Experimental Section

All commercially obtained chemicals were used as received. Tris(dibenzylidineacetone)dipalladium [$Pd_2(dba)_3$], *rac*-2, 2'-Bis(diphenyl-phosphino)-1, 1'-binaphthyl [(±)-BINAP], potassium phosphate, sodium *tert*-butoxide, lithium bis(trimethylsilyl)amide (1.0 M solution in THF) were purchased from Aldrich. 2-Dicyclohexylphosphino-2'-(N, N-dimethylamino)biphenyl was purchased from Strem Chemicals. 8-Bromo-2'-deoxygunaosine was purchased from ChemGenes. Toluene was freshly distilled from sodium and degassed with argon. THF was distilled from a sodium/benzophenone ketyl. Pyridine was freshly distilled from calcium hydride. Anhydrous 1,4-dioxane and 1, 2-dimethoxyethane (DME) were purchased from Aldrich in Sure-seal bottles and used as received. All reactions were performed in oven dried glassware and under an argon atmosphere. Melting points are uncorrected. Proton and carbon-13 NMR data were recorded at 300 or 400 and 75 or 100 MHz, respectively in $CDCl_3$ unless otherwise noted. High resolution FAB mass spectra were obtained from the University of Notre Dame Mass Spectrometry Center using nitrobenzyl alcohol as the matrix.

8-Bromo-*N*9-[3', 5'-*O*-(1, 1, 3, 3-tetrakis(isopropyl)-1, 3-disiloxanediyl)-β-D-2'-deoxyribofuranosyl]guanine. A stirred suspension of 8-bromo-2'-deoxyguanosine (2.9 g, 8.37 mmol) in dry pyridine (50 mL) was cooled 0 ^oC then 1, 3-dichloro-1, 1, 3, 3-tetreisopropyldisiloxane (2.9 g, 9.18 mmol) was added. The reaction mixture was warmed to room temperature and stirred overnight. The pyridine was evaporated under reduced pressure and the residue poured into water and extracted with chloroform. The combined organic extracts were successively washed with 10% HCl, saturated sodium bicarbonate, and brine then dried over sodium sulfate and evaporated. Purification by flash chromatography on silica, eluting with 1-2% methanol in chloroform gave the product (4.5 g, 91% yield) as a white solid, mp 194-196 ^oC; $[\alpha]^{23}_D$ +5.7 ^{o} (*c* 2.3, $CHCl_3$); ^1H NMR ($CDCl_3$) δ 12.00 (br s, 1H), 6.46 (br s, 2H), 6.15 (dd, *J* = 8.6, 3.0, 1H), 5.17 (m. 1H), 4.03 (dd, *J* = 11, 2.3, 1H), 3.93-3.84 (m, 2H), 3.28-3.22 (m, 1H), 2.54-2.45 (m, 1H), 1.14-0.93 (m, 28H); ^{13}C NMR ($CDCl_3$) δ 157.8, 153.5, 152.0, 121.6, 117.9, 85.2, 83.8, 72.9, 63.6, 38.0, 17.6, 17.5, 17.7, 17.3, 17.2, 17.0, 13.4, 13.2, 13.0, 12.6; HRMS (FAB, NBA) m/z calcd for $C_{22}H_{39}O_5N_5BrSi_2$ (M+H) 588.1673, found 588.1657.

***O*^{6}-Benzyl-8-Bromo-*N*9-[3', 5'-*O*-(1, 1, 3, 3-tetrakis(isopropyl)-1, 3-disiloxanediyl)-β-D-2'-deoxyribofuranosyl]guanine (15).** Triphenylphosphine (285 mg, 1.08 mmol) and 8-bromo-*N*9-[3', 5'-*O*-(1, 1, 3, 3-tetraisopropyl-1, 3-disiloxanediyl)-β-D-2'-deoxyribofuranosyl]guanine (0.58 g, 0.98 mmol) were placed in a dry flask and the flask was evacuated for 10 min, then refilled with argon. Anhydrous 1,4-dioxane (10 mL) and benzyl alcohol (117 mg, 1.08 mmol) were added and the resulting suspension cooled to 0 °C. Diethylazodicarboxylate (190 mg, 1.08 mmol) was added dropwise via syringe. After the addition was complete, the reaction was warmed to room temperature and stirred for 1 h, after which time it was concentrated *in vacuo* to give a yellow oil. Purification by flash chromatography on silica, eluting with 10% ethyl acetate in hexanes, gave **15** (0.51 g, 76% yield) as a white solid. mp 127-129 ^{o}C; $[\alpha]^{23}_{D}$ -11 ^{o} (*c* 1.0, $CHCl_3$); ^{1}H NMR ($CDCl_3$) δ 7.47 (d, *J* = 7.5, 2H), 7.36-7.29 (m, 3H), 6.21 (dd, *J* = 8.9, 3.4, 1H), 5.51 (s. 2H), 5.23 (dt, *J* =7.8, 6.0, 1H), 4.75 (s, 2H), 4.03-3.97 (m, 1H), 3.90-3.83 (m, 2H), 3.31-3.24 (m, 1H), 2.54-2.46 (m, 1H), 1.15-0.97 (m, 28H); ^{13}C NMR ($CDCl_3$) δ 159.79, 158.6, 154.0, 136.0, 128.3, 128.2, 128.0, 125.3, 116.2, 85.0, 83.7, 72.9, 68.0, 63.6, 37.8, 17.5, 17.4, 17.3, 17.3, 17.2, 17.1, 17.0, 169, 13.3, 13.1, 12.9, 12.5; HRMS (FAB, NBA) m/z calcd for $C_{29}H_{45}O_5N_5BrSi_2$ (M+H) 678.2143, found 678.2156.

***N*^{2}-(1, 1, 4, 4-Tetramethyldisilylazacyclopentanyl)-*O*^{6}-benzyl-8-Bromo-*N*9-[3', 5'-*O*-(1, 1, 3, 3-tetrakis(isopropyl)-1, 3-disiloxanediyl)-β-D-2'-deoxyribofuranosyl]guanine (17)**. A solution of **15** (220 mg, 0.33 mmol) in anhydrous THF (20 mL) was cooled to -78 ^{o}C, then lithium bis(trimethylsilyl)amide (1.0 M in THF, 0.65 mL, 0.65 mmol) was added over 5 min. The solution was stirred for 15 min, then 1, 2-bis(chlorodimethylsilyl)ethane (72 mg, 0.33 mmol) in THF (1 mL) was added dropwise. The reaction was warmed to room temperature and stirred for an additional 20 min. The reaction was quenched by the addition of saturated sodium bicarbonate (10 mL). The layers were separated and the aqueous phase extracted with ether. The combined organic extracts were washed with brine, dried over sodium sulfate, filtered and evaporated. Purification by flash chromatography on silica, eluting with 3% ethyl acetate in hexanes, gave **17** (226 mg, 85% yield) as white solid. mp 94-96 ^{o}C; $[\alpha]^{23}_{D}$ -13 ^{o} (*c* 1.1, $CHCl_3$); ^{1}H NMR ($CDCl_3$) δ 7.43 (d, *J* = 6.9, 2H), 7.37-7.29 (m, 3H), 6.28 (t *J* = 7.6, 1H), 5.52 (s, 2H), 4.77-4.71 (m, 1H), 4.15-4.06 (m, 1H), 3.97-3.89 (m, 2H), 3.75-3.64 (m, 1H), 2.28 (ddd, *J* = 13.5,

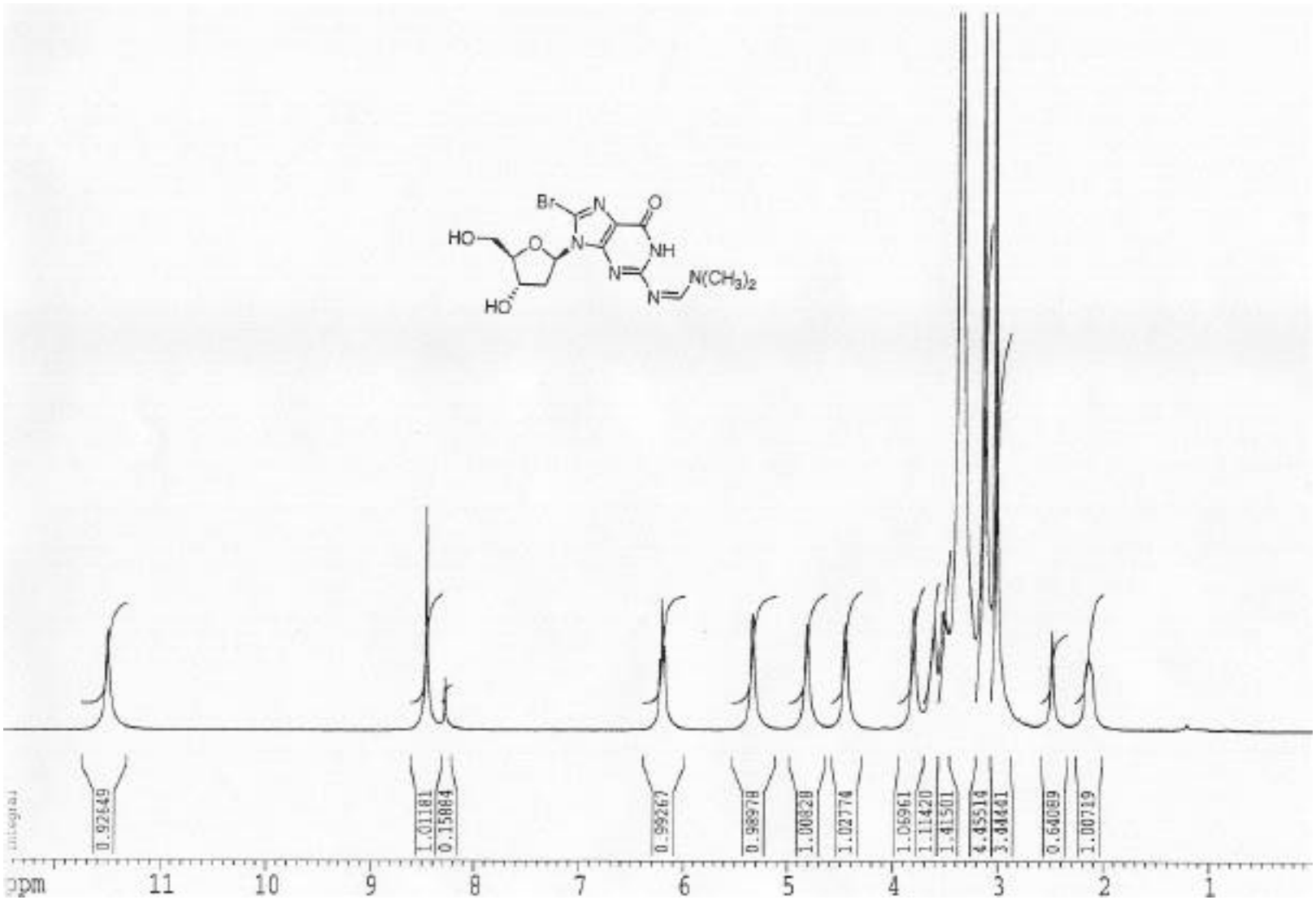
6.9, 3.0, 1H), 1.12-0.98 (m, 28H), 0.83 (s, 4H), 0.33 (s, 6H), 0.31 (s, 6H); ^{13}C NMR ($CDCl_3$) δ 160.9, 159.0, 154.7, 136.1, 128.4, 128.0, 127.7, 124.6, 116.1, 85.5, 83.8, 73.7, 68.0, 64.4, 37.4, 17.5, 17.4, 17.3, 17.2, 17.1, 17.0, 16.9, 13.4, 13.2, 13.0, 12.6, 8.2, -0.5, -0.7; HRMS (FAB, NBA) m/z calcd for $C_{35}H_{59}O_5N_5BrSi_4$ (M+H) 820.2777, found 820.2771.

***N*^{2}-(1, 1, 4, 4-Tetramethyldisilylazacyclopentanyl)-*O*^{6}-benzyl-8-(IQ)-*N*9-[3', 5'-*O*-(1, 1, 3, 3-tetrakis(isopropyl)-1, 3-disiloxanediyl)-β-D-2'-deoxyribofuranosyl]guanine (19).** To a stirred suspension of **17** (220 mg, 0.268 mmol), 2-amino-3-methylimidazo[4,5-f]-quinoline (IQ, 106 mg, 0.536 mmol), $Pd_2(dba)_3$ (25 mg, 0.0268 mmol), and BINAP (50 mg, 0.0804 mmol) in anhydrous, degassed toluene (2 mL) under an argon atmosphere, was added lithium bis(trimethylsilyl)amide (1.0 M in THF, 0.54 mL, 0.54 mmol). The reaction was heated to 100 ^{o}C and stirred for 25 min, then cooled to room temperature and quenched with saturated sodium bicarbonate (1 mL). The layers were separated and the aqueous phase extracted with ethyl acetate. The combined organic extracts were washed with brine, dried over sodium sulfate, filtered and evaporated. Purification by flash chromatography on silica, eluting with 20 % ethyl acetate in hexanes, gave **19** (170 mg, 68 %) as a yellowish solid. mp 233-235 ^{o}C; $[\alpha]^{23}_D$ -12 ^{o} (*c* 0.55, $CHCl_3$); ^{1}H NMR ($CDCl_3$) δ 9.50-8.00 (br, s 1H), 8.85 (d, *J* = 4.1, 1H), 8.60 (d, *J* = 8.3, 1H), 7.92 (d, *J* = 8.9, 1H), 7.63 (d, *J* = 8.9, 1H), 7.52 (d, *J* = 7.4, 2H), 7.46-7.36 (m, 4H), 6.66 (t, *J* = 7.7, 1H), 5.60 (s, 2H), 4.83-4.80 (m, 1H), 4.15 (dd, *J* = 10.5, 3.1, 1H), 4.07-4.02 (m, 1H), 3.99-3.88 (m, 2H), 3.83 (s, 3H), 2.32-2.24 (m, 1H), 1.15-1.05 (m, 28H), 0.85 (s, 4H), 0.37 (s, 6H), 0.35 (s, 6H); ^{13}C NMR ($CDCl_3$) δ 159.6, 154.7, 154.0, 152.6, 151.5, 148.1, 145.1, 136.9, 129.8, 129.5, 128.5, 128.4, 127.9, 127.2, 122.8, 120.6, 118.0, 112.3, 105.7, 85.5, 81.4, 74.7, 67.5, 65.1, 36.6, 28.7, 17.5, 17.4, 17.3, 17.2, 17.1, 17.0, 13.5, 13.3, 13.0, 12.7, 8.7, -0.4, -0.6; HRMS (FAB, NBA) m/z calcd for $C_{46}H_{68}O_5N_9Si_4$ (M+H) 938.4421, found 938.4456.

***O*^{6}-benzyl-8-(IQ)-*N*9-β-D-2'-deoxyribofuranosyl]guanine**. To a stirred solution of **19** (150 mg, 0.16 mmol) in THF (2 mL) was added tetrabutylammonium fluoride (0.64mmol, 1.0 M in THF). The reaction was stirred at room temperature for 1.5 h after which time the solvent was removed under reduced pressure. Purification by flash chromatography on silica, eluting with 5% methanol in chloroform, gave the product (84 mg, 95% yield) as a yellowish solid. mp 220 ^{o}C (dec); $[\alpha]^{23}_D$ +22 ^{o} (*c* 0.43, $CHCl_3$:MeOH=3:1); ^{1}H NMR (DMSO-d_6) δ 8.77 (dd, *J* = 4.2, 1.6,

1H), 8.50 (d, *J* = 8.3, 1H), 7.83 (d, *J* = 8.9, 1H), 7.76 (d, *J* = 8.9, 1H), 7.57 (d, *J* = 7.0, 2H), 7.51-7.33 (m, 4H), 7.2-5.7 (br, 3H), 6.59 (t, *J* = 7.4, 1H), 6.29 (s, 2H), 5.56 (s, 2H), 4.54-4.51 (m, 1H), 3.89-3.84 (m, 1H), 3.78-3.70 (m, 1H), 3.71 (s, 3H), 3.56 (dd, *J* = 11.6, 5.1, 1H), 3.37-3.28 (m, 1H), 2.14-2.06 (m, 1H); ^{13}C NMR (DMSO-d_6) δ 158.3, 155.9, 153.0, 152.89, 152.5, 150.7, 147.9, 144.5, 137.0, 128.9, 128.7, 128.4, 128.1, 127.9, 127.8, 122.3, 120.7, 117.0, 113.2, 104.2, 87.5, 82.3, 71.5, 66.7, 62.5, 36.0, 28.5; HRMS (FAB, NBA) m/z calcd for $C_{28}H_{28}O_4N_9$ (M+H) 554.2264, found 554.2262.

C8-IQ adduct of dG (1). Hydrogen gas was bubbled through a solution of *O*^{6}-benzyl-8-(IQ)-*N*9-β-D-2'-deoxyribofuranosyl]guanine (55 mg, 0.099 mmol) and 5% Pd/C (42 mg, 0.02 mmol) in 1:1 THF/methanol (20 mL) for 2 h. The mixture was then stirred at room temperature overnight under 1 atm of hydrogen. The catalyst was removed by filtration and the filtrate evaporated. Purification by flash chromatography on silica, eluting with 10% methanol in chloroform containing 1% NH_3, gave **1** (33 mg, 71% yield) as a yellowish solid. mp 250 ^{o}C (dec); $[\alpha]^{23}_D$ +16 ^{o} (*c* 0.55, MeOH:$CHCl_3$ =1:1); ^{1}H NMR (DMSO-d_6) δ 11.05 (br s, 1H), 8.77 (dd, *J* = 4.2, 1.7, 1H), 8.66 (d, *J* = 8.3, 1H), 7.88 (d, *J* = 8.9, 1H), 7.77 (d, *J* = 8.9, 1H), 7.57 (dd, *J* = 8.4, 4.2, 1H), 6.53-6.49 (m, 3H), 4.52-4.49 (m, 1H), 3.83-3.79 (m, 1H), 3.75 (s, 3H), 3.69 (dd, *J* = 11.5, 5.2, 1H), 3.54 (dd, *J* = 11.5, 5.6, 1H), 3.37-3.30 (m, 1H), 2.10-2.04 (m, 1H); ^{13}C NMR (DMSO-d_6) δ 154.11, 153.38, 152.81, 149.19, 148.60, 147.78, 144.55, 130.68, 129.11, 128.46, 121.75, 120.80, 117.73, 113.17, 104.05, 87.40, 81.94, 71.32, 62.34, 35.86, 28.58; HRMS (FAB, NBA) m/z calcd for $C_{21}H_{22}O_4N_9$ (M+H) 464.1795, found 464.1806.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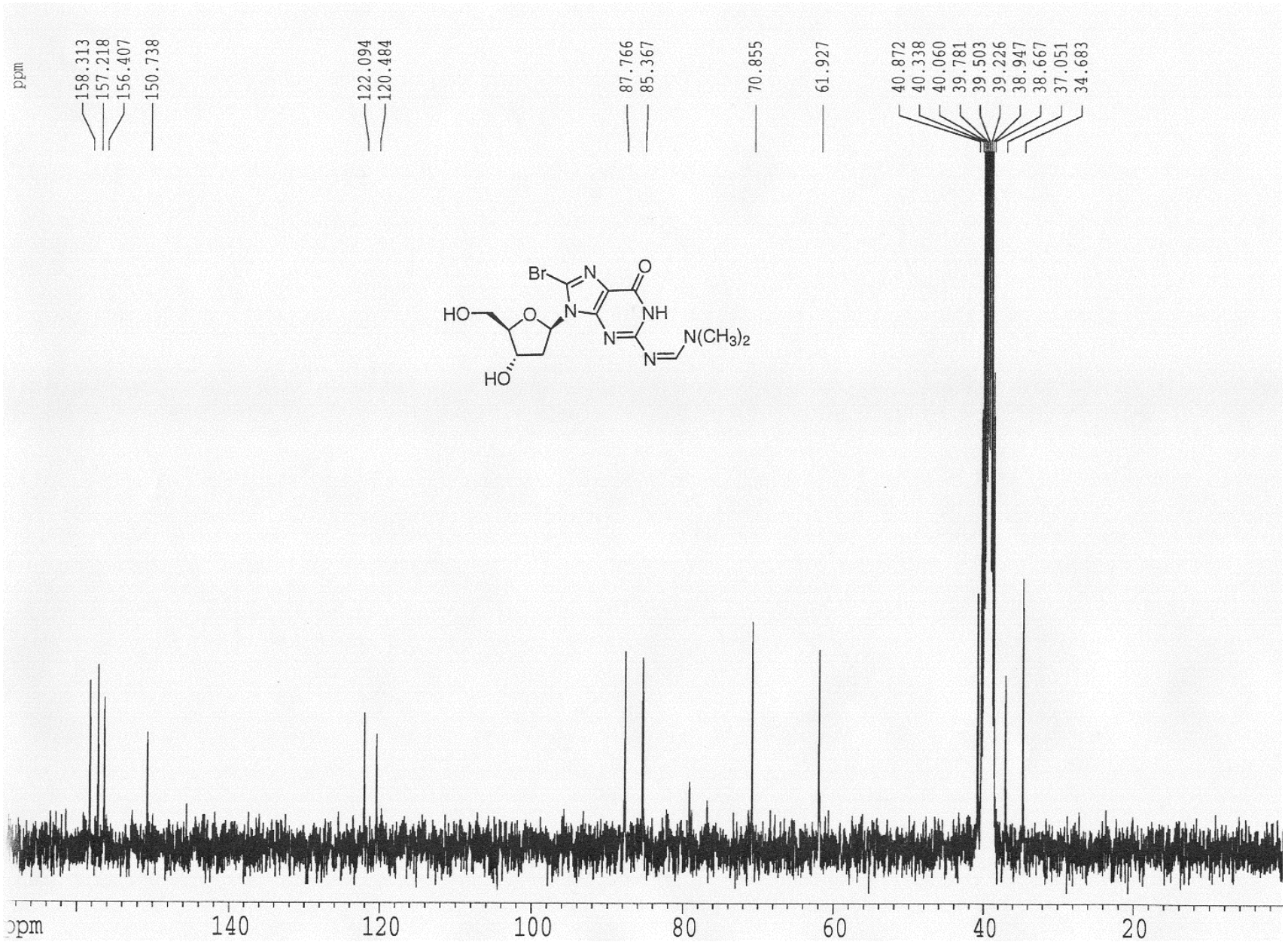
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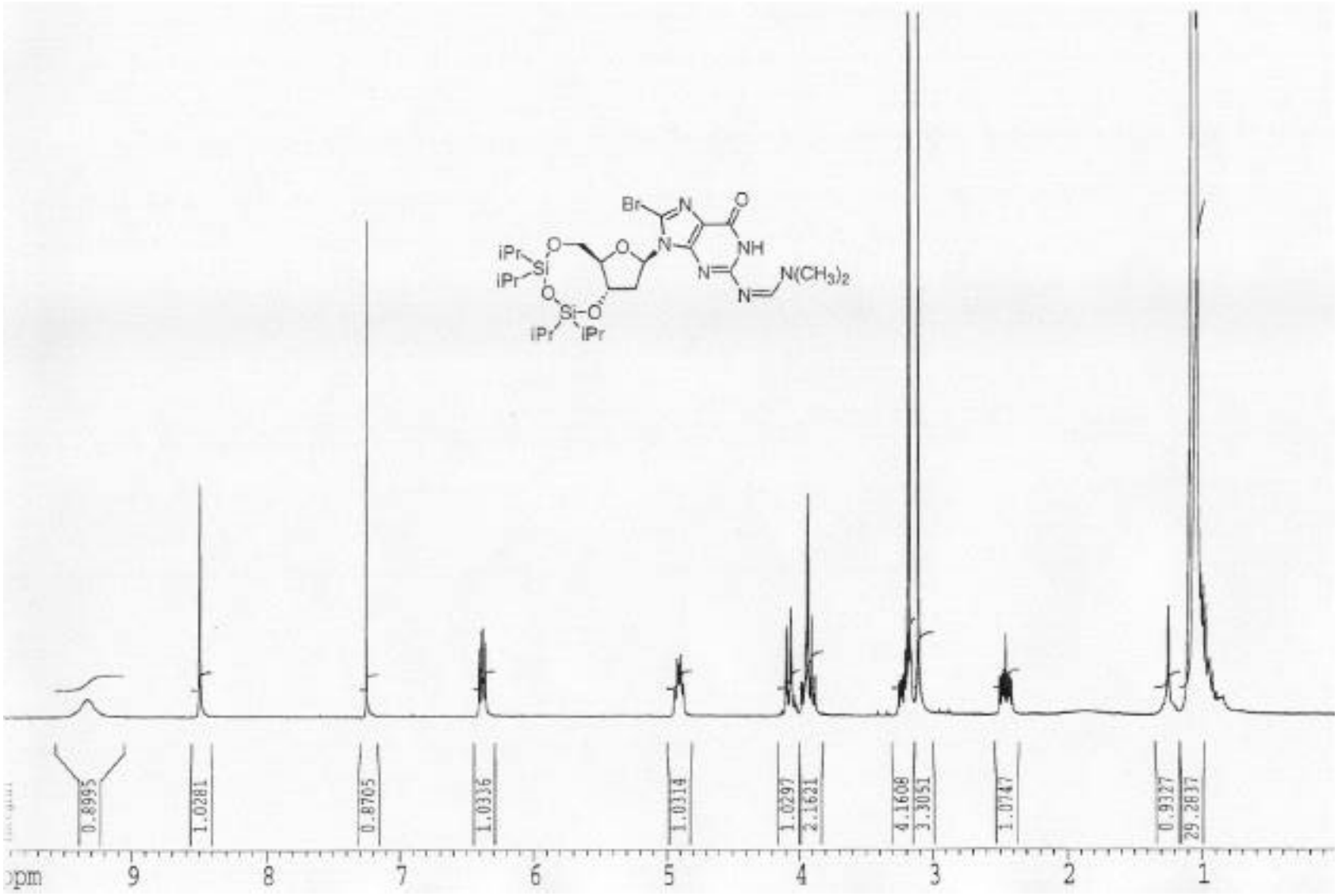
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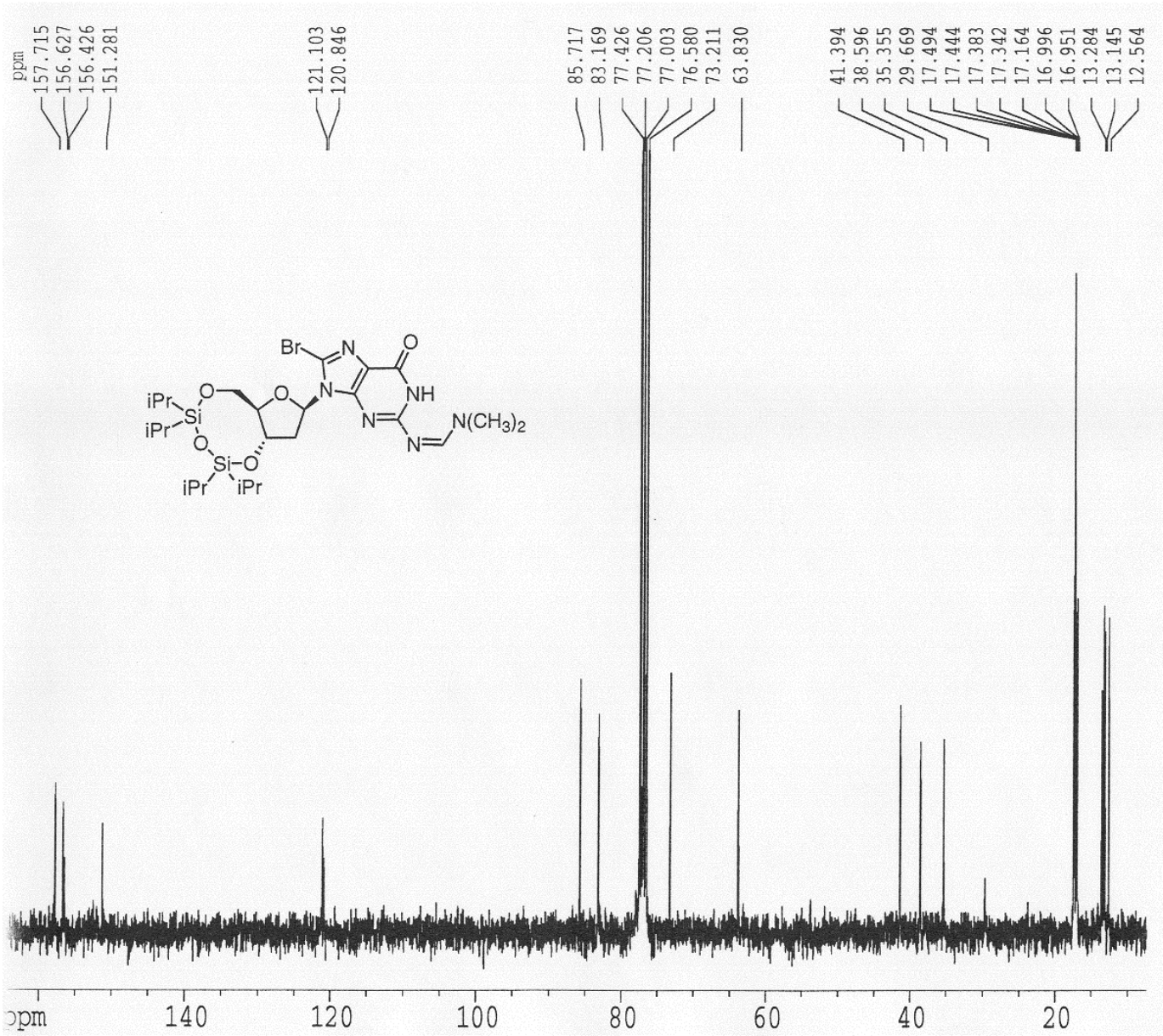
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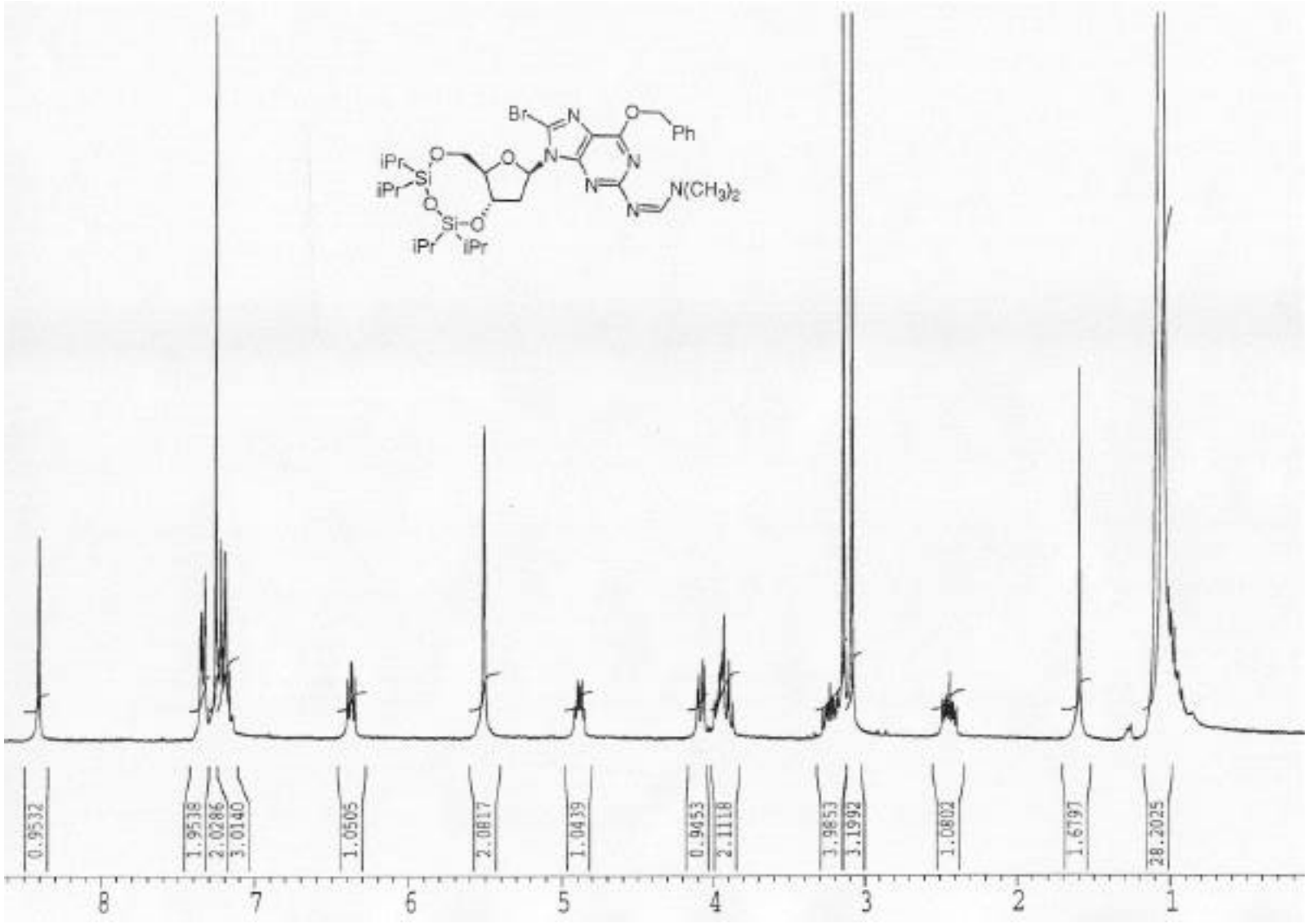
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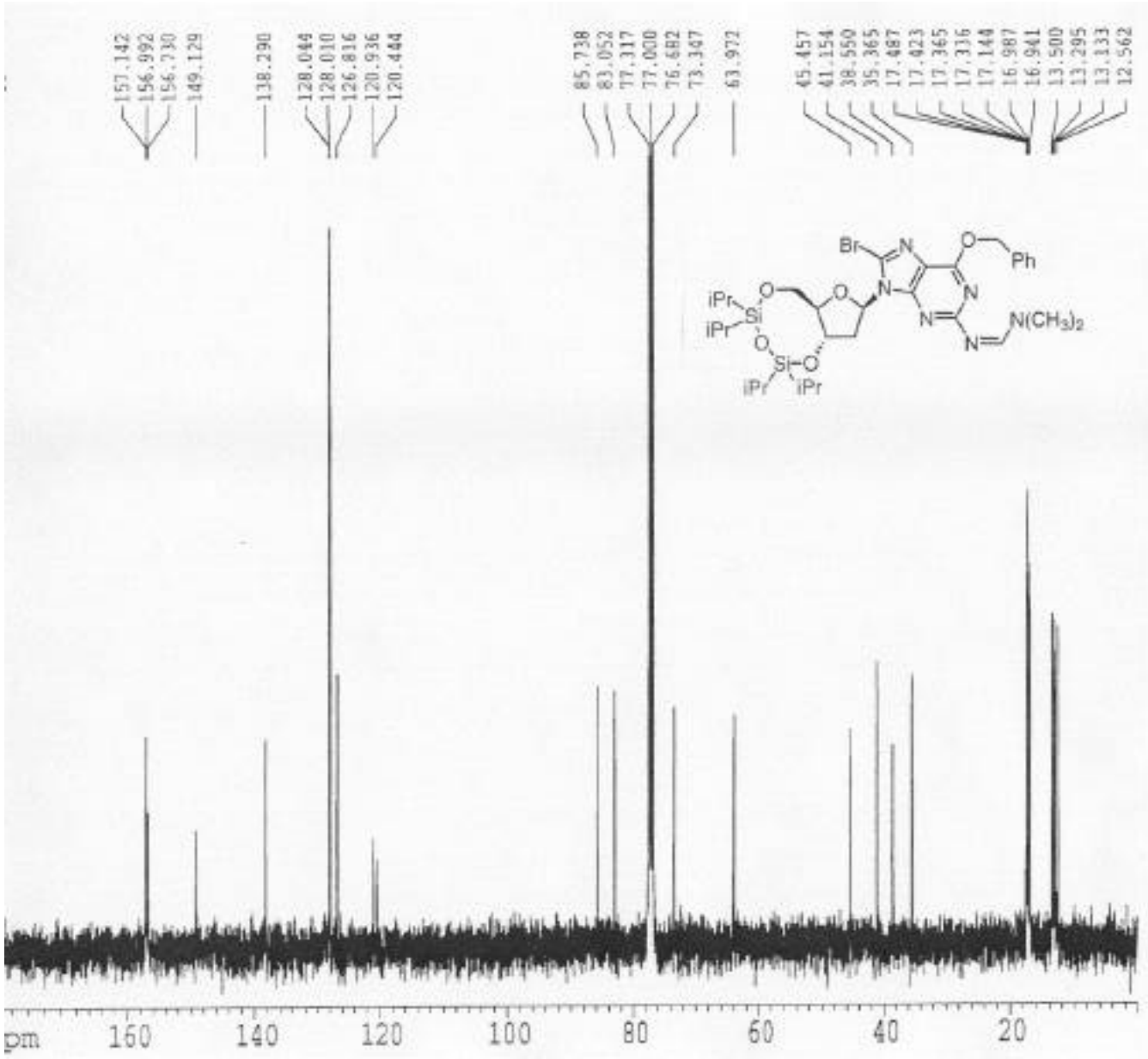
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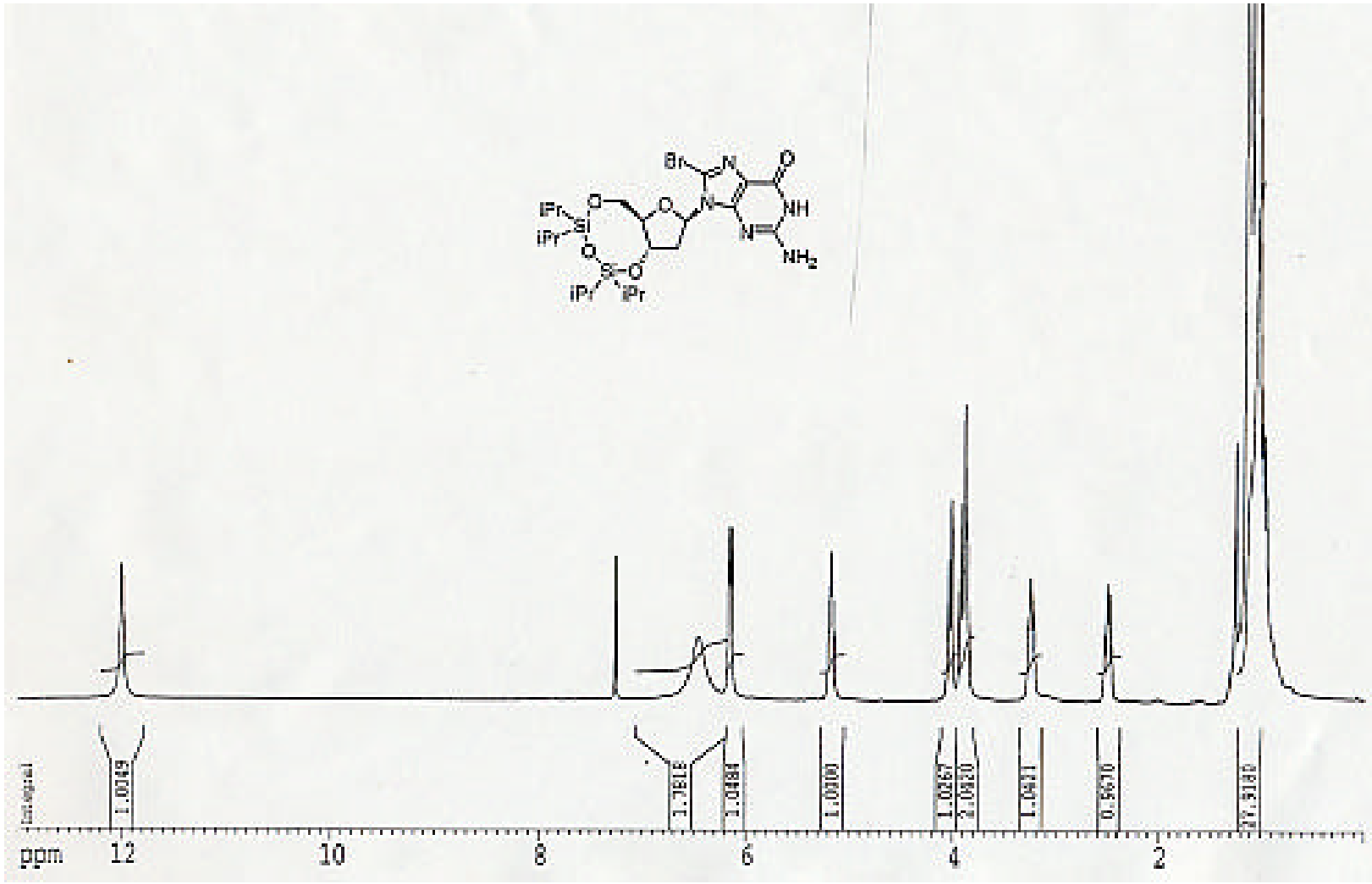
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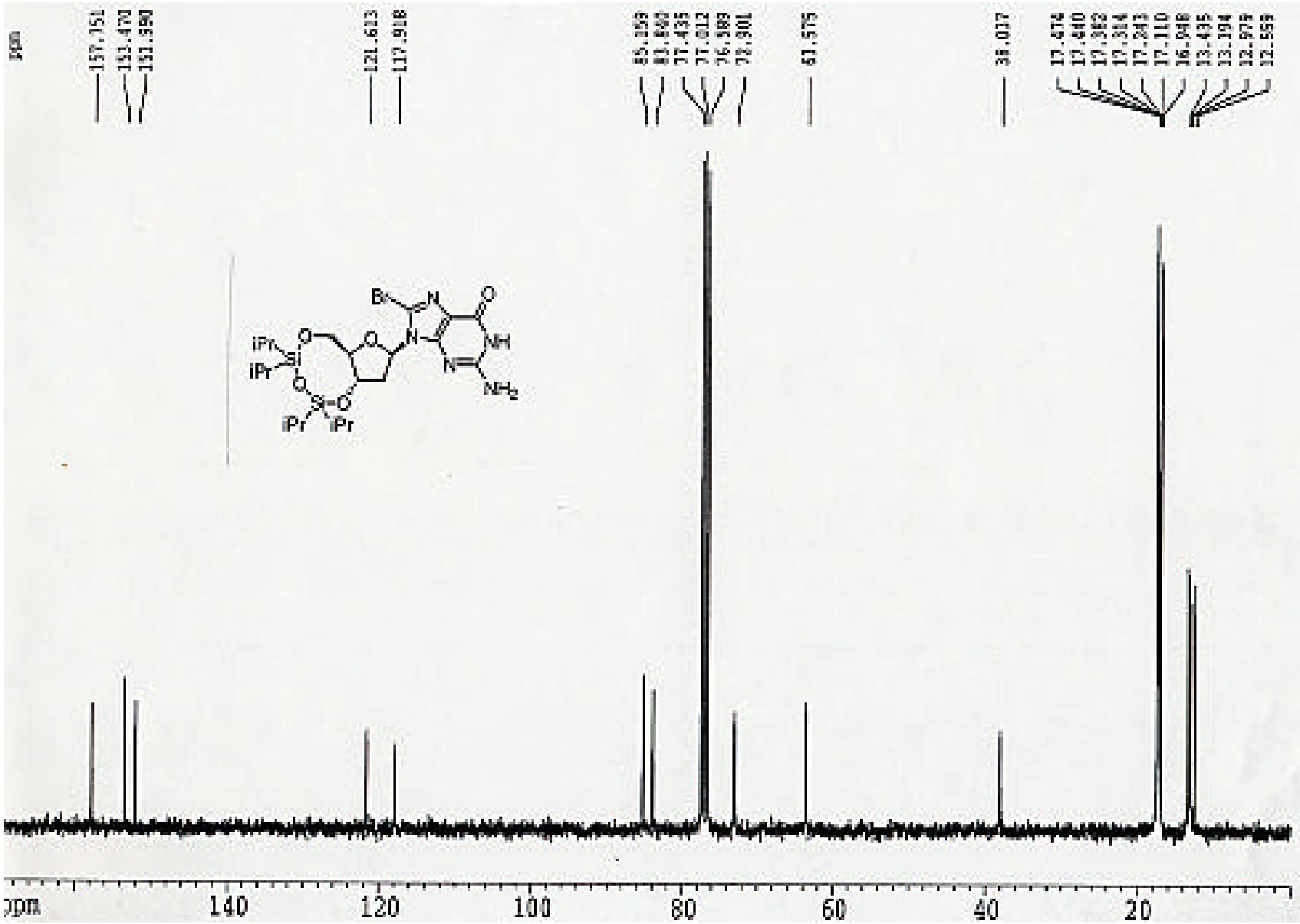
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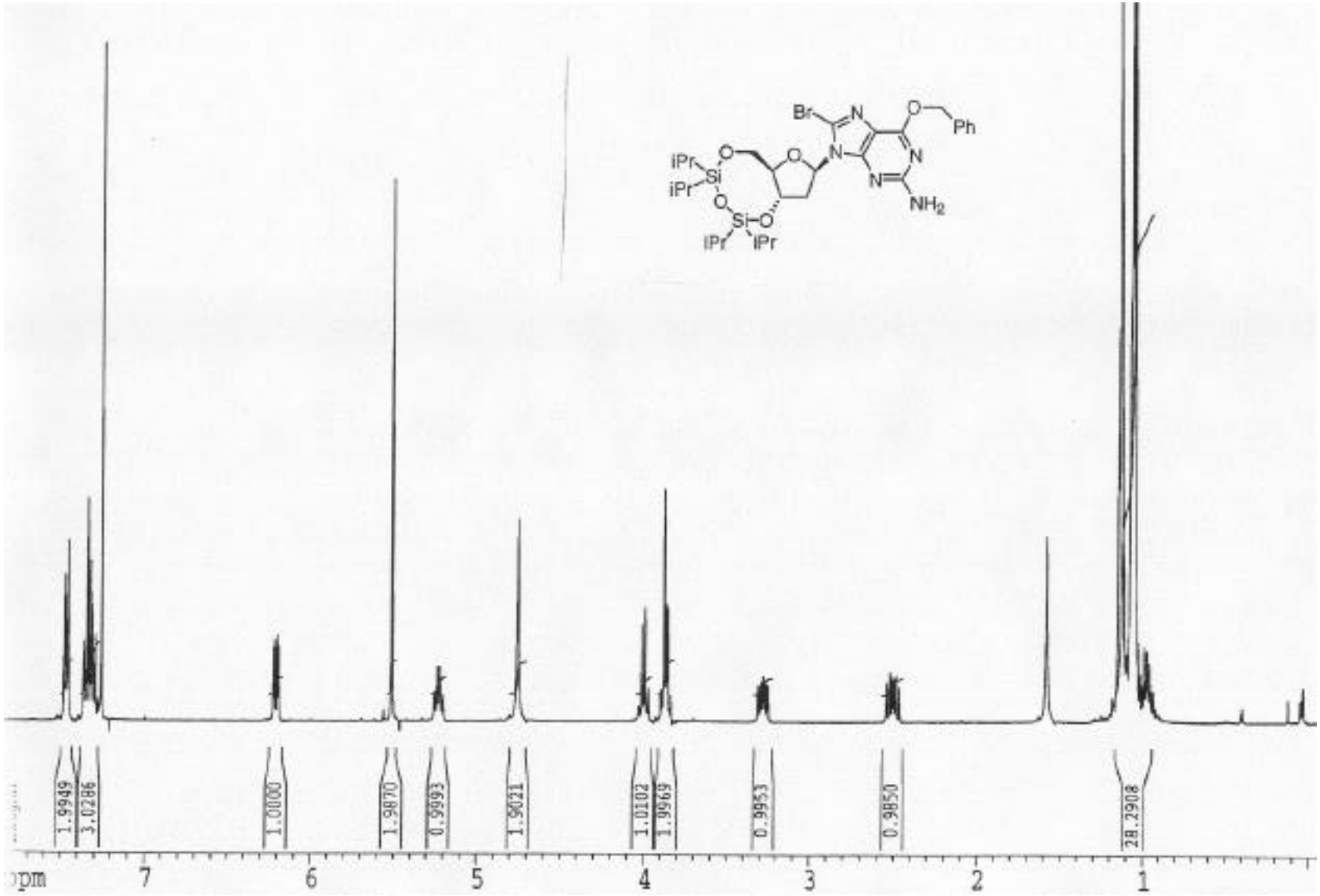
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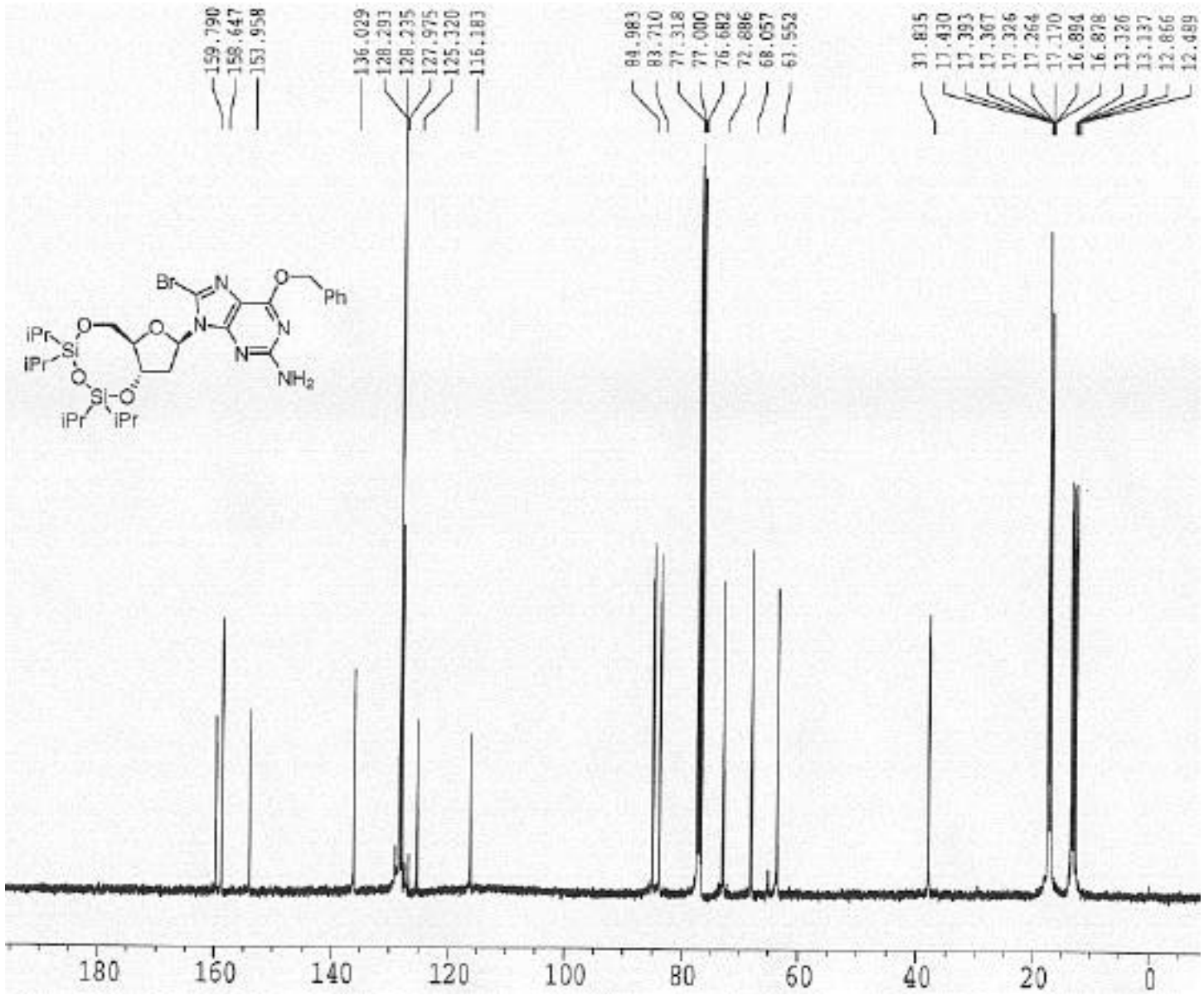
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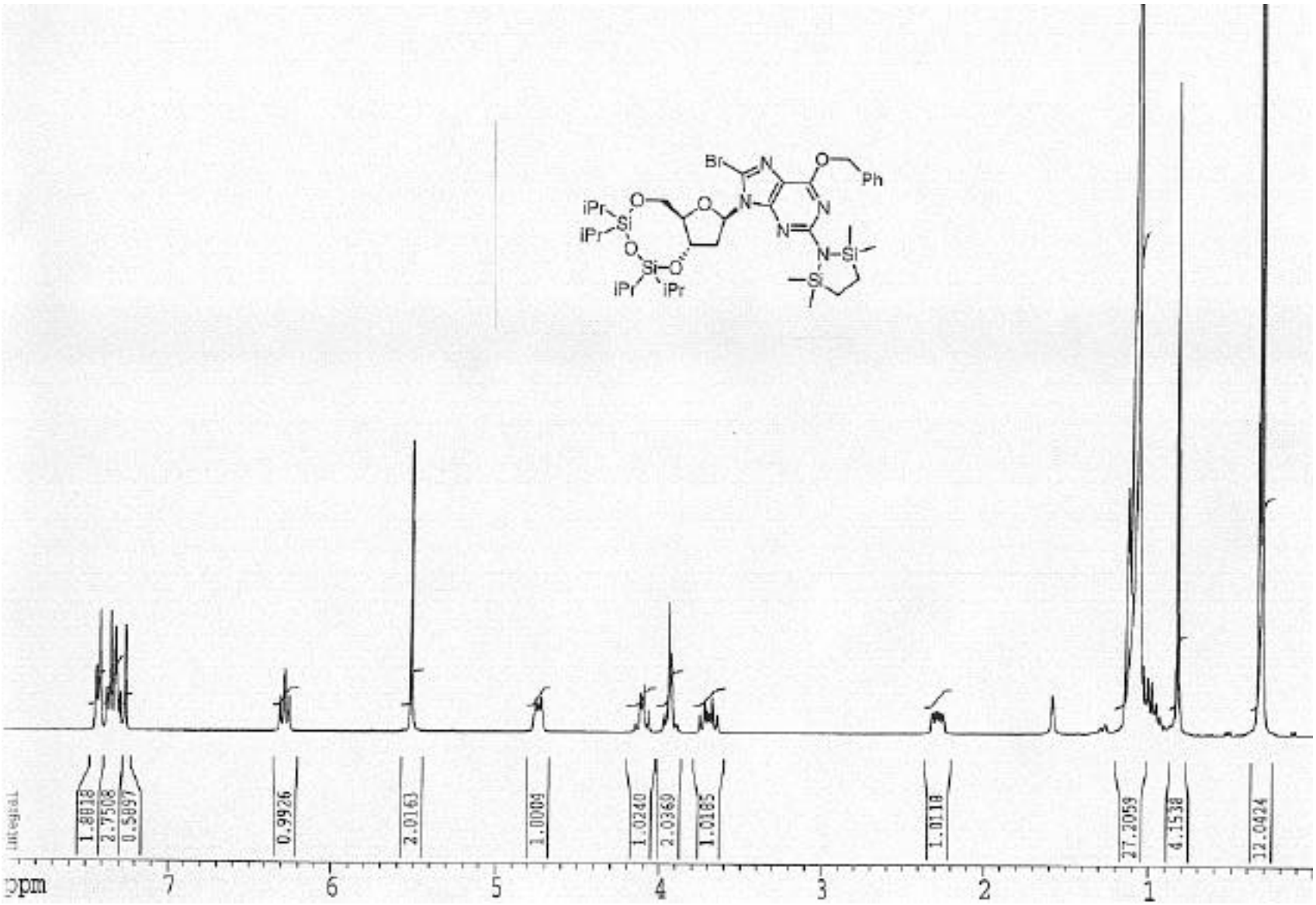
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iPr
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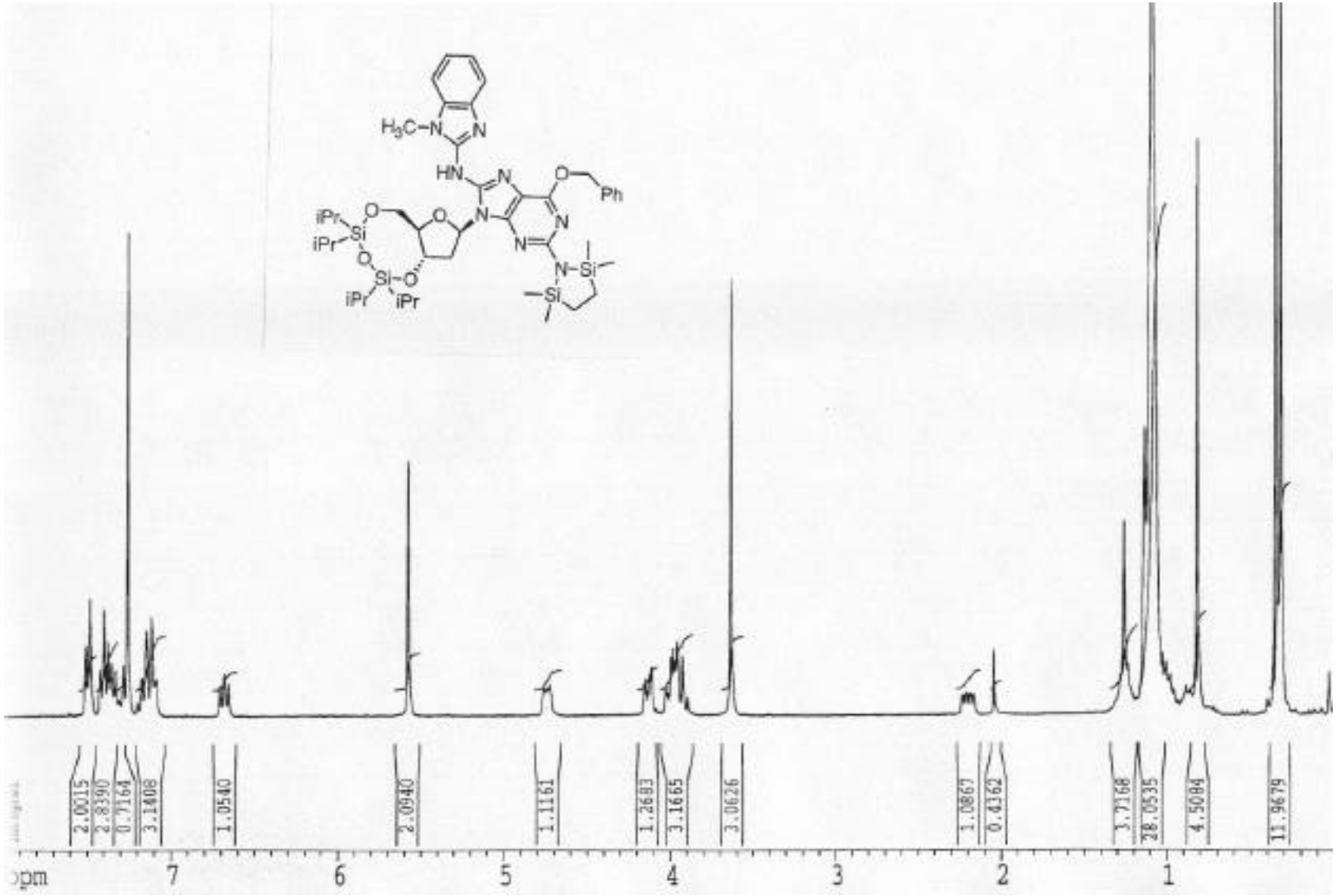
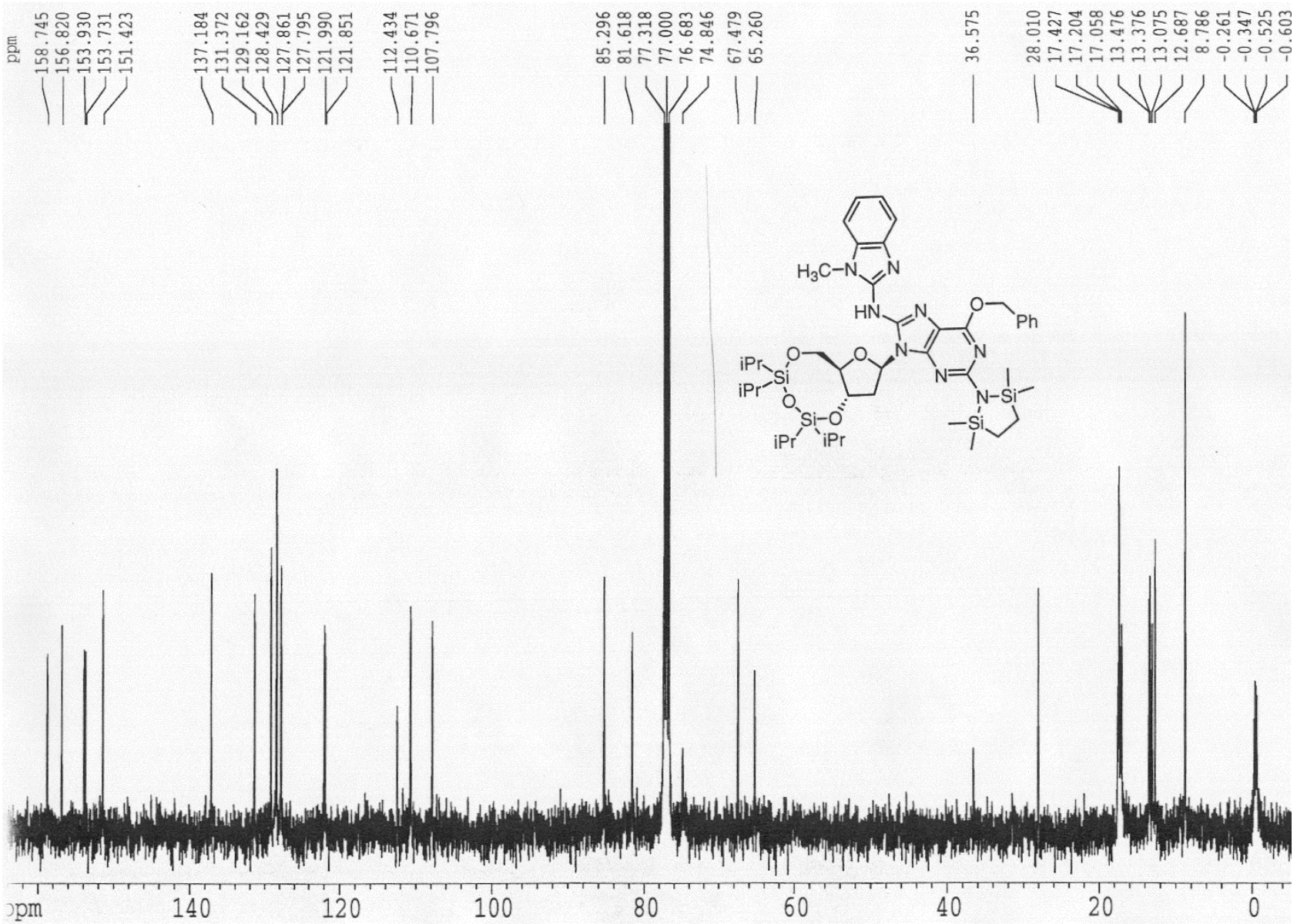
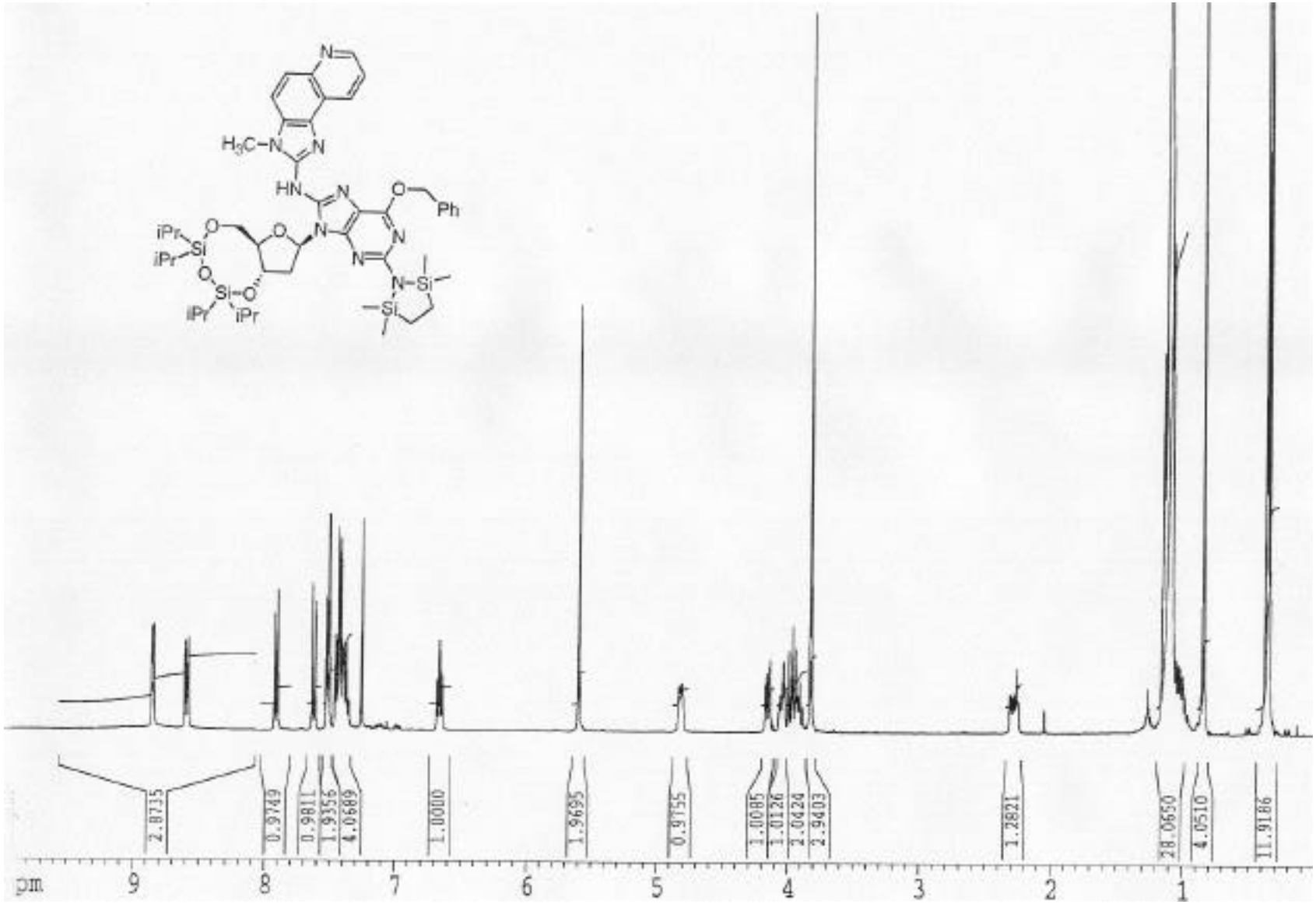
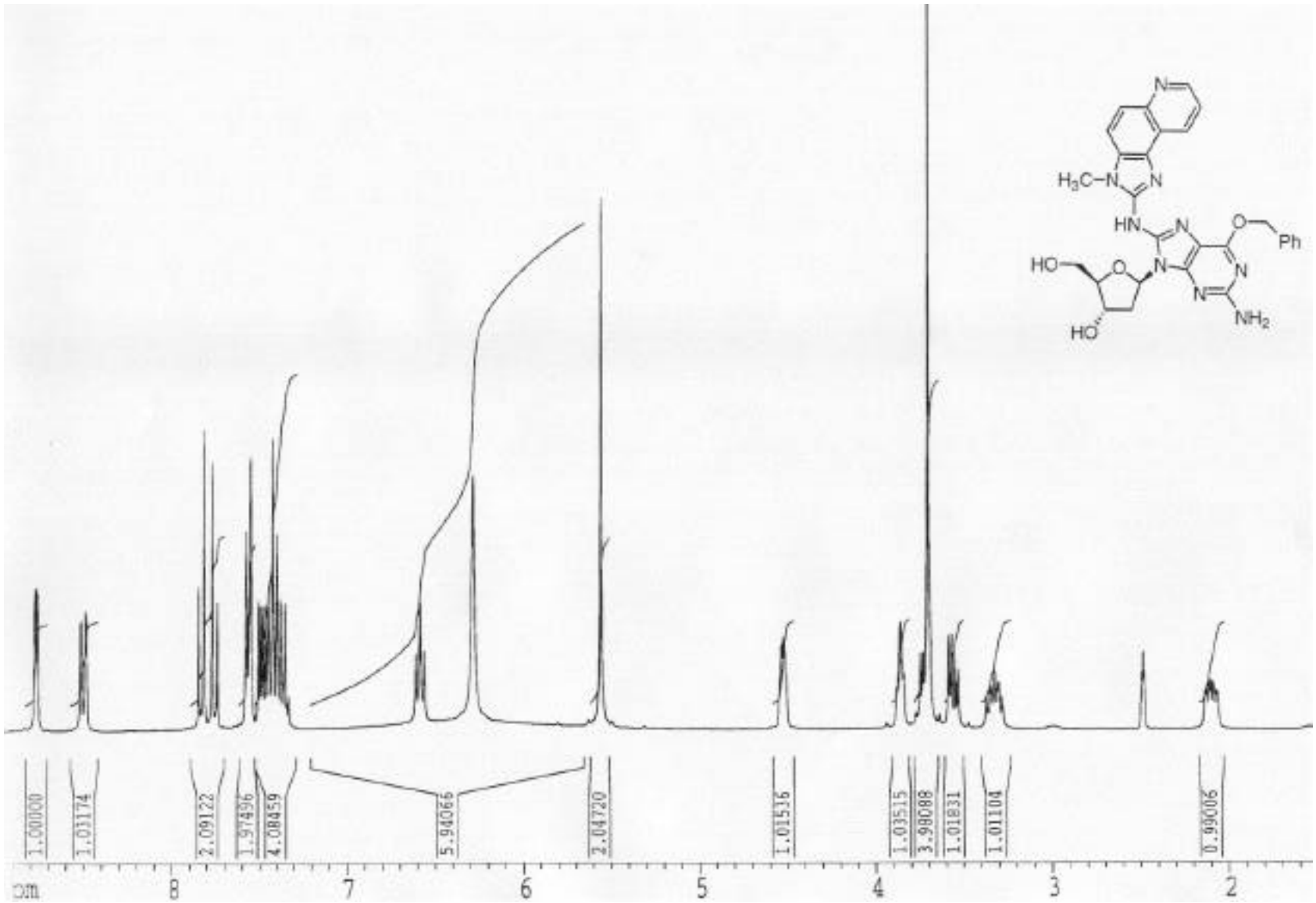
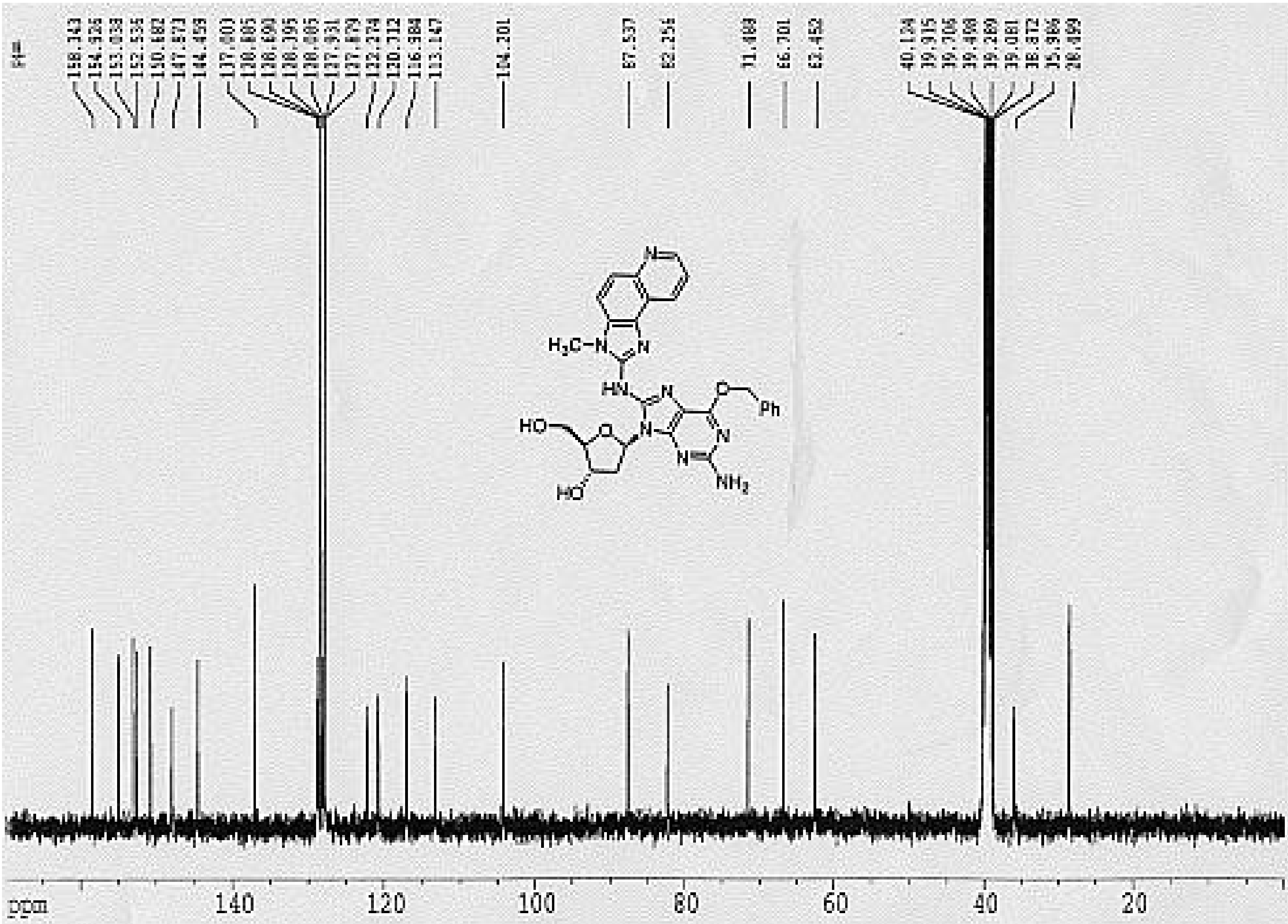

H3C-N
N
HN
N
O
Ph
iPr
iPr
Si
O
O
N
N
N
N
Si
O
Si
iPr
iPr
N-Si
Si
2.0015
2.8390
0.7164
3.1408
1.0540
2.0940
1.1161
1.2683
3.1665
3.0626
1.0867
0.4162
3.7168
28.0535
4.5084
11.9679
ppm
7
6
5
4
3
2
1


ppm
158.745
156.820
153.930
153.731
151.423
137.184
131.372
129.162
128.429
127.861
127.795
121.990
121.851
112.434
110.671
107.796
85.296
81.618
77.318
77.000
76.683
74.846
67.479
65.260
36.575
28.010
17.427
17.204
17.058
13.476
13.376
13.075
12.687
8.786
-0.261
-0.347
-0.525
-0.603
H3C–N
HN
O
Ph
iPr
Si
O
Si–O
N–Si
140
120
100
80
60
40
20
0

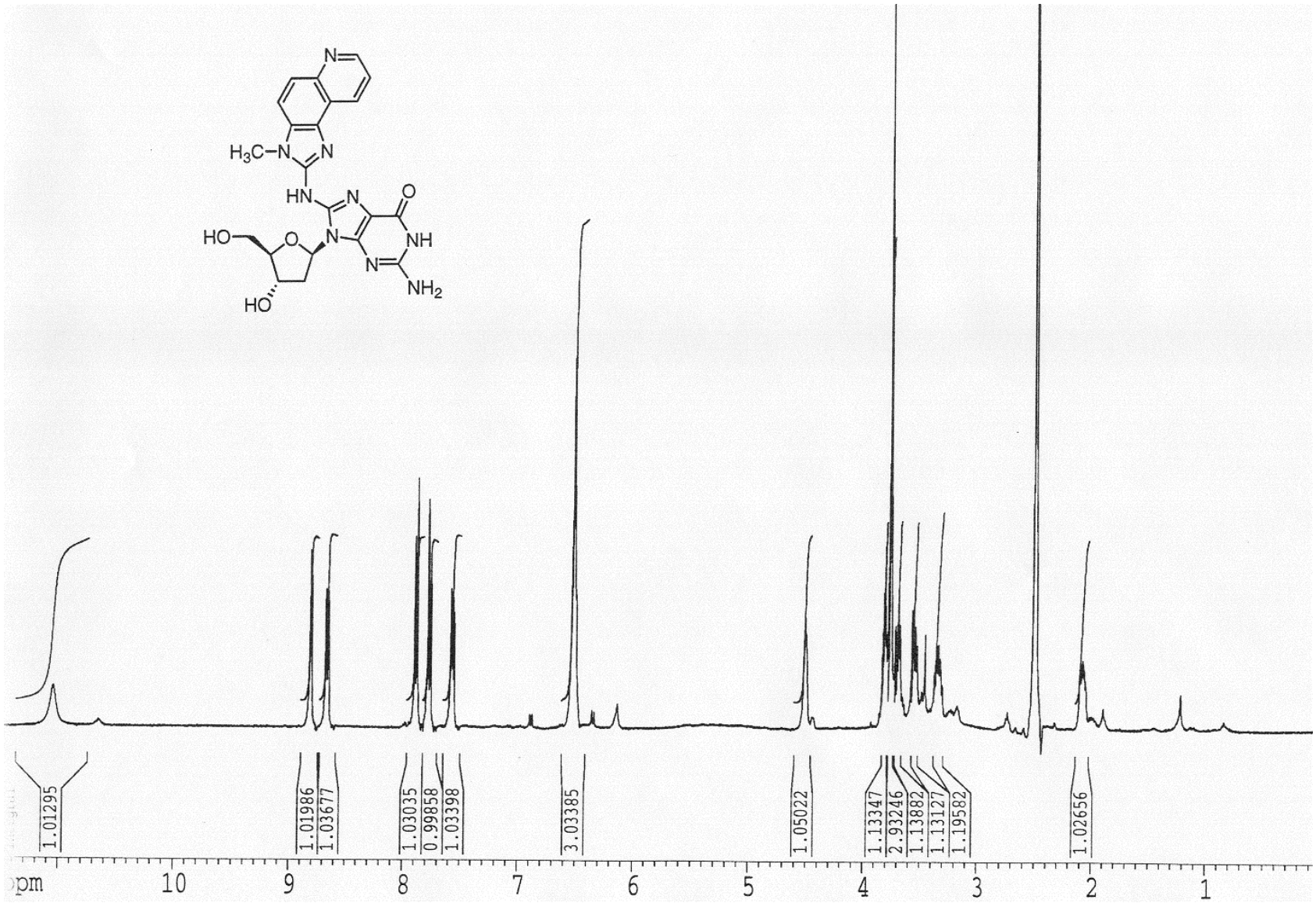

N
H3C–N
N
HN
N
O
Ph
iPr
iPr
Si
O
O
N
N
N
O
Si–O
iPr
iPr
N–Si
Si
2.8735
0.9749
0.9811
1.9356
4.0689
1.0000
1.9695
0.9755
1.0085
1.0126
2.0424
2.9403
1.2821
28.0650
4.0510
11.9186
pm
9
8
7
6
5
4
3
2
1

-0.624
-0.410
8.666
12.675
13.356
13.494
17.047
17.083
17.198
17.374
17.414
17.509
28.658
36.642
65.123
67.491
74.682
76.679
76.996
77.199
77.314
81.363
85.514
105.706
112.281
118.006
120.561
122.883
127.236
127.913
128.381
128.486
129.536
129.815
136.920
145.110
148.081
151.554
152.567
154.019
154.672
159.695
Ph
Si
iPr
HN
H_3C
150
100
50

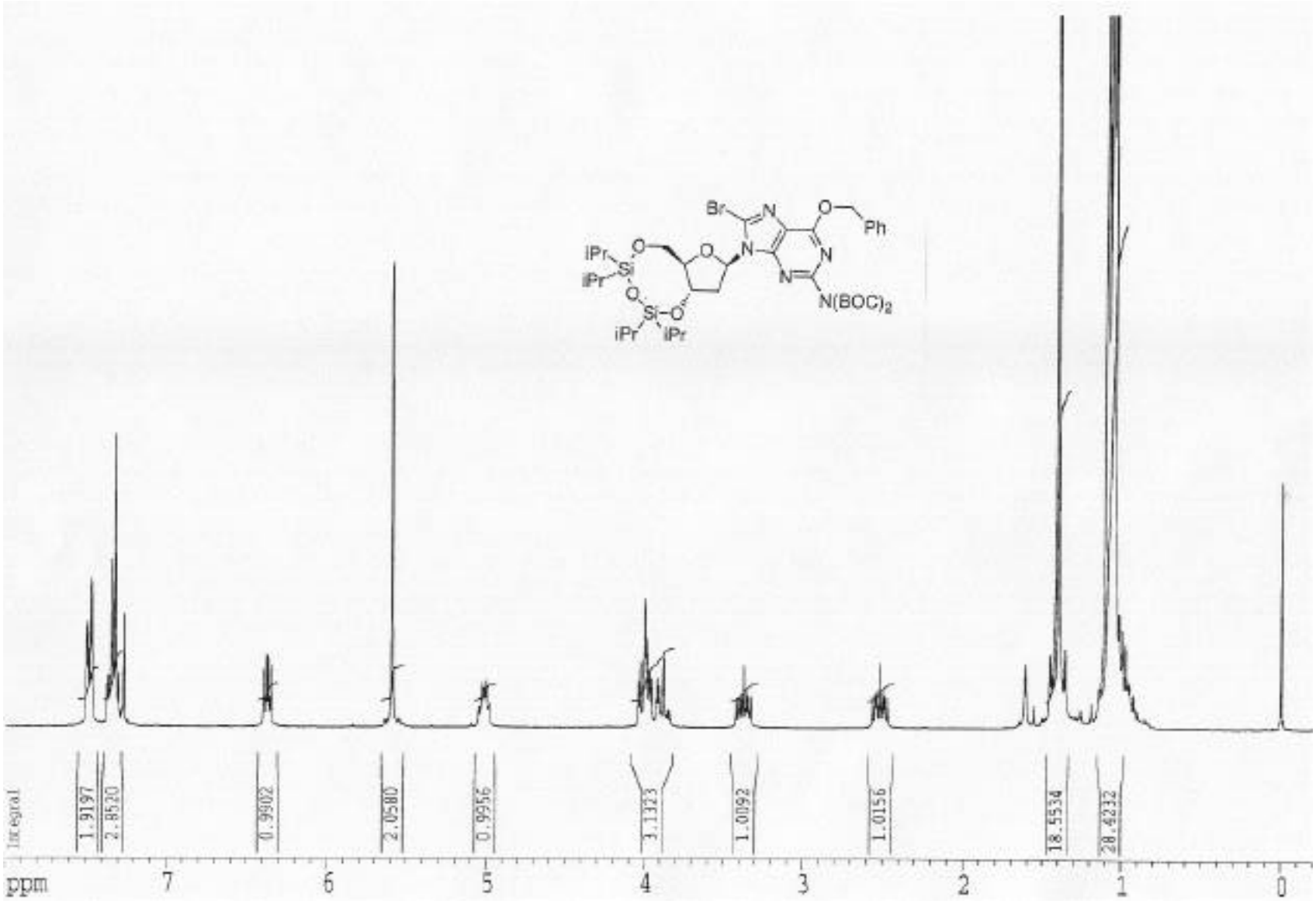

1.00000
1.01174
2.09122
1.97496
4.08459
5.94066
2.04720
1.01536
1.03515
3.98088
1.01831
1.01104
0.99006
8
7
6
5
4
3
2
N
H3C–N
N
HN
N
O
Ph
HO
O
N
N
N
N
NH2
HO

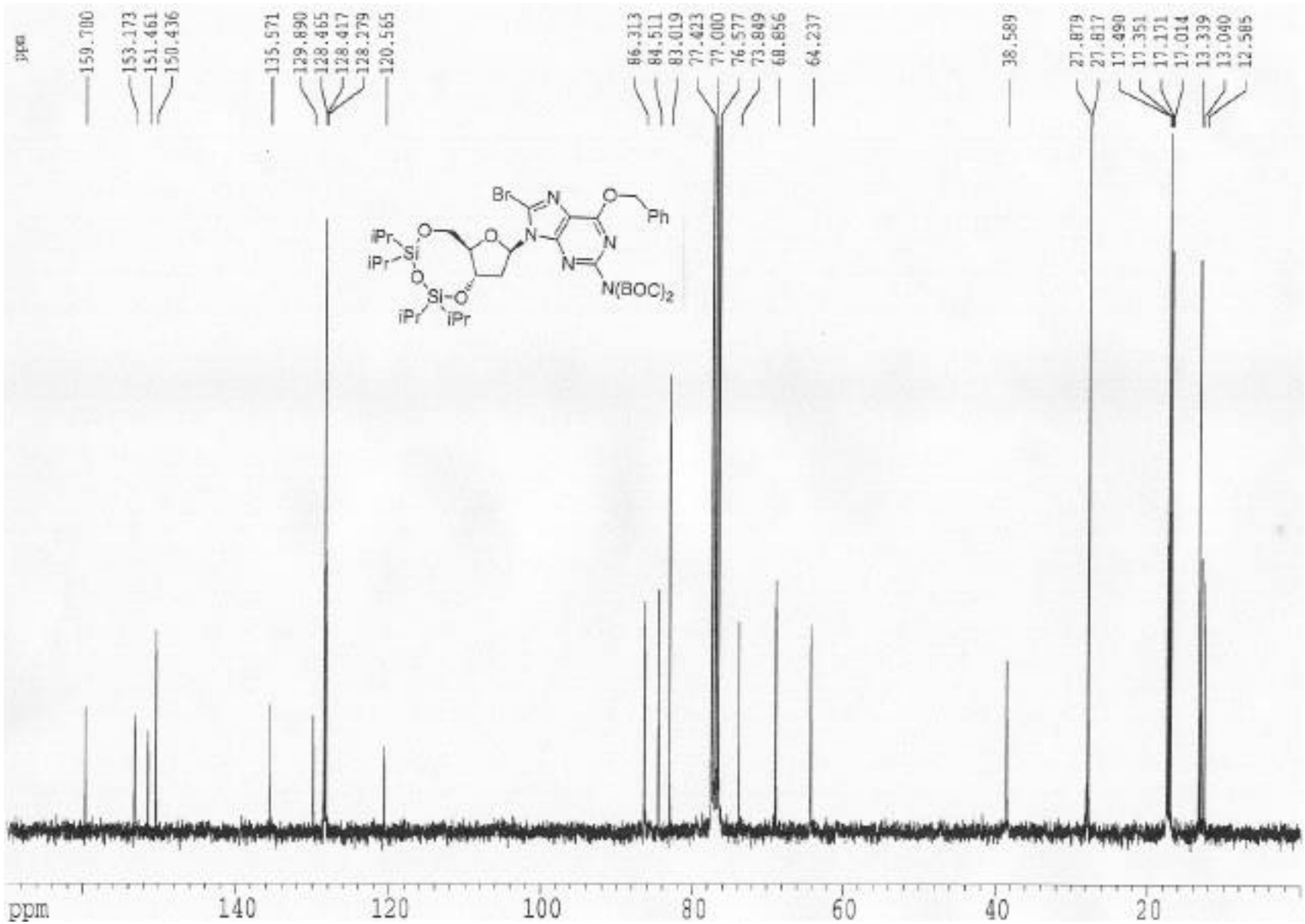


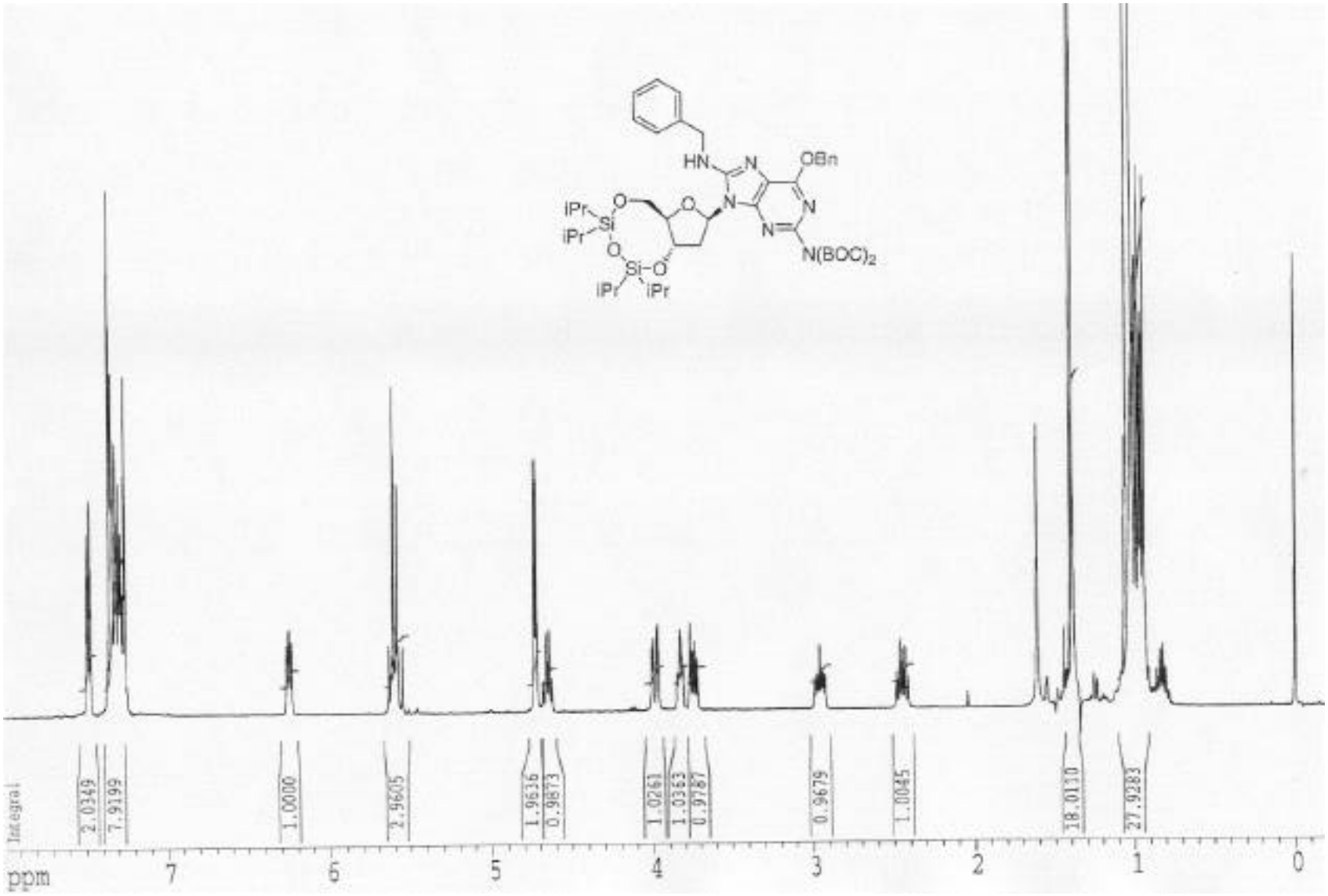
ppm
158.343
154.926
153.038
152.536
150.682
147.873
144.459
137.003
128.805
128.690
128.395
128.085
127.931
127.879
122.274
120.312
116.984
113.147
104.201
87.537
82.256
71.488
66.701
62.452
40.124
39.915
39.706
39.498
39.289
39.081
38.872
35.986
28.499
H_3C–N
N
HN
N
O
Ph
HO
O
N
N
N
HO
NH_2
ppm
140
120
100
80
60
40
20


N
H3C–N
N
HN
N
O
HO
O
N
NH
N
NH2
HO
1.01295
1.01986
1.03677
1.03035
0.99858
1.03398
3.03385
1.05022
1.13347
2.93246
1.13882
1.13127
1.19582
1.02656
ppm
10
9
8
7
6
5
4
3
2
1

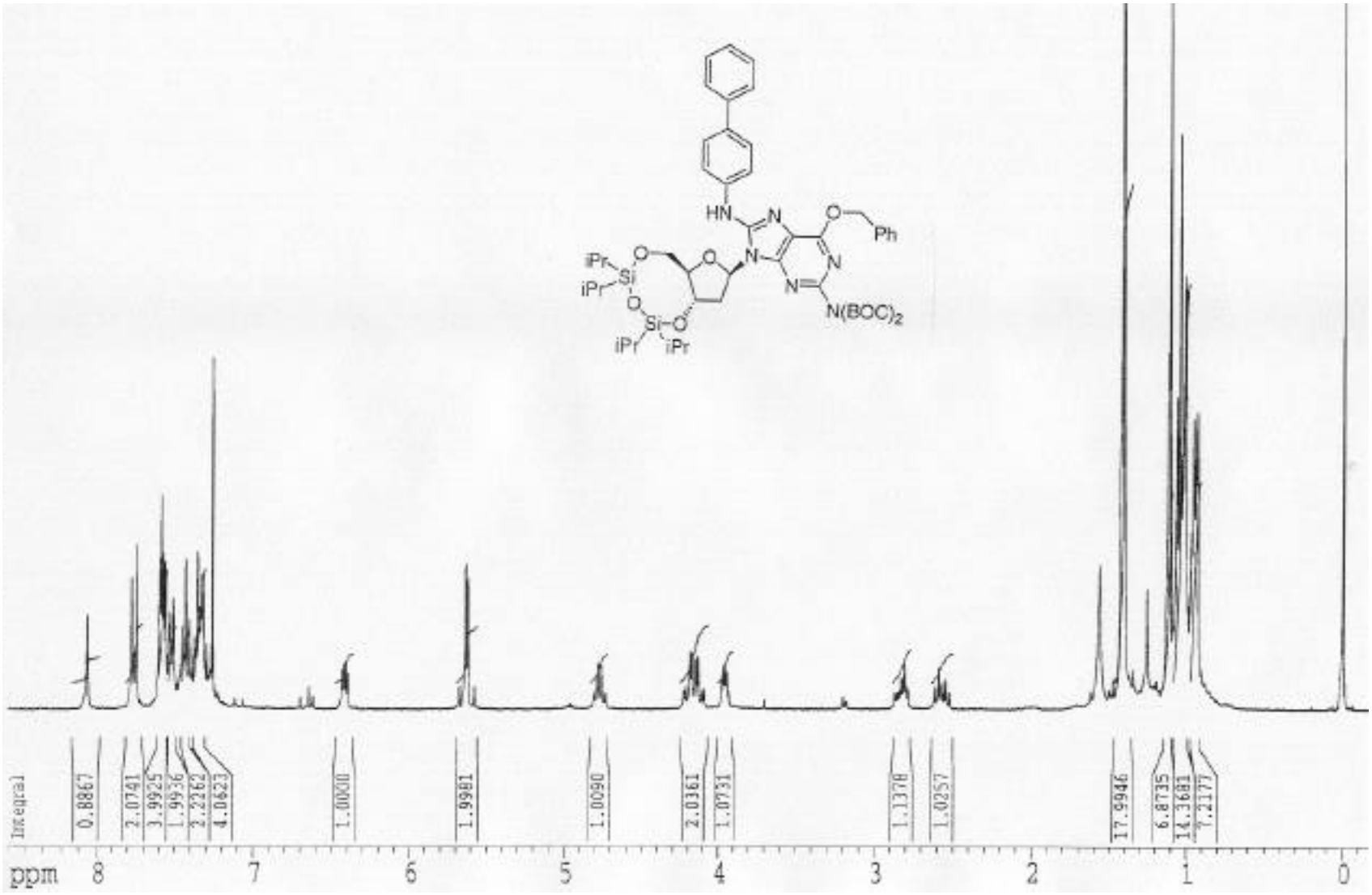
28.583
35.855
38.878
39.087
39.295
39.504
39.712
39.921
40.130
62.342
71.318
81.935
87.397
104.052
113.170
117.729
120.795
121.752
128.457
129.111
130.684
144.553
147.776
148.592
149.189
152.808
153.381
154.109
ppm
20
40
60
80
100
120
140
NH
NH2
O
N
HN
H3C
OH
HO

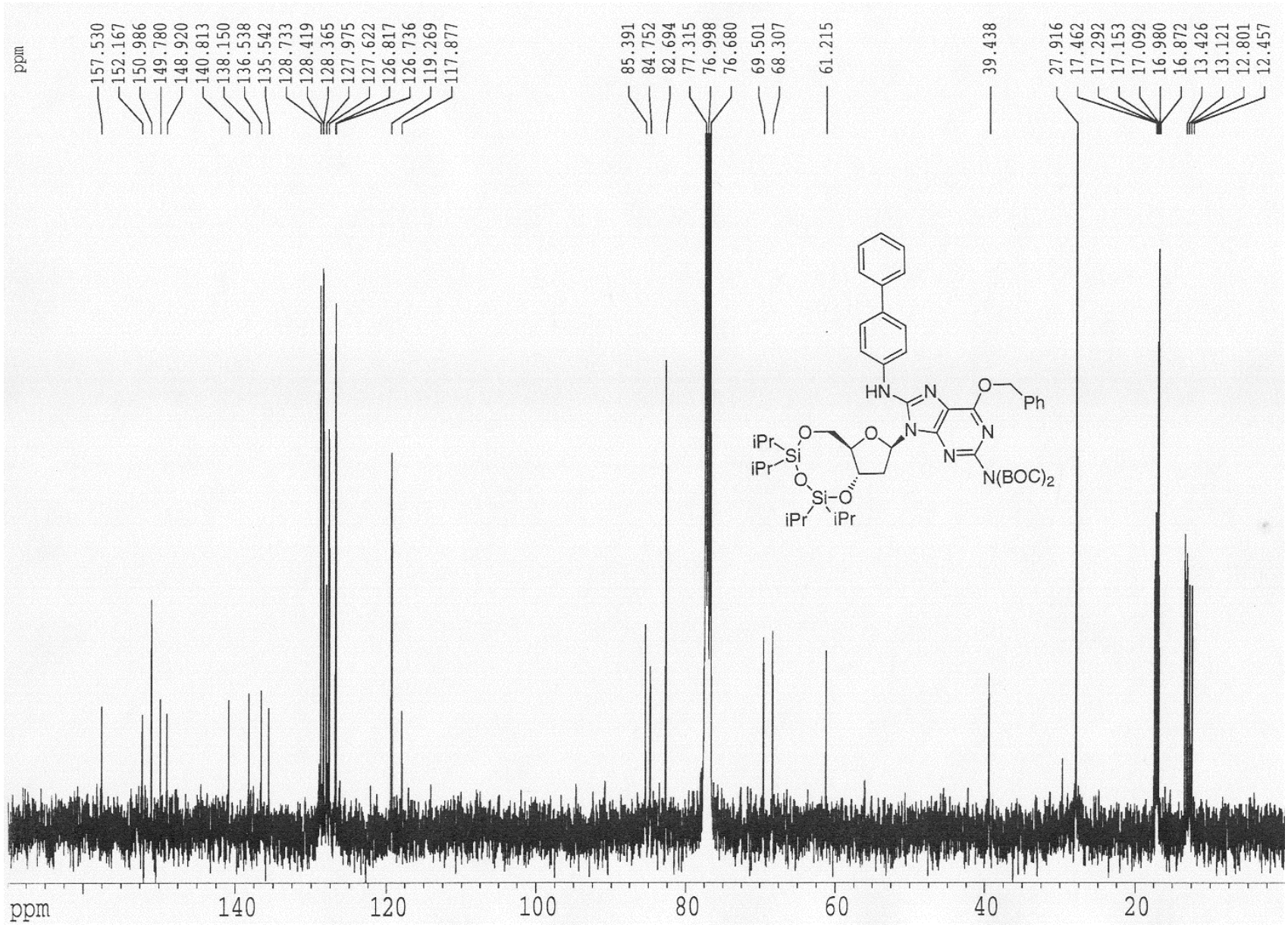

Br
N
O
Ph
iPr
Si
iPr
O
O
N
N
N(BOC)2
Si
O
iPr
iPr
Integral
1.9197
2.8520
0.9902
2.0580
0.9956
3.1333
1.0092
1.0156
18.5534
20.4232
ppm
7
6
5
4
3
2
1
0

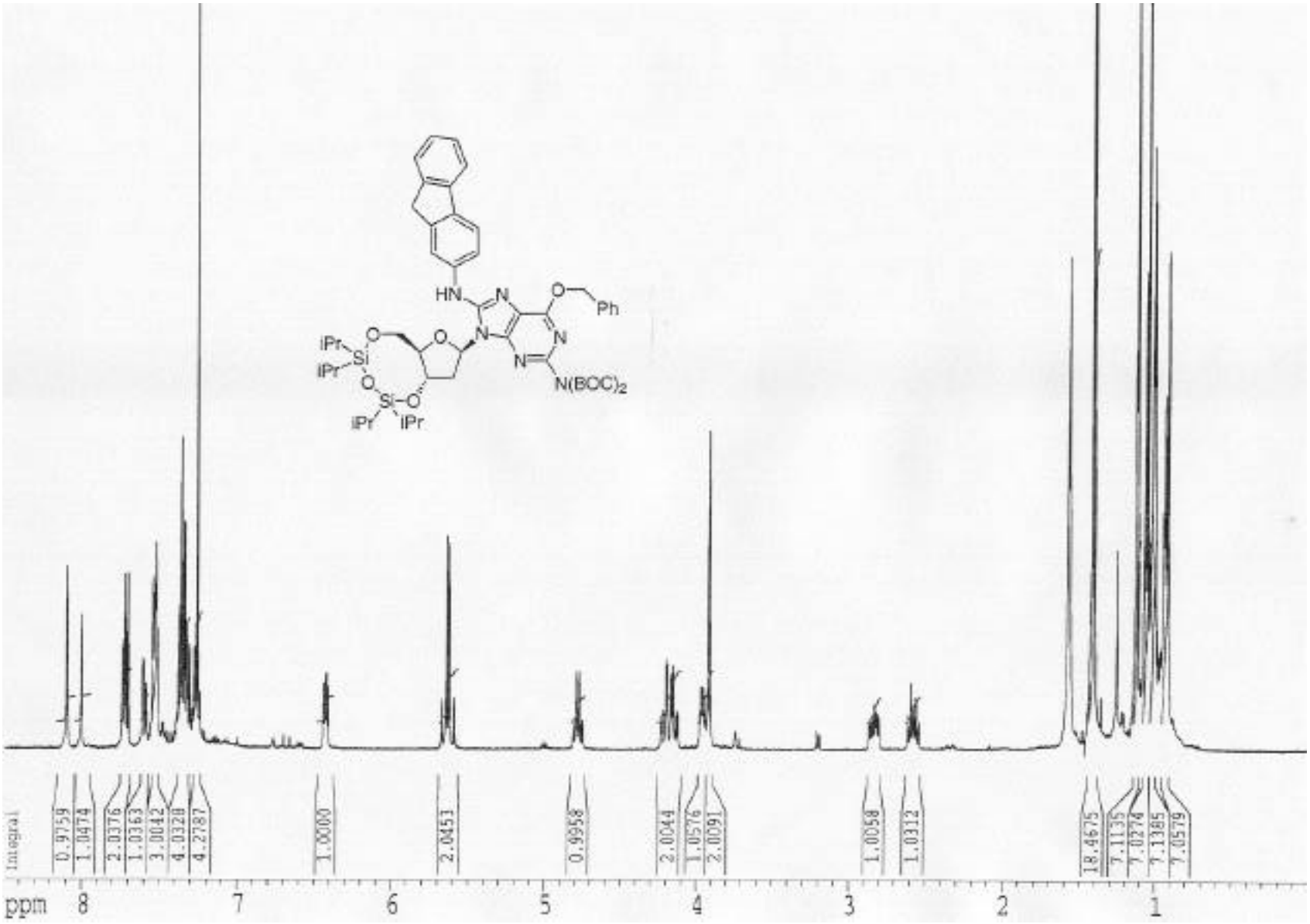

ppm
159.700
153.173
151.461
150.436
135.571
129.890
128.465
128.417
128.279
120.565
86.313
84.511
83.019
77.423
77.000
76.577
73.849
68.856
64.237
38.589
27.879
27.817
17.490
17.351
17.171
17.014
13.339
13.040
12.585
Br
N
O
Ph
iPr
Si
O
N(BOC)2
ppm
140
120
100
80
60
40
20

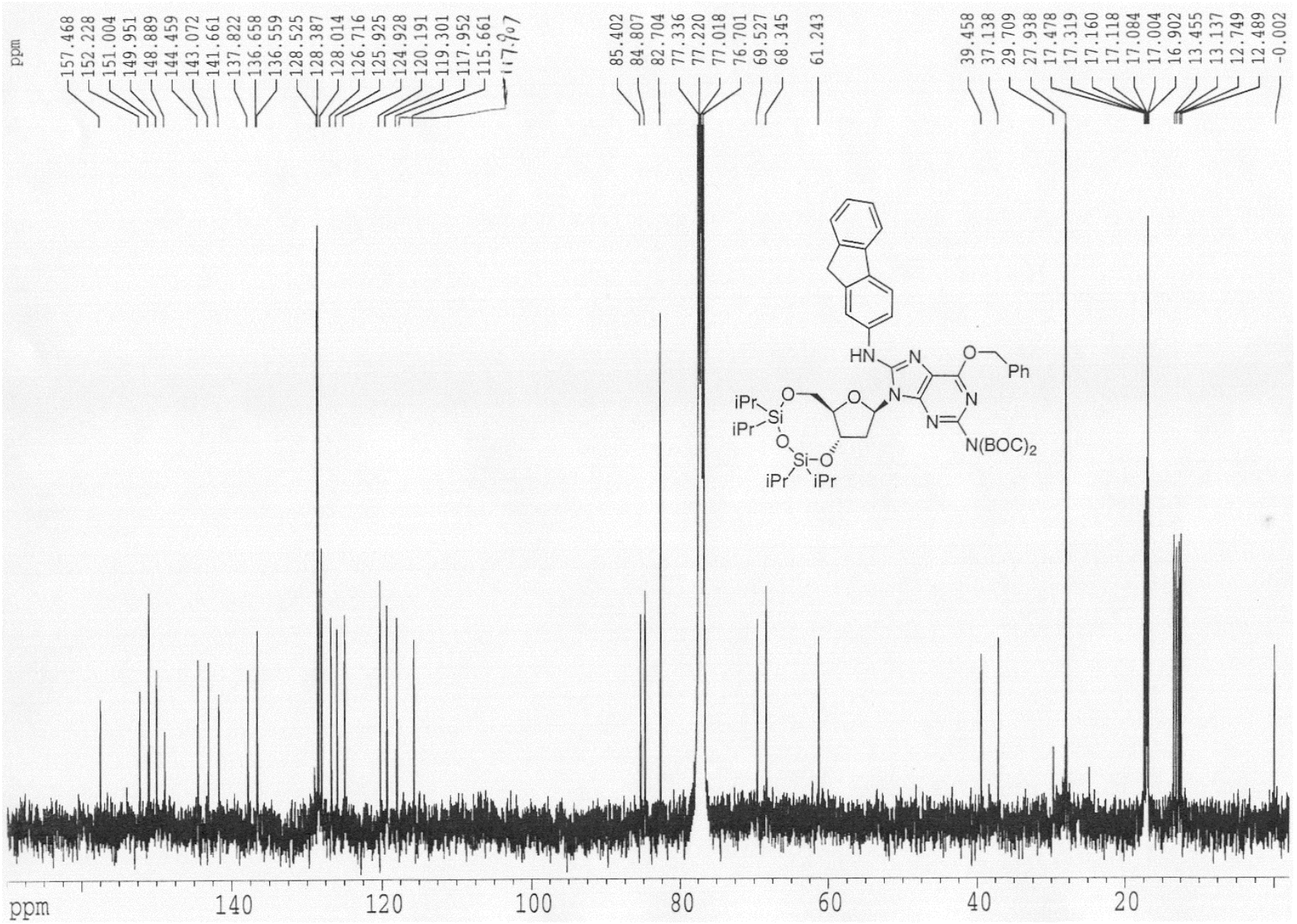

HN
N
OBn
N
N
N
N(BOC)2
O
O
iPr
Si
iPr
O
Si
O
iPr
iPr
Integral
2.0349
7.9199
1.0000
2.9605
1.9636
0.9873
1.0261
1.0353
0.9787
0.9679
1.0045
18.0110
27.9283
ppm
7
6
5
4
3
2
1
0

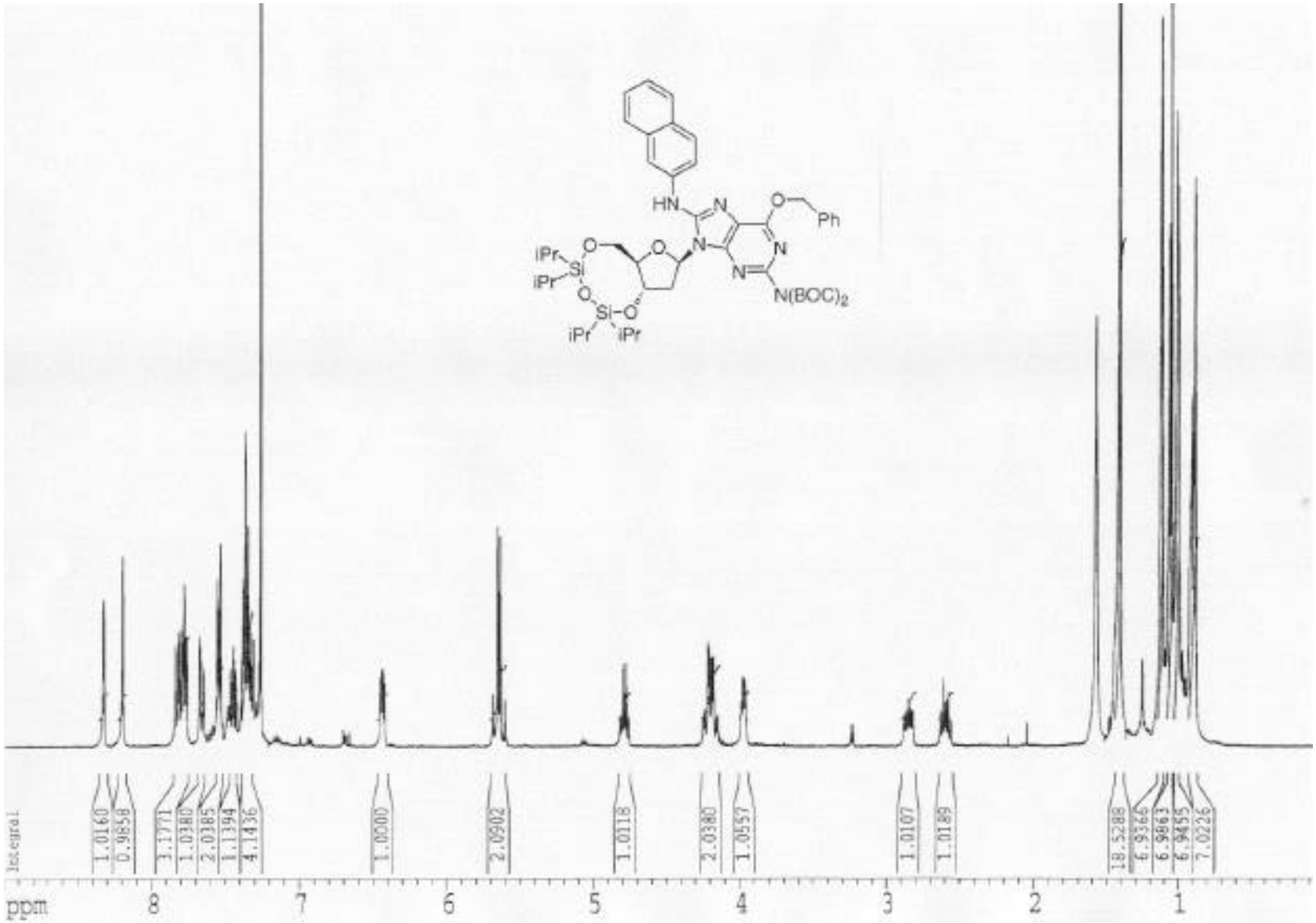
12.438
12.939
13.094
13.298
16.856
16.925
17.026
17.157
17.220
17.306
17.480
27.908
38.569
46.860
62.252
68.074
71.242
76.680
76.998
77.315
82.593
83.301
85.022
118.154
127.432
127.505
127.877
128.294
128.450
128.658
136.671
138.288
148.154
151.033
152.902
153.898
156.971
OBn
N(BOC)2
HN
iPr
Si
O
ppm
20
40
60
80
100
120
140


HN
N
O
Ph
N
N
N
N(BOC)2
iPr
iPr
Si
O
O
Si
O
iPr
iPr
Integral
0.8867
2.0741
3.9925
1.9936
2.2262
4.0623
1.0000
1.9981
1.0090
2.0361
1.0731
1.1378
1.0257
17.9946
6.8735
14.3681
7.2177
ppm
8
7
6
5
4
3
2
1
0


ppm
157.530
152.167
150.986
149.780
148.920
140.813
138.150
136.538
135.542
128.733
128.419
128.365
127.975
127.622
126.817
126.736
119.269
117.877
85.391
84.752
82.694
77.315
76.998
76.680
69.501
68.307
61.215
39.438
27.916
17.462
17.292
17.153
17.092
16.980
16.872
13.426
13.121
12.801
12.457
HN
N
O
Ph
N
N
N
N(BOC)2
O
O
iPr
iPr
Si
O
Si
O
iPr
iPr
ppm
140
120
100
80
60
40
20


HN
N
O
Ph
N
N
N
N(BOC)2
O
Si
iPr
iPr
O
O
Si
O
iPr
iPr
Integral
0.9759
1.0474
2.0376
1.0363
3.0042
4.0328
4.2787
1.0000
2.0453
0.9958
2.0044
1.0576
2.0091
1.0058
1.0312
18.4675
7.1136
7.0274
7.1385
7.0579
ppm
8
7
6
5
4
3
2
1


ppm
157.468
152.228
151.004
149.951
148.889
144.459
143.072
141.661
137.822
136.658
136.559
128.525
128.387
128.014
126.716
125.925
124.928
120.191
119.301
117.952
115.661
85.402
84.807
82.704
77.336
77.220
77.018
76.701
69.527
68.345
61.243
39.458
37.138
29.709
27.938
17.478
17.319
17.160
17.118
17.084
17.004
16.902
13.455
13.137
12.749
12.489
-0.002
HN
N
O
Ph
iPr
Si
iPr
O
O
Si
O
iPr
iPr
N
N
N(BOC)2
ppm
140
120
100
80
60
40
20


HN
N
O
Ph
N
N
N
N(BOC)2
O
iPr
Si
iPr
O
O
Si
O
iPr
iPr
Integral
1.0160
0.9858
3.1771
1.0380
2.0385
1.1394
4.1436
1.0000
2.0902
1.0118
2.0380
1.0557
1.0107
1.0189
18.5288
6.9364
6.9963
6.9455
7.0226
ppm
8
7
6
5
4
3
2
1

12.464
12.678
13.102
13.424
16.874
16.973
17.015
17.089
17.286
17.445
27.912
39.493
61.145
68.376
69.400
76.681
76.999
77.202
77.316
82.732
84.895
85.392
114.646
117.867
119.896
124.256
126.316
127.469
127.517
128.010
128.395
128.447
128.696
129.938
134.153
136.205
136.503
148.969
149.747
150.973
152.178
157.567
Ph
N(BOC)2
HN
iPr
Si
O
ppm
20
40
60
80
100
120
140