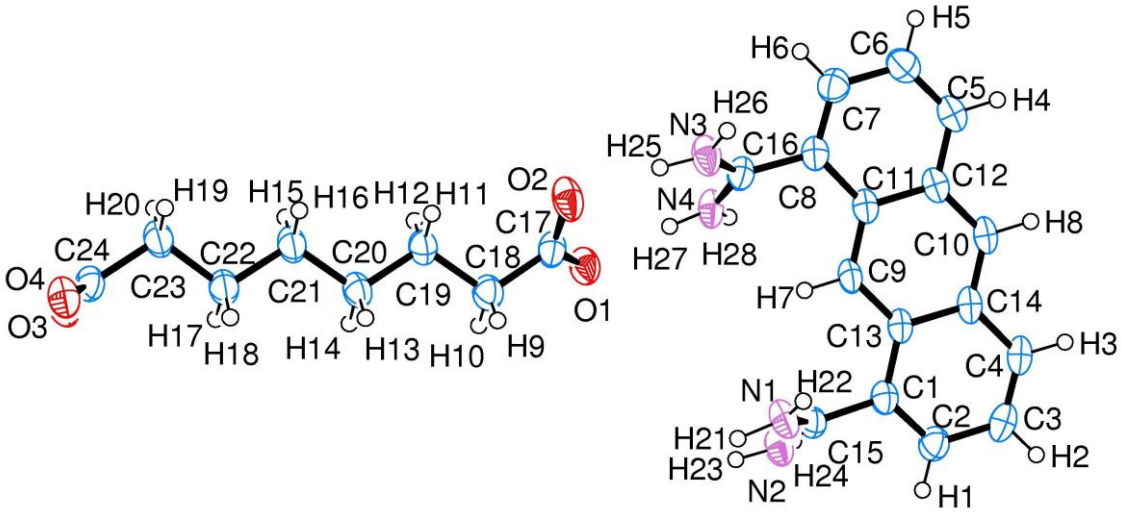


Fig. S32. ORTEP drawing of **1c•3b**. Water molecules are omitted for clarity.

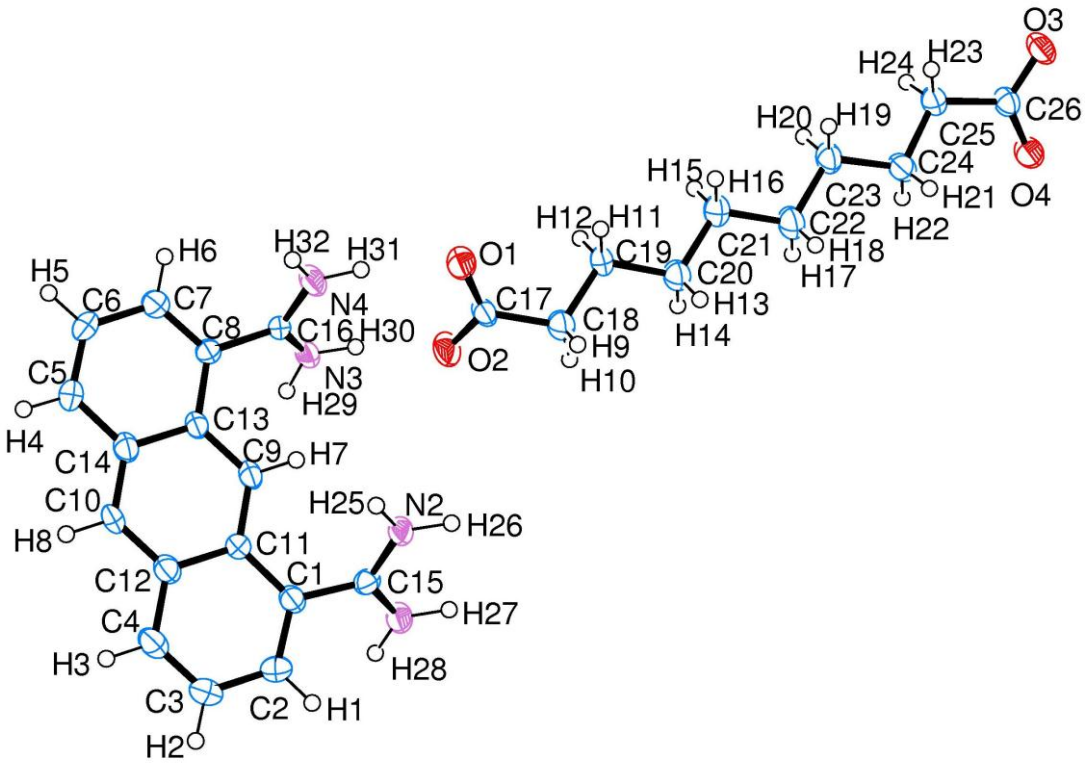
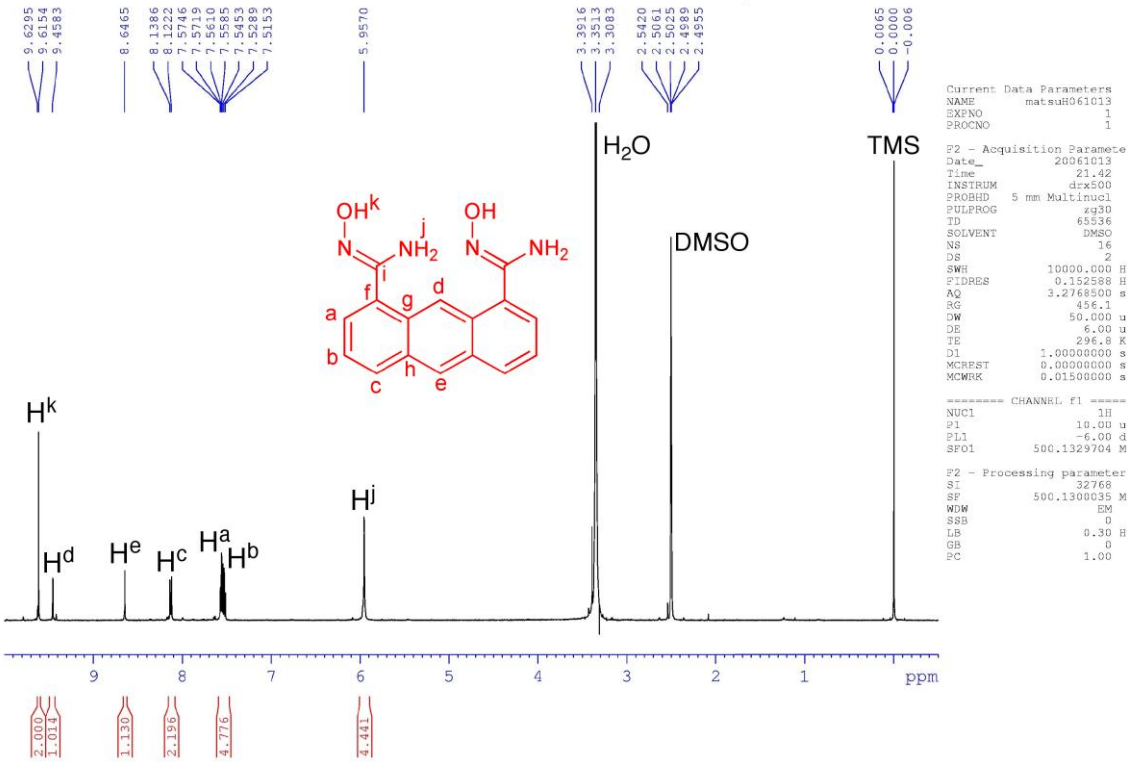


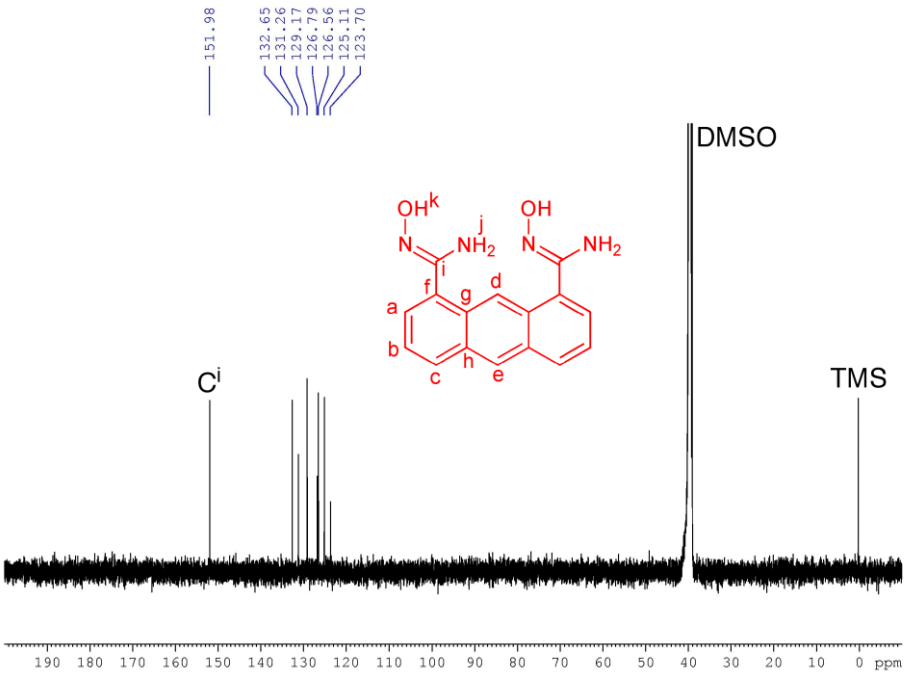
Fig. S33. ORTEP drawing of **1c•3c**.

^{1}H NMR spectrum (DMSO-d_6)



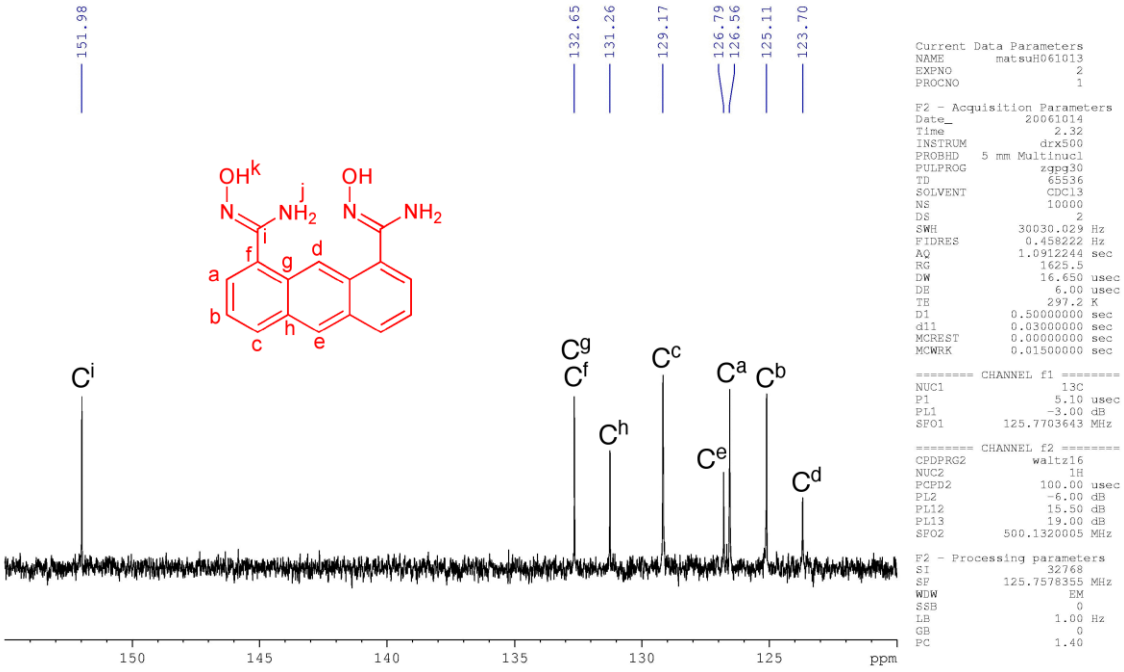

```
Current Data Parameters
NAME         matsuH061013
EXPNO                   1
PROCNO                  1

F2 - Acquisition Parameters
Date_              20061013
Time                 21.42
INSTRUM              drx500
PROBHD   5 mm Multinucl
PULPROG              zg30
TD                  65536
SOLVENT              DMSO
NS                     16
DS                      2
SWH             10000.000 H
FIDRES              0.152588 H
AQ                  3.2768500 s
RG                      456.1
DW                   50.000 u
DE                    6.00 u
TE                   296.8 K
D1                1.00000000 s
MCREST            0.00000000 s
MCWRK             0.01500000 s

======== CHANNEL f1 =====
NUC1                     1H
P1                    10.00 u
PL1                   -6.00 d
SFO1             500.1329704 M

F2 - Processing parameter
SI                      32768
SF               500.1300035 M
WDW                     EM
SSB                      0
LB                    0.30 H
GB                       0
PC                    1.00
```

^{13}C NMR spectrum (DMSO-d_6)



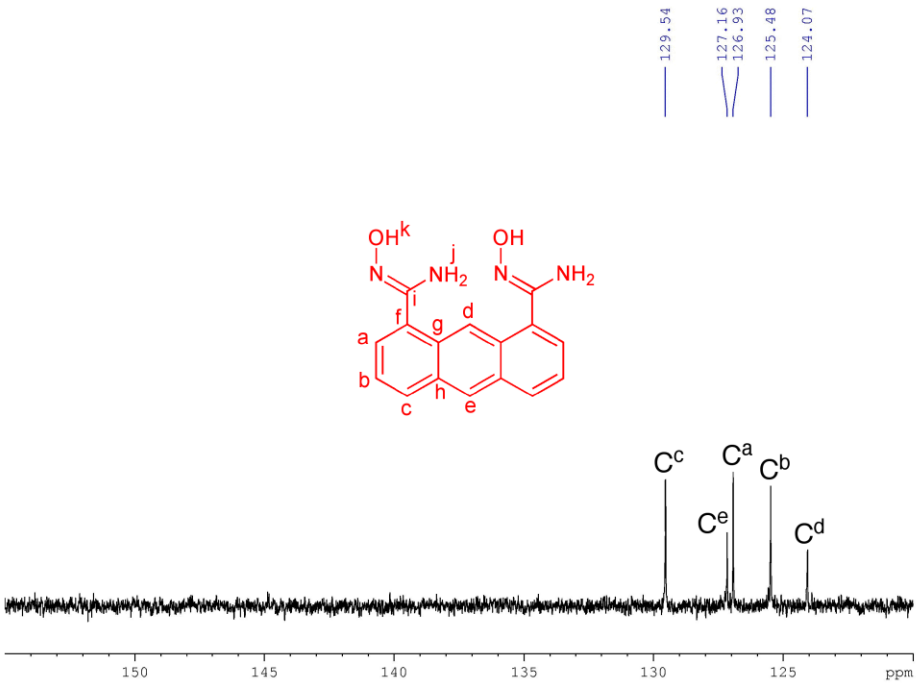

```
Current Data Parameters
NAME         matsuH061013
EXPNO                   2
PROCNO                  1

F2 - Acquisition Parameters
Date_              20061014
Time                  2.32
INSTRUM              drx500
PROBHD   5 mm Multinucl
PULPROG             zgpg30
TD                  65536
SOLVENT              CDCl3
NS                     10000
DS                      2
SWH             30030.029 Hz
FIDRES              0.458222 Hz
AQ                  1.0912244 sec
RG                      1625.5
DW                   16.650 usec
DE                    6.00 usec
TE                   297.2 K
D1                0.50000000 sec
d11               0.03000000 sec
MCREST            0.00000000 sec
MCWRK             0.01500000 sec

======== CHANNEL f1 ========
NUC1                    13C
P1                    5.10 usec
PL1                   -3.00 dB
SFO1            125.7703643 MHz

======== CHANNEL f2 ========
CPDPRG2             waltz16
NUC2                    1H
PCPD2                100.00 usec
PL2                   -6.00 dB
PL12                  15.50 dB
PL13                  19.00 dB
SFO2             500.1320005 MHz

F2 - Processing parameters
SI                      32768
SF              125.7578355 MHz
WDW                     EM
SSB                      0
LB                    1.00 Hz
GB                       0
PC                    1.40
```

^{13}C NMR spectrum (DMSO-d_6)

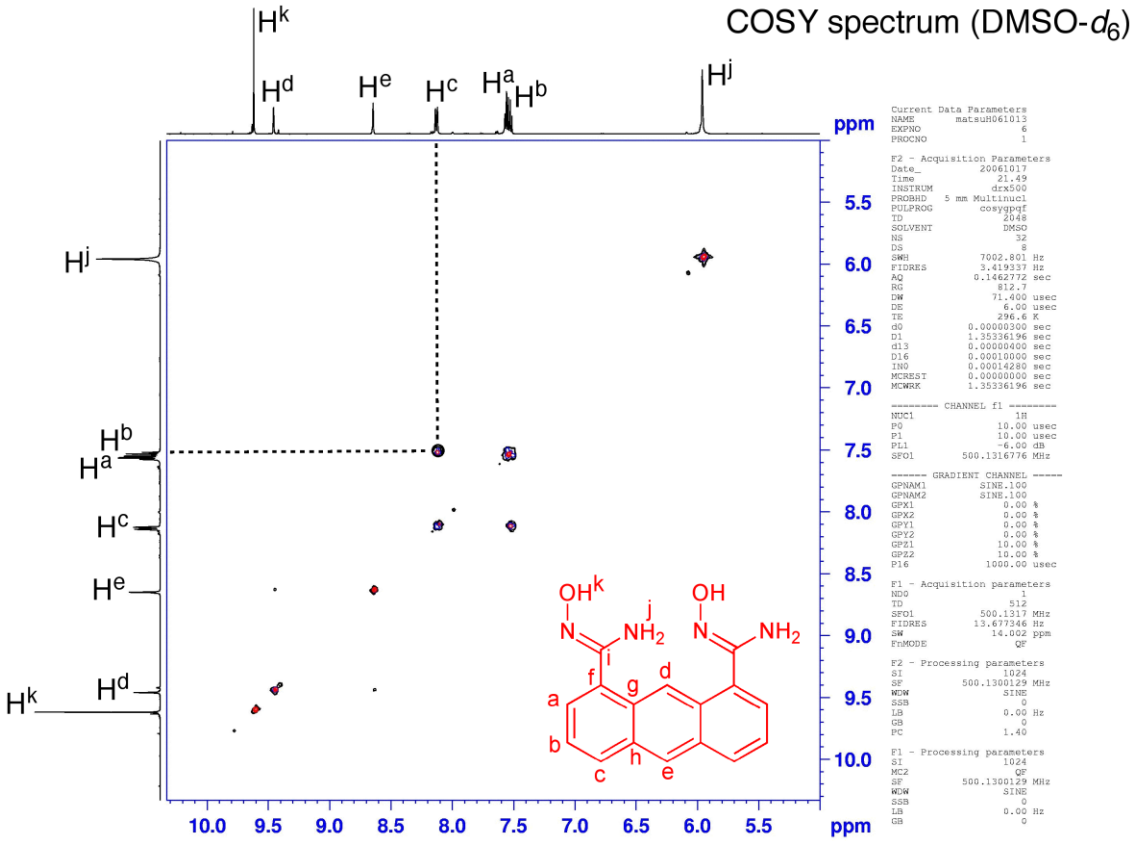


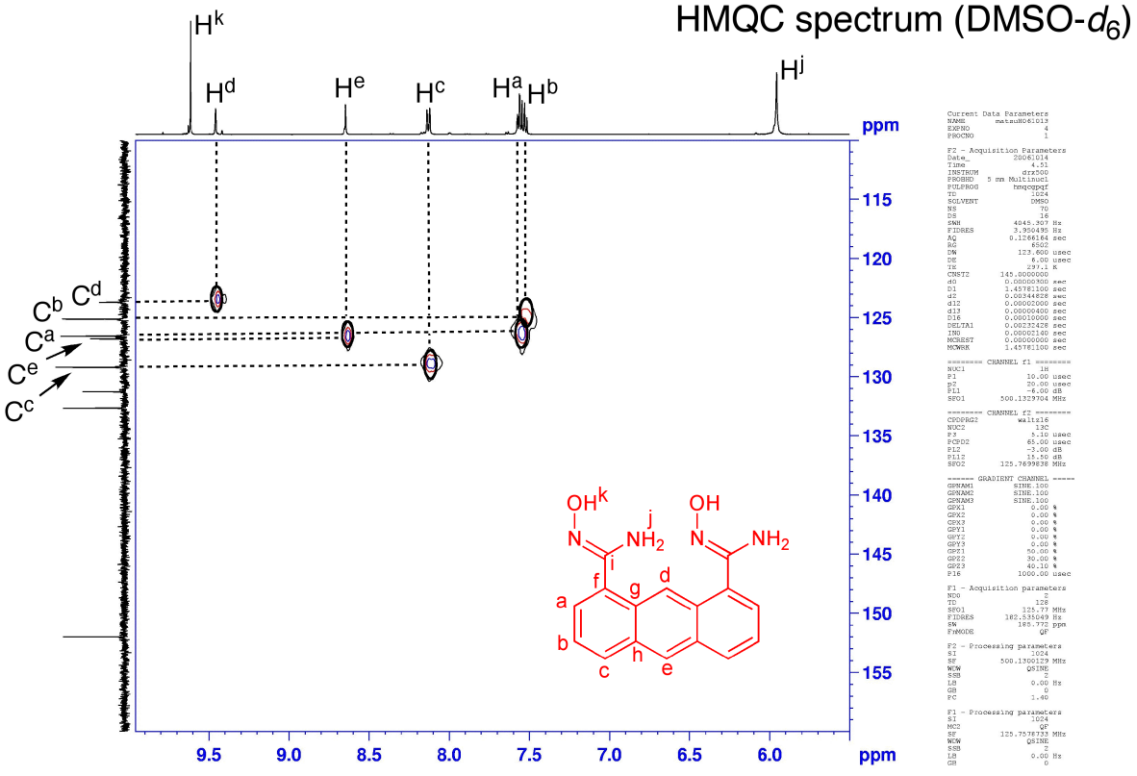

```
Current Data Parameters
NAME          matsuH061013
EXPNO                    2
PROCNO                   1

F2 - Acquisition Parameters
Date_              20061014
Time                   2.32
INSTRUM              drx500
PROBHD   5 mm Multinucl
PULPROG              zgpg30
TD                    65536
SOLVENT               CDCl3
NS                    10000
DS                        2
SWH               30030.029 Hz
FIDRES             0.458222 Hz
AQ                 1.0912244 sec
RG                    1625.5
DW                   16.650 usec
DE                    6.00 usec
TE                   297.2 K
D1                0.50000000 sec
d11               0.03000000 sec
MCREST            0.00000000 sec
MCWRK             0.01500000 sec

======== CHANNEL f1 ========
NUC1                    13C
P1                     5.10 usec
PL1                   -3.00 dB
SFO1           125.7703643 MHz

======== CHANNEL f2 ========
CPDPRG2              waltz16
NUC2                    1H
PCPD2                100.00 usec
PL2                   -6.00 dB
PL12                  15.50 dB
PL13                  19.00 dB
SFO2           500.1320005 MHz

F2 - Processing parameters
SI                       32768
SF             125.7578355 MHz
WDW                         EM
SSB                          0
LB                        1.00 Hz
GB                           0
PC                        1.40
```

DEPT spectrum (DMSO-d_6)

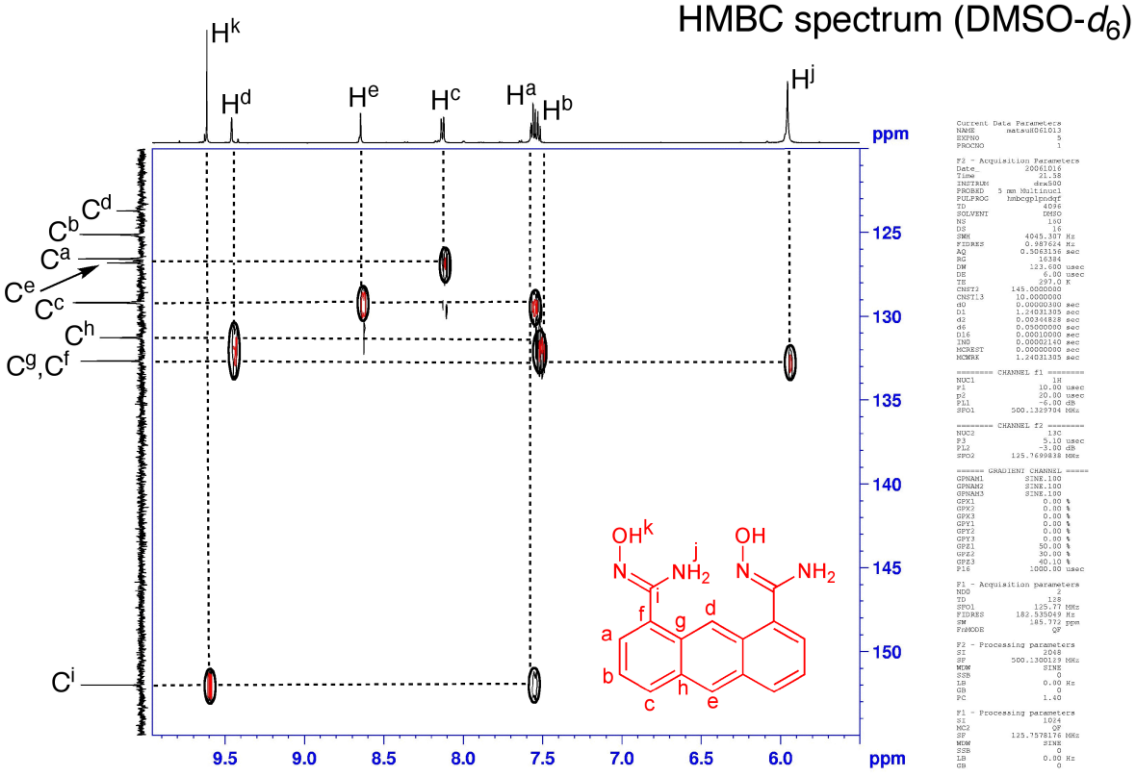



```
Current Data Parameters
NAME          matsuH061013
EXPNO                    3
PROCNO                   1

F2 - Acquisition Parameters
Date_              20061014
Time                   4.49
INSTRUM              drx500
PROBHD   5 mm Multinucl
PULPROG             dept135
TD                    65536
SOLVENT               CDCl3
NS                     5000
DS                        2
SWH               30030.029 Hz
FIDRES             0.458222 Hz
AQ                 1.0912244 sec
RG                    4597.6
DW                   16.650 usec
DE                    6.00 usec
TE                   297.1 K
CNST2             145.0000000
D1                0.50000000 sec
d2                0.00344828 sec
d12               0.00002000 sec
DELTA             0.00000649 sec
MCREST            0.00000000 sec
MCWRK             0.01500000 sec

======== CHANNEL f1 ========
NUC1                    13C
P1                     5.10 usec
p2                    10.20 usec
PL1                   -3.00 dB
SFO1           125.7703643 MHz

-------- CHANNEL f2 --------
CPDPRG2              waltz16
NUC2                    1H
P3                    10.00 usec
p4                    20.00 usec
PCPD2                100.00 usec
PL2                   -6.00 dB
PL12                  15.50 dB
SFO2           500.1320005 MHz

F2 - Processing parameters
SI                       32768
SF             125.7577890 MHz
WDW                         EM
SSB                          0
LB                        1.00 Hz
GB                           0
PC                        1.40
```


COSY spectrum (DMSO-d_6)
H^k
H^d
H^e
H^c
H^a
H^b
H^j
ppm
10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 ppm


HMQC spectrum (DMSO-d_6)
H^k
H^d
H^e
H^c
H^a
H^b
H^j
C^b C^d
C^a
C^e
C^c
ppm
115 120 125 130 135 140 145 150 155
9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 ppm

HMBC spectrum (DMSO-d_6)



Current Data Parameters
NAME matsu061013
EXPNO 5
PROCNO 1

F2 - Acquisition Parameters
Date_ 20061016
Time 21.58
INSTRUM dra600
PROBHD 5 mm Multinuc1
PULPROG hmbcgplpndqf
TD 4096
SOLVENT DMSO
NS 140
DS 16
SWH 4045.307 Hz
FIDRES 0.987624 Hz
AQ 0.5063136 sec
RG 16384
DW 123.600 usec
DE 6.00 usec
TE 297.0 K
CNST2 145.0000000
CNST13 10.0000000
d0 0.00000300 sec
D1 1.24031305 sec
d2 0.00364828 sec
d6 0.05000000 sec
D16 0.00010000 sec
ING 0.00002140 sec
MCREST 0.00000000 sec
MCWRK 1.24031305 sec

======== CHANNEL f1 ========
NUC1 1H
P1 10.00 usec
p2 20.00 usec
PL1 -6.00 dB
SFO1 500.1329704 MHz

======== CHANNEL f2 ========
NUC2 13C
P3 5.10 usec
PL2 -3.00 dB
SFO2 125.7699838 MHz

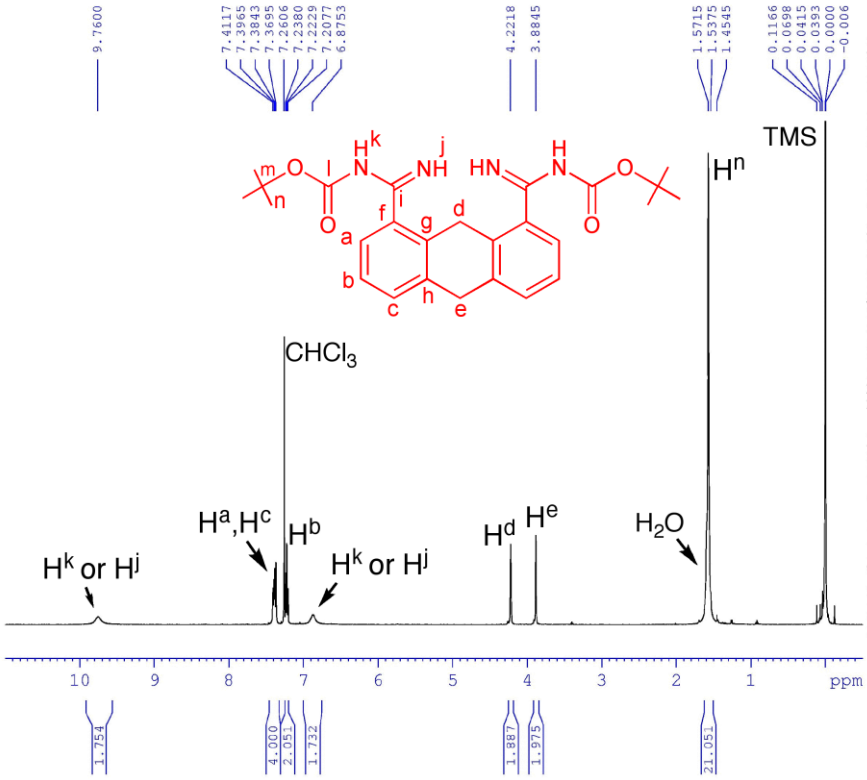
====== GRADIENT CHANNEL =====
GPNAM1 SINE.100
GPNAM2 SINE.100
GPNAM3 SINE.100
GPX1 0.00 %
GPX2 0.00 %
GPX3 0.00 %
GPY1 0.00 %
GPY2 0.00 %
GPY3 0.00 %
GPZ1 50.00 %
GPZ2 30.00 %
GPZ3 40.10 %
P16 1000.00 usec

F1 - Acquisition parameters
ND0 2
TD 128
SFO1 125.77 MHz
FIDRES 182.535049 Hz
SW 185.772 ppm
FnMODE QF

F2 - Processing parameters
SI 2048
SF 500.1300139 MHz
WDW SINE
SSB 0
LB 0.00 Hz
GB 0
PC 1.40

F1 - Processing parameters
SI 1024
MC2 QF
SF 125.7578176 MHz
WDW SINE
SSB 0
LB 0.00 Hz
GB 0

^{1}H NMR spectrum ($CDCl_3$)

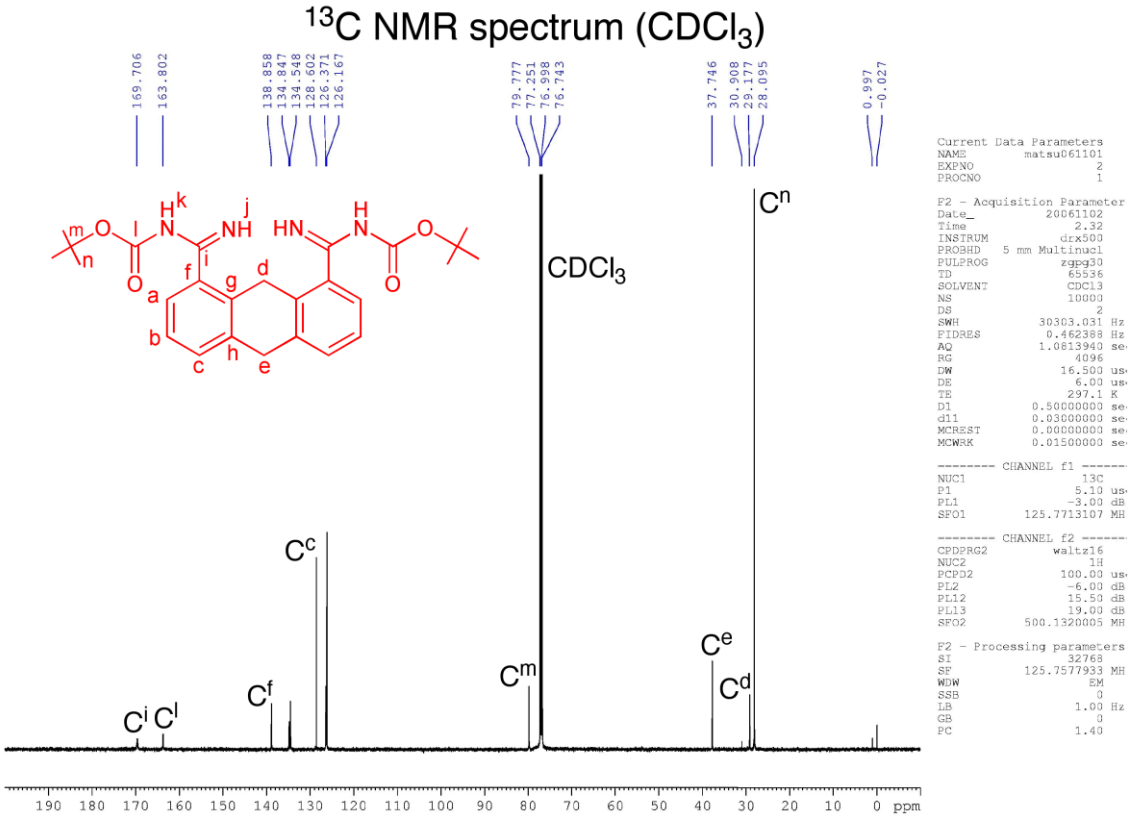


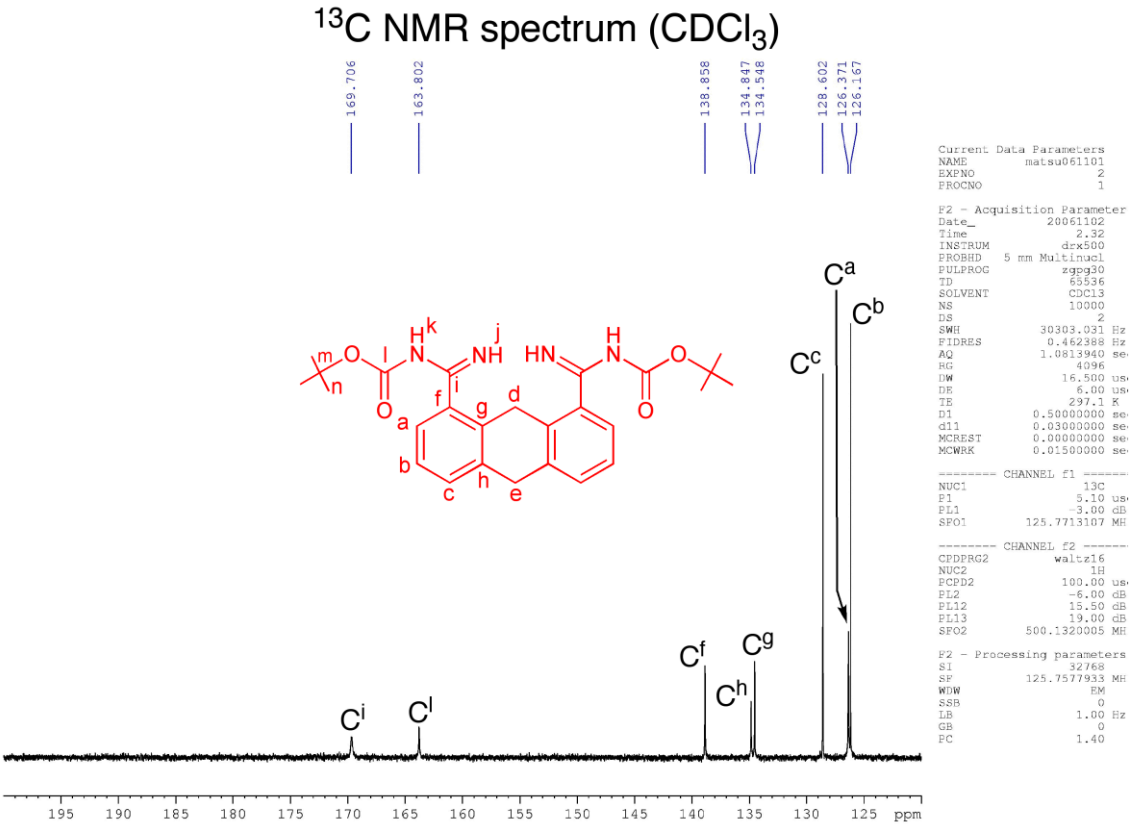
Current Data Parameters
NAME matsu061101
EXPNO 7
PROCNO 1

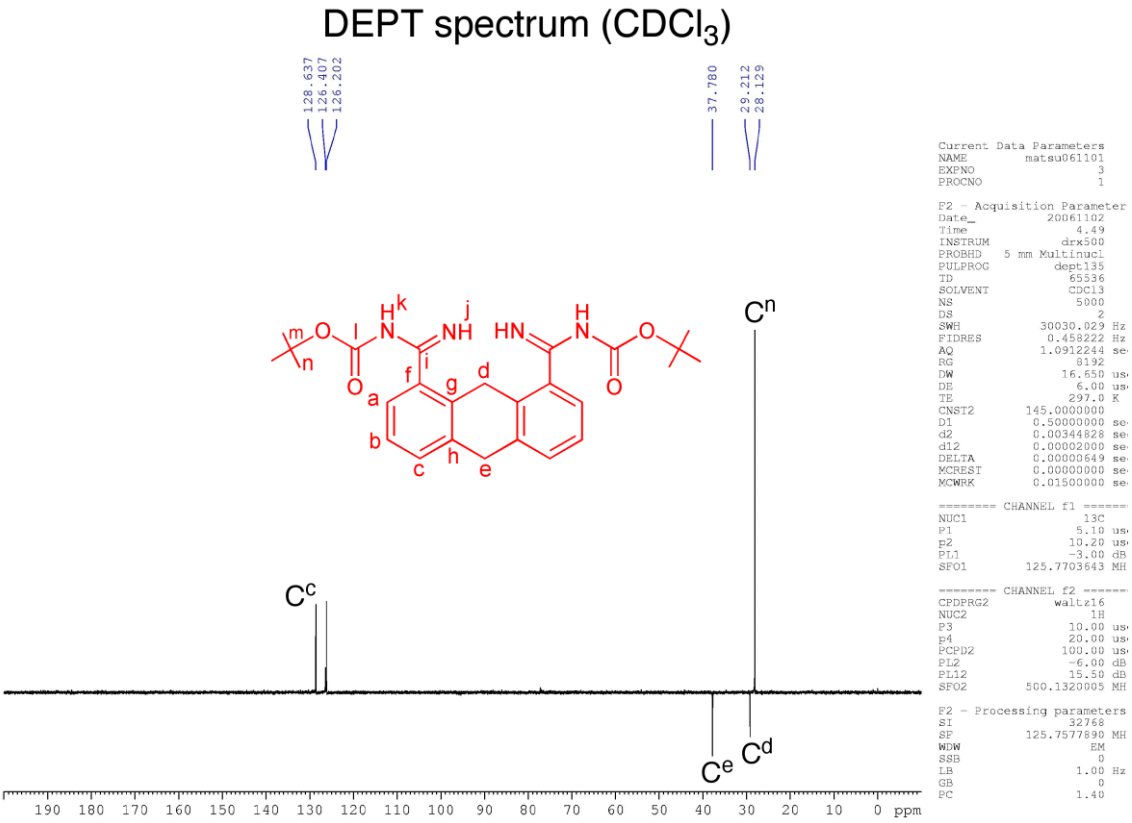
F2 - Acquisition Parameters
Date_ 20090310
Time 14.07
INSTRUM drx500
PROBHD 5 mm Multinuc1
PULPROG zg30
TD 65536
SOLVENT CDCl3
NS 16
DS 2
SWH 10330.578 Hz
FIDRES 0.157632 Hz
AQ 3.1719923 sec
RG 574.7
DW 48.400 usec
DE 6.00 usec
TE 298.0 K
D1 1.00000000 sec
MCREST 0.00000000 sec
MCWRK 0.01500000 sec

======== CHANNEL f1 ========
NUC1 1H
P1 10.00 usec
PL1 -6.00 dB
SFO1 500.1330885 MHz

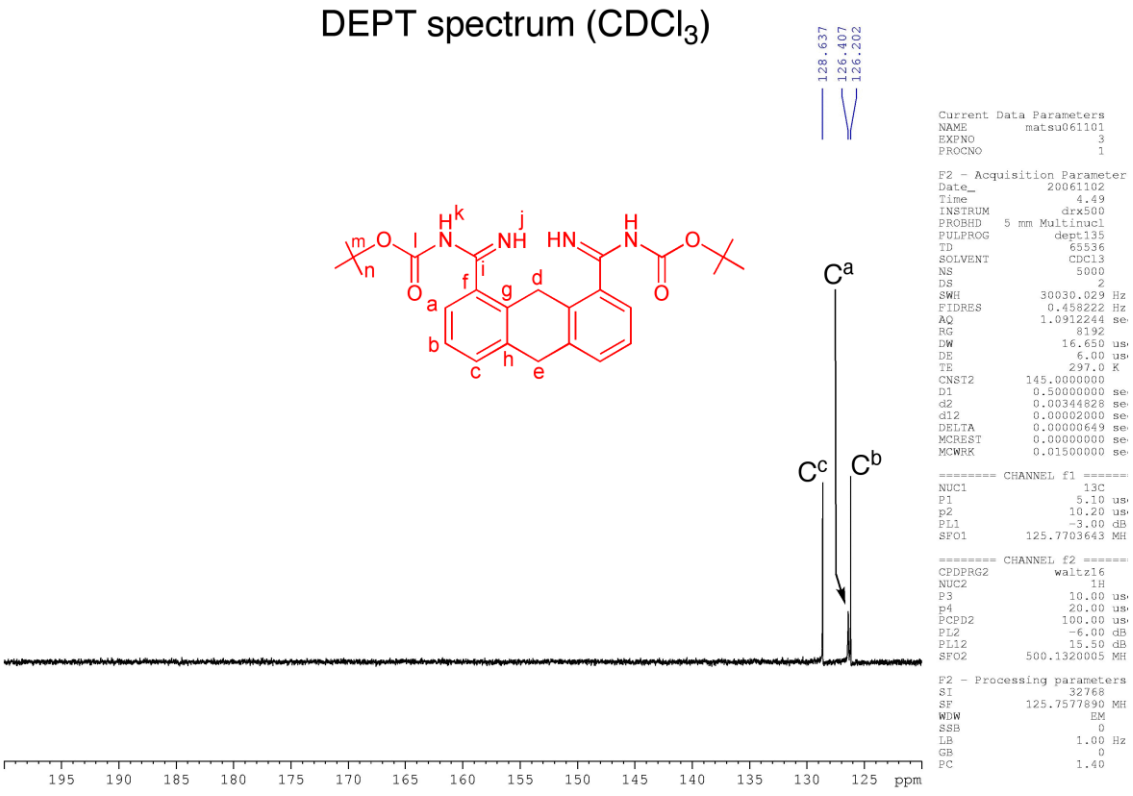
F2 - Processing parameters
SI 32768
SF 500.1300131 MHz
WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 1.00


13C NMR spectrum (CDCl3)
169.706
163.802
138.858
134.847
134.548
128.602
126.371
126.167
79.777
77.251
76.998
76.743
37.746
30.908
29.177
28.095
0.997
-0.027
CDCl3
Cn
Cc
Ce
Cm
Cf
Cd
Ci Cl
190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 ppm
Current Data Parameters
NAME matsu061101
EXPNO 2
PROCNO 1
F2 - Acquisition Parameter
Date_ 20061102
Time 2.32
INSTRUM drx500
PROBHD 5 mm Multinucl
PULPROG zgpg30
TD 65536
SOLVENT CDCl3
NS 10000
DS 2
SWH 30303.031 Hz
FIDRES 0.462388 Hz
AQ 1.0813940 se
RG 4096
DW 16.500 us
DE 6.00 us
TE 297.1 K
D1 0.50000000 se
d11 0.03000000 se
MCREST 0.00000000 se
MCWRK 0.01500000 se
-------- CHANNEL f1 ------
NUC1 13C
P1 5.10 us
PL1 -3.00 dB
SFO1 125.7713107 MH
-------- CHANNEL f2 ------
CPDPRG2 waltz16
NUC2 1H
PCPD2 100.00 us
PL2 -6.00 dB
PL12 15.50 dB
PL13 19.00 dB
SFO2 500.1320005 MH
F2 - Processing parameters
SI 32768
SF 125.7577933 MH
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40


13C NMR spectrum (CDCl3)
169.706
163.802
138.858
134.847
134.548
128.602
126.371
126.167
Ca
Cb
Cc
Cf
Cg
Ch
Ci
Cl
195 190 185 180 175 170 165 160 155 150 145 140 135 130 125 ppm
Current Data Parameters
NAME matsu061101
EXPNO 2
PROCNO 1
F2 - Acquisition Parameter
Date_ 20061102
Time 2.32
INSTRUM drx500
PROBHD 5 mm Multinucl
PULPROG zgpg30
TD 65536
SOLVENT CDCl3
NS 10000
DS 2
SWH 30303.031 Hz
FIDRES 0.462388 Hz
AQ 1.0813940 se
RG 4096
DW 16.500 us
DE 6.00 us
TE 297.1 K
D1 0.50000000 se
d11 0.03000000 se
MCREST 0.00000000 se
MCWRK 0.01500000 se
-------- CHANNEL f1 ------
NUC1 13C
P1 5.10 us
PL1 -3.00 dB
SFO1 125.7713107 MH
-------- CHANNEL f2 ------
CPDPRG2 waltz16
NUC2 1H
PCPD2 100.00 us
PL2 -6.00 dB
PL12 15.50 dB
PL13 19.00 dB
SFO2 500.1320005 MH
F2 - Processing parameters
SI 32768
SF 125.7577933 MH
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

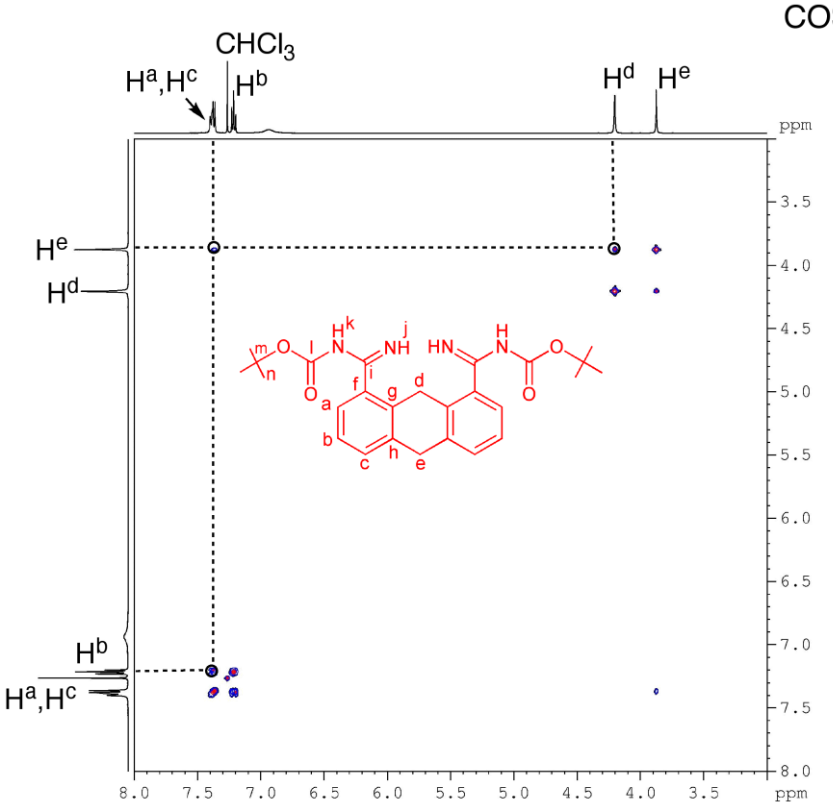



```
Current Data Parameters
NAME          matsu061101
EXPNO                   3
PROCNO                  1

F2 - Acquisition Parameter
Date_           20061102
Time                4.49
INSTRUM          drx500
PROBHD   5 mm Multinucl
PULPROG         dept135
TD                65536
SOLVENT           CDCl3
NS                 5000
DS                    2
SWH           30030.029 Hz
FIDRES        0.458222 Hz
AQ            1.0912244 se
RG                 8192
DW               16.650 us
DE                6.00 us
TE              297.0 K
CNST2        145.0000000
D1             0.50000000 se
d2             0.00344828 se
d12            0.00002000 se
DELTA          0.00000649 se
MCREST         0.00000000 se
MCWRK          0.01500000 se

======== CHANNEL f1 ======
NUC1                13C
P1                 5.10 us
p2                10.20 us
PL1               -3.00 dB
SFO1        125.7703643 MH

======== CHANNEL f2 ======
CPDPRG2         waltz16
NUC2                1H
P3                10.00 us
p4                20.00 us
PCPD2            100.00 us
PL2               -6.00 dB
PL12              15.50 dB
SFO2        500.1320005 MH

F2 - Processing parameters
SI                 32768
SF          125.7577890 MH
WDW                   EM
SSB                    0
LB                  1.00 Hz
GB                     0
PC                  1.40
```




```
Current Data Parameters
NAME          matsu061101
EXPNO                   3
PROCNO                  1

F2 - Acquisition Parameter
Date_           20061102
Time                4.49
INSTRUM          drx500
PROBHD   5 mm Multinucl
PULPROG         dept135
TD                65536
SOLVENT           CDCl3
NS                 5000
DS                    2
SWH           30030.029 Hz
FIDRES        0.458222 Hz
AQ            1.0912244 se
RG                 8192
DW               16.650 us
DE                6.00 us
TE              297.0 K
CNST2        145.0000000
D1             0.50000000 se
d2             0.00344828 se
d12            0.00002000 se
DELTA          0.00000649 se
MCREST         0.00000000 se
MCWRK          0.01500000 se

======== CHANNEL f1 ======
NUC1                13C
P1                 5.10 us
p2                10.20 us
PL1               -3.00 dB
SFO1        125.7703643 MH

======== CHANNEL f2 ======
CPDPRG2         waltz16
NUC2                1H
P3                10.00 us
p4                20.00 us
PCPD2            100.00 us
PL2               -6.00 dB
PL12              15.50 dB
SFO2        500.1320005 MH

F2 - Processing parameters
SI                 32768
SF          125.7577890 MH
WDW                   EM
SSB                    0
LB                  1.00 Hz
GB                     0
PC                  1.40
```

COSY spectrum ($CDCl_3$)



NAME matsu061101
EXPNO 4
PROCNO 1

F2 - Acquisition Parameters
Date_ 20061102
Time 4.50
INSTRUM drx500
PROBHD 5 mm Multinuc1
PULPROG cosygpqf
TD 2048
SOLVENT DMSO
NS 16
DS 8
SWH 4699.248 Hz
FIDRES 2.294555 Hz
AQ 0.2179572 sec
RG 322.5
DW 106.400 usec
DE 6.00 usec
TE 296.9 K
d0 0.00000300 sec
D1 1.35336196 sec
d13 0.00000400 sec
D16 0.00010000 sec
IN0 0.00021280 sec
MCREST 0.00000000 sec
MCWRK 1.35336196 sec

======== CHANNEL f1 ========
NUC1 1H
P0 10.00 usec
P1 10.00 usec
PL1 -6.00 dB
SFO1 500.1328279 MHz

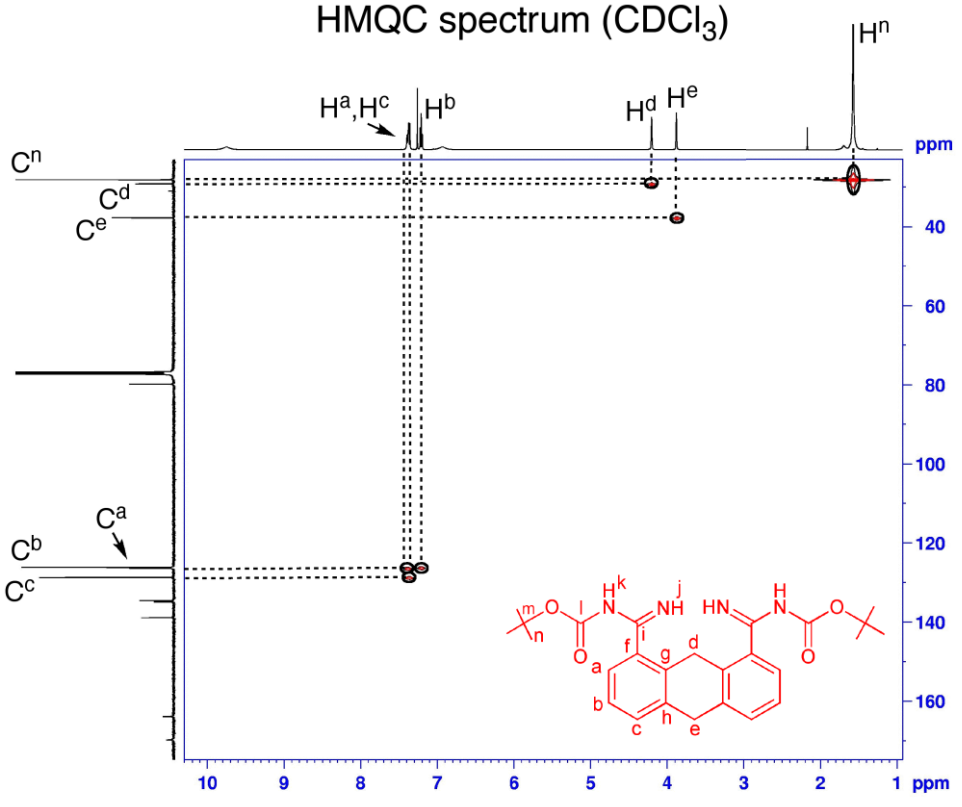
====== GRADIENT CHANNEL =====
GPNAM1 SINE.100
GPNAM2 SINE.100
GPX1 0.00 %
GPX2 0.00 %
GPY1 0.00 %
GPY2 0.00 %
GPZ1 10.00 %
GPZ2 10.00 %
P16 1000.00 usec

F1 - Acquisition parameters
ND0 1
TD 512
SFO1 500.1328 MHz
FIDRES 9.178219 Hz
SW 9.396 ppm
FnMODE QF

F2 - Processing parameters
SI 1024
SF 500.1300129 MHz
WDW SINE
SSB 0
LB 0.00 Hz
GB 0
PC 1.40

F1 - Processing parameters
SI 1024
MC2 QF
SF 500.1300129 MHz
WDW SINE
SSB 0
LB 0.00 Hz
GB 0

HMQC spectrum ($CDCl_3$)



Current Data Parameters
NAME matsu061101
EXPNO 5
PROCNO 1

F2 - Acquisition Parameters
Date_ 20061102
Time 21.52
INSTRUM drx500
PROBHD 5 mm Multinuc1
PULPROG hmqcgpqf
TD 1024
SOLVENT CDCl3
NS 16
DS 16
SWH 4699.248 Hz
FIDRES 4.589109 Hz
AQ 0.1090036 sec
RG 3649.1
DW 106.400 usec
DE 6.00 usec
TE 296.8 K
CNST2 145.0000000
d0 0.00000300 sec
D1 1.45781100 sec
d2 0.00344828 sec
d12 0.00002000 sec
d13 0.00000400 sec
D16 0.00010000 sec
DELTA1 0.00232428 sec
IN0 0.00002620 sec
MCREST 0.00000000 sec
MCWRK 1.45781100 sec

======== CHANNEL f1 ========
NUC1 1H
P1 10.00 usec
p2 20.00 usec
PL1 -6.00 dB
SFO1 500.1328279 MHz

======== CHANNEL f2 ========
CPDPRG2 waltz16
NUC2 13C
P3 5.10 usec
PCPD2 65.00 usec
PL2 -3.00 dB
PL12 15.50 dB
SFO2 125.7702099 MHz

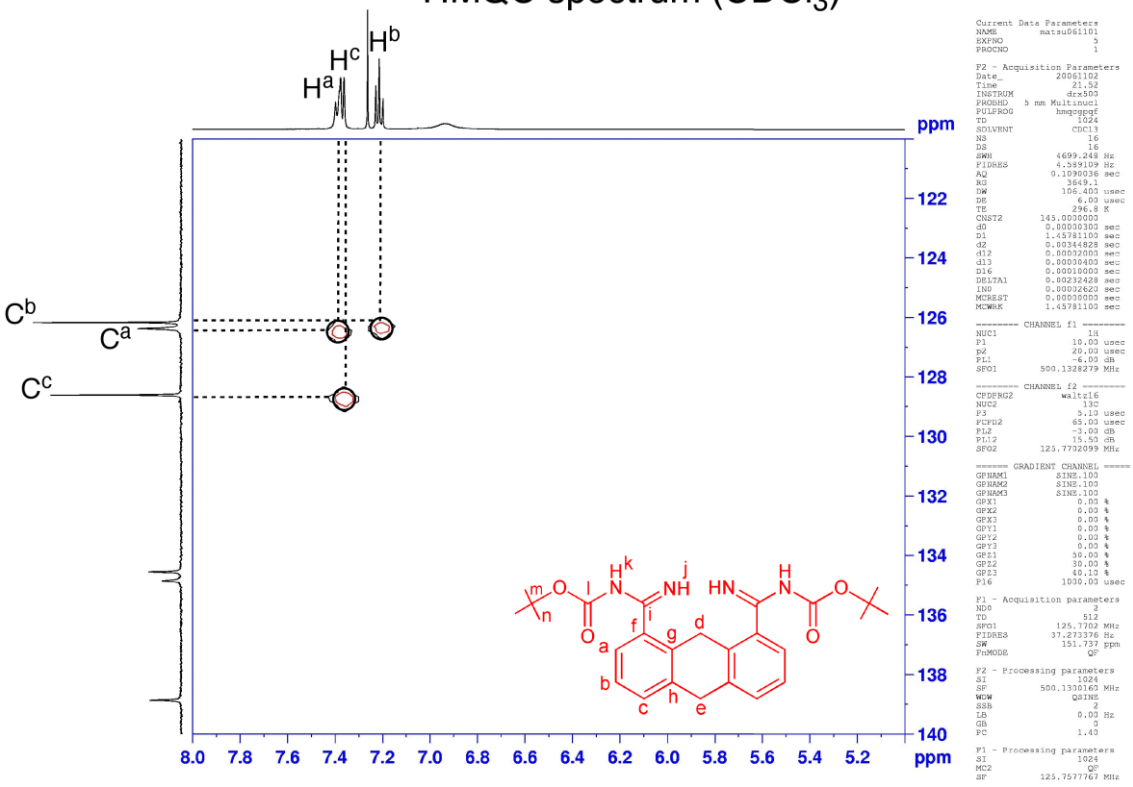
====== GRADIENT CHANNEL =====
GPNAM1 SINE.100
GPNAM2 SINE.100
GPNAM3 SINE.100
GPX1 0.00 %
GPX2 0.00 %
GPX3 0.00 %
GPY1 0.00 %
GPY2 0.00 %
GPY3 0.00 %
GPZ1 50.00 %
GPZ2 30.00 %
GPZ3 40.10 %
P16 1000.00 usec

F1 - Acquisition parameters
ND0 2
TD 512
SFO1 125.7702 MHz
FIDRES 37.273376 Hz
SW 151.737 ppm
FnMODE QF

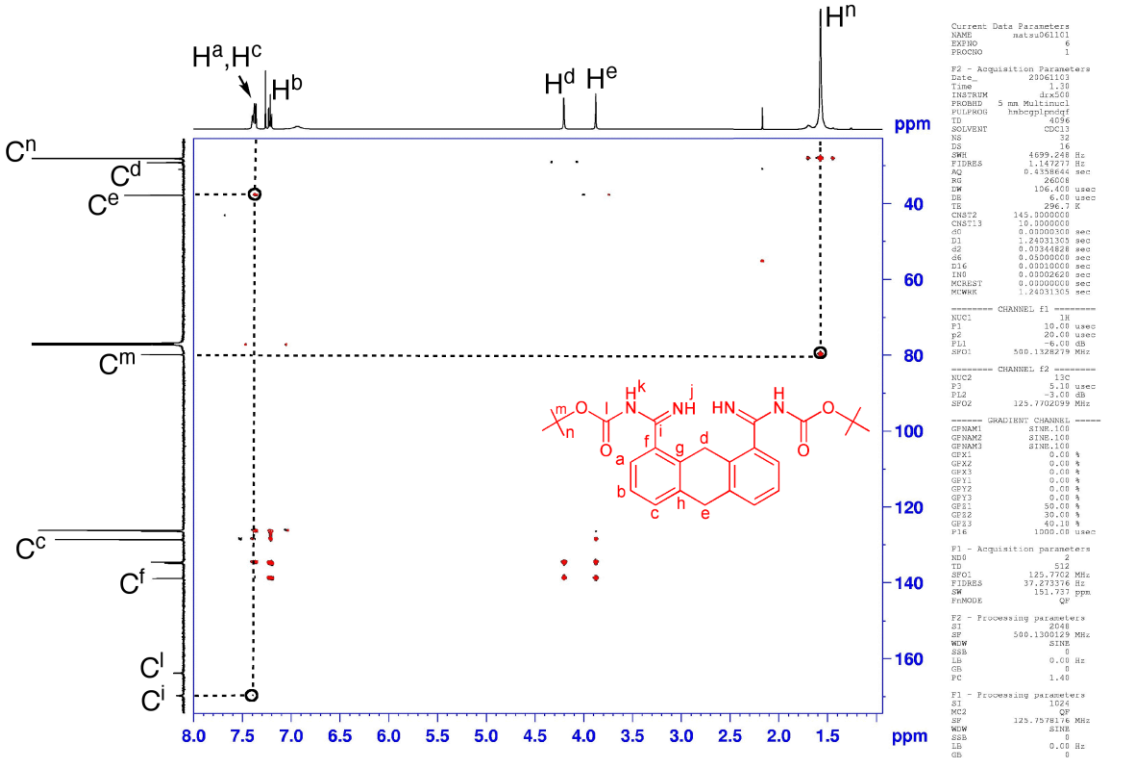
F2 - Processing parameters
SI 1024
SF 500.1300160 MHz
WDW QSINE
SSB 2
LB 0.00 Hz
GB 0
PC 1.40

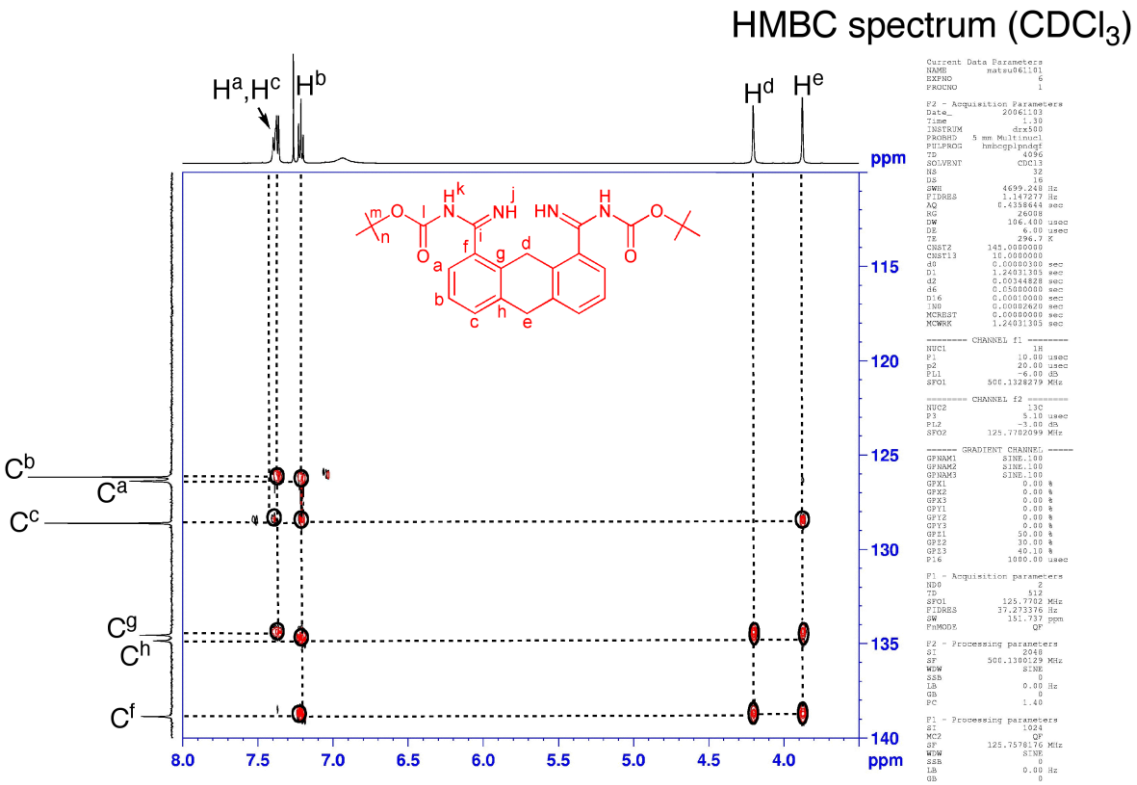
F1 - Processing parameters
SI 1024
MC2 QF
SF 125.7577767 MHz

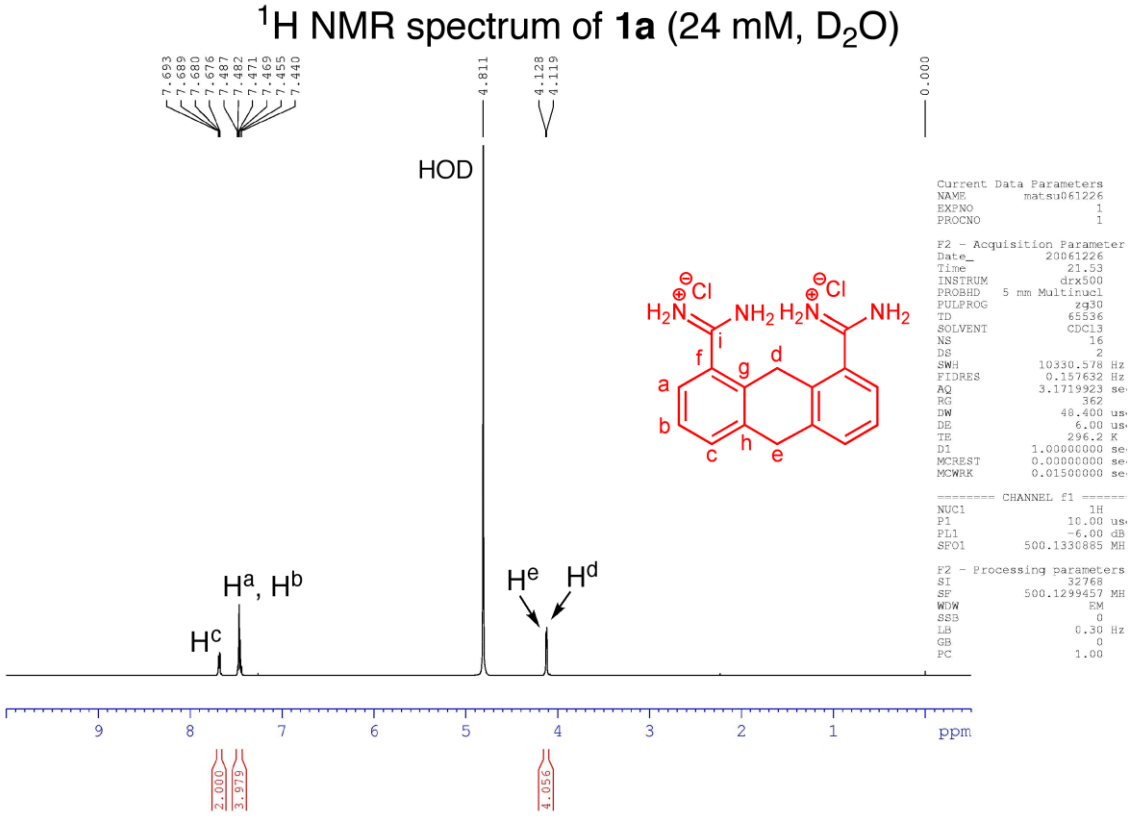
HMQC spectrum ($CDCl_3$)



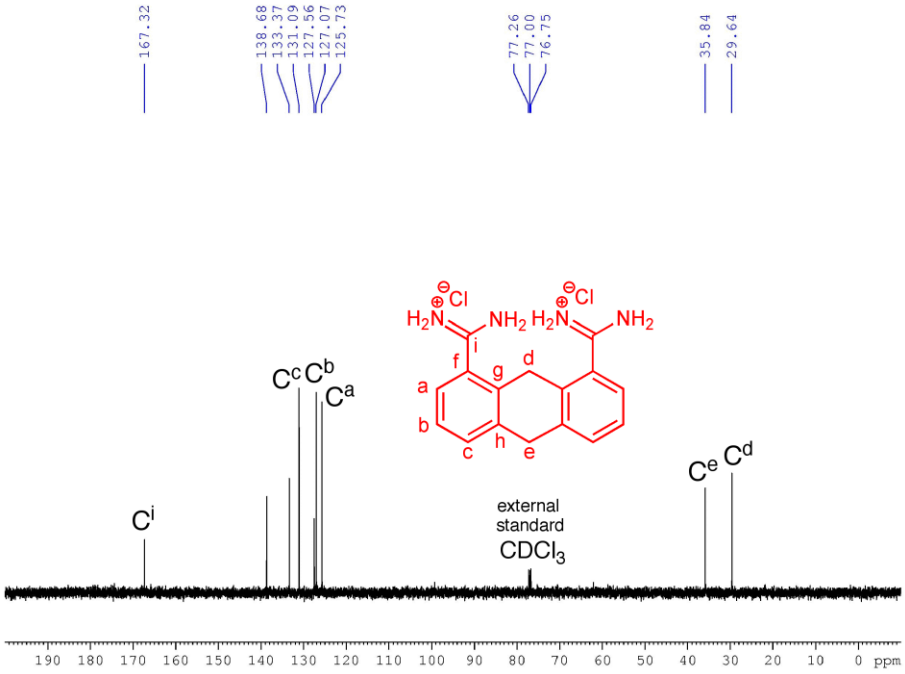
HMBC spectrum ($CDCl_3$)




HMBC spectrum ($CDCl_3$)
H^a,H^c
H^b
H^d
H^e
ppm
C^b
C^a
C^c
C^g
C^h
C^f
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
115
120
125
130
135
140

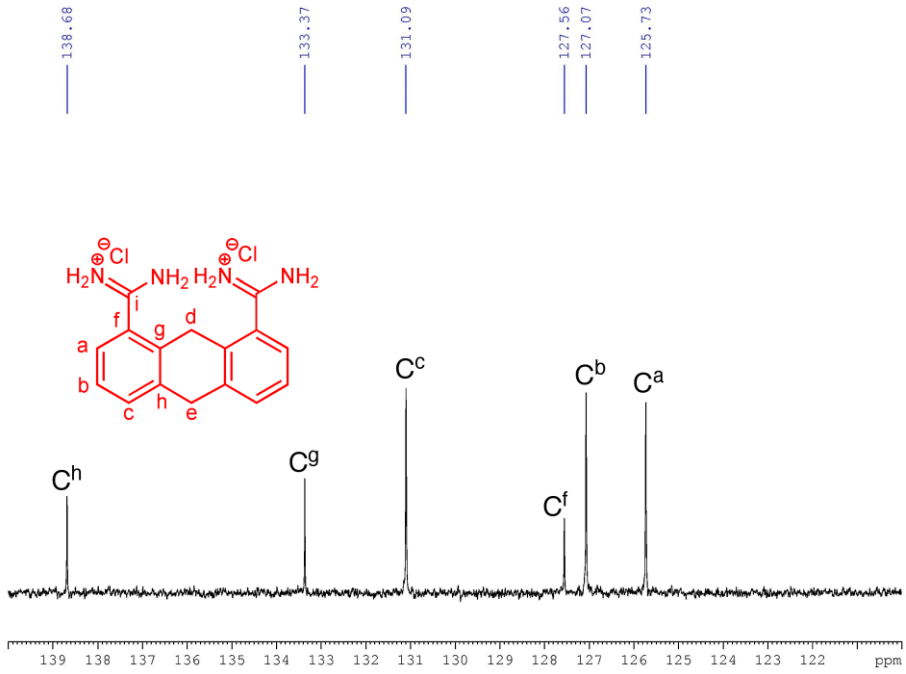

^{1}H NMR spectrum of **1a** (24 mM, D_2O)
7.693
7.689
7.680
7.676
7.487
7.482
7.471
7.469
7.455
7.440
4.811
4.128
4.119
0.000
HOD
H^a, H^b
H^c
H^e
H^d
9
8
7
6
5
4
3
2
1
ppm
2.000
3.979
4.056

^{13}C NMR spectrum of **1a** (24 mM, D_2O)



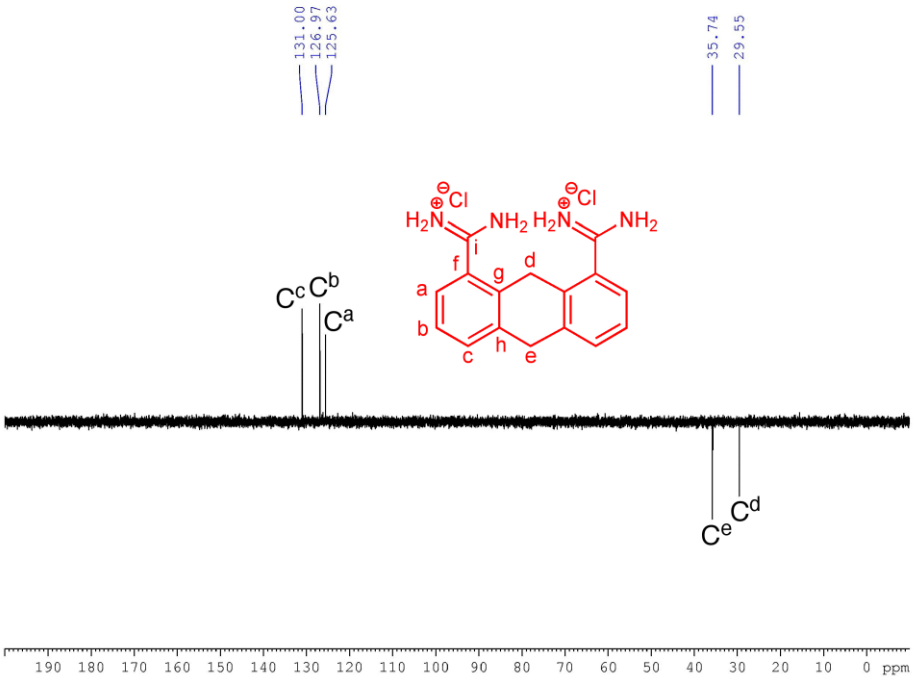

```
Current Data Parameters
NAME                 matsu061226
EXPNO                          2
PROCNO                         1

F2 - Acquisition Parameters
Date_               20061226
Time                   23.21
INSTRUM                drx500
PROBHD   5 mm Multinucl
PULPROG                zgpg30
TD                      65536
SOLVENT                 CDCl3
NS                       2000
DS                          2
SWH                27777.777 Hz
FIDRES              0.423855 Hz
AQ                  1.1796980 sec
RG                     1290.2
DW                     18.000 usec
DE                      6.00 usec
TE                     296.9 K
D1                  0.50000000 sec
d11                 0.03000000 sec
MCREST              0.00000000 sec
MCWRK               0.01500000 sec

======== CHANNEL f1 ========
NUC1                      13C
P1                       5.10 usec
PL1                     -3.00 dB
SFO1               125.7702099 MHz

======== CHANNEL f2 ========
CPDPRG2                waltz16
NUC2                       1H
PCPD2                  100.00 usec
PL2                     -6.00 dB
PL12                    15.50 dB
PL13                    19.00 dB
SFO2               500.1320005 MHz

F2 - Processing parameters
SI                         32768
SF                 125.7577775 MHz
WDW                           EM
SSB                            0
LB                          1.00 Hz
GB                             0
PC                          1.40
```

^{13}C NMR spectrum of **1a** (24 mM, D_2O)




```
Current Data Parameters
NAME                 matsu061226
EXPNO                          2
PROCNO                         1

F2 - Acquisition Parameters
Date_               20061226
Time                   23.21
INSTRUM                drx500
PROBHD   5 mm Multinucl
PULPROG                zgpg30
TD                      65536
SOLVENT                 CDCl3
NS                       2000
DS                          2
SWH                27777.777 Hz
FIDRES              0.423855 Hz
AQ                  1.1796980 sec
RG                     1290.2
DW                     18.000 usec
DE                      6.00 usec
TE                     296.9 K
D1                  0.50000000 sec
d11                 0.03000000 sec
MCREST              0.00000000 sec
MCWRK               0.01500000 sec

-------- CHANNEL f1 --------
NUC1                      13C
P1                       5.10 usec
PL1                     -3.00 dB
SFO1               125.7702099 MHz

-------- CHANNEL f2 --------
CPDPRG2                waltz16
NUC2                       1H
PCPD2                  100.00 usec
PL2                     -6.00 dB
PL12                    15.50 dB
PL13                    19.00 dB
SFO2               500.1320005 MHz

F2 - Processing parameters
SI                         32768
SF                 125.7577775 MHz
WDW                           EM
SSB                            0
LB                          1.00 Hz
GB                             0
PC                          1.40
```

DEPT spectrum of **1a** (24 mM, D_2O)



Current Data Parameters
NAME matsu061226
EXPNO 3
PROCNO 1

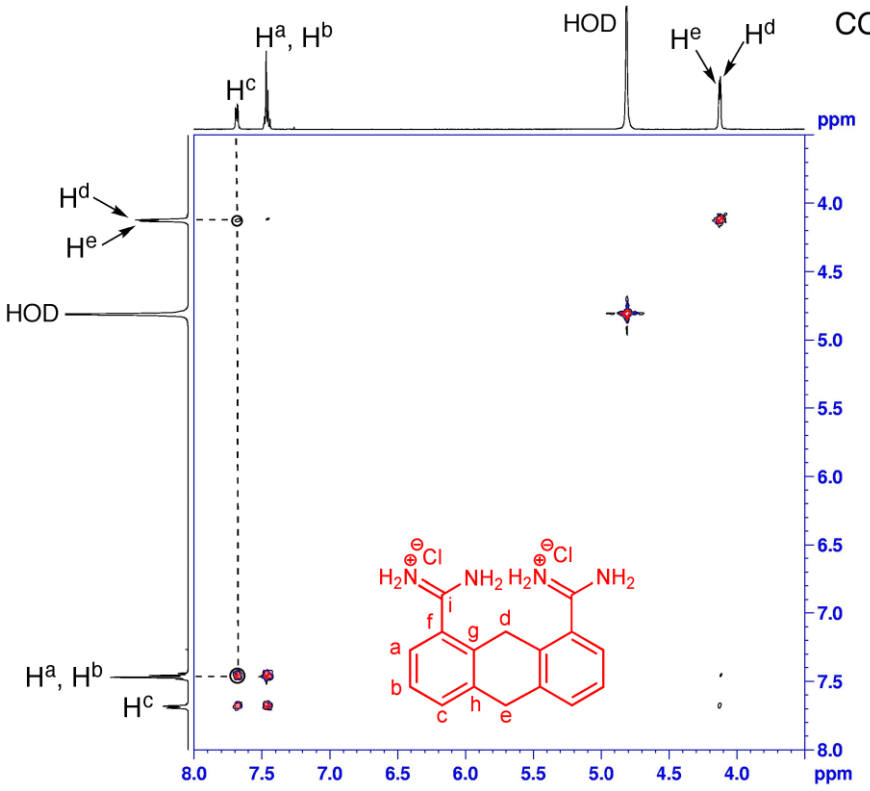
F2 - Acquisition Parameters
Date_ 20061226
Time 23.49
INSTRUM drx500
PROBHD 5 mm Multinucl
PULPROG dept135
TD 65536
SOLVENT CDCl3
NS 1000
DS 2
SWH 30030.029 Hz
FIDRES 0.458222 Hz
AQ 1.0912244 sec
RG 9195.2
DW 16.650 usec
DE 6.00 usec
TE 296.8 K
CNST2 145.0000000
D1 0.50000000 sec
d2 0.00344828 sec
d12 0.00002000 sec
DELTA 0.00000649 sec
MCREST 0.00000000 sec
MCWRK 0.01500000 sec

======== CHANNEL f1 ========
NUC1 13C
P1 5.10 usec
p2 10.20 usec
PL1 -3.00 dB
SFO1 125.7703643 MHz

======== CHANNEL f2 ========
CPDPRG2 waltz16
NUC2 1H
P3 10.00 usec
p4 20.00 usec
PCPD2 100.00 usec
PL2 -6.00 dB
PL12 15.50 dB
SFO2 500.1320005 MHz

F2 - Processing parameters
SI 32768
SF 125.7577890 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

COSY spectrum of **1a** (24 mM, D_2O)



Current Data Parameters
NAME matsu061226
EXPNO 4
PROCNO 1

F2 - Acquisition Parameters
Date_ 20061226
Time 23.49
INSTRUM drx500
PROBHD 5 mm Multinucl
PULPROG cosygpqf
TD 2048
SOLVENT D2O
NS 4
DS 8
SWH 4006.410 Hz
FIDRES 1.956255 Hz
AQ 0.2556404 sec
RG 2298.8
DW 124.800 usec
DE 6.00 usec
TE 296.7 K
d0 0.00000300 sec
D1 1.35336196 sec
d13 0.00000400 sec
D16 0.00010000 sec
IN0 0.00024960 sec
MCREST 0.00000000 sec
MCWRK 1.35336196 sec

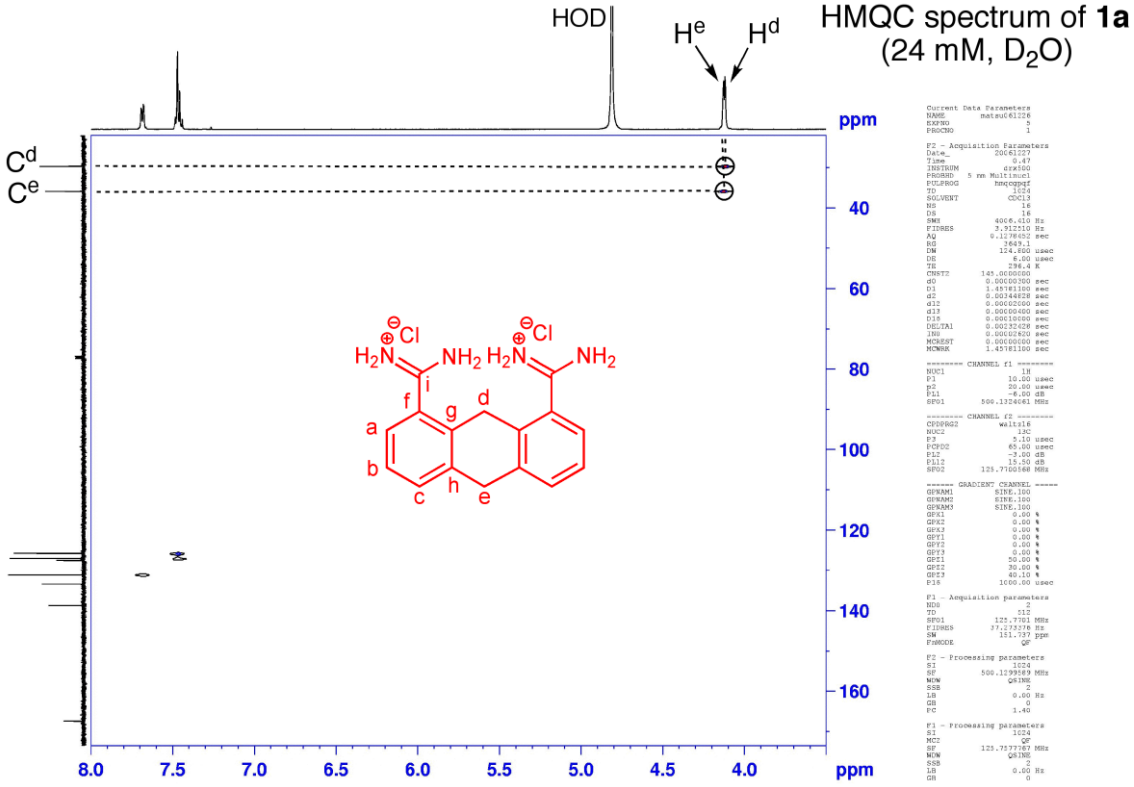
======== CHANNEL f1 ========
NUC1 1H
P0 10.00 usec
P1 10.00 usec
PL1 -6.00 dB
SFO1 500.1324061 MHz

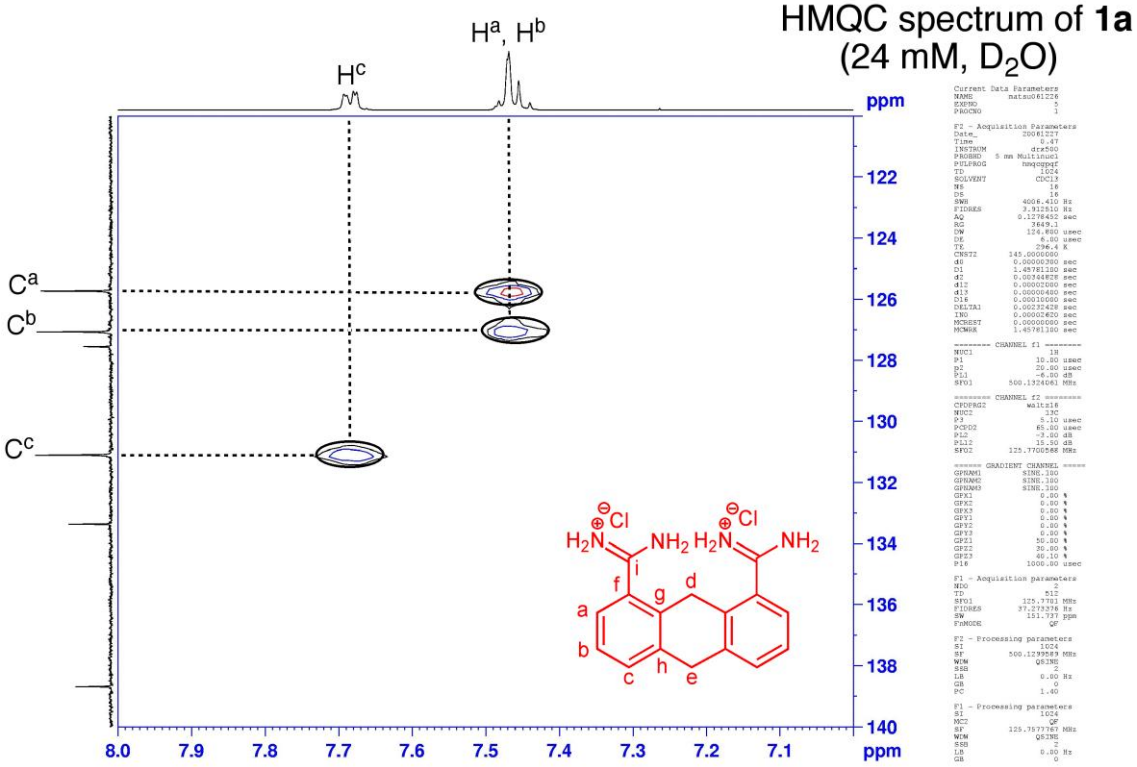
====== GRADIENT CHANNEL =====
GPNAM1 SINE.100
GPNAM2 SINE.100
GPX1 0.00 %
GPX2 0.00 %
GPY1 0.00 %
GPY2 0.00 %
GPZ1 10.00 %
GPZ2 10.00 %
P16 1000.00 usec

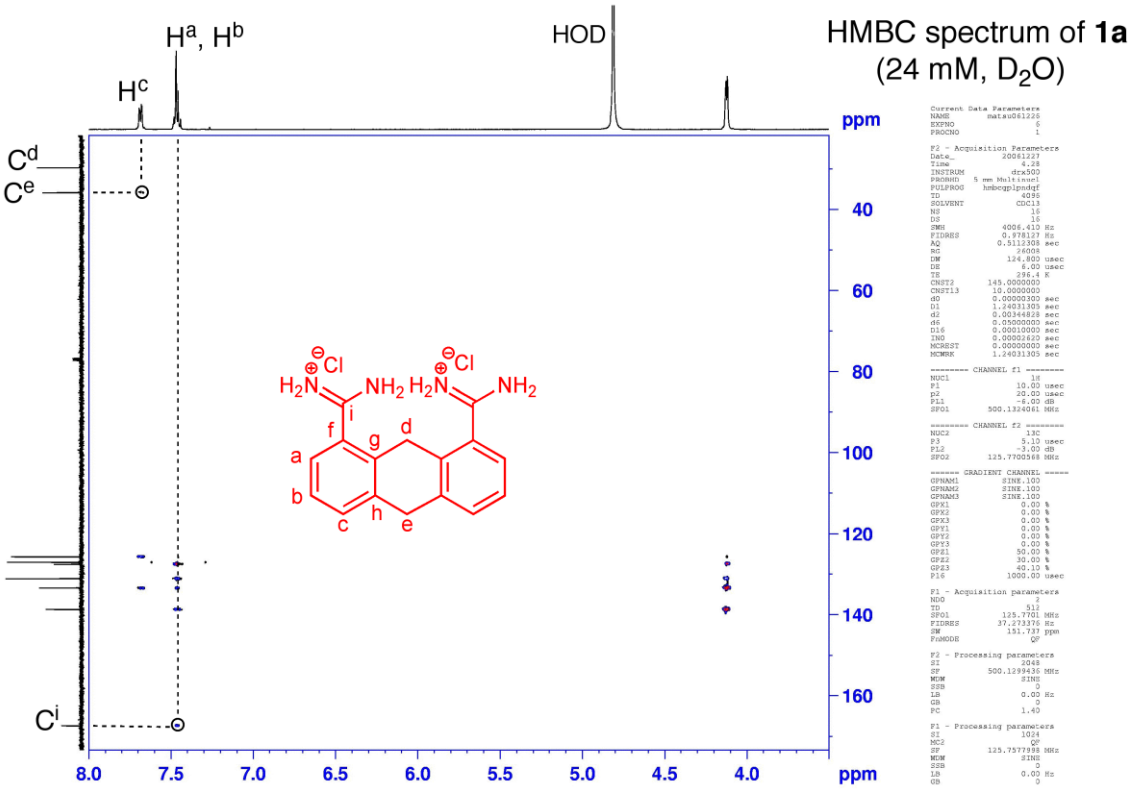
F1 - Acquisition parameters
ND0 1
TD 512
SFO1 500.1324 MHz
FIDRES 7.825020 Hz
SW 8.011 ppm
FnMODE QF

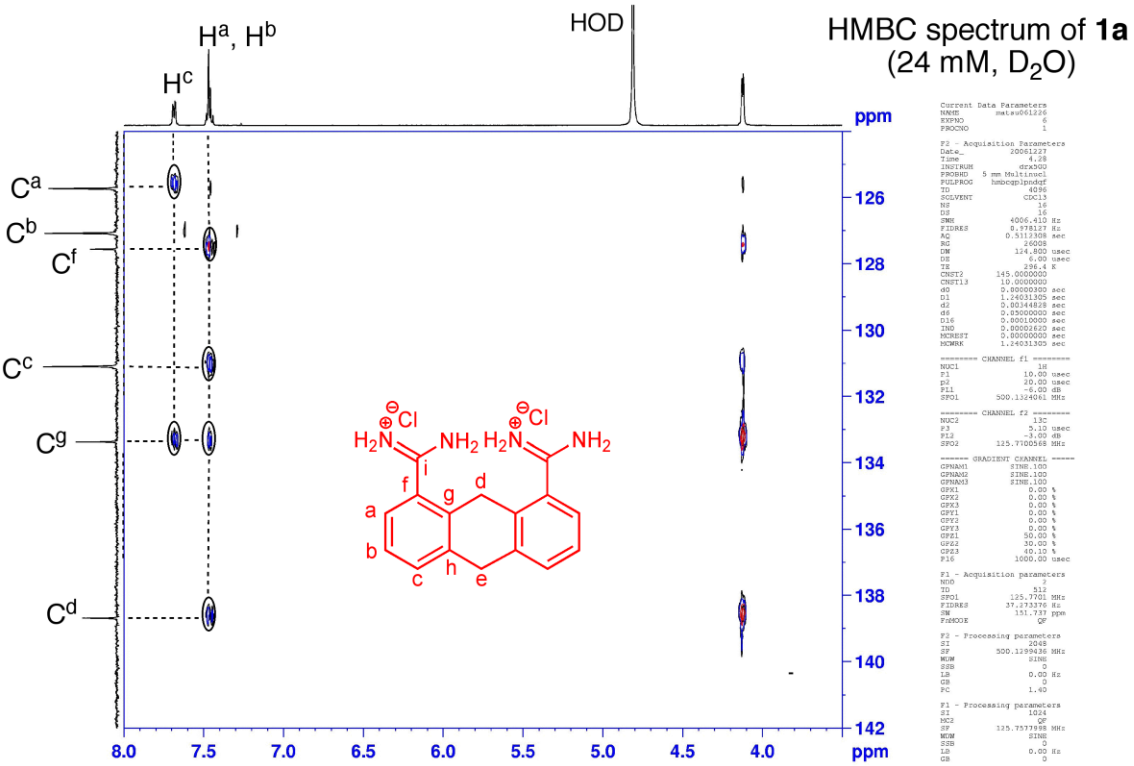
F2 - Processing parameters
SI 1024
SF 500.1299489 MHz
WDW SINE
SSB 0
LB 0.00 Hz
GB 0
PC 1.40

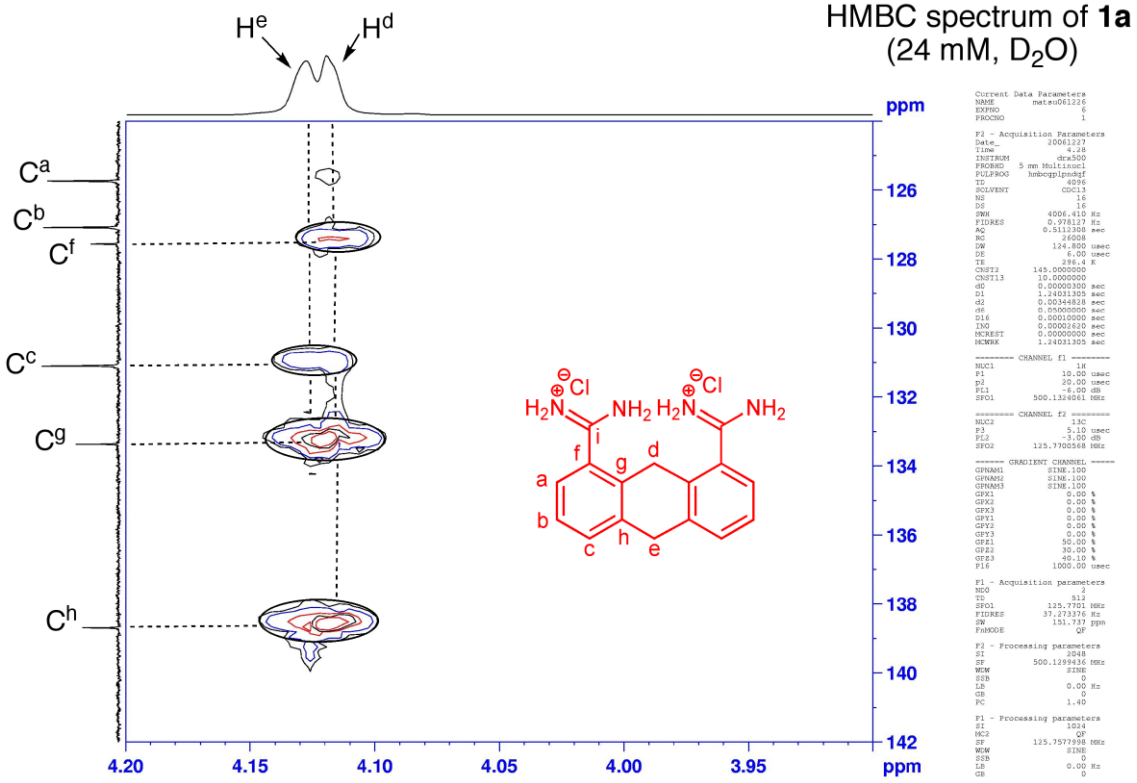
F1 - Processing parameters
SI 1024
MC2 QF
SF 500.1299489 MHz
WDW SINE
SSB 0
LB 0.00 Hz
GB 0

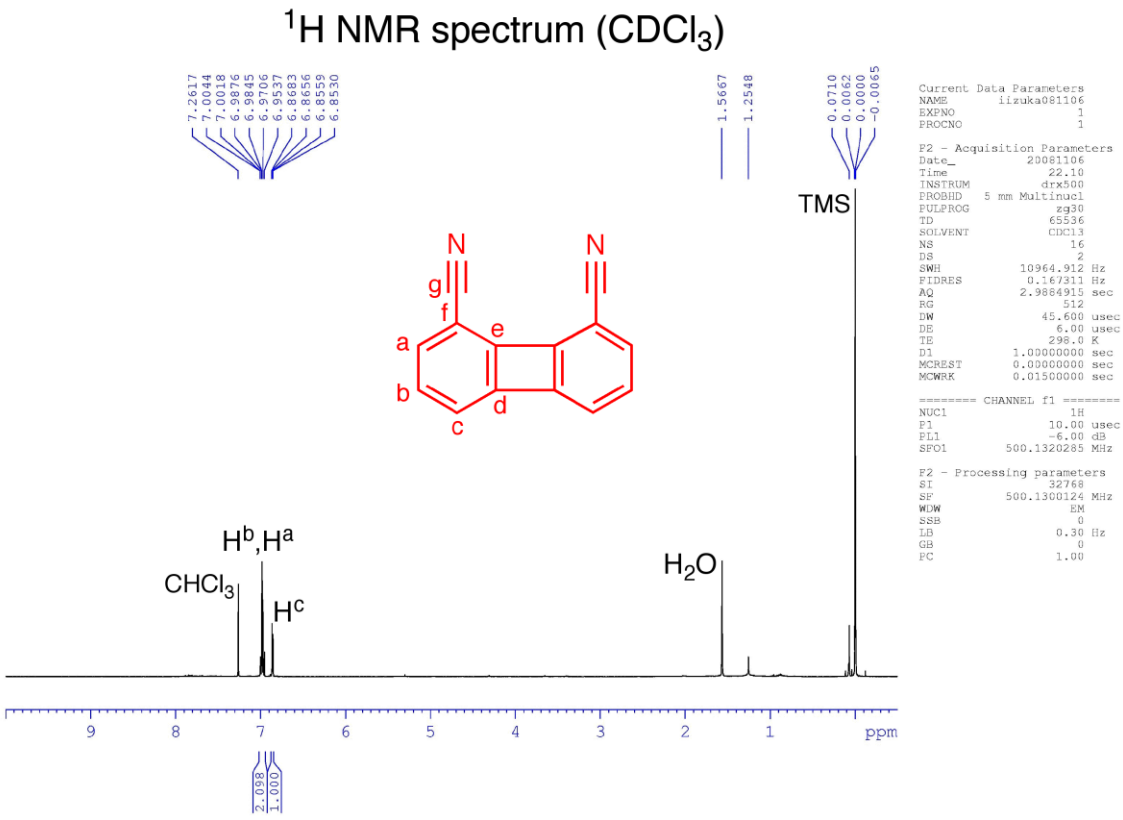

HMQC spectrum of 1a
(24 mM, D2O)
HOD
He
Hd
Cd
Ce
ppm
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
40
60
80
100
120
140
160


HMQC spectrum of 1a
(24 mM, D2O)
Ha, Hb
Hc
Ca
Cb
Cc
ppm
8.0
7.9
7.8
7.7
7.6
7.5
7.4
7.3
7.2
7.1
122
124
126
128
130
132
134
136
138
140

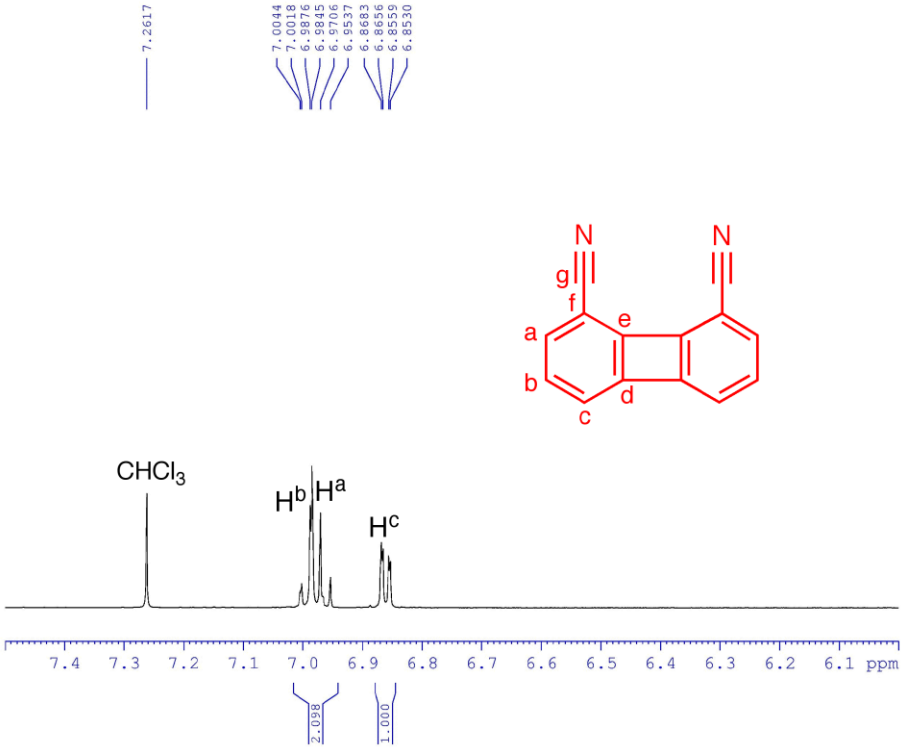





HMBC spectrum of 1a
(24 mM, D2O)
He
Hd
Ca
Cb
Cf
Cc
Cg
Ch
ppm
126
128
130
132
134
136
138
140
142
4.20
4.15
4.10
4.05
4.00
3.95
ppm
Cl
NH2
a
b
c
d
e
f
g
h
i

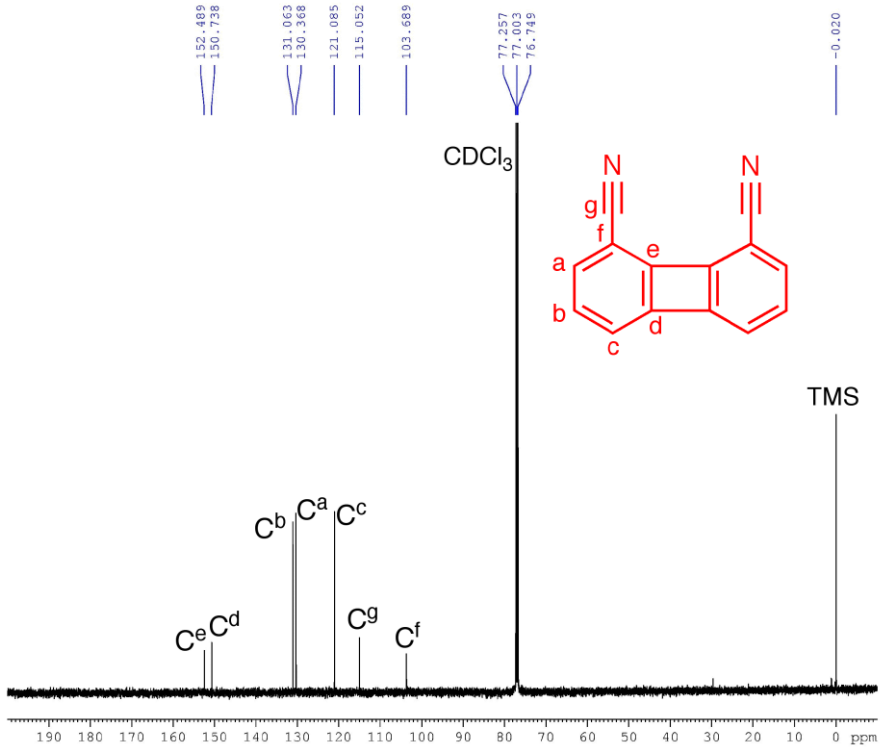

1H NMR spectrum (CDCl3)
7.2617
7.0044
7.0018
6.9876
6.9845
6.9706
6.9537
6.8683
6.8656
6.8559
6.8530
1.5667
1.2548
0.0710
0.0062
0.0000
-0.0065
TMS
N
N
a
b
c
d
e
f
g
Hb,Ha
CHCl3
Hc
H2O
9
8
7
6
5
4
3
2
1
ppm
2.098
1.000

^{1}H NMR spectrum ($CDCl_3$)



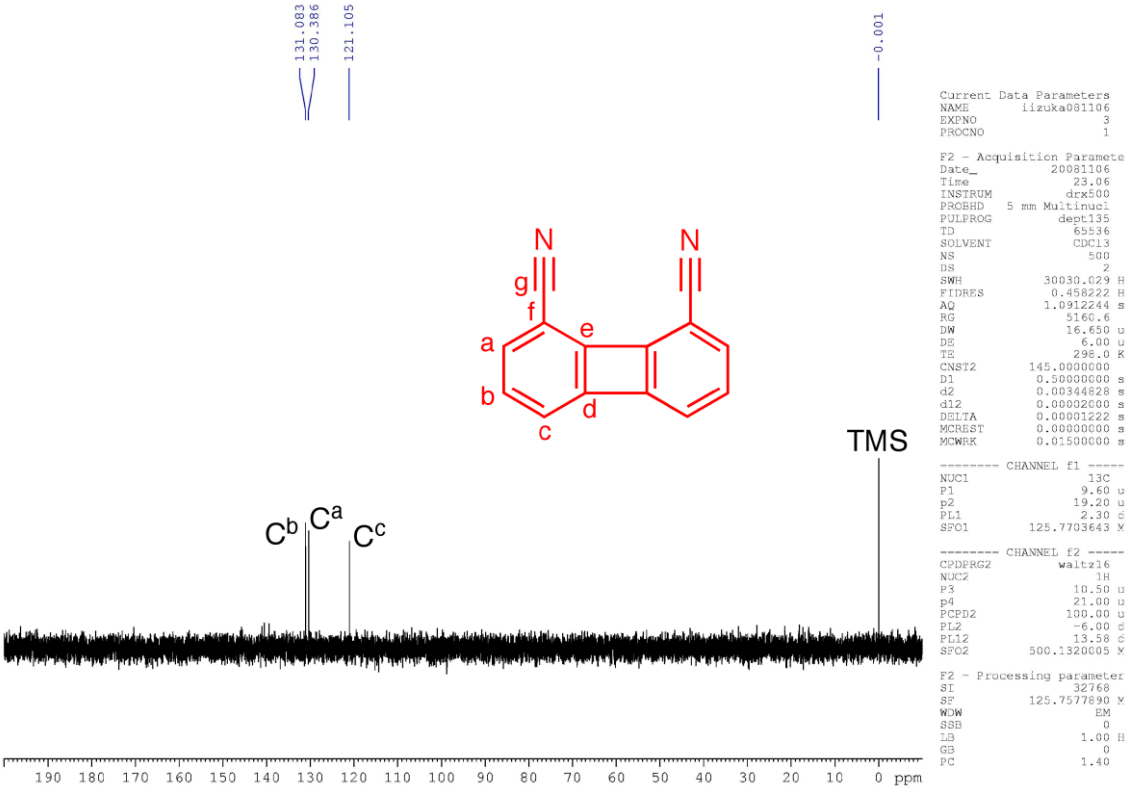

```
Current Data Parameters
NAME         iizuka081106
EXPNO                   1
PROCNO                  1

F2 - Acquisition Parameters
Date_          20081106
Time             22.10
INSTRUM         drx500
PROBHD   5 mm Multinucl
PULPROG           zg30
TD               65536
SOLVENT          CDCl3
NS                  16
DS                   2
SWH          10964.912 Hz
FIDRES        0.167311 Hz
AQ            2.9884915 sec
RG                 512
DW              45.600 usec
DE               6.00 usec
TE              298.0 K
D1           1.00000000 sec
MCREST       0.00000000 sec
MCWRK        0.01500000 sec

======== CHANNEL f1 ========
NUC1                 1H
P1                10.00 usec
PL1               -6.00 dB
SFO1        500.1320285 MHz

F2 - Processing parameters
SI                  32768
SF          500.1300124 MHz
WDW                    EM
SSB                     0
LB                 0.30 Hz
GB                      0
PC                   1.00
```

^{13}C NMR spectrum ($CDCl_3$)




```
Current Data Parameters
NAME         iizuka081106
EXPNO                   2
PROCNO                  1

F2 - Acquisition Paramete
Date_          20081106
Time             22.51
INSTRUM         drx500
PROBHD   5 mm Multinucl
PULPROG         zgpg30
TD               65536
SOLVENT          CDCl3
NS                1000
DS                   2
SWH          27777.777 H
FIDRES        0.423855 H
AQ            1.1796980 s
RG               5160.6
DW              18.000 u
DE               6.00 u
TE              298.0 K
D1           0.50000000 s
d11          0.03000000 s
MCREST       0.00000000 s
MCWRK        0.01500000 s

-------- CHANNEL f1 -----
NUC1                13C
P1                9.60 u
PL1               2.30 d
SFO1        125.7703858 M

-------- CHANNEL f2 -----
CPDPRG2         waltz16
NUC2                1H
PCPD2           100.00 u
PL2              -6.00 d
PL12             13.58 d
PL13             16.50 d
SFO2        500.1320005 M

F2 - Processing parameter
SI                  32768
SF          125.7577915 M
WDW                    EM
SSB                     0
LB                 1.00 H
GB                      0
PC                   1.40
```

DEPT spectrum ($CDCl_3$)



Current Data Parameters
NAME lizuka081106
EXPNO 3
PROCNO 1

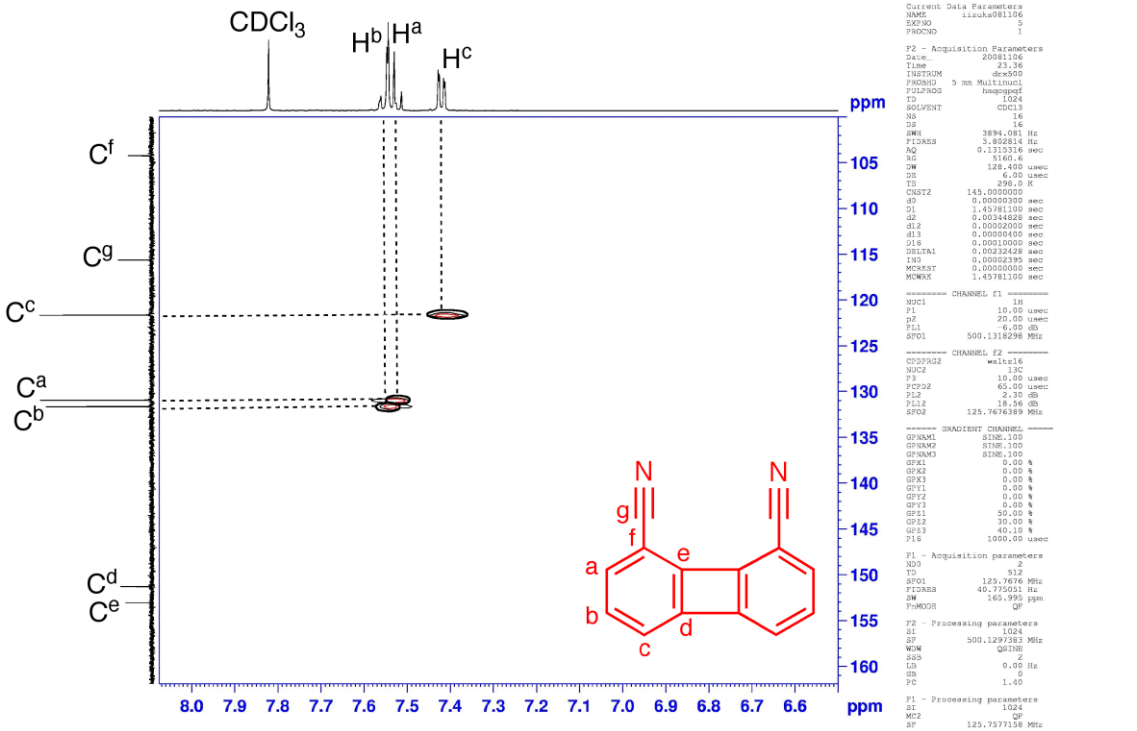
F2 - Acquisition Parameters
Date_ 20081106
Time 23.06
INSTRUM drx500
PROBHD 5 mm Multinucl
PULPROG dept135
TD 65536
SOLVENT CDCl3
NS 500
DS 2
SWH 30030.029 Hz
FIDRES 0.458222 Hz
AQ 1.0912244 sec
RG 5160.6
DW 16.650 usec
DE 6.00 usec
TE 298.0 K
CNST2 145.0000000
D1 0.50000000 sec
d2 0.00344828 sec
d12 0.00002000 sec
DELTA 0.00001222 sec
MCREST 0.00000000 sec
MCWRK 0.01500000 sec

-------- CHANNEL f1 -----
NUC1 13C
P1 9.60 usec
p2 19.20 usec
PL1 2.30 dB
SFO1 125.7703643 MHz

-------- CHANNEL f2 -----
CPDPRG2 waltz16
NUC2 1H
P3 10.50 usec
p4 21.00 usec
PCPD2 100.00 usec
PL2 -6.00 dB
PL12 13.58 dB
SFO2 500.1320005 MHz

F2 - Processing parameter
SI 32768
SF 125.7577890 MHz
WDW EM
SSB 0
LB 1.00 Hz
GB 0
PC 1.40

HMQC spectrum ($CDCl_3$)



Current Data Parameters
NAME lizuka081106
EXPNO 5
PROCNO 1

F2 - Acquisition Parameters
Date_ 20081106
Time 23.36
INSTRUM drx500
PROBHD 5 mm Multinucl
PULPROG hsqcgpqf
TD 1024
SOLVENT CDCl3
NS 16
DS 16
SWH 3894.081 Hz
FIDRES 3.802814 Hz
AQ 0.1315316 sec
RG 5160.6
DW 128.400 usec
DE 6.00 usec
TE 298.0 K
CNST2 145.0000000
d0 0.00000300 sec
D1 1.45781100 sec
d2 0.00344828 sec
d12 0.00002000 sec
d13 0.00000400 sec
D16 0.00010000 sec
DELTA1 0.00232428 sec
IN0 0.00002395 sec
MCREST 0.00000000 sec
MCWRK 1.45781100 sec

======== CHANNEL f1 ========
NUC1 1H
P1 10.00 usec
p2 20.00 usec
PL1 6.00 dB
SFO1 500.1318296 MHz

======== CHANNEL f2 ========
CPDPRG2 waltz16
NUC2 13C
P3 10.00 usec
PCPD2 65.00 usec
PL2 2.30 dB
PL12 18.56 dB
SFO2 125.7676389 MHz

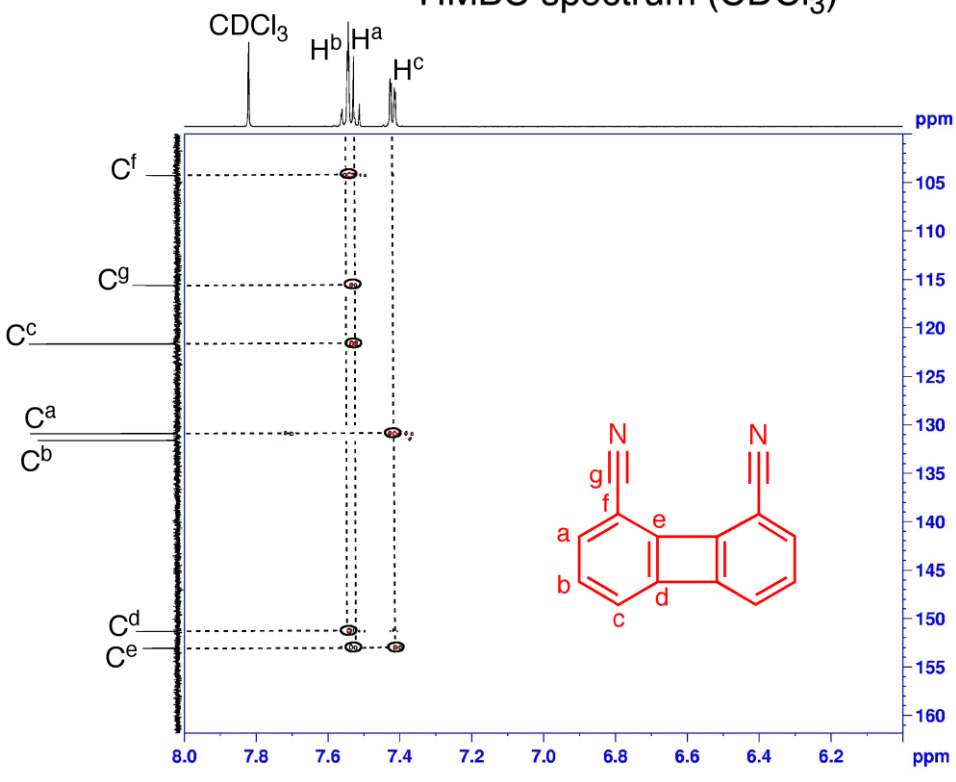
====== GRADIENT CHANNEL =====
GPNAM1 SINE.100
GPNAM2 SINE.100
GPNAM3 SINE.100
GPZ1 0.00 %
GPZ2 0.00 %
GPZ3 0.00 %
GPY1 0.00 %
GPY2 0.00 %
GPY3 0.00 %
GPZ1 50.00 %
GPZ2 30.00 %
GPZ3 40.10 %
P16 1000.00 usec

F1 - Acquisition parameters
ND0 2
TD 512
SFO1 125.7676 MHz
FIDRES 40.775051 Hz
SW 165.995 ppm
FnMODE QF

F2 - Processing parameters
SI 1024
SF 500.1297383 MHz
WDW QSINE
SSB 2
LB 0.00 Hz
GB 0
PC 1.40

F1 - Processing parameters
SI 1024
MC2 QF
SF 125.7577158 MHz

HMBC spectrum ($CDCl_3$)



Current Data Parameters
NAME izuka081106
EXPNO 6
PROCNO 1

F2 - Acquisition Parameters
Date_ 20081107
Time 3.17
INSTRUM drx500
PROBHD 5 mm Multinucl
PULPROG hmbcgplpndqf
TD 4096
SOLVENT CDCl3
NS 20
DS 16
SWH 3894.081 Hz
FIDRES 0.950703 Hz
AQ 0.5259764 sec
RG 20642.5
DW 128.400 usec
DE 6.00 usec
TE 298.0 K
CNST2 145.0000000
CNST13 10.0000000
d0 0.00000300 sec
D1 1.24031305 sec
d2 0.00344828 sec
d6 0.03000000 sec
D16 0.00010000 sec
IN0 0.00002395 sec
MCREST 0.00000000 sec
MCWRK 1.24031305 sec

======== CHANNEL f1 ========
NUC1 1H
P1 10.00 usec
p2 20.00 usec
PL1 -6.00 dB
SFO1 500.1318298 MHz

======== CHANNEL f2 ========
NUC2 13C
P3 10.80 usec
PL2 2.30 dB
SFO2 125.7676389 MHz

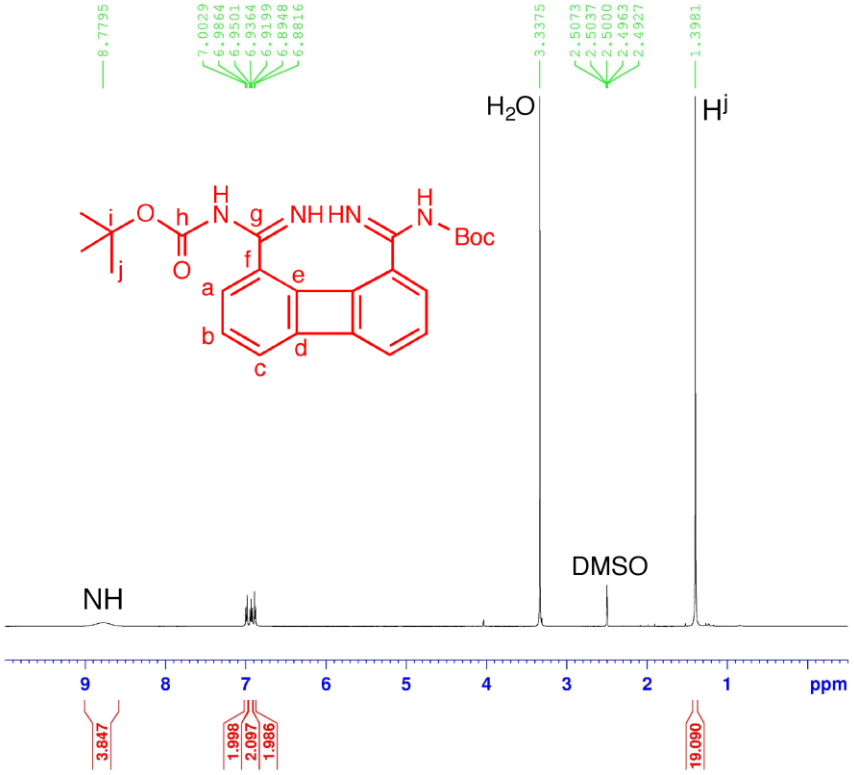
------ GRADIENT CHANNEL -----
GPNAM1 SINE.100
GPNAM2 SINE.100
GPNAM3 SINE.100
GPX1 0.00 %
GPX2 0.00 %
GPX3 0.00 %
GPY1 0.00 %
GPY2 0.00 %
GPY3 0.00 %
GPZ1 50.00 %
GPZ2 30.00 %
GPZ3 40.10 %
P16 1000.00 usec

F1 - Acquisition parameters
ND0 2
TD 512
SFO1 125.7676 MHz
FIDRES 40.779031 Hz
SW 165.995 ppm
FnMODE QF

F2 - Processing parameters
SI 2048
SF 500.1297362 MHz
WDW SINE
SSB 0
LB 0.00 Hz
GB 0
PC 1.40

F1 - Processing parameters
SI 1024
MC2 QF
SF 125.7577275 MHz
WDW SINE
SSB 0
LB 0.00 Hz
GB 0

^{1}H NMR spectrum (DMSO-d_6)



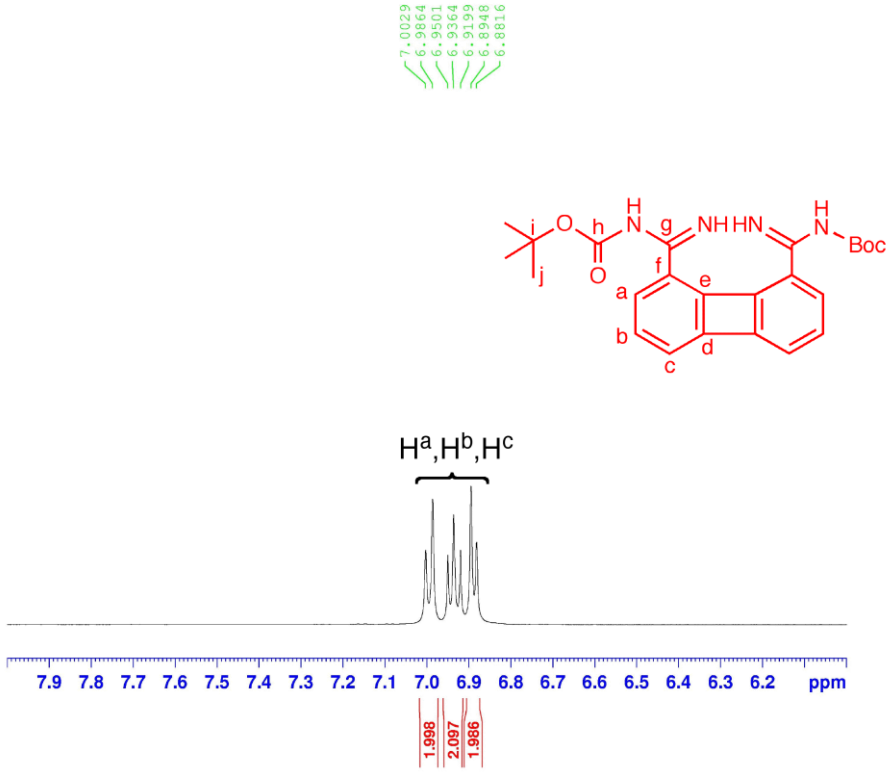
Current Data Parameters
NAME 56J-229
EXPNO 2
PROCNO 1

F2 - Acquisition Parameters
Date_ 20110127
Time 17.40
INSTRUM spect
PROBHD 5 mm PABBO BB-
PULPROG zg30
TD 65536
SOLVENT DMSO
NS 16
DS 2
SWH 10330.578 Hz
FIDRES 0.157632 Hz
AQ 3.1719923 sec
RG 76.73
DW 48.400 usec
DE 6.50 usec
TE 298.2 K
D1 1.00000000 sec

======== CHANNEL f1 ========
NUC1 1H
P1 11.00 usec
PLW1 20.51199913 W
SFO1 500.1330885 MHz

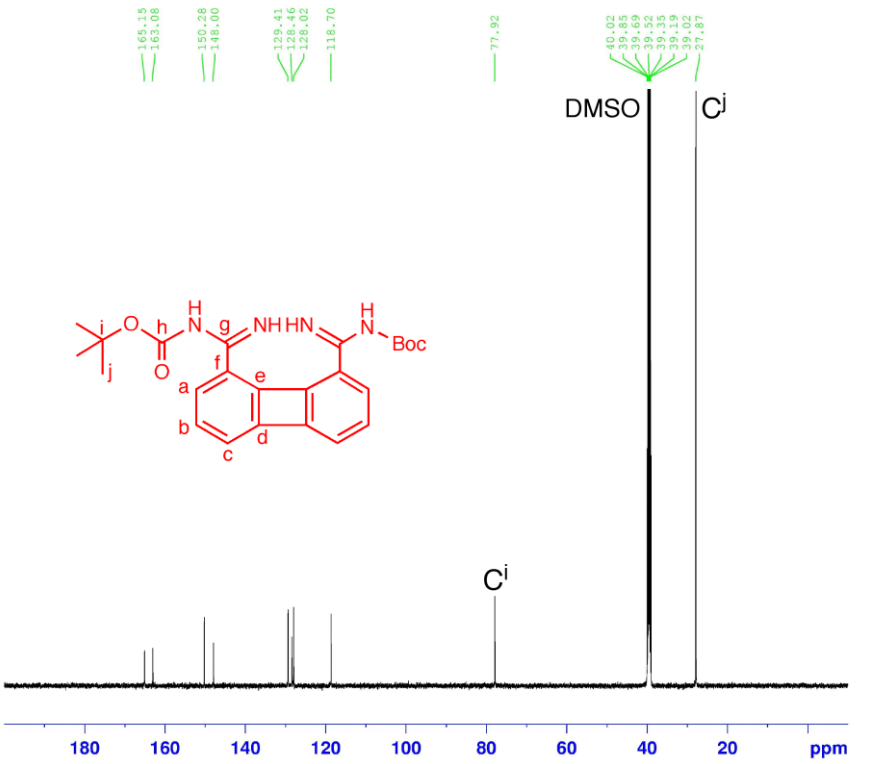
F2 - Processing parameters
SI 65536
SF 500.1300045 MHz
WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 1.00

^{1}H NMR spectrum (DMSO-d_6)



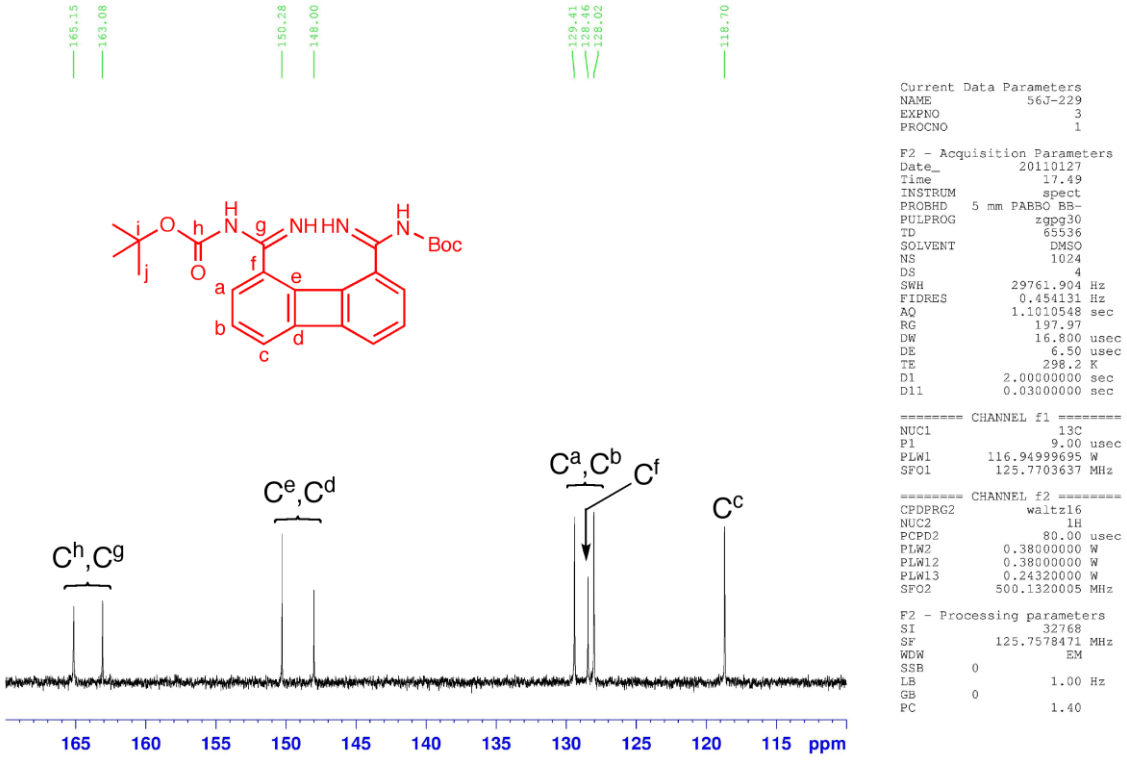

```
Current Data Parameters
NAME                  56J-229
EXPNO                       2
PROCNO                      1

F2 - Acquisition Parameters
Date_              20110127
Time                  17.40
INSTRUM               spect
PROBHD    5 mm PABBO BB-
PULPROG                zg30
TD                    65536
SOLVENT                DMSO
NS                       16
DS                        2
SWH               10330.578 Hz
FIDRES             0.157632 Hz
AQ                 3.1719923 sec
RG                     76.73
DW                    48.400 usec
DE                     6.50 usec
TE                    298.2 K
D1                 1.00000000 sec

======== CHANNEL f1 ========
NUC1                      1H
P1                     11.00 usec
PLW1             20.51199913 W
SFO1            500.1330885 MHz

F2 - Processing parameters
SI                    65536
SF               500.1300045 MHz
WDW                      EM
SSB          0
LB                     0.30 Hz
GB           0
PC                     1.00
```

^{13}C NMR spectrum (DMSO-d_6)



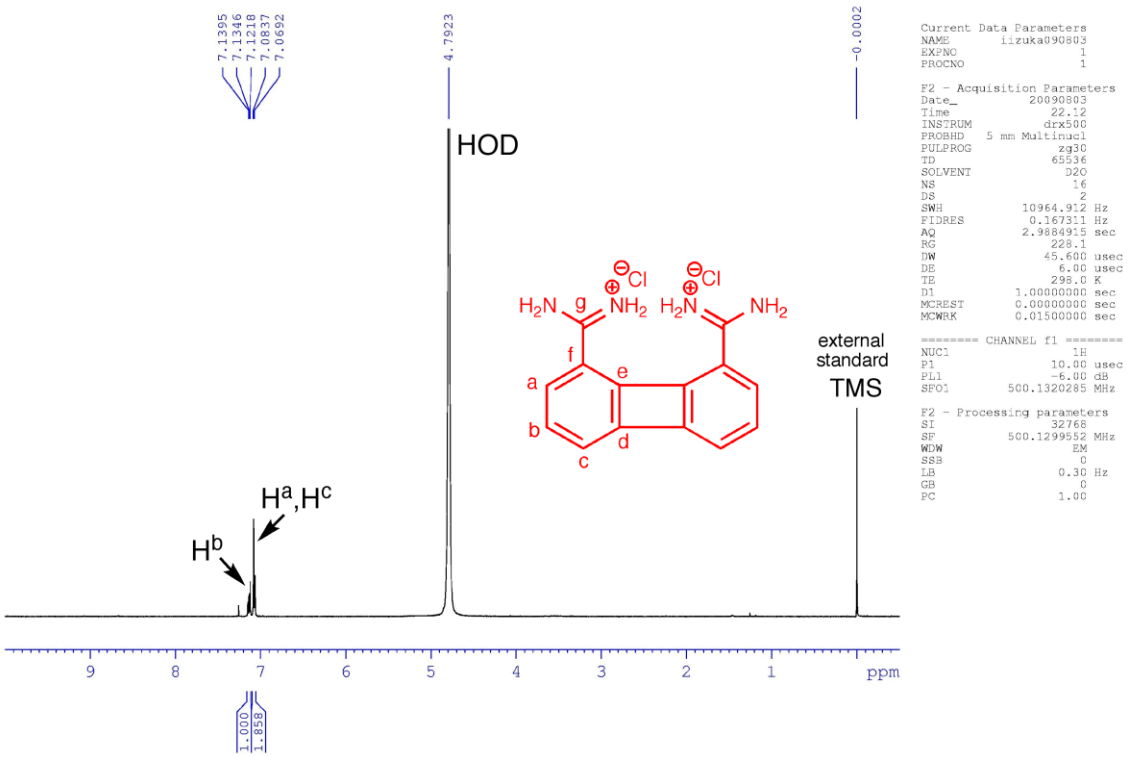

```
Current Data Parameters
NAME                  56J-229
EXPNO                       3
PROCNO                      1

F2 - Acquisition Parameters
Date_              20110127
Time                  17.49
INSTRUM               spect
PROBHD    5 mm PABBO BB-
PULPROG             zgpg30
TD                    65536
SOLVENT                DMSO
NS                      1024
DS                        4
SWH               29761.904 Hz
FIDRES             0.454131 Hz
AQ                 1.1010548 sec
RG                    197.97
DW                    16.800 usec
DE                     6.50 usec
TE                    298.2 K
D1                 2.00000000 sec
D11                0.03000000 sec

======== CHANNEL f1 ========
NUC1                      13C
P1                     9.00 usec
PLW1            116.94999695 W
SFO1             125.7703637 MHz

======== CHANNEL f2 ========
CPDPRG2              waltz16
NUC2                      1H
PCPD2                  80.00 usec
PLW2             0.38000000 W
PLW12            0.38000000 W
PLW13            0.24320000 W
SFO2            500.1320005 MHz

F2 - Processing parameters
SI                    32768
SF               125.7578471 MHz
WDW                      EM
SSB          0
LB                     1.00 Hz
GB           0
PC                     1.40
```

^{13}C NMR spectrum (DMSO-d_6)



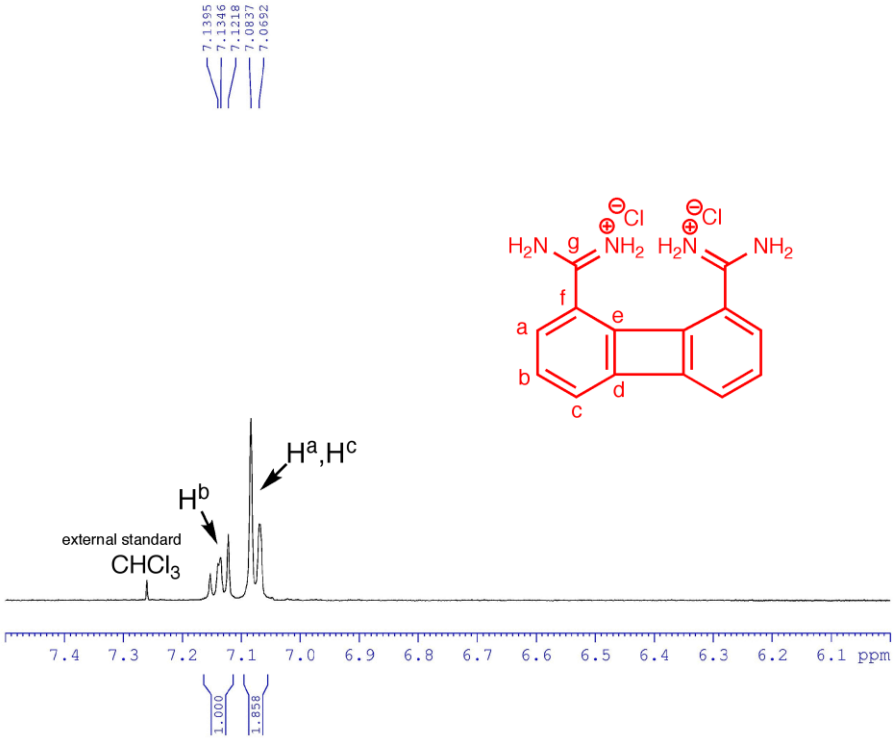

```
Current Data Parameters
NAME                  56J-229
EXPNO                       3
PROCNO                      1

F2 - Acquisition Parameters
Date_                20110127
Time                    17.49
INSTRUM                 spect
PROBHD    5 mm PABBO BB-
PULPROG                zgpg30
TD                      65536
SOLVENT                  DMSO
NS                       1024
DS                          4
SWH                29761.904 Hz
FIDRES             0.454131 Hz
AQ                 1.1010548 sec
RG                     197.97
DW                   16.800 usec
DE                    6.50 usec
TE                    298.2 K
D1                2.00000000 sec
D11               0.03000000 sec

======== CHANNEL f1 ========
NUC1                      13C
P1                      9.00 usec
PLW1           116.94999695 W
SFO1           125.7703637 MHz

======== CHANNEL f2 ========
CPDPRG2                waltz16
NUC2                       1H
PCPD2                  80.00 usec
PLW2              0.38000000 W
PLW12             0.38000000 W
PLW13             0.24320000 W
SFO2           500.1320005 MHz

F2 - Processing parameters
SI                        32768
SF                125.7578471 MHz
WDW                          EM
SSB                0
LB                     1.00 Hz
GB                 0
PC                     1.40
```

^{1}H NMR spectrum of **1b** (D_2O)



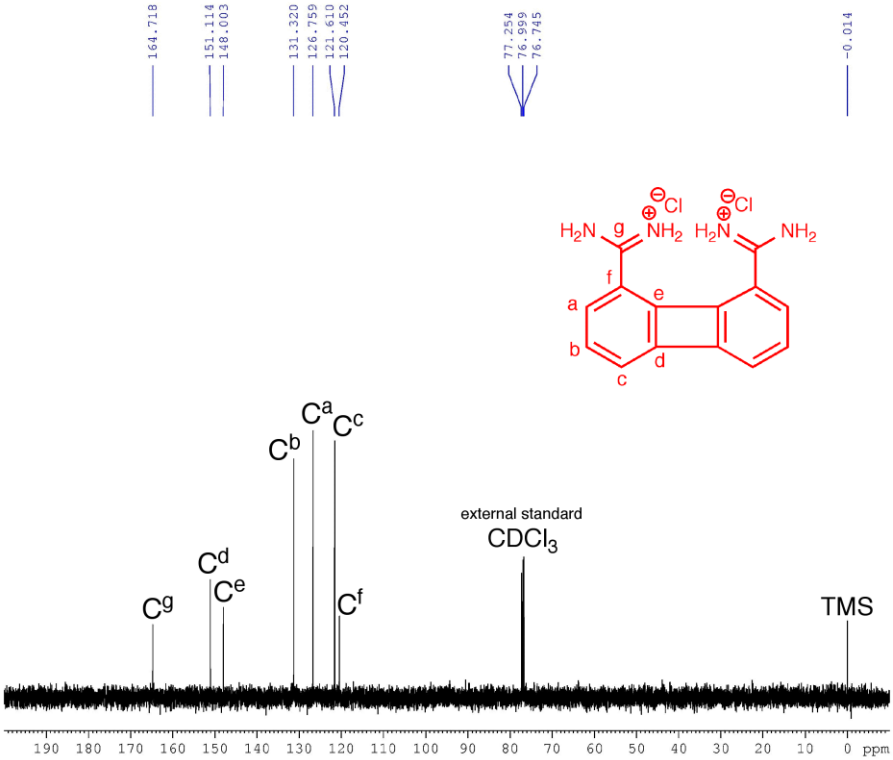

```
Current Data Parameters
NAME          iizuka090803
EXPNO                    1
PROCNO                   1

F2 - Acquisition Parameters
Date_             20090803
Time                 22.12
INSTRUM            drx500
PROBHD    5 mm Multinucl
PULPROG               zg30
TD                   65536
SOLVENT                D2O
NS                      16
DS                       2
SWH              10964.912 Hz
FIDRES            0.167311 Hz
AQ                2.9884915 sec
RG                    228.1
DW                  45.600 usec
DE                   6.00 usec
TE                   298.0 K
D1               1.00000000 sec
MCREST           0.00000000 sec
MCWRK            0.01500000 sec

======== CHANNEL f1 ========
NUC1                      1H
P1                     10.00 usec
PL1                    -6.00 dB
SFO1          500.1320285 MHz

F2 - Processing parameters
SI                       32768
SF               500.1299552 MHz
WDW                         EM
SSB                          0
LB                    0.30 Hz
GB                           0
PC                    1.00
```

^{1}H NMR spectrum of **1b** (D_2O)



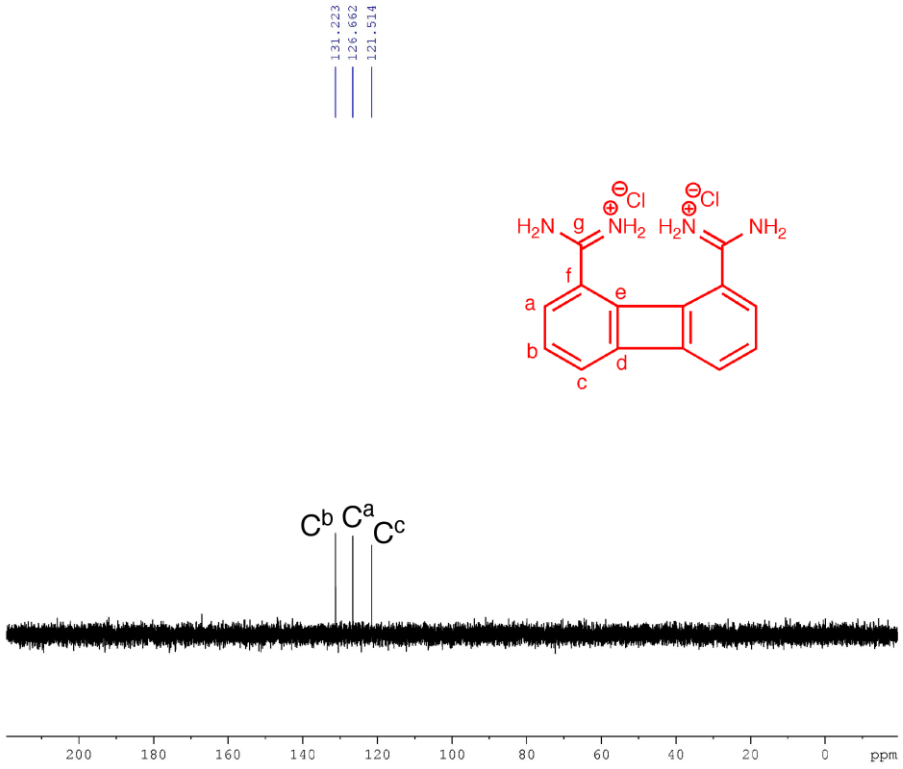

```
Current Data Parameters
NAME         iizuka090803
EXPNO                   1
PROCNO                  1

F2 - Acquisition Parameters
Date_              20090803
Time                 22.12
INSTRUM             drx500
PROBHD   5 mm Multinucl
PULPROG              zg30
TD                  65536
SOLVENT               D2O
NS                     16
DS                      2
SWH             10964.912 Hz
FIDRES          0.167311 Hz
AQ              2.9884915 sec
RG                  228.1
DW                 45.600 usec
DE                  6.00 usec
TE                298.0 K
D1              1.00000000 sec
MCREST          0.00000000 sec
MCWRK           0.01500000 sec

======== CHANNEL f1 ========
NUC1                   1H
P1                 10.00 usec
PL1                -6.00 dB
SFO1          500.1320285 MHz

F2 - Processing parameters
SI                  32768
SF            500.1299552 MHz
WDW                    EM
SSB                     0
LB                   0.30 Hz
GB                      0
PC                   1.00
```

^{13}C NMR spectrum of **1b** (D_2O)



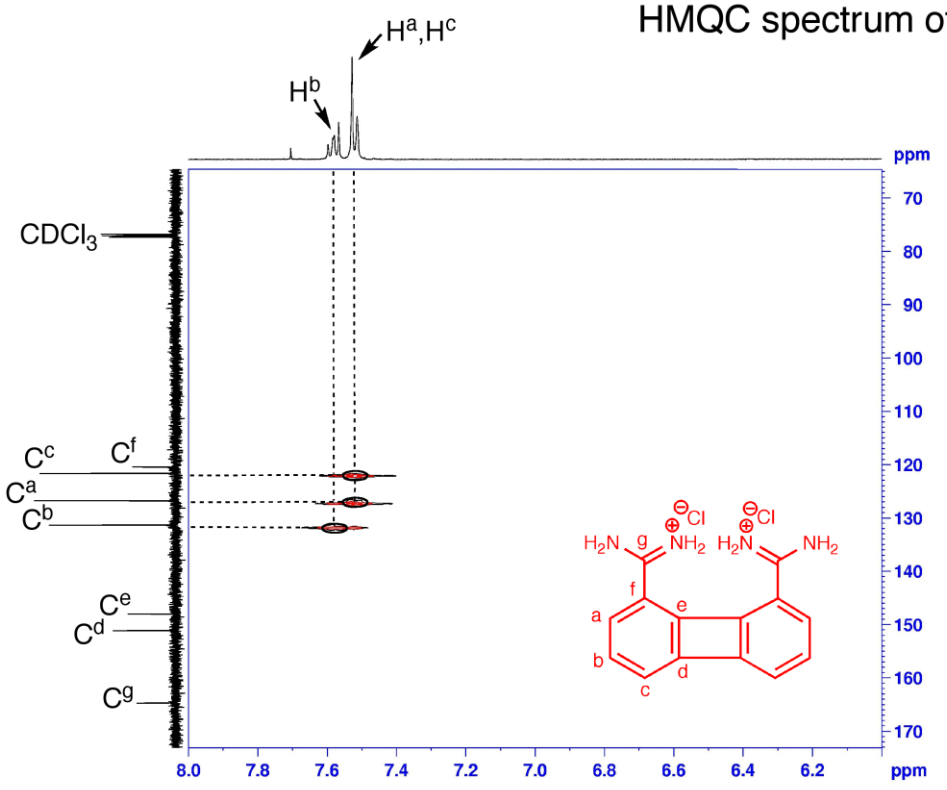

```
Current Data Parameters
NAME         iizuka090803
EXPNO                   2
PROCNO                  1

F2 - Acquisition Paramete
Date_              20090803
Time                 22.15
INSTRUM             drx500
PROBHD   5 mm Multinucl
PULPROG            zgpg30
TD                  65536
SOLVENT             CDCl3
NS                  22341
DS                      2
SWH             27777.777 H
FIDRES          0.423855 H
AQ              1.1796980 s
RG                   4096
DW                 18.000 u
DE                  6.00 u
TE                298.2 K
D1              0.50000000 s
d11             0.03000000 s
MCREST          0.00000000 s
MCWRK           0.01500000 s

======== CHANNEL f1 =====
NUC1                  13C
P1                  9.60 u
PL1                 2.30 d
SFO1          125.7703858 M

======== CHANNEL f2 =====
CPDPRG2           waltz16
NUC2                   1H
PCPD2              100.00 u
PL2                -6.00 d
PL12               13.58 d
PL13               16.50 d
SFO2          500.1320005 M

F2 - Processing parameter
SI                  32768
SF            125.7577770 M
WDW                    EM
SSB                     0
LB                   1.00 H
GB                      0
PC                   1.40
```

DEPT spectrum of **1b** (D_2O)



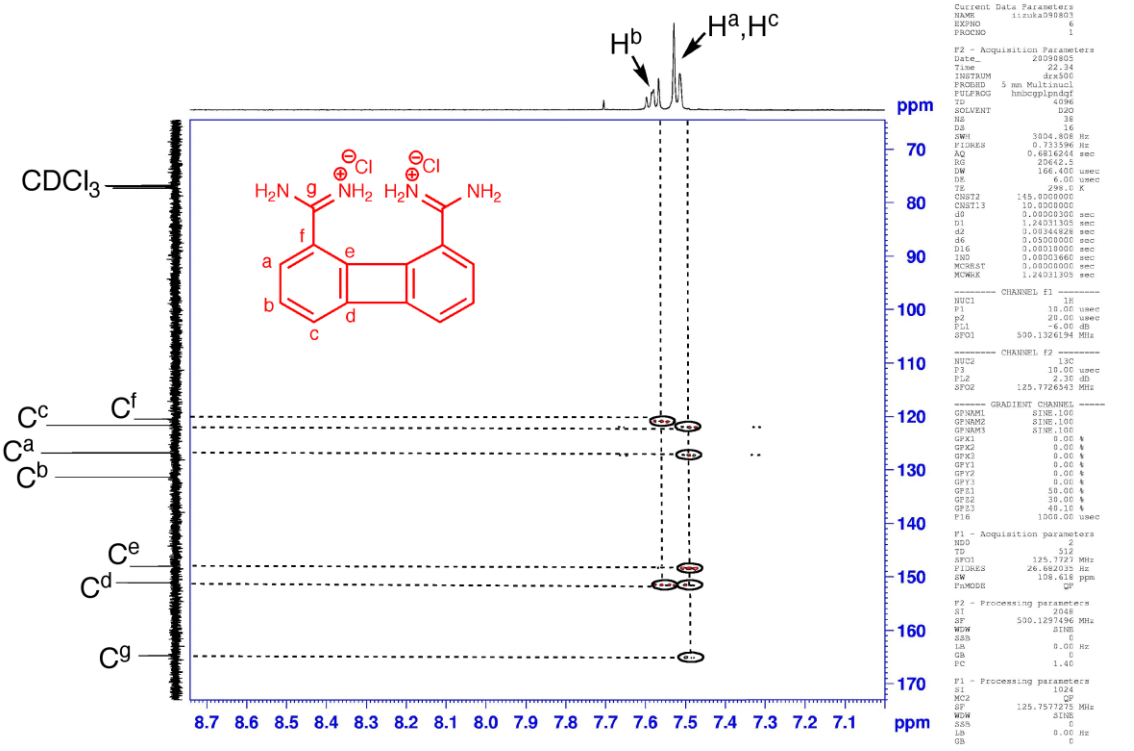

```
Current Data Parameters
NAME                 iizuka090803
EXPNO                           3
PROCNO                          1

F2 - Acquisition Paramete
Date_                    20090805
Time                         2.58
INSTRUM                    drx500
PROBHD    5 mm Multinucl
PULPROG                   dept135
TD                          65536
SOLVENT                       D2O
NS                            12000
DS                                2
SWH                      30030.029 H
FIDRES                    0.458222 H
AQ                        1.0912244 s
RG                          10321.3
DW                           16.650 u
DE                            6.00 u
TE                          298.0 K
CNST2                  145.0000000
D1                       0.50000000 s
d2                       0.00344828 s
d12                      0.00002000 s
DELTA                    0.00001222 s
MCREST                   0.00000000 s
MCWRK                    0.01500000 s

========= CHANNEL f1 =====
NUC1                          13C
P1                           9.60 u
p2                          19.20 u
PL1                          2.30 c
SFO1                125.7703643 M

======== CHANNEL f2 =====
CPDPRG2                   waltz16
NUC2                           1H
P3                          10.50 u
p4                          21.00 u
PCPD2                      100.00 u
PL2                         -6.00 c
PL12                        13.58 c
SFO2                500.1320005 M

F2 - Processing parameter
SI                            32768
SF                  125.7577890 M
WDW                              EM
SSB                               0
LB                           1.00 H
GB                                0
PC                           1.40
```

HMQC spectrum of **1b** (D_2O)

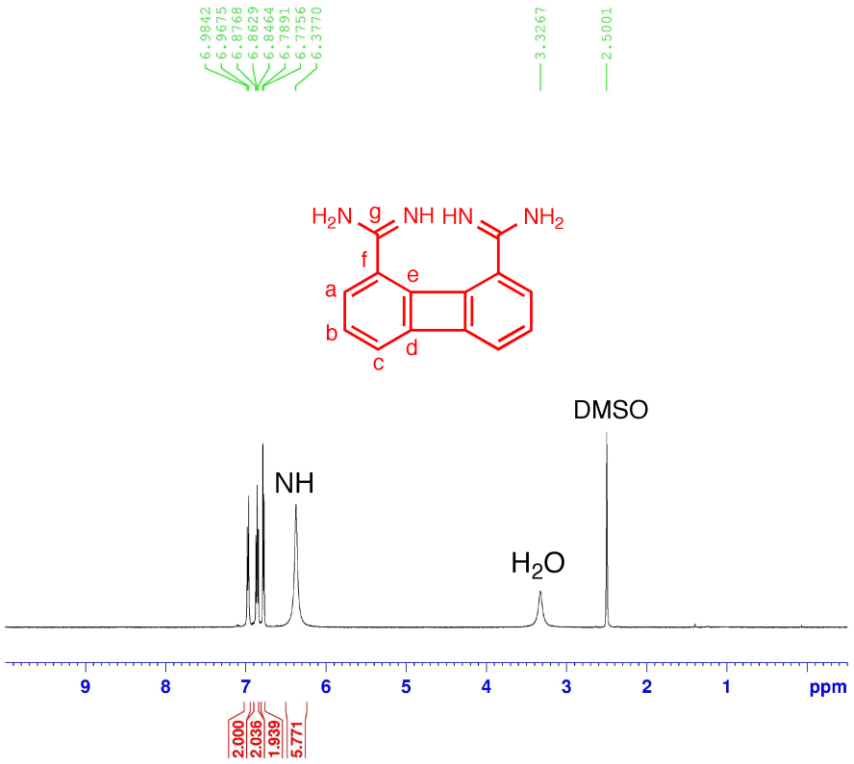



```
Current Data Parameters
NAME                 iizuka090803
EXPNO                           4
PROCNO                          1

F2 - Acquisition Parameters
Date_                    20090805
Time                         2.59
INSTRUM                    drx500
PROBHD    5 mm Multinucl
PULPROG                 hmqcgpqf
TD                           1024
SOLVENT                       D2O
NS                              28
DS                              16
SWH                     3004.808 Hz
FIDRES                  2.934382 Hz
AQ                     0.1704436 sec
RG                          9160.6
DW                        166.400 usec
DE                          6.00 usec
TE                          298.0 K
CNST2                  145.0000000
d0                     0.00000300 sec
D1                     1.45781100 sec
d2                     0.00344828 sec
d12                    0.00002000 sec
d13                    0.00000400 sec
D16                    0.00018000 sec
DELTA1                 0.00232428 sec
IN0                    0.00003660 sec
MCREST                 0.00000000 sec
MCWRK                  1.45781100 sec

======== CHANNEL f1 ========
NUC1                           1H
P1                          10.00 usec
p2                          20.00 usec
PL1                         -6.00 dB
SFO1                500.1326194 MHz

======== CHANNEL f2 ========
CPDPRG2                   waltz16
NUC2                          13C
P3                          10.00 usec
PCPD2                       65.00 usec
PL2                          2.30 dB
PL12                        18.56 dB
SFO2                125.7726543 MHz

====== GRADIENT CHANNEL =====
GPNAM1                   SINE.100
GPNAM2                   SINE.100
GPNAM3                   SINE.100
GPX1                         8.00 %
GPX2                         8.00 %
GPX3                         8.00 %
GPY1                         8.00 %
GPY2                         8.00 %
GPY3                         8.00 %
GPZ1                        50.00 %
GPZ2                        30.00 %
GPZ3                        40.10 %
P16                       1000.00 usec

F1 - Acquisition parameters
ND0                              2
TD                             512
SFO1                125.7727 MHz
FIDRES                  26.682035 Hz
SW                         198.618 ppm
FnMODE                         QF

F2 - Processing parameters
SI                            1024
SF                  500.1297383 MHz
WDW                         QSINE
SSB                               2
LB                           0.00 Hz
GB                                0
PC                           1.40

F1 - Processing parameters
SI                            1024
MC2                             QF
SF                  125.7577158 MHz
```

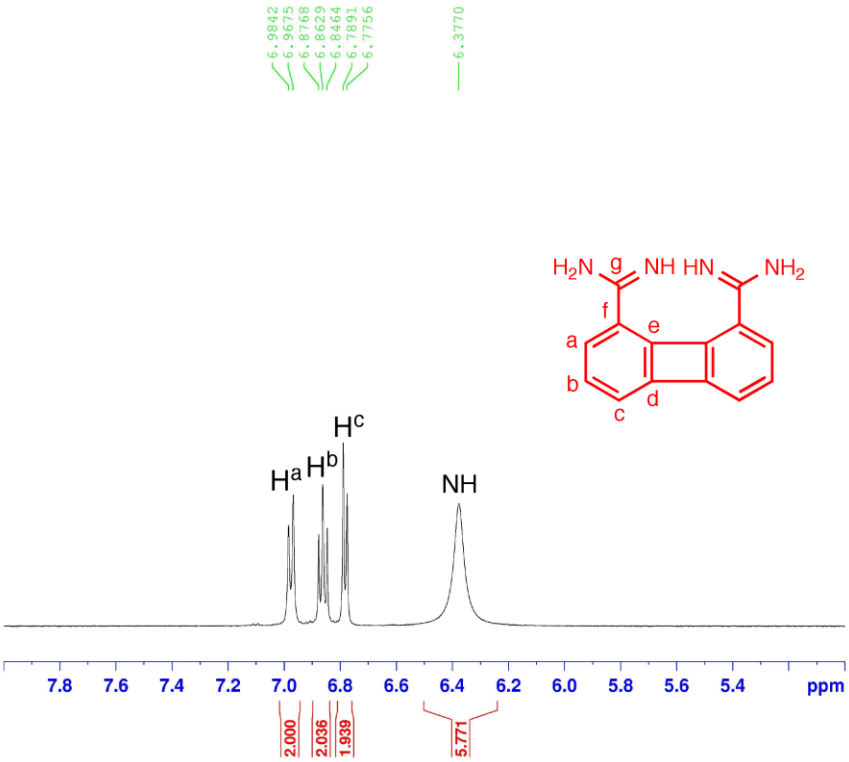
HMBC spectrum of **1b** (D_2O)



^{1}H NMR spectrum (DMSO-d_6)

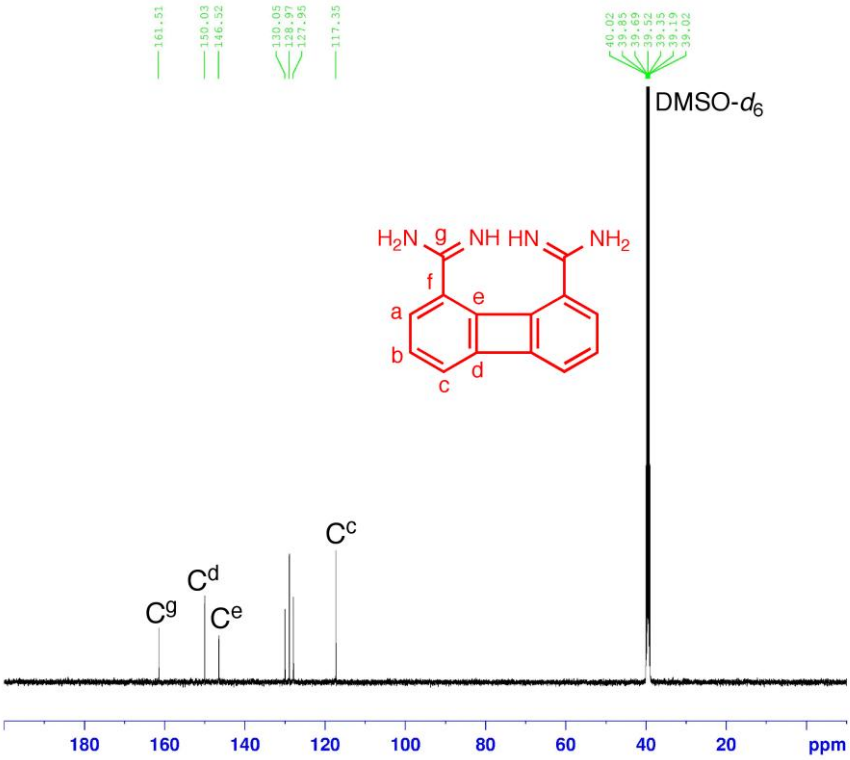


^{1}H NMR spectrum (DMSO-d_6)



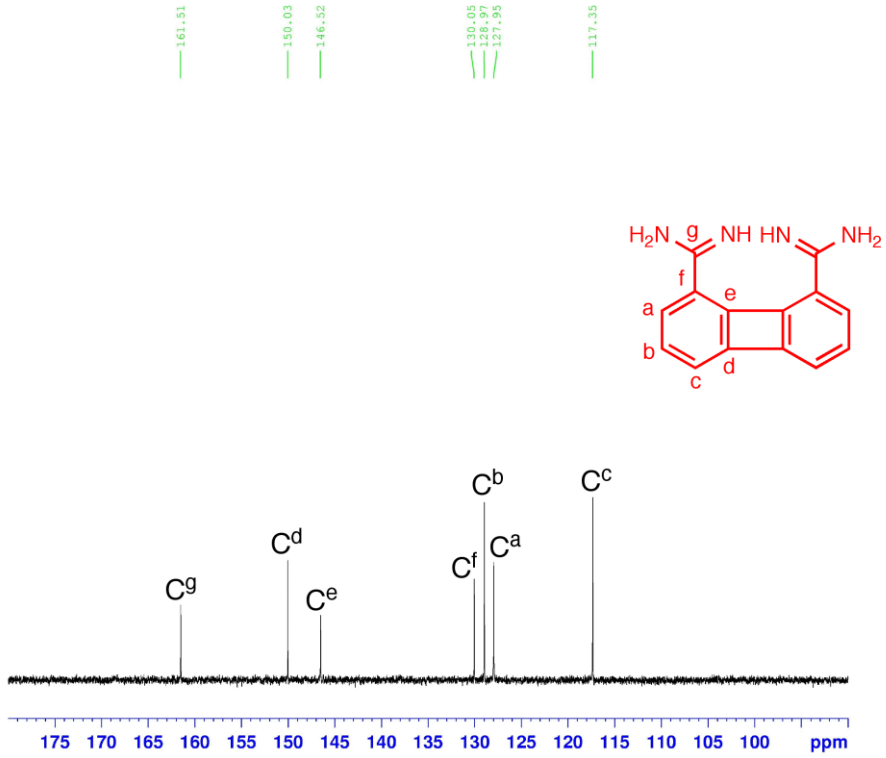

```
Current Data Parameters
NAME       iizuka110822
EXPNO                 1
PROCNO                1

F2 - Acquisition Parameters
Date_           20110822
Time              20.45
INSTRUM           spect
PROBHD   5 mm PABBO BB-
PULPROG            zg30
TD                65536
SOLVENT            DMSO
NS                    1
DS                    2
SWH           10000.000 Hz
FIDRES            0.152588 Hz
AQ             3.2768500 sec
RG                154.97
DW                50.000 usec
DE                 6.50 usec
TE                298.2 K
D1             1.00000000 sec

======== CHANNEL f1 ========
NUC1                  1H
P1                 11.00 usec
PLW1          20.51199913 W
SFO1         500.1324086 MHz

F2 - Processing parameters
SI                65536
SF           500.1300053 MHz
WDW                  EM
SSB       0
LB                0.30 Hz
GB        0
PC                1.00
```

^{13}C NMR spectrum (DMSO-d_6)



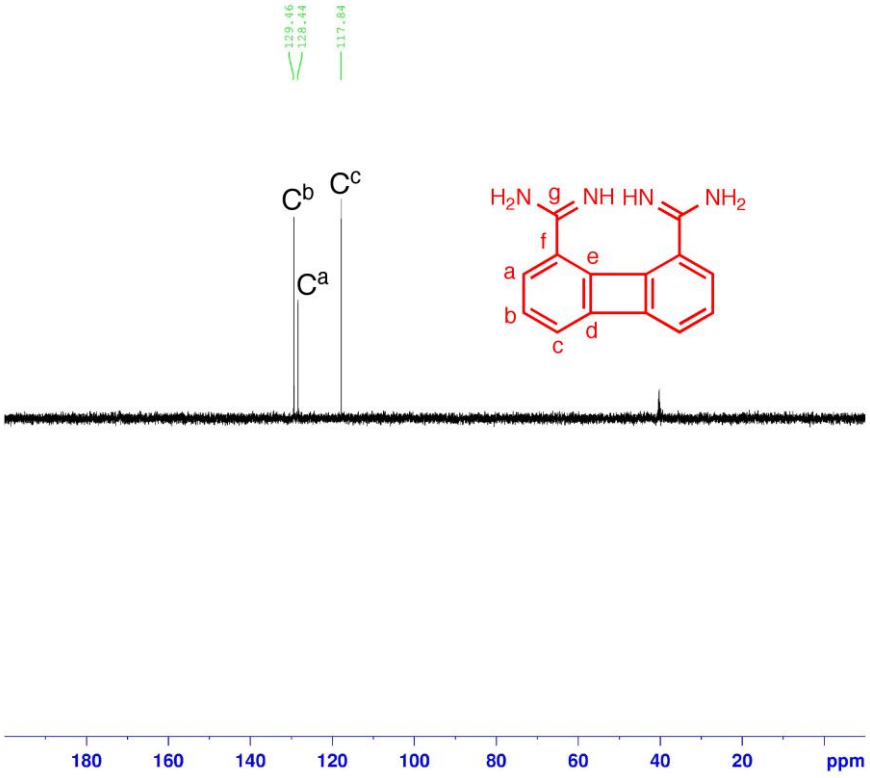

```
Current Data Parameters
NAME       iizuka110822
EXPNO                 2
PROCNO                1

F2 - Acquisition Parameters
Date_           20110822
Time              21.58
INSTRUM           spect
PROBHD   5 mm PABBO BB-
PULPROG          zgpg30
TD                65536
SOLVENT            DMSO
NS                 1000
DS                    4
SWH           29761.904 Hz
FIDRES            0.454131 Hz
AQ             1.1010548 sec
RG                197.97
DW                16.800 usec
DE                 6.50 usec
TE                298.8 K
D1             2.00000000 sec
D11            0.03000000 sec

======== CHANNEL f1 ========
NUC1                 13C
P1                 9.00 usec
PLW1         116.94999695 W
SFO1          125.7703637 MHz

======== CHANNEL f2 ========
CPDPRG2          waltz16
NUC2                  1H
PCPD2              80.00 usec
PLW2          0.38000000 W
PLW12         0.38000000 W
PLW13         0.24320000 W
SFO2         500.1320005 MHz

F2 - Processing parameters
SI                32768
SF           125.7578504 MHz
WDW                  EM
SSB       0
LB                1.00 Hz
GB        0
PC                1.40
```

^{13}C NMR spectrum (DMSO-d_6)



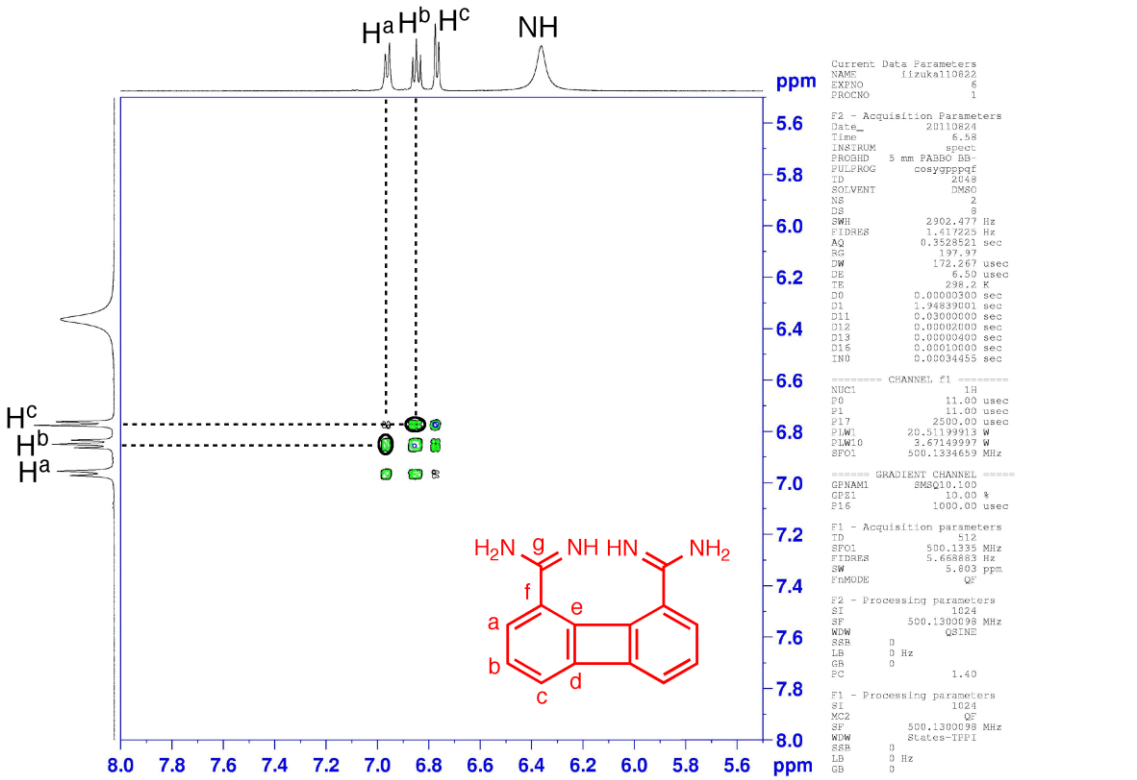

```
Current Data Parameters
NAME          iizuka110822
EXPNO                    2
PROCNO                   1

F2 - Acquisition Parameters
Date_              20110822
Time                 21.58
INSTRUM              spect
PROBHD    5 mm PABBO BB-
PULPROG             zgpg30
TD                   65536
SOLVENT               DMSO
NS                    1000
DS                       4
SWH             29761.904 Hz
FIDRES          0.454131 Hz
AQ              1.1010548 sec
RG                  197.97
DW                16.800 usec
DE                 6.50 usec
TE                 298.8 K
D1              2.00000000 sec
D11             0.03000000 sec

======== CHANNEL f1 ========
NUC1                   13C
P1                  9.00 usec
PLW1         116.94999695 W
SFO1          125.7703637 MHz

======== CHANNEL f2 ========
CPDPRG2            waltz16
NUC2                    1H
PCPD2              80.00 usec
PLW2           0.38000000 W
PLW12          0.38000000 W
PLW13          0.24320000 W
SFO2          500.1320005 MHz

F2 - Processing parameters
SI                   32768
SF            125.7578504 MHz
WDW                     EM
SSB                      0
LB                  1.00 Hz
GB                       0
PC                  1.40
```

DEPT spectrum (DMSO-d_6)

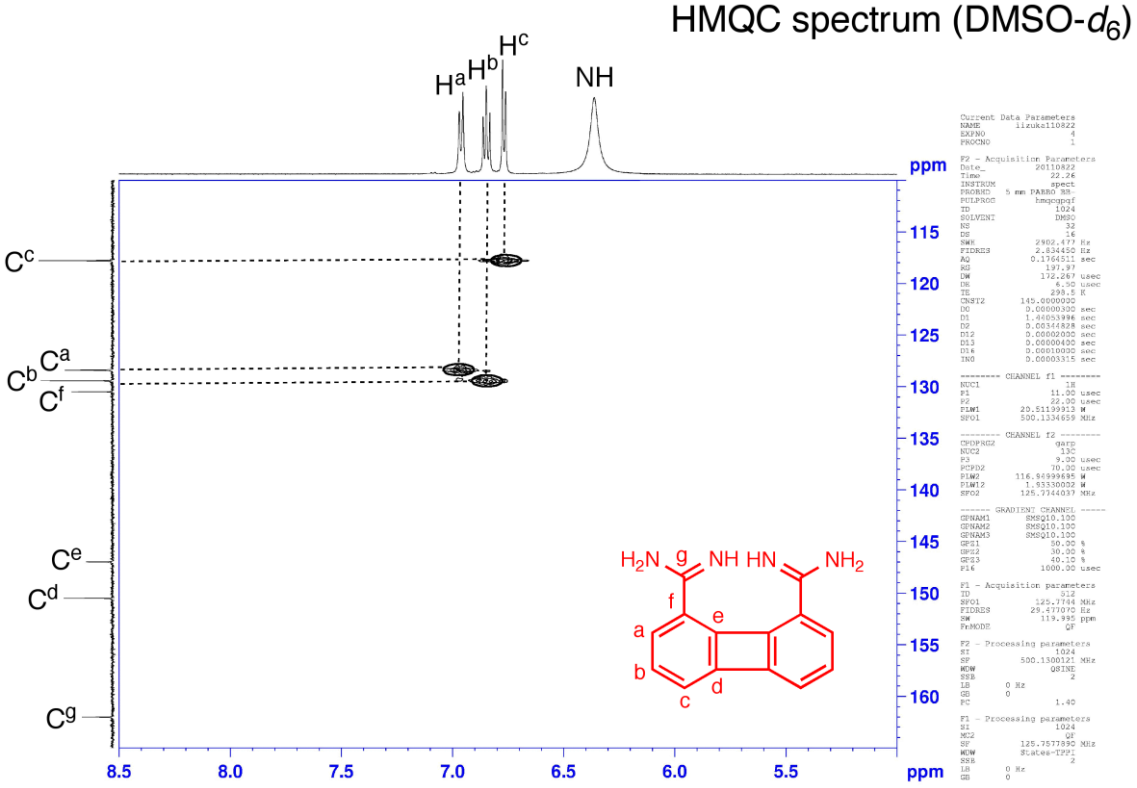


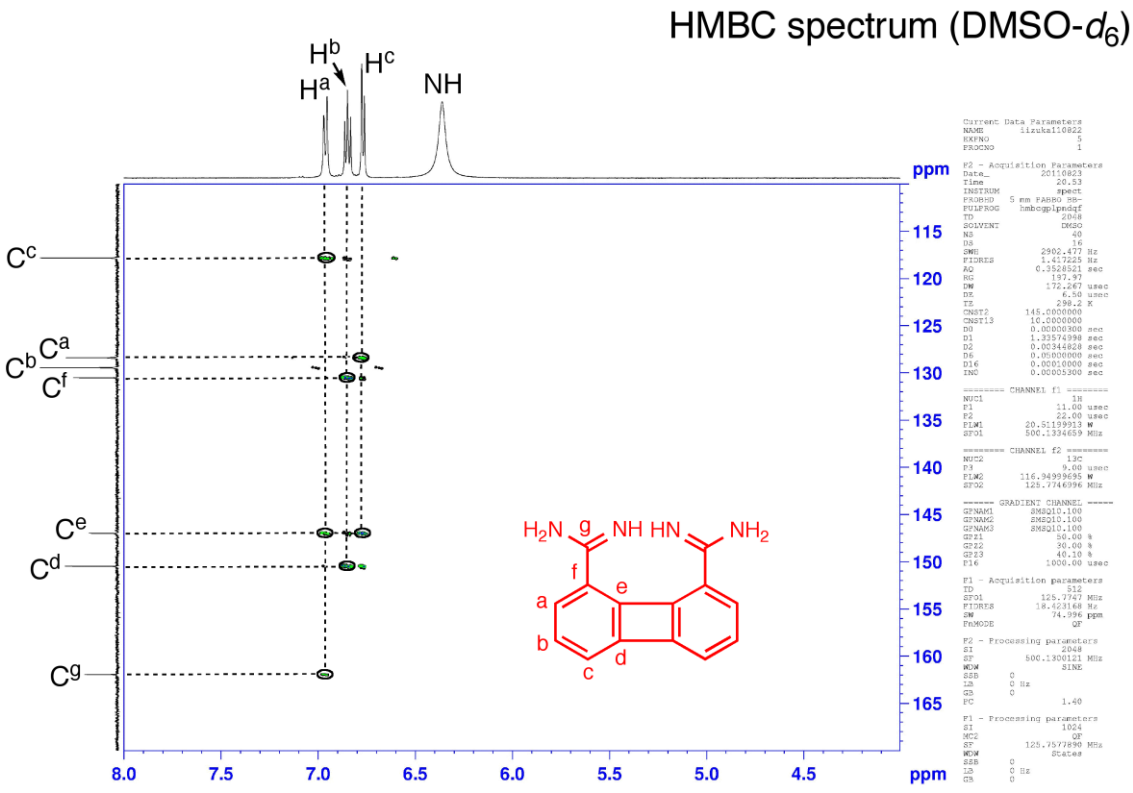

```
Current Data Parameters
NAME          iizuka110822
EXPNO                    3
PROCNO                   1

F2 - Acquisition Parameters
Date_              20110822
Time                 22.24
INSTRUM              spect
PROBHD    5 mm PABBO BB-
PULPROG          deptsp135
TD                   65536
SOLVENT               DMSO
NS                     500
DS                       4
SWH             29761.904 Hz
FIDRES          0.454131 Hz
AQ              1.1010548 sec
RG                  197.97
DW                16.800 usec
DE                 6.50 usec
TE                 298.2 K
CNST2          145.0000000
D1              2.00000000 sec
D2              0.00344828 sec
D12             0.00002000 sec

======== CHANNEL f1 ========
NUC1                   13C
P1                  9.00 usec
P13               2000.00 usec
PLW0       0 W
PLW1         116.94999695 W
SFO1          125.7694289 MHz
SPNAM5        Crp60comp.4
SPOAL5              0.500
SPOFFS5    0 Hz
SPW5         14.47399998 W

======== CHANNEL f2 ========
CPDPRG2            waltz16
NUC2                    1H
P3                 80.00 usec
P4                160.00 usec
PCPD2              80.00 usec
PLW2           0.38000000 W
PLW12          0.38000000 W
SFO2          500.1315995 MHz

F2 - Processing parameters
SI                   32768
SF            125.7577890 MHz
WDW                     EM
SSB                      0
LB                  1.00 Hz
GB                       0
PC                  1.40
```

COSY spectrum (DMSO-d_6)



HMQC spectrum (DMSO-d_6)




HMBC spectrum (DMSO-d_6)
H^b
H^a
H^c
NH
C^c
C^b
C^a
C^f
C^e
C^d
C^g
H_2N
g
NH HN
NH_2
f
e
a
b
d
c
ppm
115
120
125
130
135
140
145
150
155
160
165
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
ppm