Electronic Supplementary Material (ESI) for Green Chemistry.

Supporting Information

In situ CO_2 Capture and Transformation to Cyclic Carbonate using Flue Gas

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Contents

1. Experimental

1.1 Materials.

All chemicals were purchased from commercial sources and used without further treatment. Flue gas was purchased from Lanzhou Yulon Gas Co., Ltd where the flue gas is directly supplied from a coal power group and the Flue gas is CO_2 (19.4%), O_2 (1.8%), N_2 (78.8%).

1.2 Synthesis of $PP_{1,4}Br$.

For the synthesis of the $PP_{1,4}Br$, 10 mmol of 1-methylpiperidine (990 mg) and 100 mL ethanol were put into the round-bottomed flask, which was stirred in oil bath at 40 °C. Then 1.2 mmol n-butyl bromide (164 mg) was slowly added to the above solution, and the solution was continuously stirred for 24 h. Then, the ethanol and excess n-butyl bromide were removed by distillation at 80 °C using a rotary evaporator. It was then crystallized in acetone to produce a white powder. The resultant solid was washed 3 times with acetone to obtain pure $PP_{1,4}Br$.

1.3 Coupling reactions of CO_2 and epoxides.

Styrene oxide (1200 mg, 10 mmol), $PP_{1,4}Br$ (100 mg, 0.42 mmol) and $ZnCl_2$ (20.4 mg, 0.15 mmol) were put into a glass tube which was placed in a 100 ml stainless-steel autoclave. After sealing, the autoclave was purged and charged with flue gas (typically 3.0 MPa). The reaction mixture was stirred at 60 °C for 18 h. After the reaction finished, the autoclave was cooled to room temperature and the excess gas was carefully released. Subsequently, the reaction mixture was diluted with 35 mL ethyl acetate and the reaction mixture was separated by centrifugation and analyzed quantitatively by a gas chromatograph (Agilent 7890A) equipped with a capillary column (HP-5) and an FID detector with biphenyl as an internal standard. The crude reaction mixture was concentrated by rot-vap and purified by column chromatography on a silica gel column to give the desired products.

1.4 Catalyst recycling experiments.

The used $PP_{1,4}Br/ZnCl_2$ was separated by centrifugation and washed 3 times with ethyl acetate, followed dry at 60 °C for 4 h. The catalyst was then used for the next catalytic run. Styrene oxide (1200 mg, 10 mmol), $PP_{1,4}Br/ZnCl_2$ (120 mg) were added into a glass tube which was placed in a 100 mL stainless-steel autoclave. Other steps were the same as the coupling reactions of CO_2 and epoxides.

1.5 Investigation of the water-resistance.

Styrene oxide (1200 mg, 10 mmol), $PP_{1,4}Br$ (100 mg, 0.42 mmol), quantified deionized water (1 mmol, 2 mmol, 3 mmol, 4 mmol, 5 mmol, 7 mmol, 10 mmol, 15 mmol) and $ZnCl_2$ (20.4 mg, 0.15 mmol) were put into a glass tube which was placed in a 100 mL stainless-steel autoclave. The other steps are the same as the coupling reactions of CO_2 and epoxides.

1.6 Scaled-up reactions of coupling reactions of CO_2 and epoxides.

$PP_{1,4}Br$ (4170 mg, 17.74 mmol), $ZnCl_2$ (851 mg, 6.26 mmol) and ethylene carbonate (325 g, 3.69 mol) were put into a glass tube which was placed in a 2 L stainless-steel autoclave. The mixture was stirred at 60 °C for 1 h. After stopping heating, the autoclave was cooled to room temperature. Styrene oxide (50 g, 0.417 mol) was added into the autoclave, and the reactor was purged and charged with flue gas up to 2.6 MPa followed by the addition of N_2 to 4.6 MPa. Afterward, the reaction mixture was heated and stirred at 60 °C for 18 h. After the reaction completed, the reactor was allowed to cool to room temperature. The gas was analyzed quantitatively for CO_2 by a gas chromatograph (Agilent 7890A) equipped with a capillary column (C Mol Sieve) and an TCD detector. The actual amount of CO_2 captured and converted is obtained from the mass after and before the reaction. The yield is obtained by dividing the amount of CO_2 actually captured and converted by the total amount of carbon dioxide injected.

$PP_{1,4}Br$ (6000 mg, 25.52 mmol), $ZnCl_2$ (1200 mg, 8.82 mmol) and ethylene carbonate (296 g, 3.36 mol) were put into a glass tube which was placed in a 2 L stainless-steel autoclave. The mixture was stirred at 60 °C for 1 h. After stopping heating, the autoclave was cooled to room temperature. Styrene oxide (72 g, 0.6 mol) was added into the autoclave. The reactor was purged and charged with flue gas up to 3.7 MPa. The other steps are the same as above.

$PP_{1,4}Br$ (7500 mg, 31.90 mmol), $ZnCl_2$ (1500 mg, 11.03 mmol) and ethylene carbonate (270 g, 3.07 mol) were put into a glass tube which was placed in a 2 L stainless-steel autoclave. The mixture was stirred at 60 °C for 1 h. After stopping heating, the autoclave was cooled to room temperature. Styrene oxide (90 g, 0.750 mmol) was added into the autoclave. The reactor was purged and charged with flue gas up to 4.8 MPa. The other steps are the same as above.

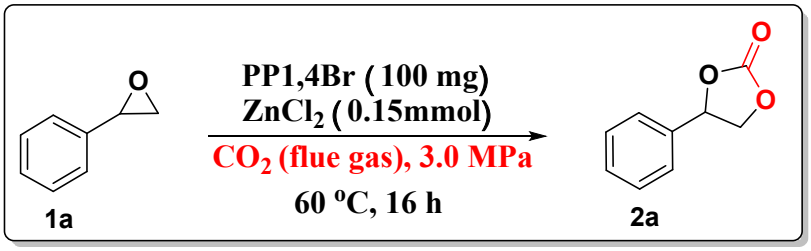
1.7 Analytic measurements

The liquid nuclear magnetic resonance spectra (NMR) were recorded on a Bruker AvanceTM III 400 MHz in deuterated chloroform unless otherwise noted. Data are reported in parts per million (ppm) as follows: chemical shift, multiplicity (s = singlet, d = doublet, t = triplet, q = quartet, quint = quintet, m = multiplet, dd = doublet of doublet and br = broad signal), coupling constant in Hz and integration.

The contents of Zn in the catalysts after use were measured by continuous light source atomic absorption spectrometer (CS AAS), using the Jena contrAA700.

2. Supplementary Table

Table S1. Screening of the pressure. [a]



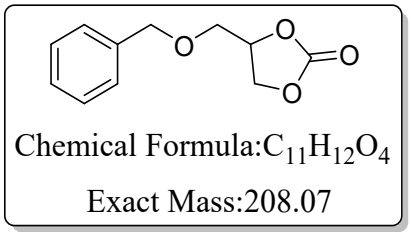
Entry	Pressure (MPa)	Yield(%)[b]
1	2	57
2	3	90
3	4	90

[a] Reaction conditions: 10 mmol substrates, $ZnCl_2$ (0.15 mmol), 100 mg catalyst, 3.0 MPa flue gas, 60 °C, 18 h. [b] The yield of the product 2a were calculated by GC.

Table S2. Zn contents.

Cycle	Zn contents (mg)
1	9.8
2	9.8
3	9.8
4	9.5

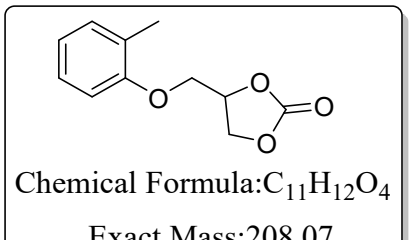
3. NMR data of the products



Chemical Formula:$C_{11}H_{12}O_4$

Exact Mass:208.07

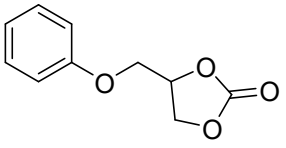
4-((benzyloxy)methyl)-1,3-dioxolan-2-one (2b)[1]: white solid, 2.060 g, 99% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 7.40 – 7.29 (m, 5H), 4.82 (m, 1H), 4.60 (q, J = 12.0 Hz, 2H), 4.52 – 4.36 (m, 2H), 3.67 (m, 2H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 155.18, 137.15, 128.69, 128.21, 127.87, 75.10, 73.79, 68.90, 66.40.



Chemical Formula:$C_{11}H_{12}O_4$

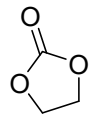
Exact Mass:208.07

4-((o-tolyloxy)methyl)-1,3-dioxolan-2-one (2c): white solid, 1.903 g, 91% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 7.15 (d, *J* = 2.6 Hz, 2H), 6.96 – 6.72 (m, 2H), 5.04 (dd, *J* = 8.2, 5.2 Hz, 1H), 4.68 – 4.53 (m, 2H), 4.18 (m, 2H), 2.21 (s, 3H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 155.87, 154.94, 131.18, 127.18, 126.99, 121.75, 110.95, 74.36, 67.12, 66.35, 16.06.



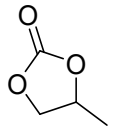
Chemical Formula:$C_{10}H_{10}O_4$
Exact Mass:194.06

4-(phenoxymethyl)-1,3-dioxolan-2-one (2d)[1]: white solid, 1.488 g, 76% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 7.35 – 7.27 (m, 2H), 7.02 (dd, *J* = 10.6, 4.1 Hz, 1H), 6.91 (m, 2H), 5.09 – 4.96 (m, 1H), 4.66 – 4.48 (m, 2H), 4.19 (m, 2H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 157.86, 154.81, 129.81, 122.09, 114.71, 74.24, 66.96, 66.34.



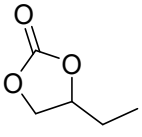
Chemical Formula:$C_3H_4O_3$
Exact Mass:88.02

1,3-dioxolan-2-one (2e)[2]: white solid, 0.880 g, 100% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 4.54 (s, 4H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 155.60, 64.74.



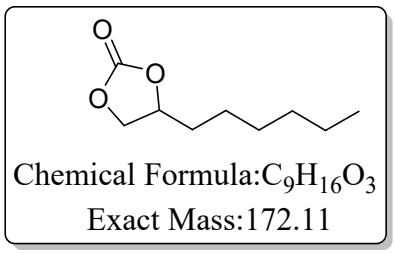
Chemical Formula:$C_4H_6O_3$
Exact Mass:102.03

4-methyl-1,3-dioxolan-2-one (2f)[1]: colorless liquid, 0.992 g, 95% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 4.93 – 4.80 (m, 1H), 4.57 (dd, *J* = 8.3, 7.8 Hz, 1H), 4.04 (dd, *J* = 8.4, 7.3 Hz, 1H), 1.50 (d, *J* = 6.3 Hz, 3H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 155.14, 73.64, 70.75, 19.53.

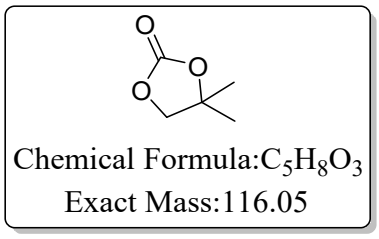


Chemical Formula:$C_5H_8O_3$
Exact Mass:116.05

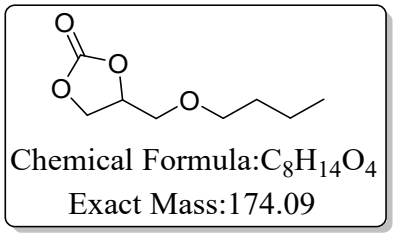
4-ethyl-1,3-dioxolan-2-one (2g)[1]: colorless liquid, 1.163 g, 98% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 4.80 – 4.63 (m, 1H), 4.59 – 4.49 (m, 1H), 4.10 (dd, *J* = 8.4, 7.0 Hz, 1H), 1.96 – 1.68 (m, 2H), 1.04 (t, *J* = 7.5 Hz, 3H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 155.23, 78.12, 69.12, 27.03, 8.58.



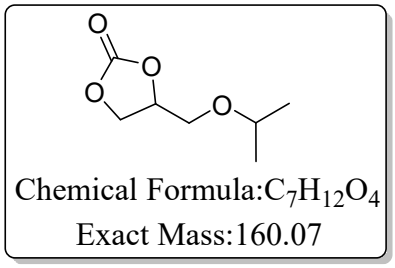
4-hexyl-1,3-dioxolan-2-one (2h)[1]: colorless liquid, 1.142 g, 66% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 4.71 (m, 1H), 4.53 (t, *J* = 8.1 Hz, 1H), 4.07 (dd, *J* = 8.3, 7.3 Hz, 1H), 1.91 – 1.61 (m, 2H), 1.53 – 1.23 (m, 8H), 0.89 (t, *J* = 6.9 Hz, 3H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 155.22, 77.48, 69.51, 33.99, 31.62, 28.90, 24.43, 22.56, 14.10.



4,4-dimethyl-1,3-dioxolan-2-one (2i)[1]: colorless liquid, 0.370 g, 32% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 4.16 (s, 2H), 1.53 (s, 6H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 154.73, 81.81, 75.51, 26.18.

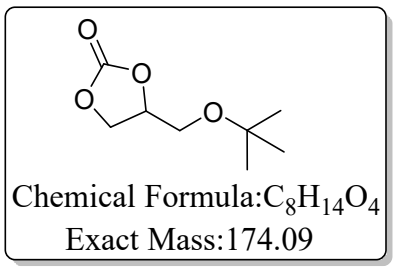


4-(butoxymethyl)-1,3-dioxolan-2-one (2j)[3]: yellowish liquid, 1.704 g, 97% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 4.91 – 4.74 (m, 1H), 4.50 (t, *J* = 8.3 Hz, 1H), 4.40 (dd, *J* = 8.3, 6.1 Hz, 1H), 3.64 (m, 2H), 3.51 (t, *J* = 6.5 Hz, 2H), 1.56 (m, 2H), 1.36 (m, 2H), 0.92 (t, *J* = 7.4 Hz, 3H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 155.12, 75.24, 71.97, 69.71, 66.40, 31.60, 19.23, 13.92.



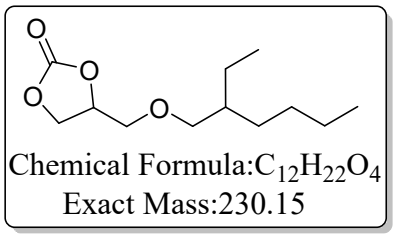
Chemical Formula:$C_7H_{12}O_4$
Exact Mass:160.07

4-(isopropoxymethyl)-1,3-dioxolan-2-one (2k)[2]: colorless liquid, 1.522 g, 95% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 4.81 (m, 1H), 4.50 (t, *J* = 8.3 Hz, 1H), 4.39 (dd, *J* = 8.2, 6.1 Hz, 1H), 3.73 – 3.54 (m, 3H), 1.17 (d, *J* = 6.0 Hz, 6H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 155.20, 75.29, 73.04, 67.21, 66.53, 21.98, 21.88.



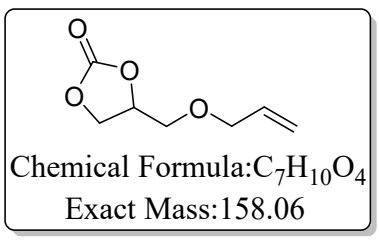
Chemical Formula:$C_8H_{14}O_4$
Exact Mass:174.09

4-(tert-butoxymethyl)-1,3-dioxolan-2-one (2l)[1]: colorless liquid, 1.612 g, 92% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 4.87 – 4.68 (m, 1H), 4.48 (t, *J* = 8.2 Hz, 1H), 4.39 (dd, *J* = 8.2, 5.8 Hz, 1H), 3.58 (m, 2H), 1.20 (s, 9H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 155.29, 75.29, 73.98, 66.66, 61.38, 27.39.



Chemical Formula:$C_{12}H_{22}O_4$
Exact Mass:230.15

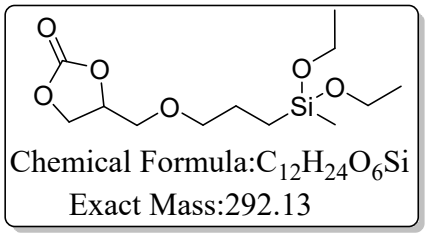
4-(((2-ethylhexyl)oxy)methyl)-1,3-dioxolan-2-one (2m)[3]: colorless liquid, 2.126 g, 92% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 4.81 (m, 1H), 4.49 (t, *J* = 8.3 Hz, 1H), 4.39 (m, 1H), 3.63 (m, 2H), 3.44 – 3.29 (m, 2H), 1.50 (m, 1H), 1.41 – 1.17 (m, 8H), 0.88 (dt, *J* = 10.1, 7.0 Hz, 6H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 155.11, 75.21, 74.97, 74.94, 70.02, 66.43, 39.70, 30.54, 30.51, 29.16, 23.86, 23.13, 14.18, 11.17, 11.16.



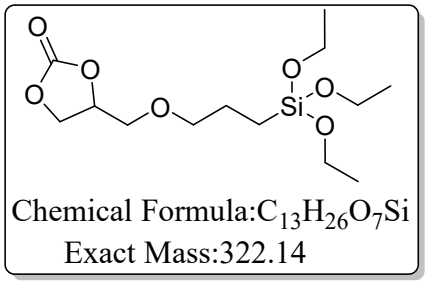
Chemical Formula:$C_7H_{10}O_4$
Exact Mass:158.06

4-((allyloxy)methyl)-1,3-dioxolan-2-one (2n)[1]: yellowish liquid, 1.550 g, 98% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 5.88 (m, 1H), 5.26 (m, 2H), 4.98 – 4.74 (m, 1H), 4.51 (t, *J* = 8.4 Hz, 1H), 4.50 – 4.33 (m, 1H), 4.17

– 4.01 (m, 2H), 3.66 (m, 2H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 155.05, 133.77, 118.04, 75.14, 72.70, 68.95, 66.39.

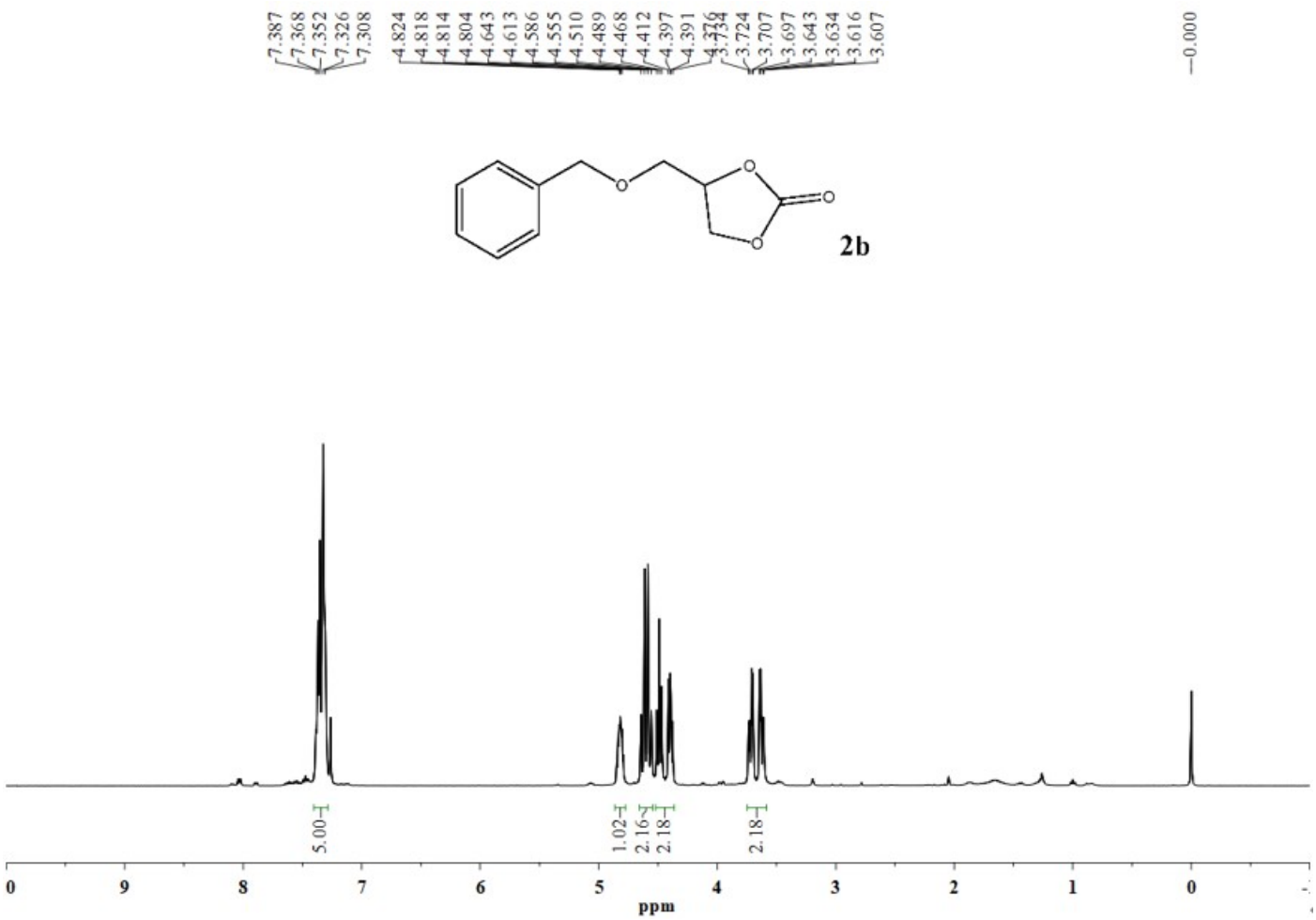


4-((3-(diethoxy(methyl)silyl)propoxy)methyl)-1,3-dioxolan-2-one (2o)[1]: yellowish liquid, 0.876 g, 60% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 5.04 – 4.67 (m, 1H), 4.60 – 4.29 (m, 2H), 3.88 – 3.39 (m, 8H), 1.80 – 1.58 (m, 2H), 1.23 (dd, *J* = 14.9, 7.9 Hz, 6H), 0.61 (dd, *J* = 10.0, 6.8 Hz, 2H), 0.12 (s, 3H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 155.06, 75.16, 74.45, 69.68, 66.41, 58.23, 23.02, 18.48, 9.87, -4.86.



4-((3-(triethoxysilyl)propoxy)methyl)-1,3-dioxolan-2-one (2p)[1]: yellowish liquid, 0.879 g, 55% yield. **^{1}H NMR** (400 MHz, $CDCl_3$) δ 4.84 – 4.75 (m, 1H), 4.50 (t, *J* = 8.3 Hz, 1H), 4.40 (dd, *J* = 8.2, 6.2 Hz, 1H), 3.82 (q, *J* = 7.0 Hz, 6H), 3.65 (dd, *J* = 17.7, 3.9 Hz, 2H), 3.49 (m, 2H), 1.81 – 1.63 (m, 2H), 1.23 (t, *J* = 7.0 Hz, 9H), 0.64 (dd, *J* = 9.5, 7.1 Hz, 2H). **^{13}C NMR** (101 MHz, $CDCl_3$) δ 155.06, 75.14, 74.20, 69.66, 66.42, 58.50, 22.97, 18.39, 6.44.

4. NMR spectra of the products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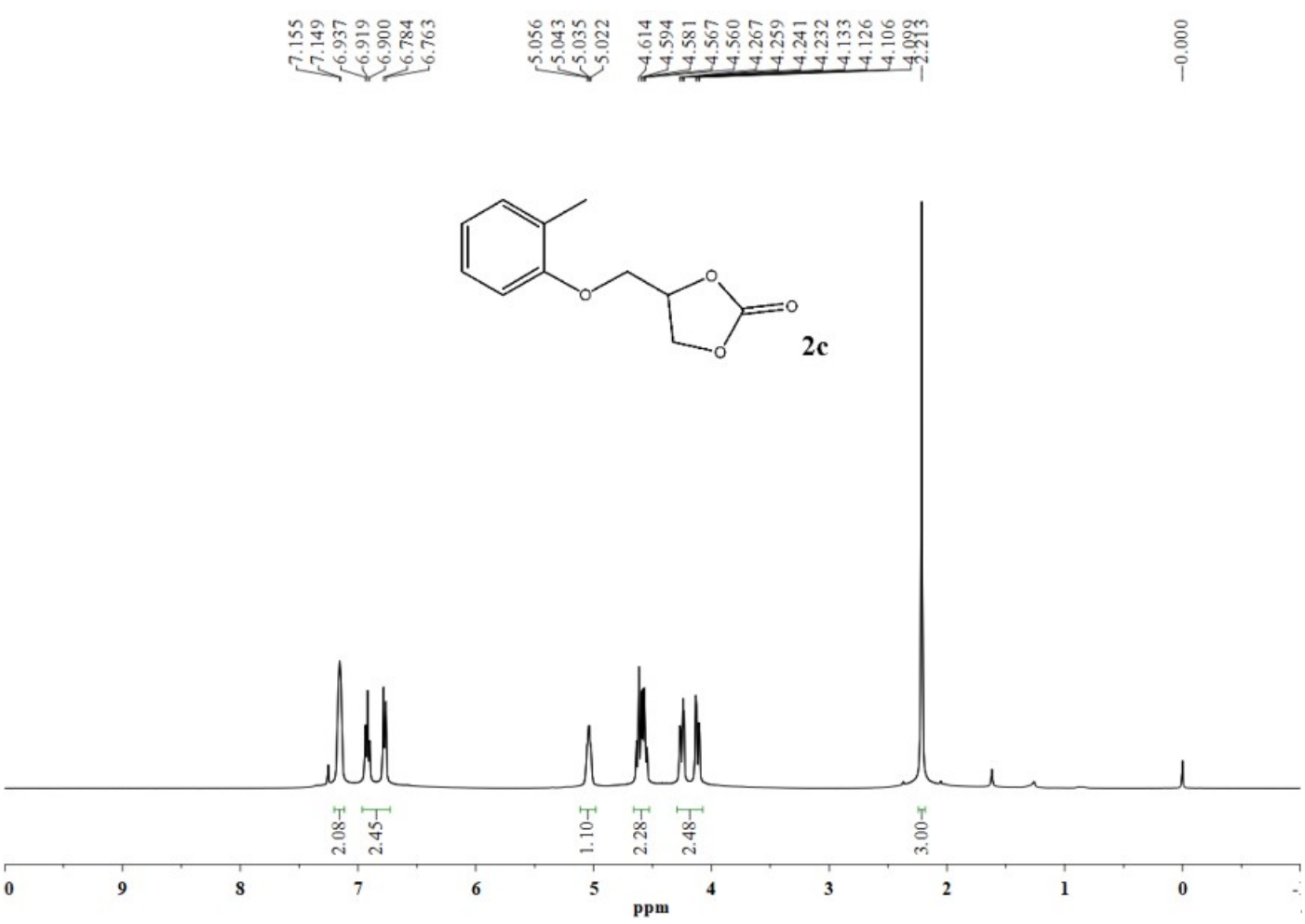
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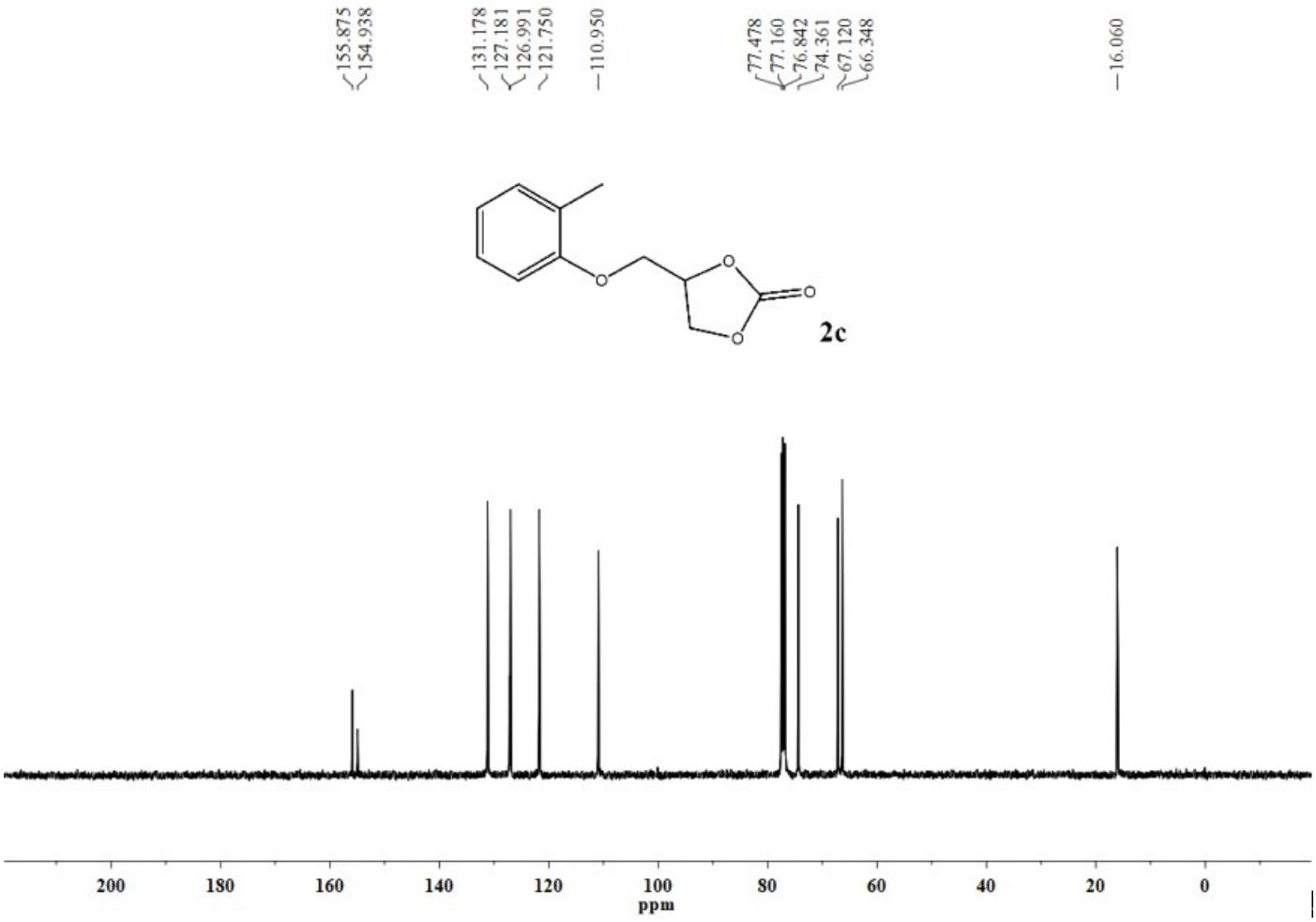
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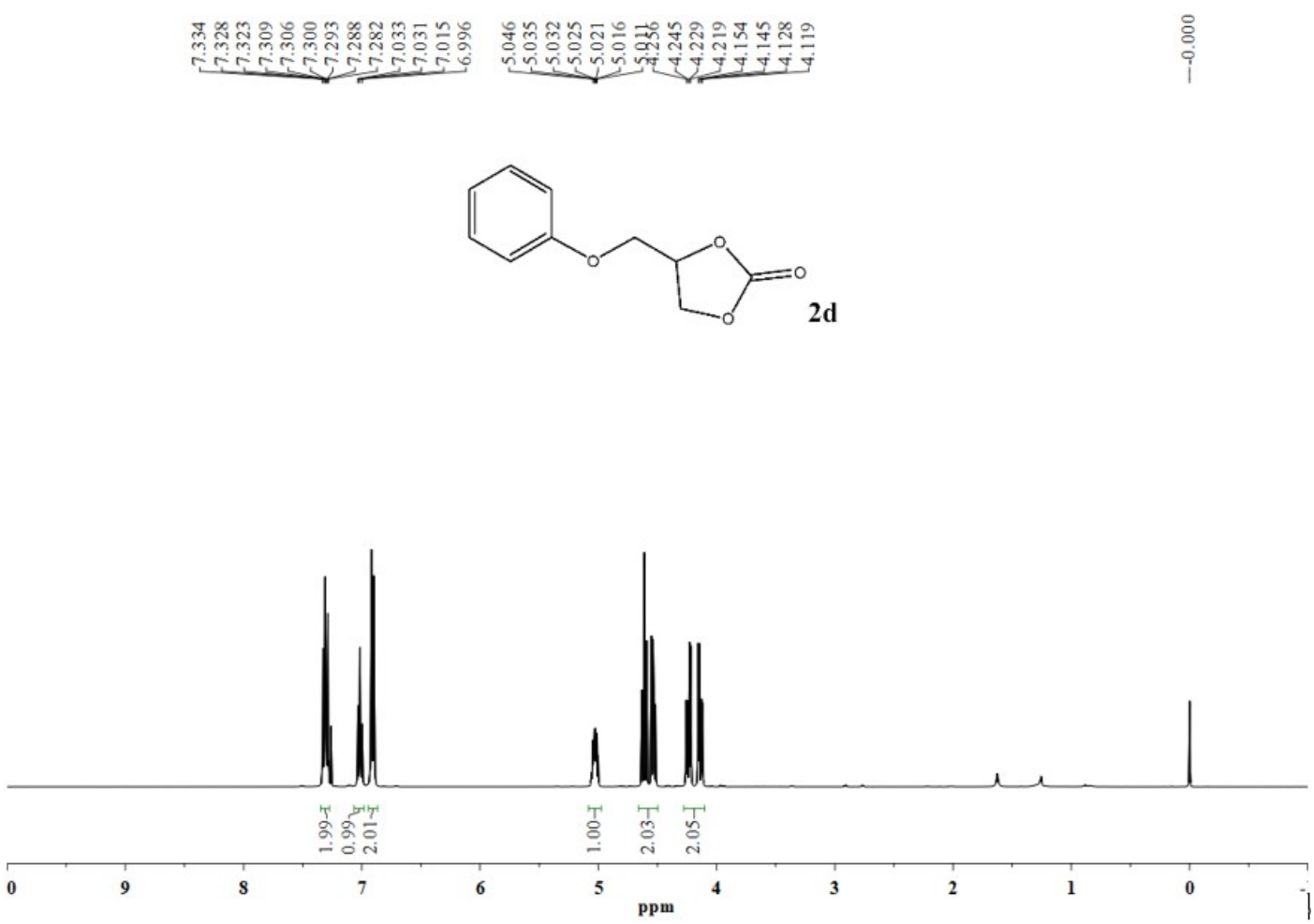
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0.99
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1.00
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2.05
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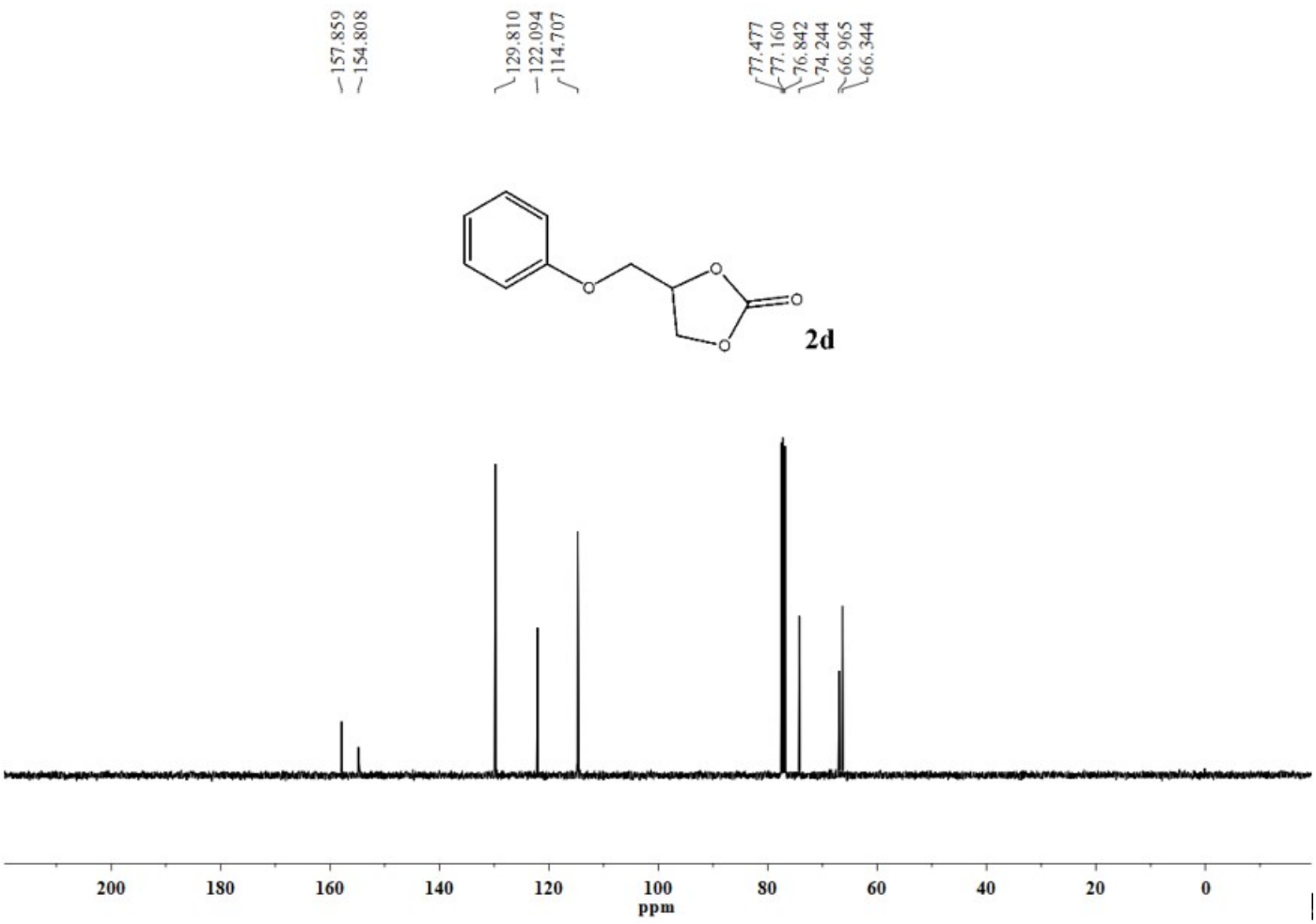
2d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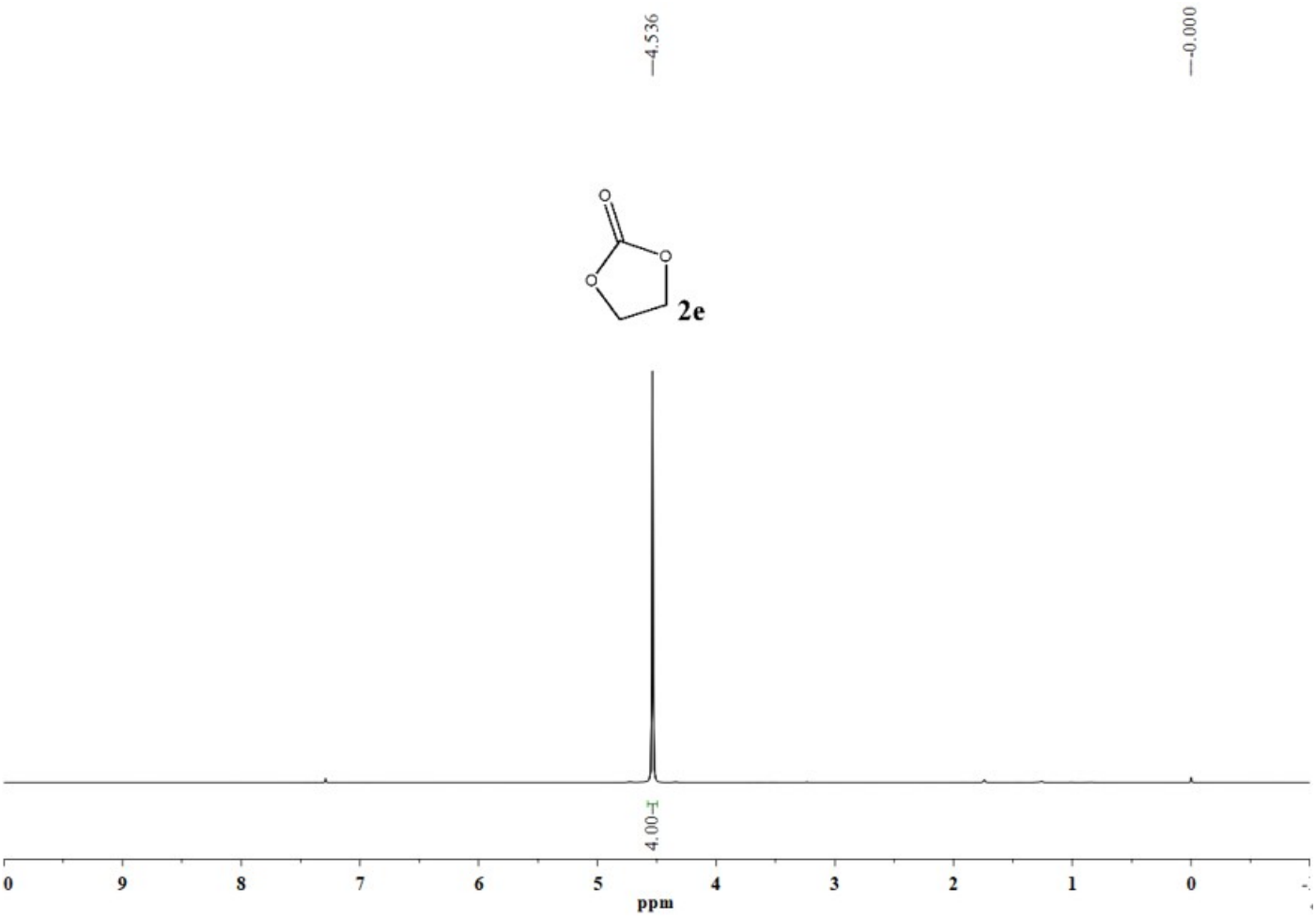
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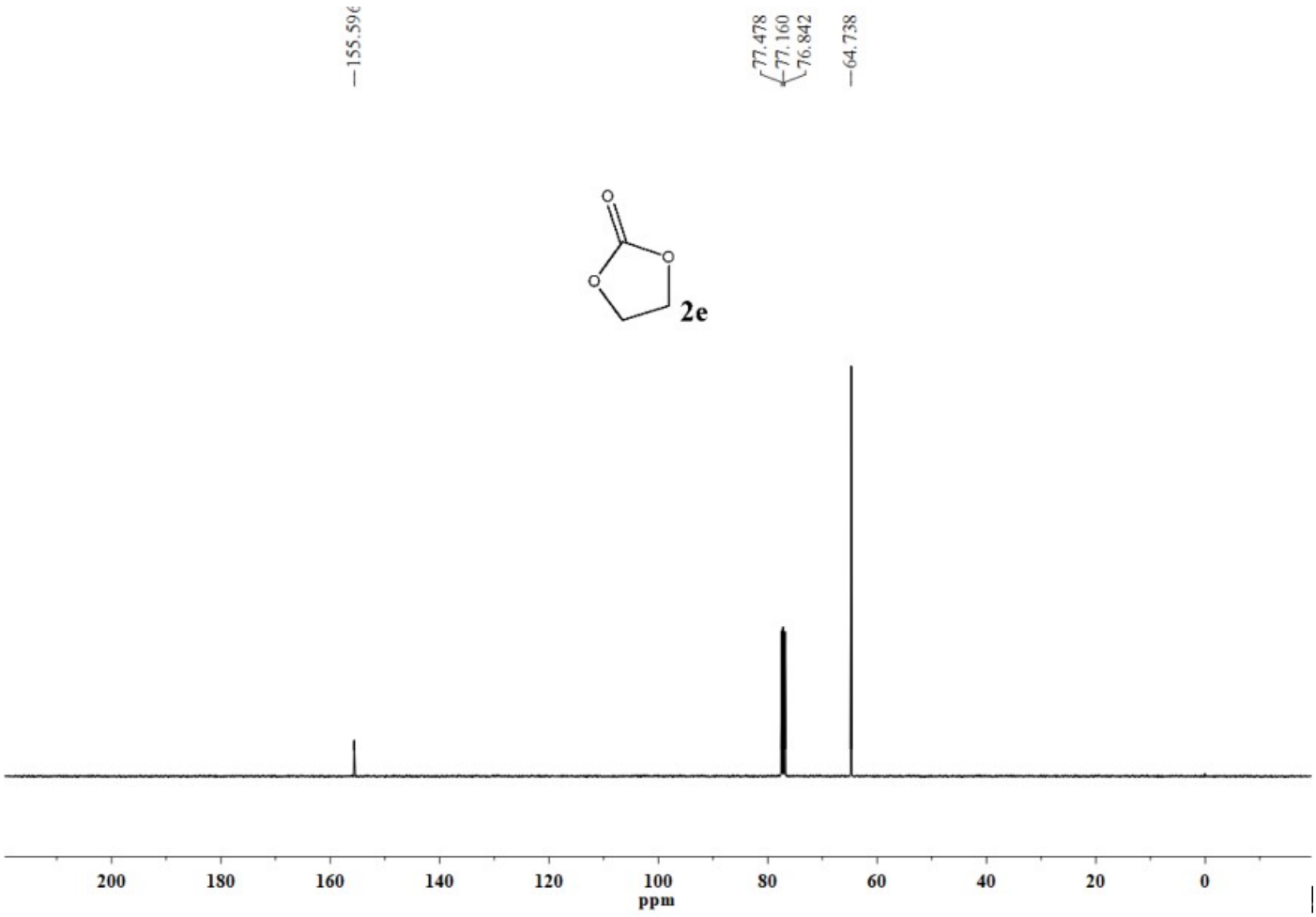
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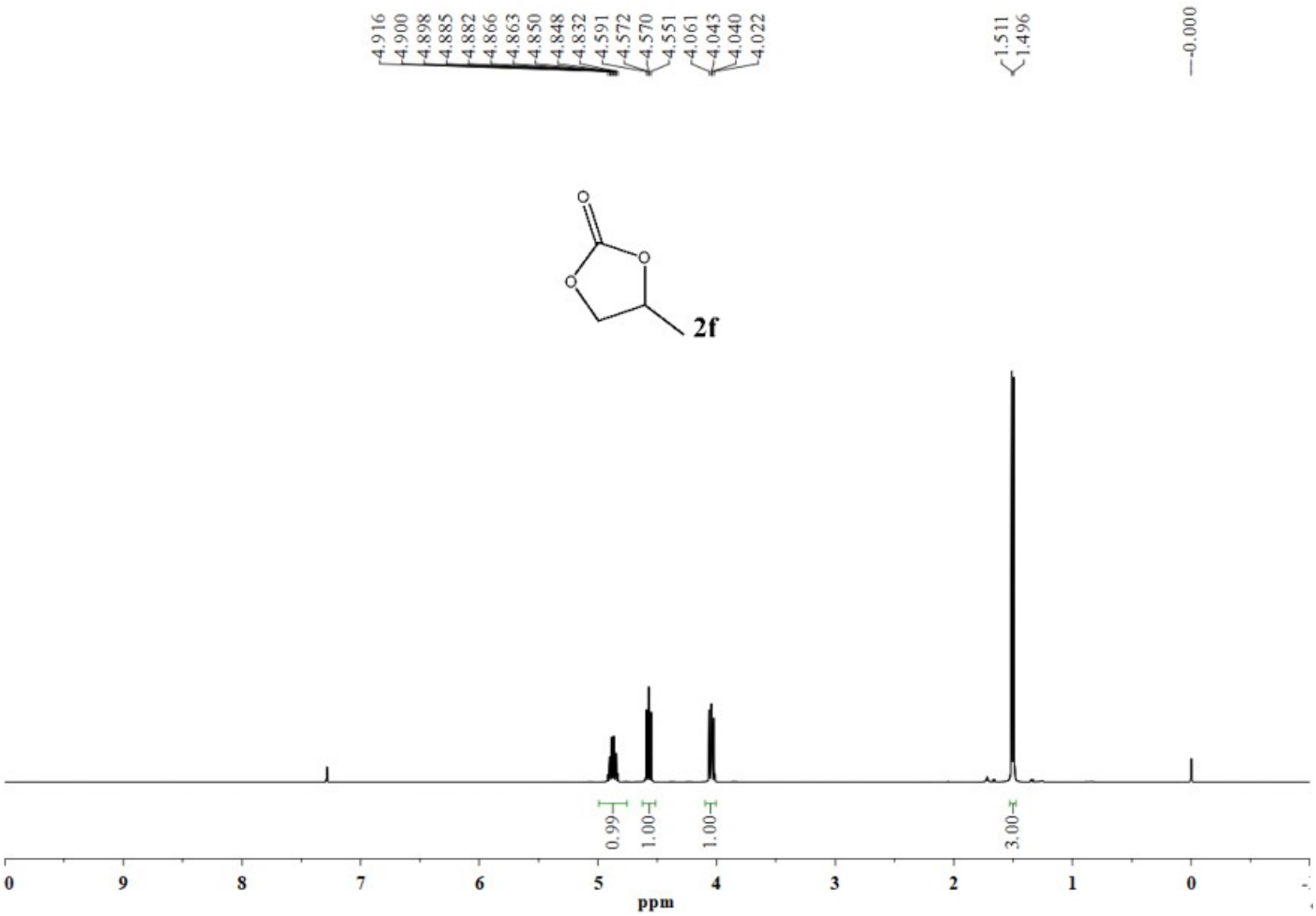
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4.898
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4.866
4.863
4.850
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4.832
4.591
4.572
4.570
4.551
4.061
4.043
4.040
4.022
1.511
1.496
0.000
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1.00
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8
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6
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2
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0
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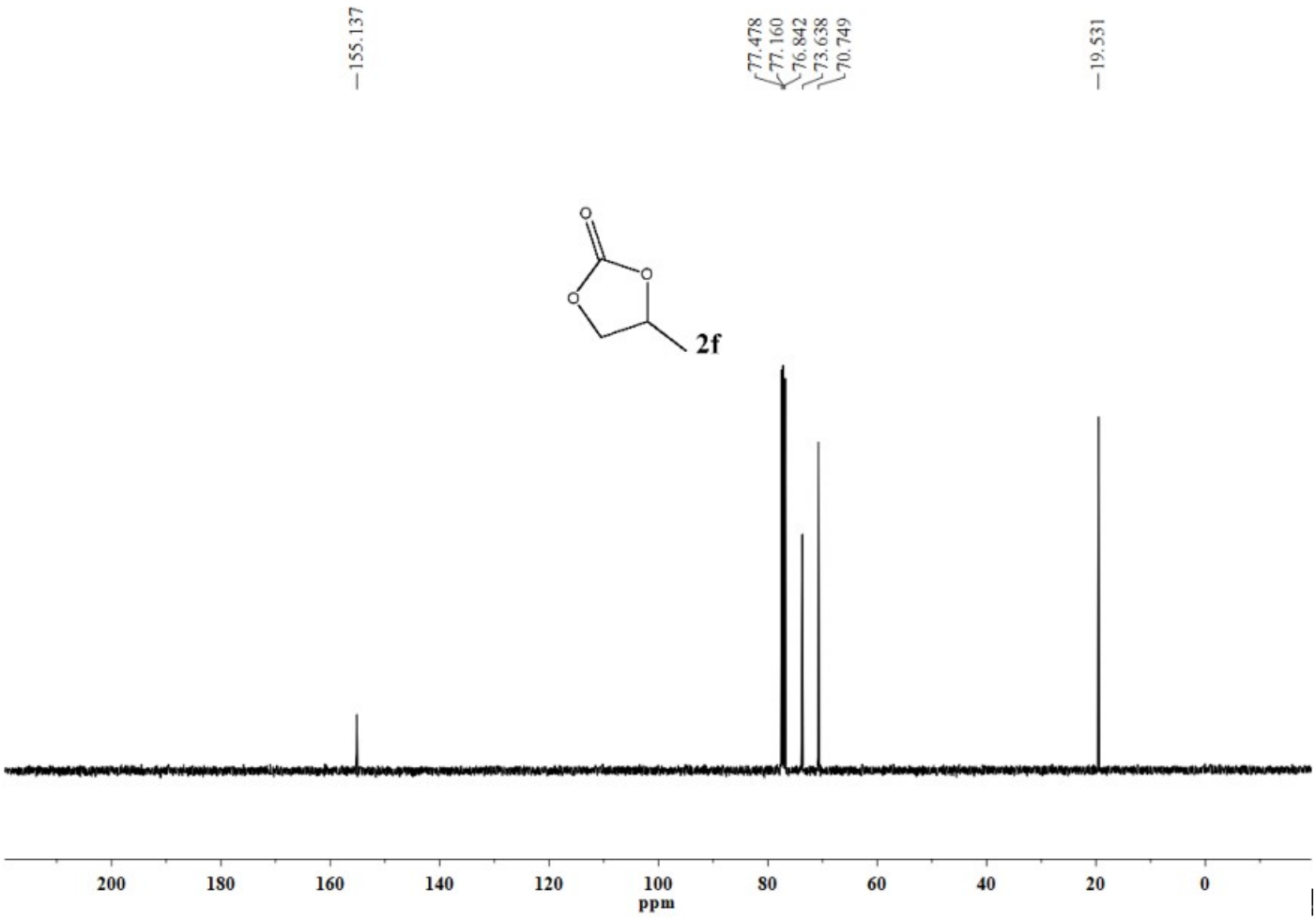
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20
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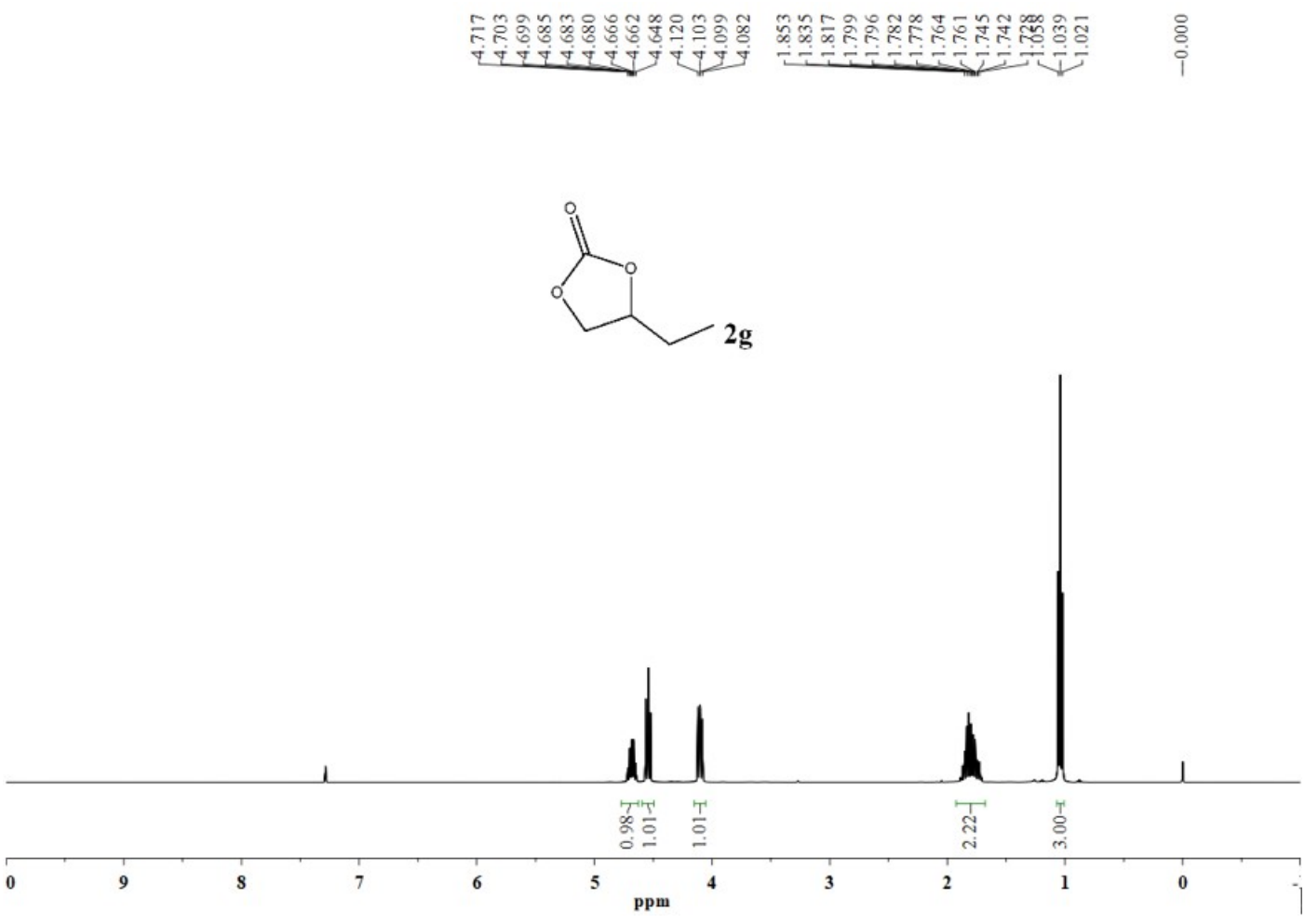
ppm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4.703
4.699
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4.683
4.680
4.666
4.662
4.648
4.120
4.103
4.099
4.082
1.853
1.835
1.817
1.799
1.796
1.782
1.778
1.764
1.761
1.745
1.742
1.039
1.021
0.000
2g
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1.01
2.22
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8
7
6
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4
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2
1
0
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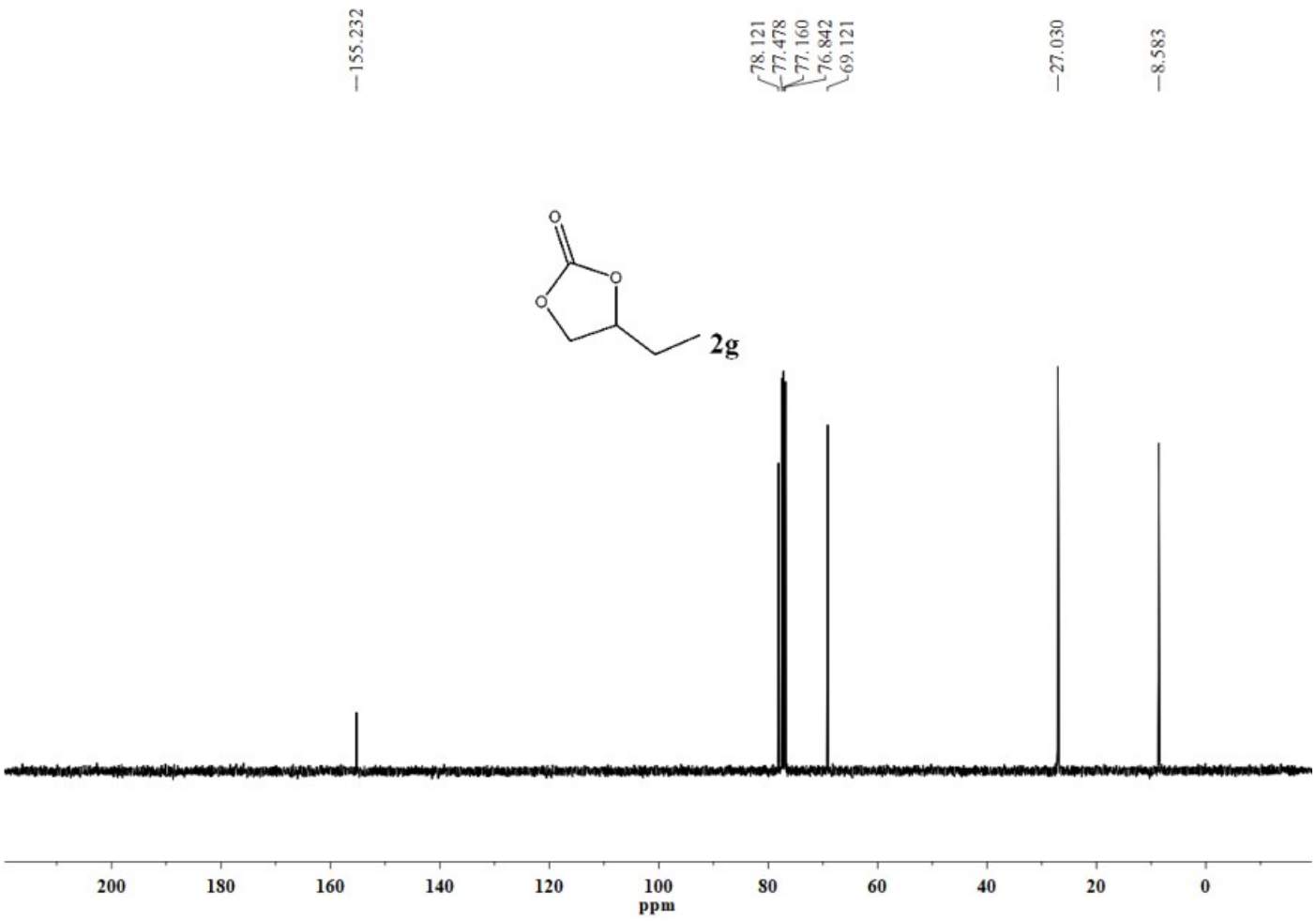
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40
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0
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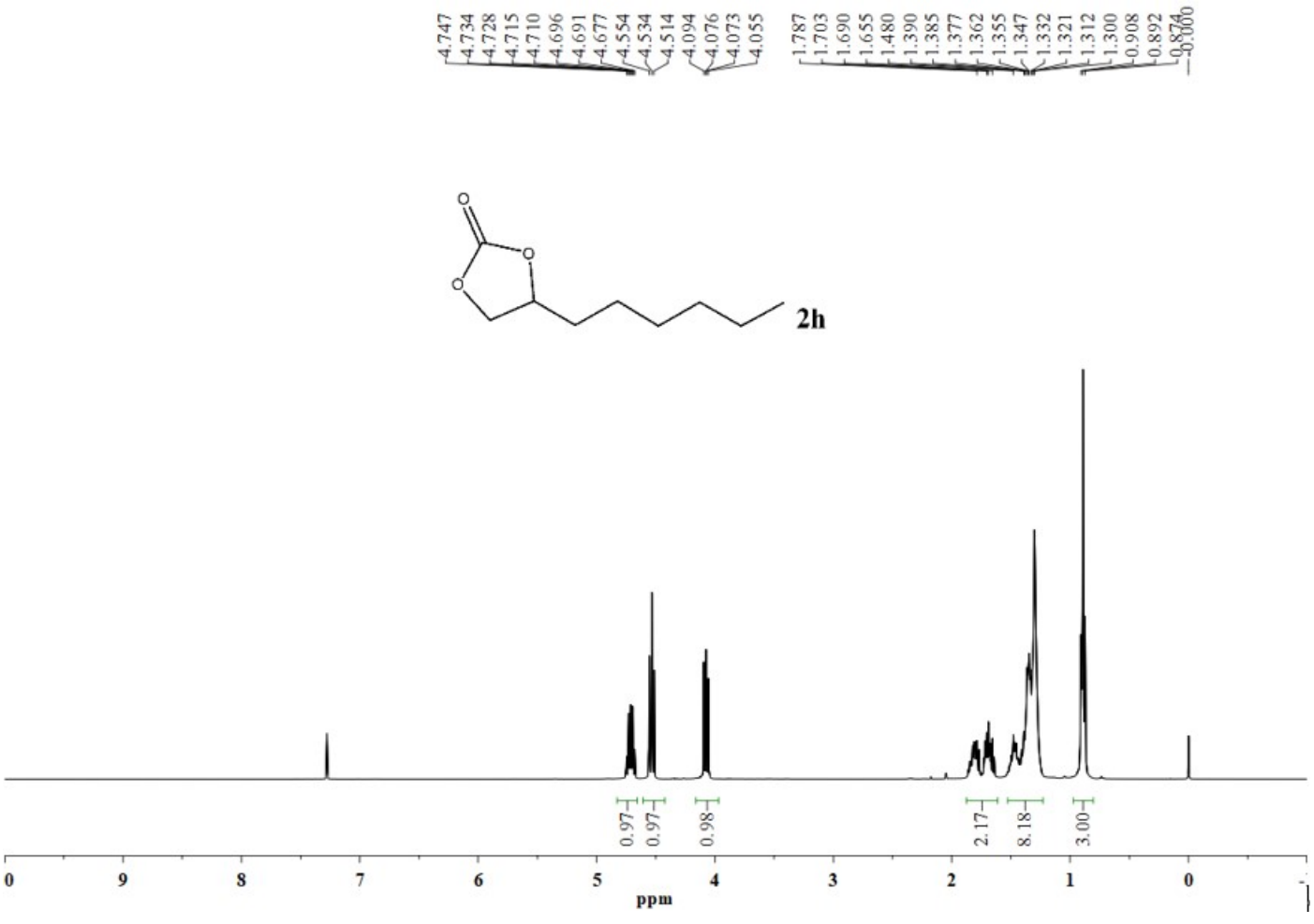
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4.728
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4.696
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4.677
4.554
4.534
4.514
4.094
4.076
4.073
4.055
1.787
1.703
1.690
1.655
1.480
1.390
1.385
1.377
1.362
1.355
1.347
1.332
1.321
1.312
1.300
0.908
0.892
0.874
0.000
2h
0.97
0.97
0.98
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3.00
0
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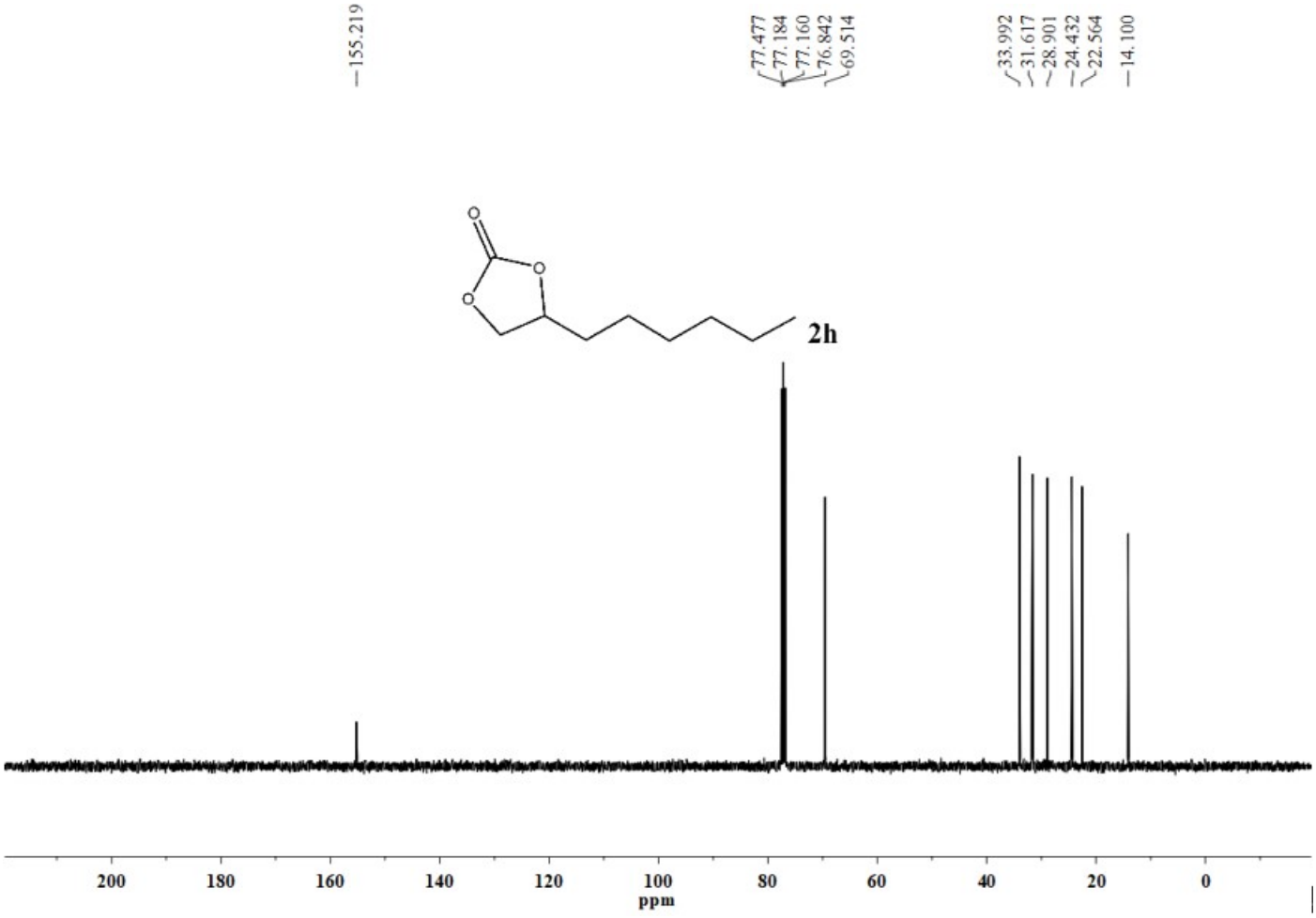
8
7
6
5
4
3
2
1
0
ppm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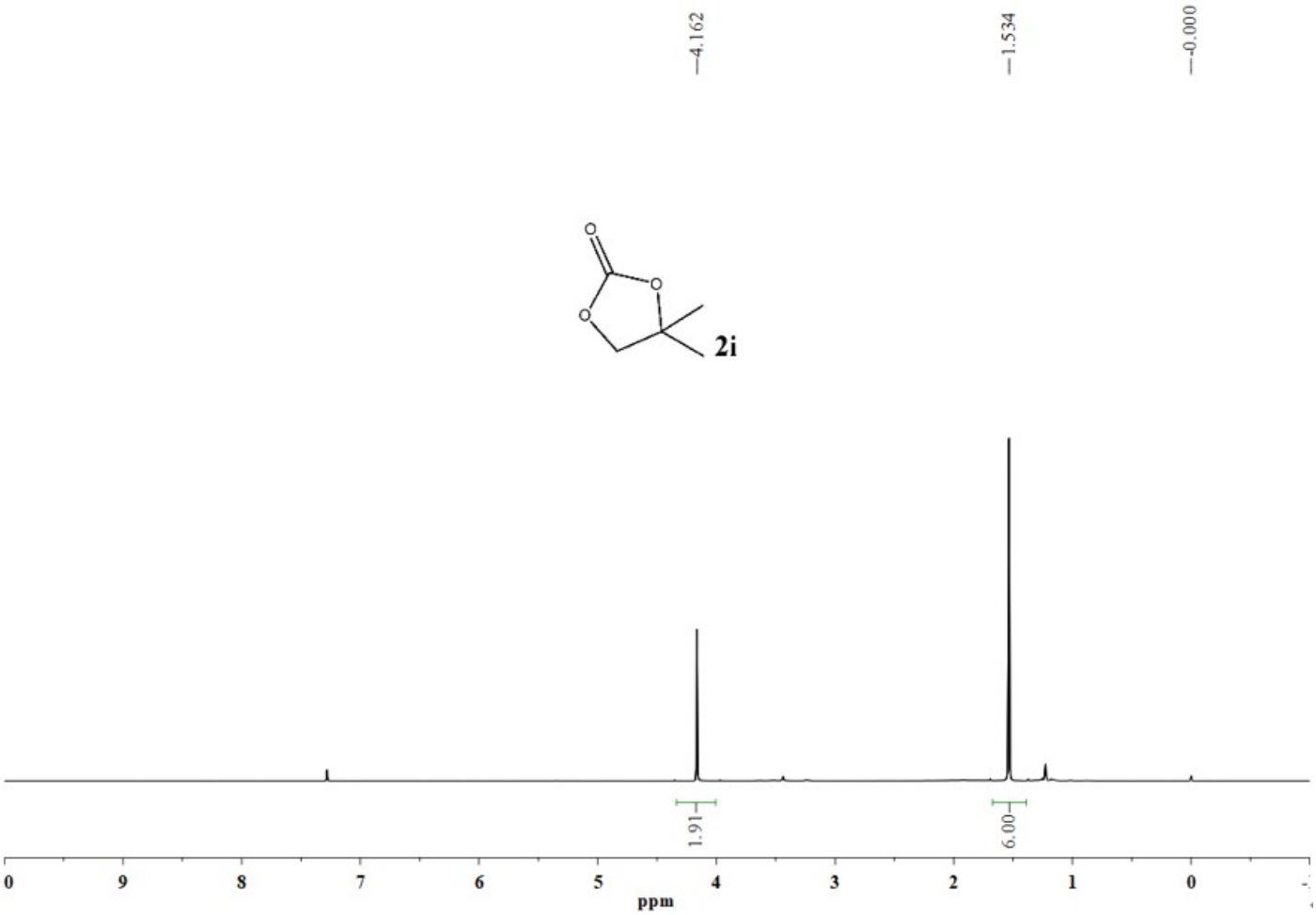
76.842
69.514
33.992
31.617
28.901
24.432
22.564
14.100
2h
200
180
160
140
120
100
80
60
40
20
0
ppm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8
7
6
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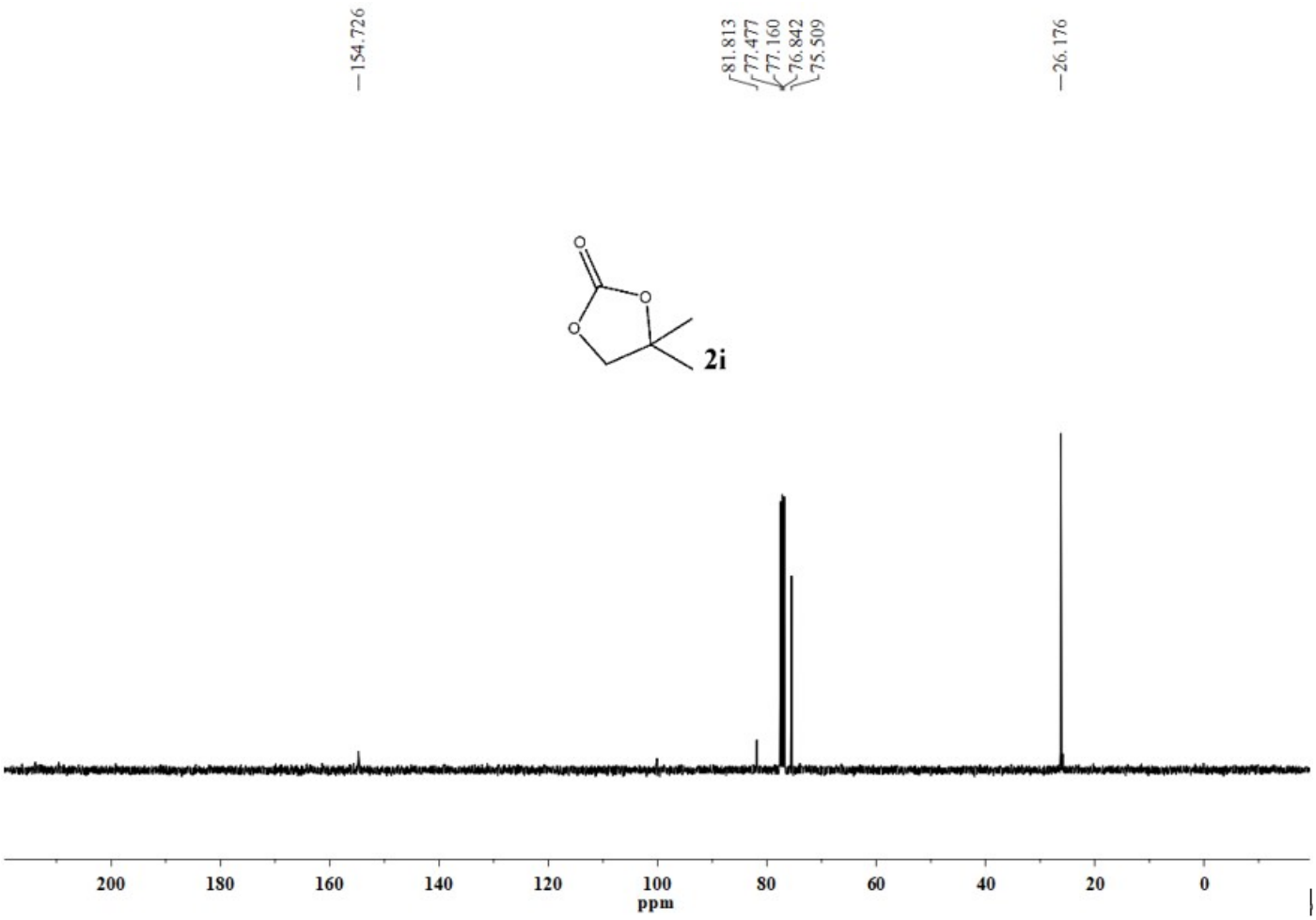
3
2
1
0
ppm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81.813
77.477
77.160
76.842
75.509
26.176
2i
200
180
160
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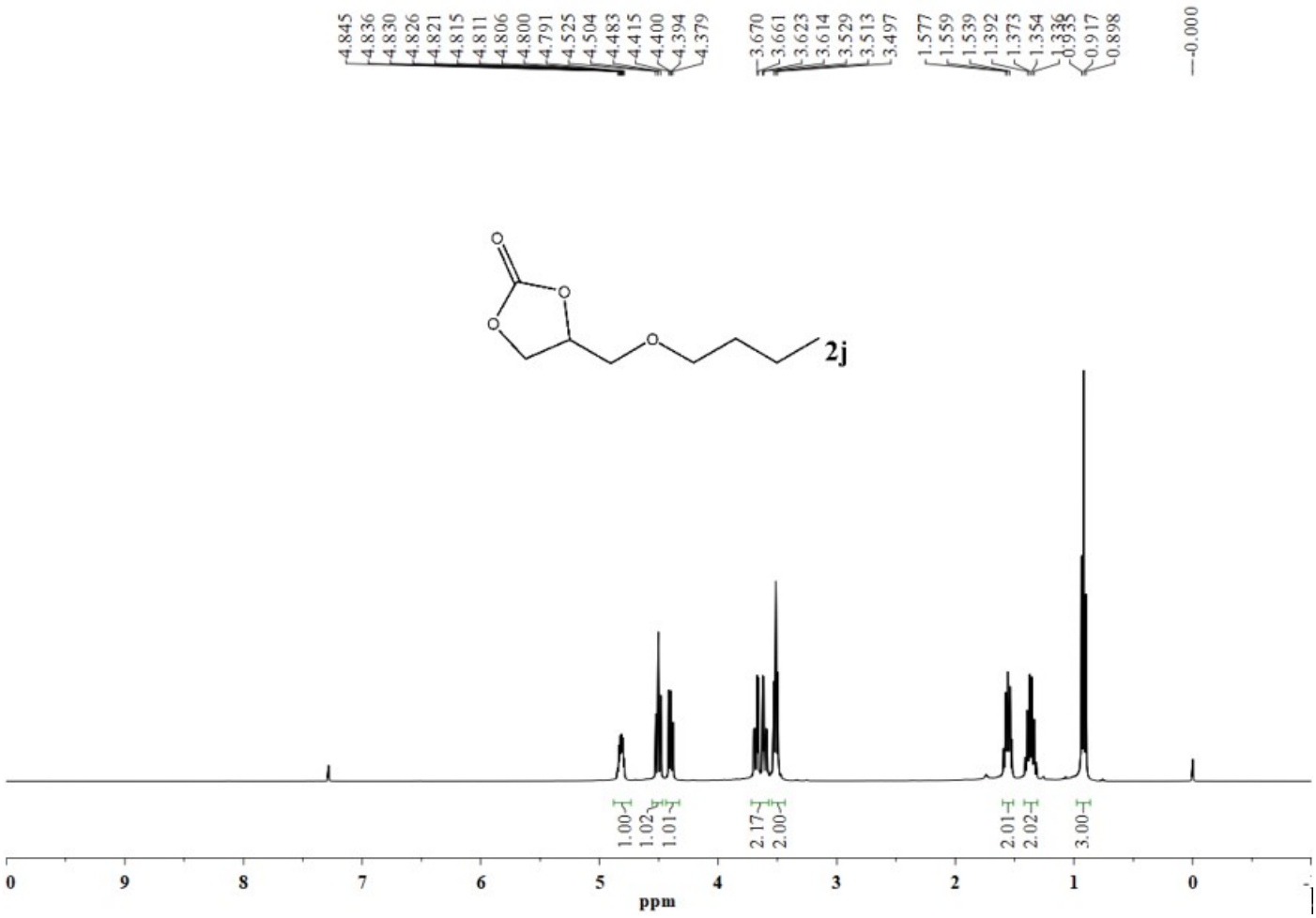
120
100
80
60
40
20
0
ppm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4.525
4.504
4.483
4.415
4.400
4.394
4.379
3.670
3.661
3.623
3.614
3.529
3.513
3.497
1.577
1.559
1.539
1.392
1.373
1.354
1.336
0.935
0.917
0.898
-0.000
2j
1.00
1.02
1.01
2.17
2.00
2.01
2.02
3.00
0
9
8
7
6
5
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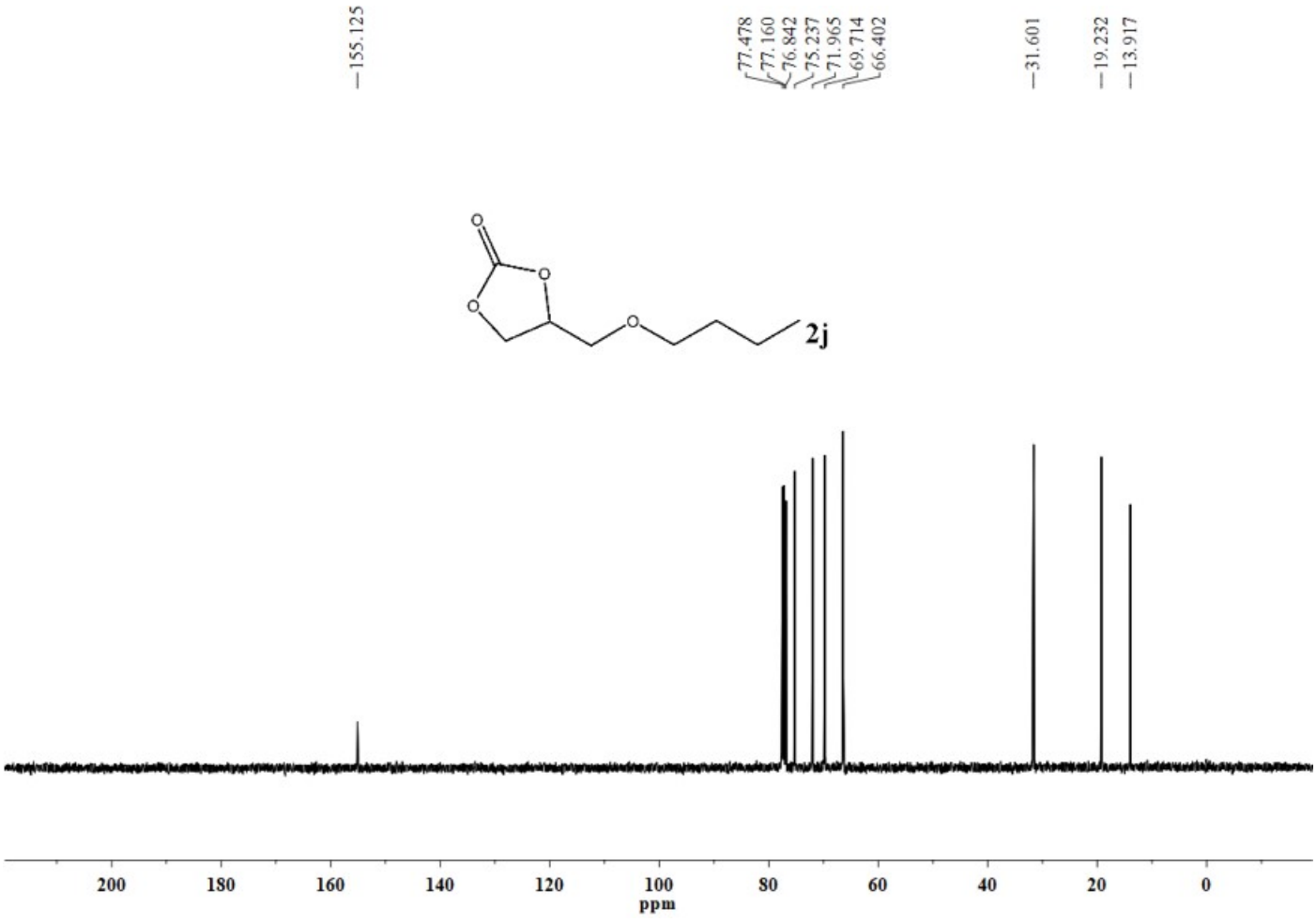
3
2
1
0
ppm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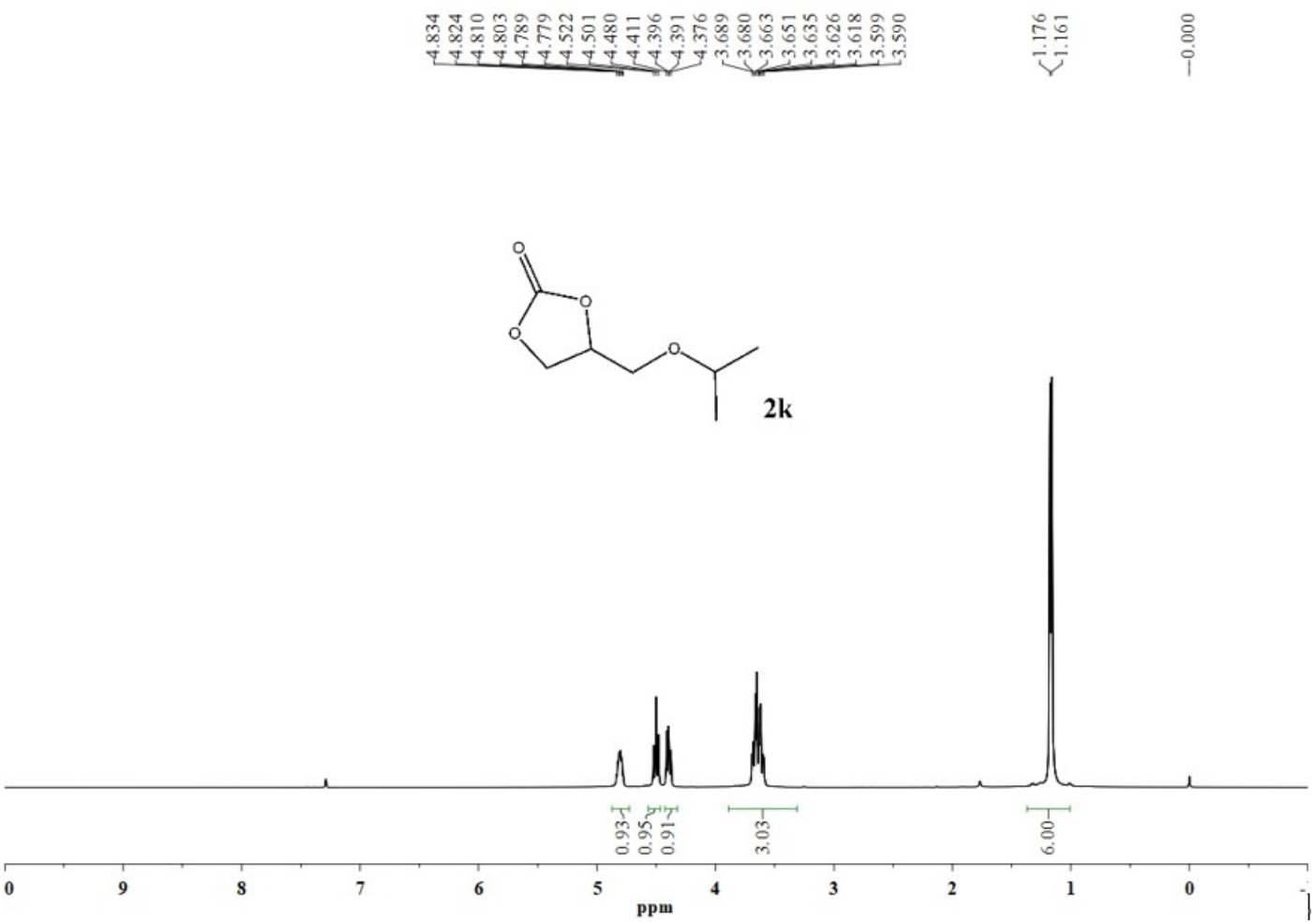
77.160
76.842
75.237
71.965
69.714
66.402
31.601
19.232
13.917
2j
200
180
160
140
120
100
80
60
40
20
0
ppm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4.810
4.803
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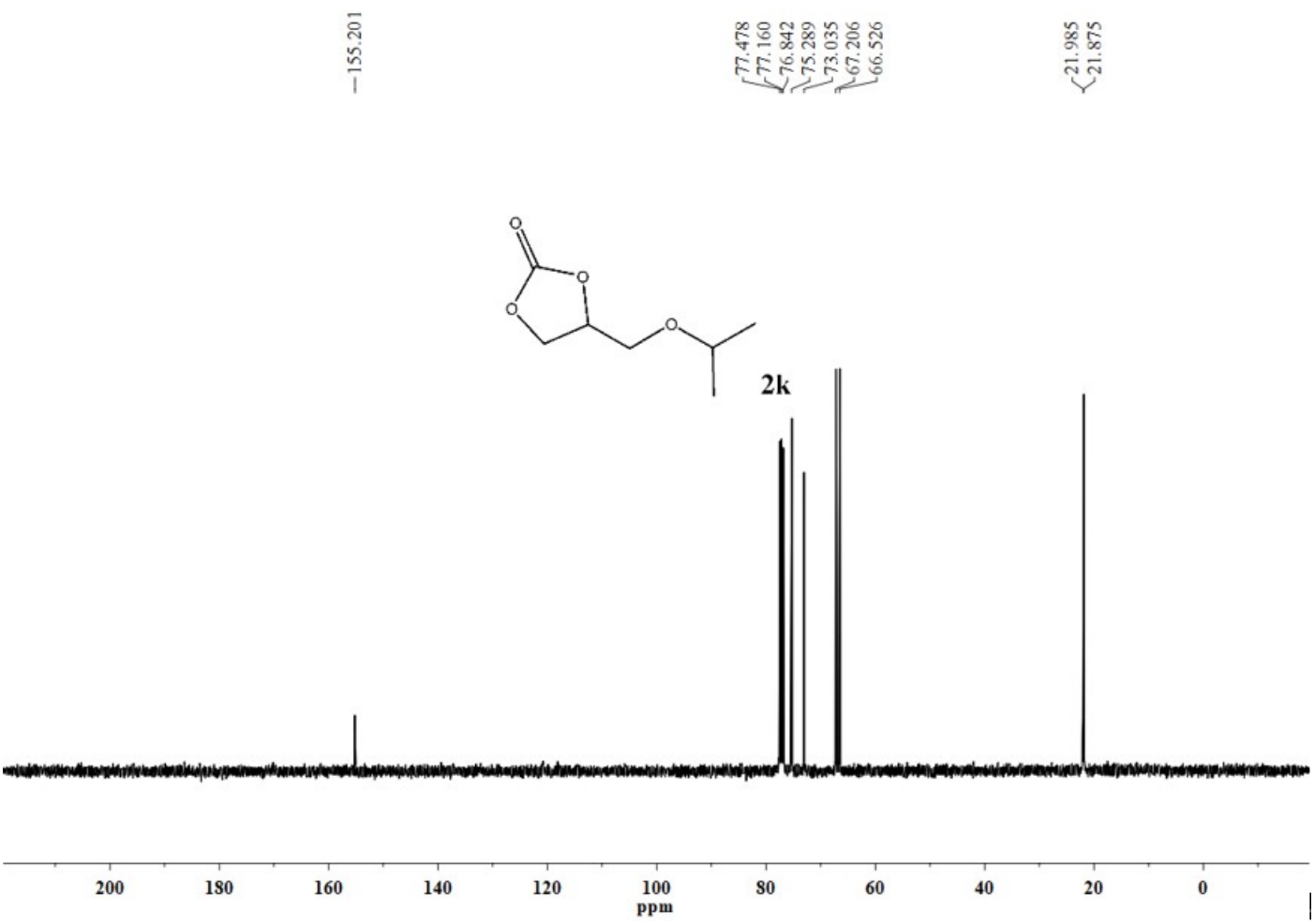
4.522
4.501
4.480
4.411
4.396
4.391
4.376
3.689
3.680
3.663
3.651
3.635
3.626
3.618
3.599
3.590
1.176
1.161
0.000
2k
0.93
0.95
0.91
3.03
6.00
ppm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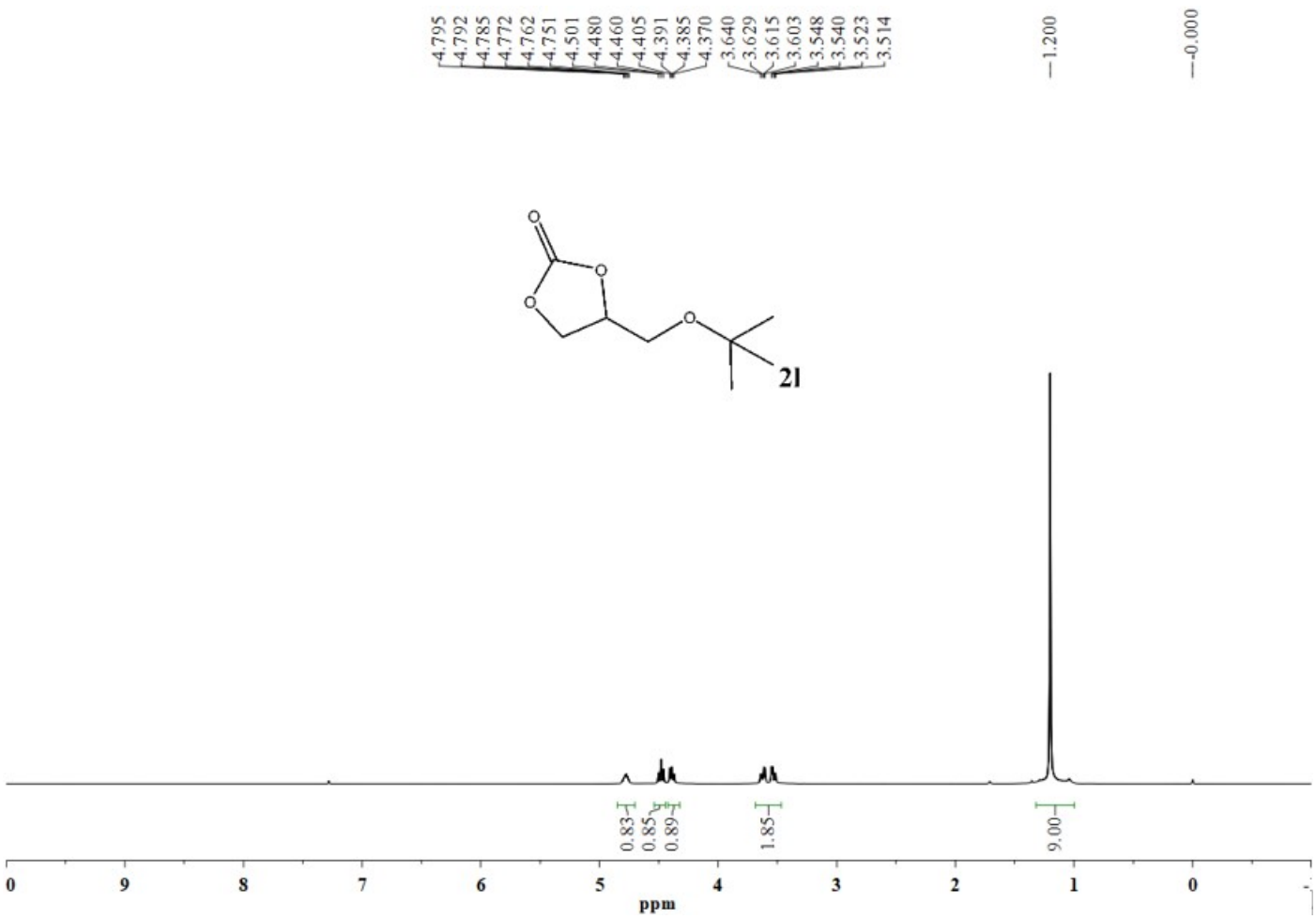
73.035
67.206
66.526
21.985
21.875
2k
ppm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4.501
4.480
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4.405
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4.385
4.370
3.640
3.629
3.615
3.603
3.548
3.540
3.523
3.514
1.200
0.000
21
0.83
0.85
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1.85
9.00
0
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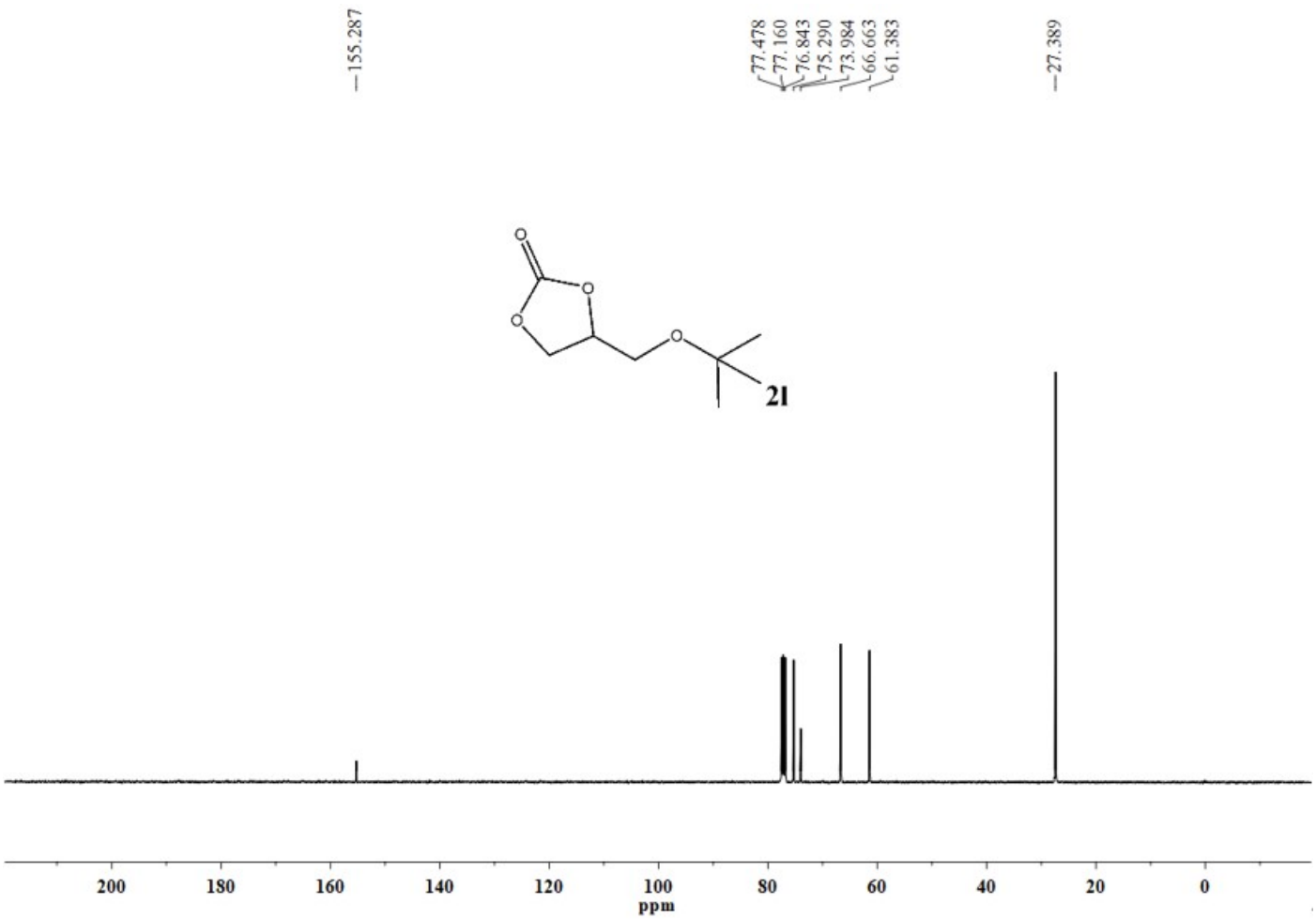
8
7
6
5
4
3
2
1
0
ppm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73.984
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61.383
27.389
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200
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120
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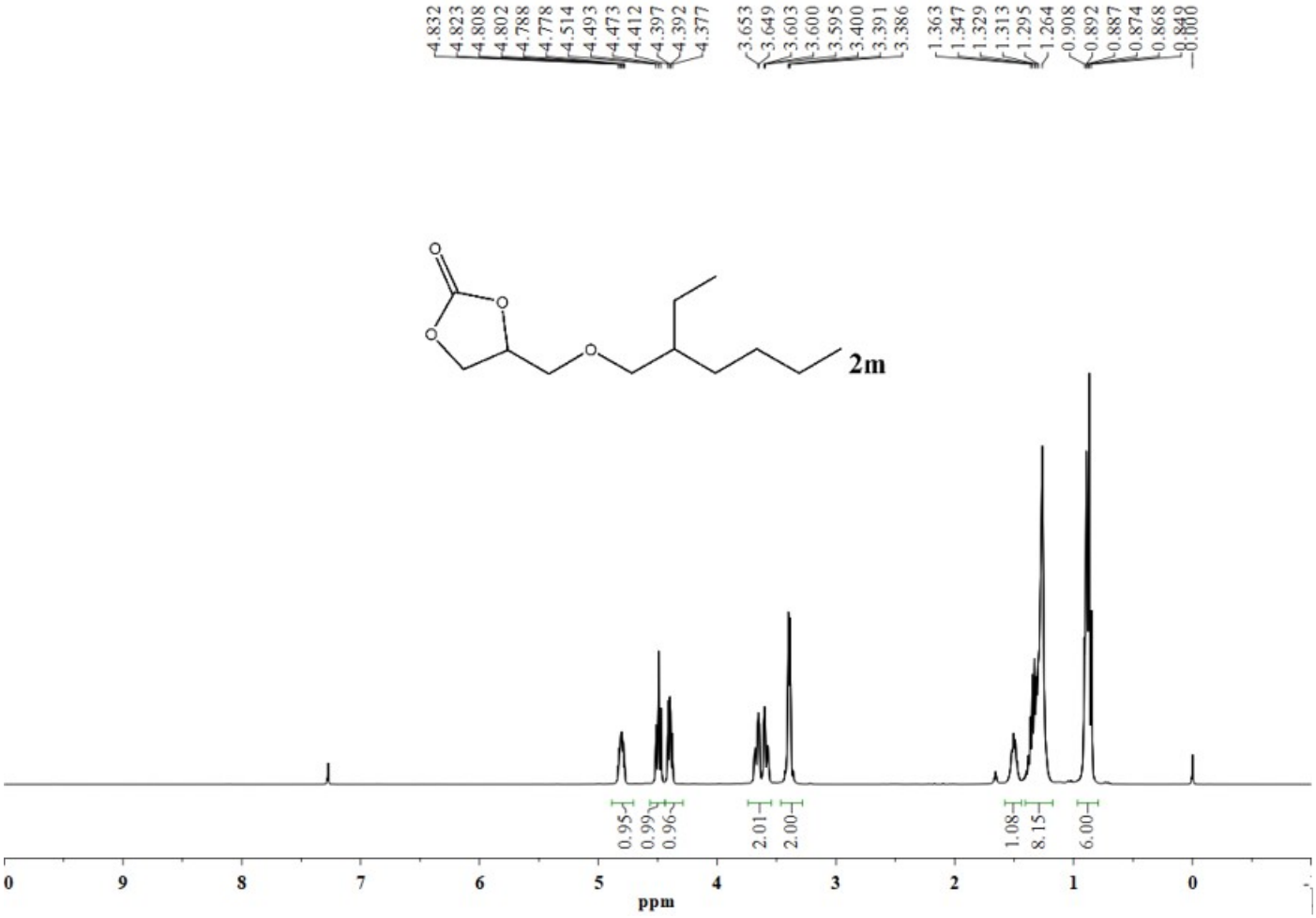
60
40
20
0
ppm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4.412
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4.377
3.653
3.649
3.603
3.600
3.595
3.400
3.391
3.386
1.363
1.347
1.329
1.313
1.295
1.264
0.908
0.892
0.887
0.874
0.868
0.849
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2m
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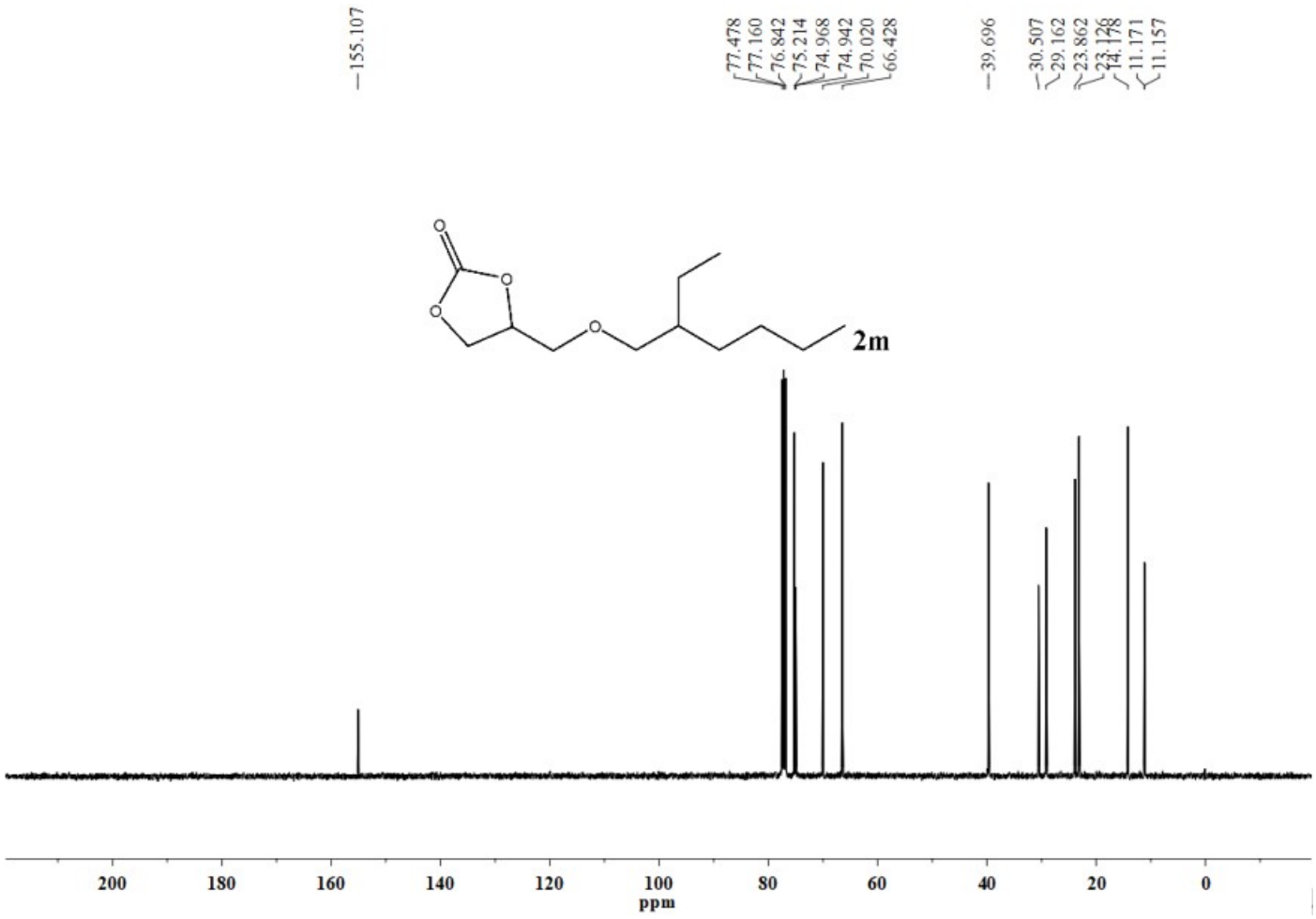
2.00
1.08
8.15
6.00
ppm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74.968
74.942
70.020
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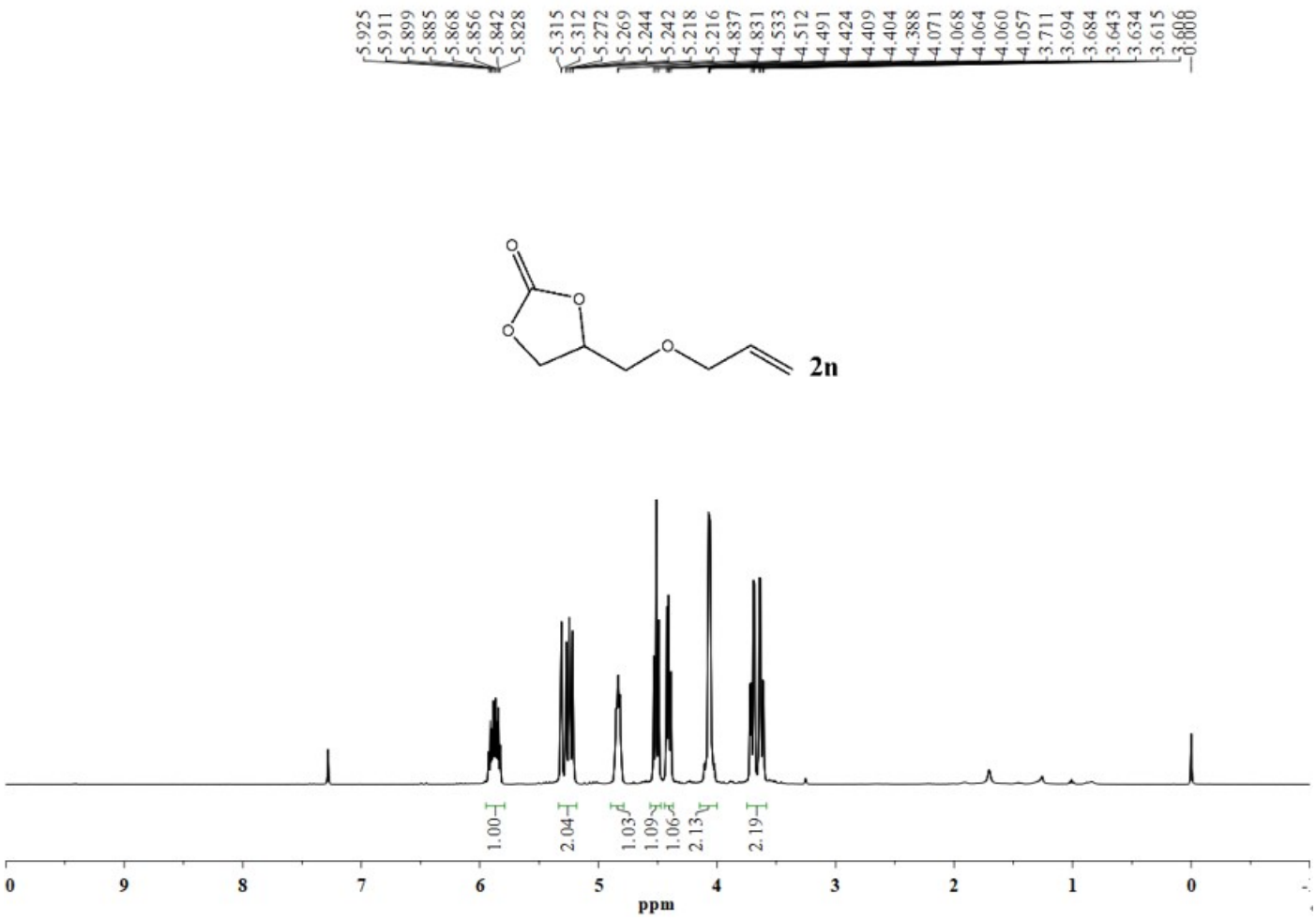
14.178
11.171
11.157
2m
ppm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5.856
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5.315
5.312
5.272
5.269
5.244
5.242
5.218
5.216
4.837
4.831
4.533
4.512
4.491
4.424
4.409
4.404
4.388
4.071
4.068
4.064
4.060
4.057
3.711
3.694
3.684
3.643
3.634
3.615
3.606
0.000
2n
1.00
2.04
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1.06
2.13
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6
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2
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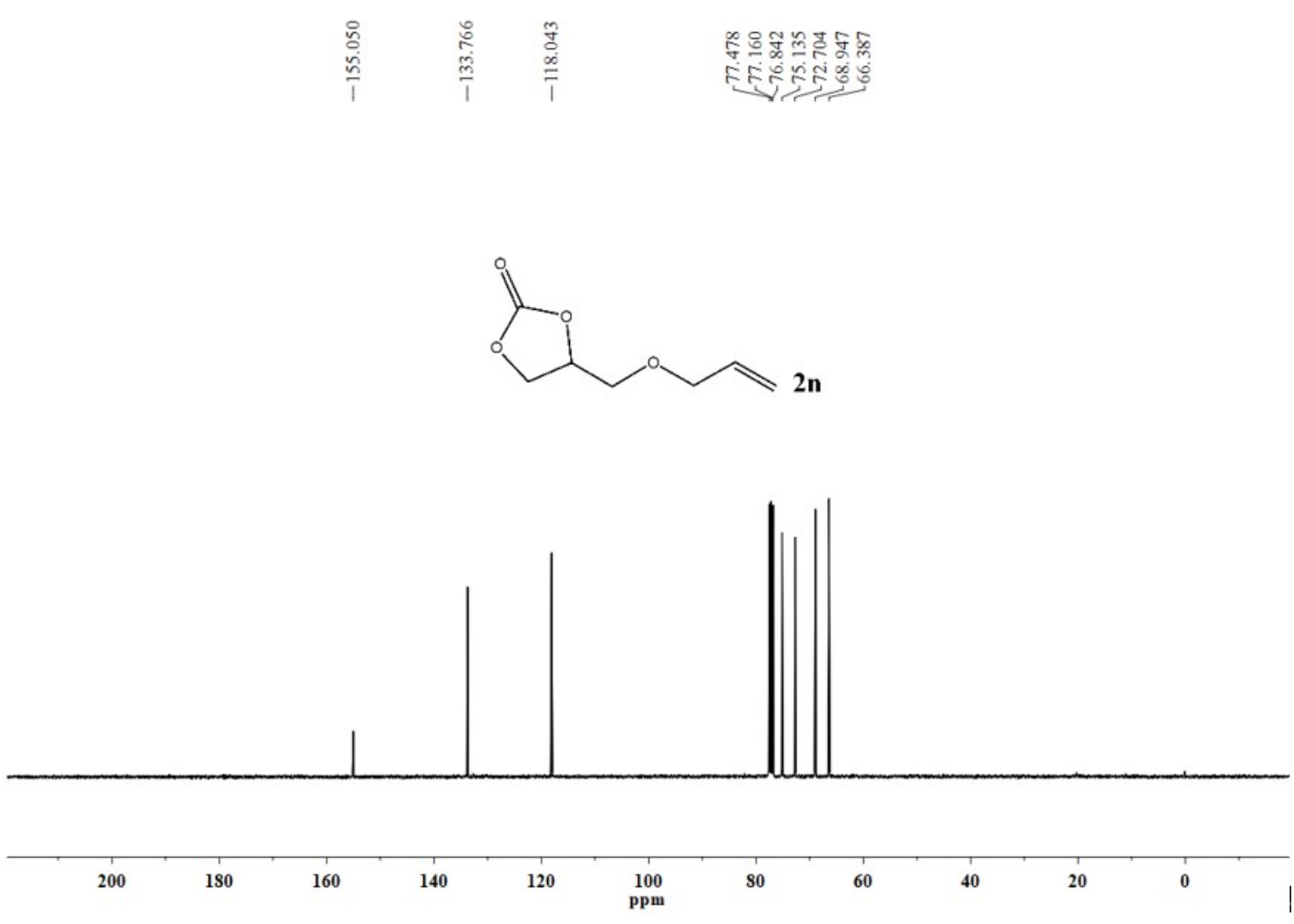
0
ppm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77.160
76.842
75.135
72.704
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66.387
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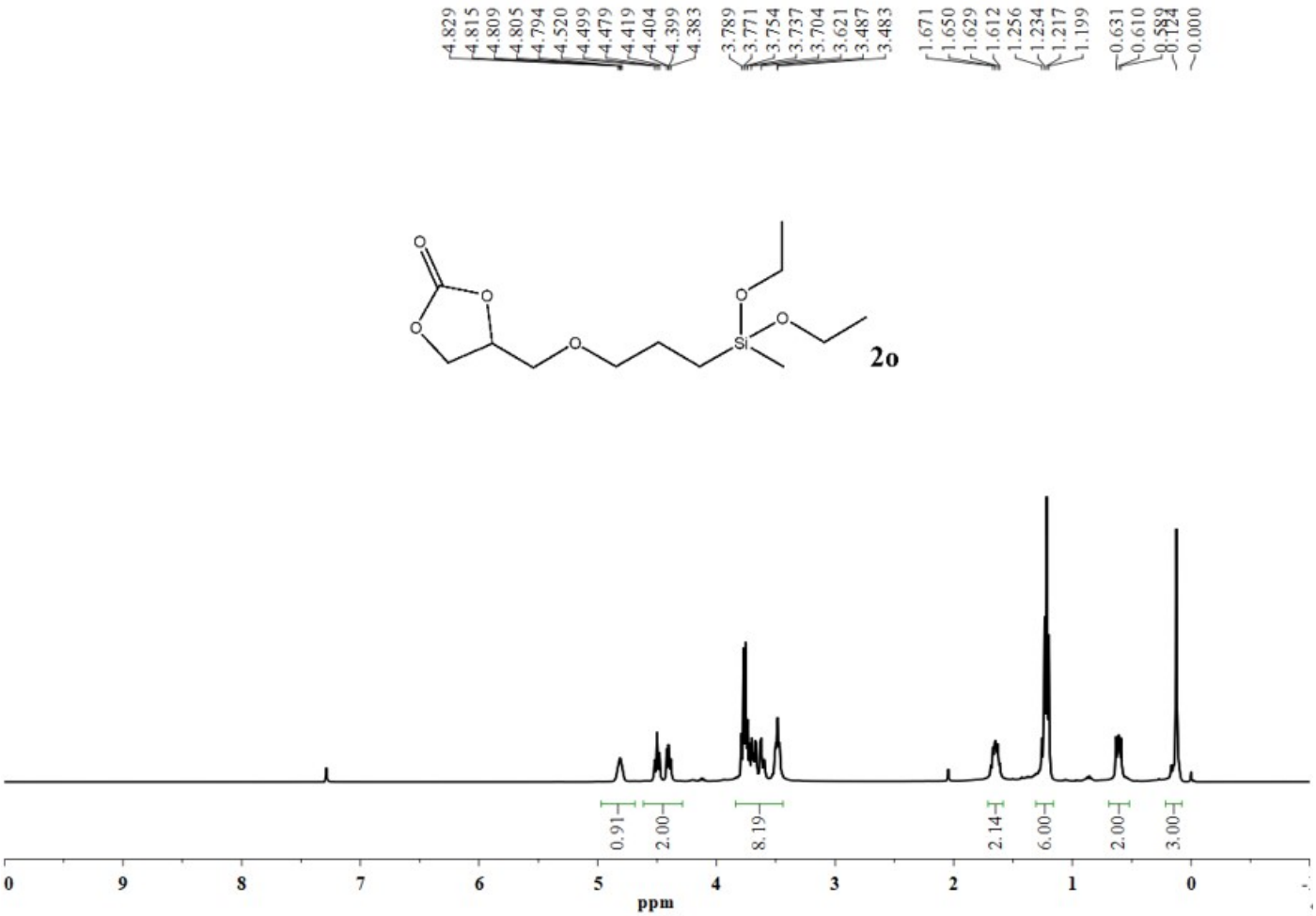
120
100
80
60
40
20
0
ppm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4.809
4.805
4.794
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4.499
4.479
4.419
4.404
4.399
4.383
3.789
3.771
3.754
3.737
3.704
3.621
3.487
3.483
1.671
1.650
1.629
1.612
1.256
1.234
1.217
1.199
0.631
0.610
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0.000
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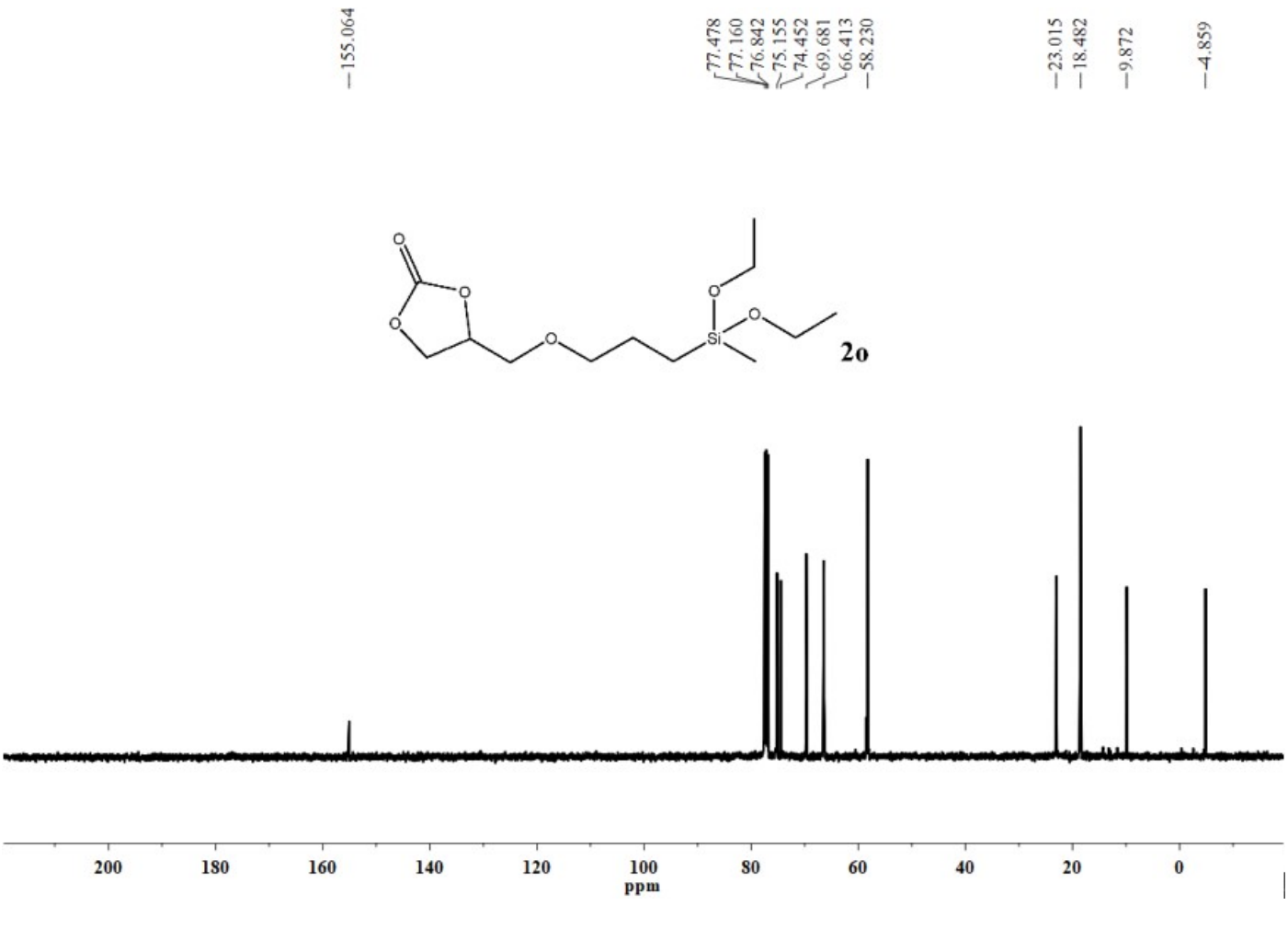
2.14
6.00
2.00
3.00
ppm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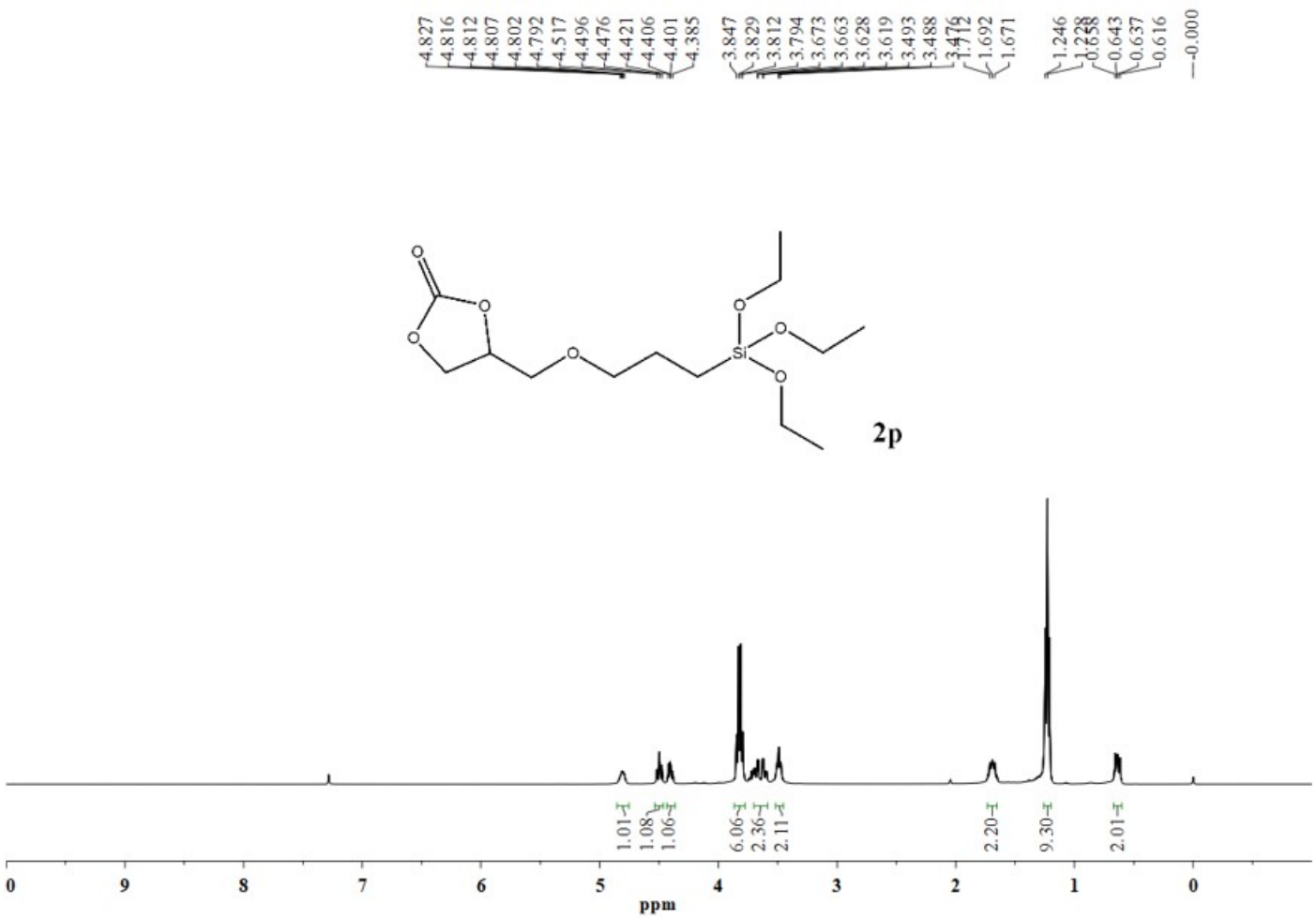
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4.496
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4.401
4.385
3.847
3.829
3.812
3.794
3.673
3.663
3.628
3.619
3.493
3.488
1.692
1.671
1.246
0.658
0.643
0.637
0.616
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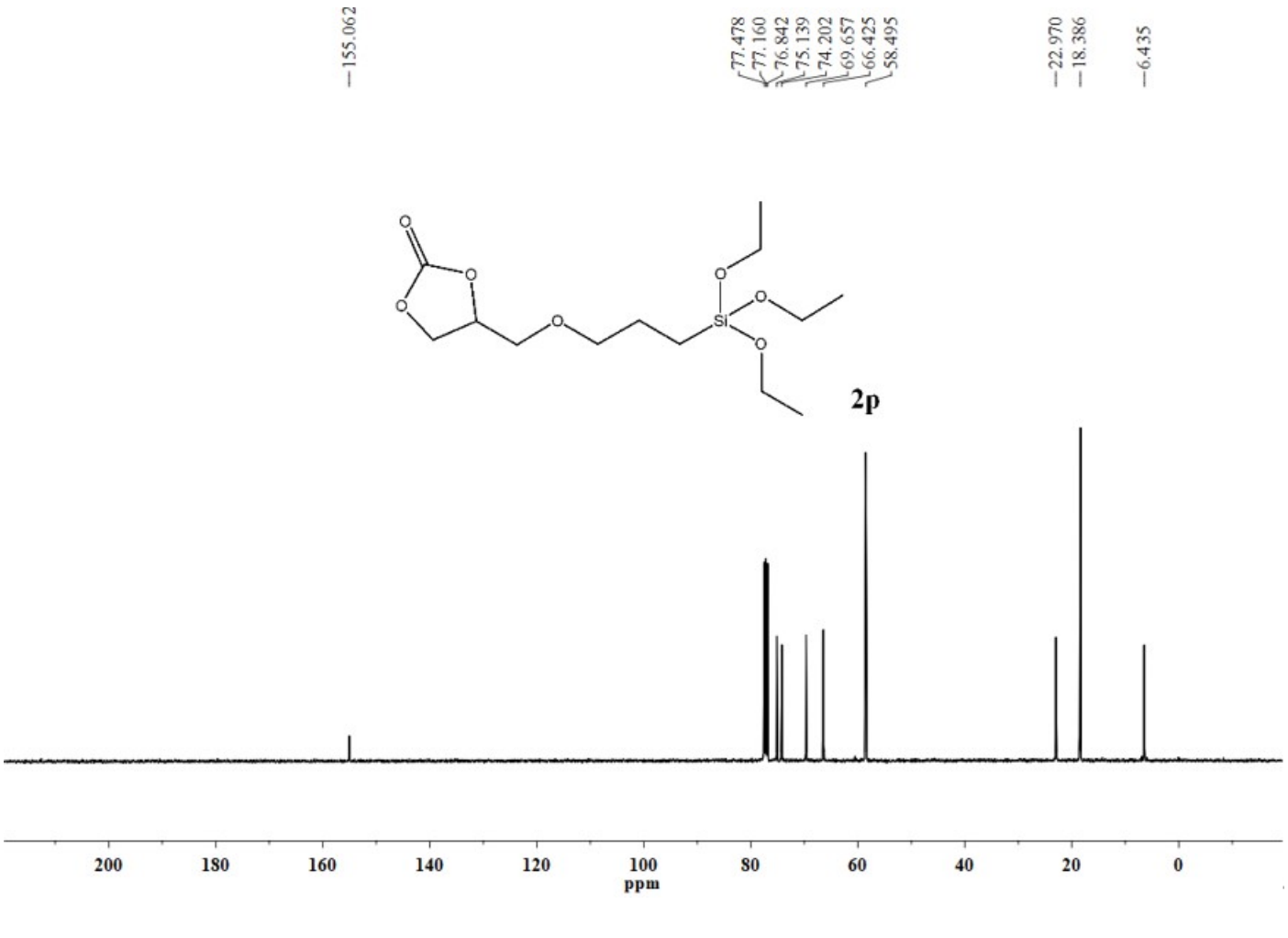
2p
1.01
1.08
1.06
6.06
2.36
2.11
2.20
9.30
2.01
ppm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77.160
76.842
75.139
74.202
69.657
66.425
58.495
22.970
18.386
6.435
2p
ppm

5. References

1. X. Cui, X. Dai, A.-E. Surkus, K. Junge, C. Kreyenschulte, G. Agostini, N. Rockstroh and M. Beller, *Chin. J. Catal.*, 2019, **40**, 1679-1685.
2. R. Ma, L.-N. He and Y.-B. Zhou, *Green Chem.*, 2016, **18**, 226-231.
3. C. Chen and A. Zhang, *New J. Chem.*, 2021, **45**, 20155-20163.