Electronic Supplementary Material (ESI) for New Journal of Chemistry.
This journal is © The Royal Society of Chemistry and the Centre National de la Recherche Scientifique 2019

Supporting information

Magnetic graphene oxide/NHC catalyzed aerobic direct amidation and cross-dehydrogenative coupling of aldehydes

Heshmatollah Sepahvand,[a] Elnaz Ghasemi*,[a] Mohammad Sharbati,[b] Melika Sadat Mohammadi,[c] Maqsoud Arshadi Pirlar[c]

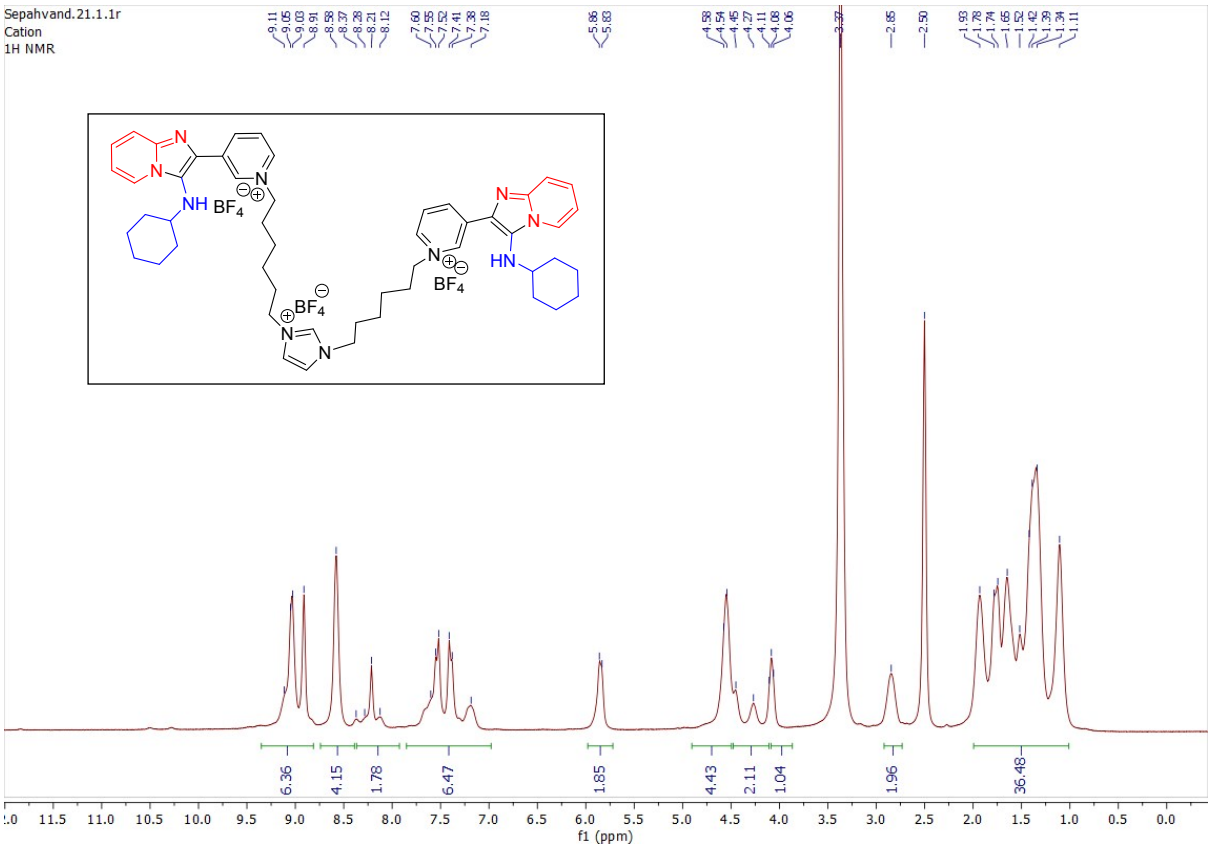
[a]*Department of Chemistry, Tabriz Branch, Islamic Azad University, Tabriz, Iran*
[b]*School of Metallurgy and Materials Engineering, College of Engineering, University of Tehran, Tehran, Iran*
[c]*Department of Physic, Shahid Beheshti University, Tehran, Iran*

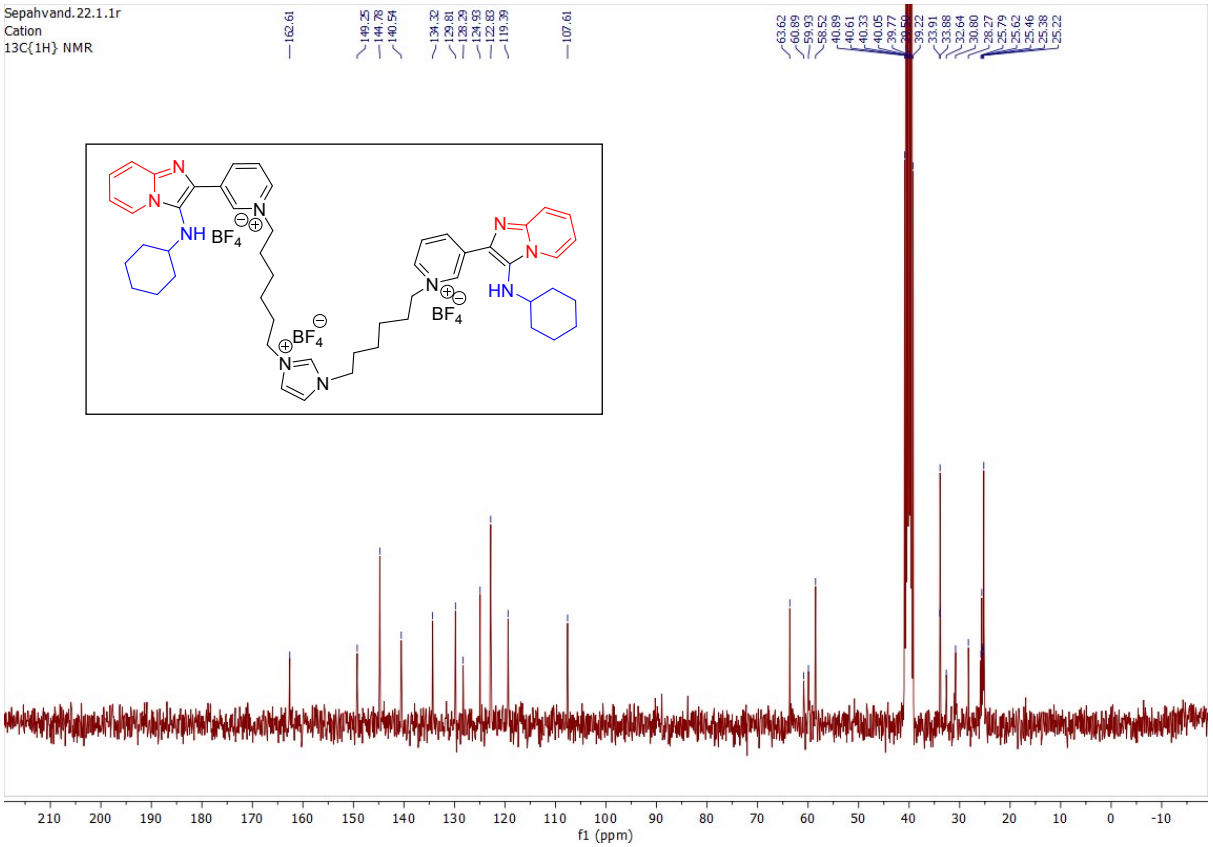
Synthesis of the magnetic graphene oxide (GO)

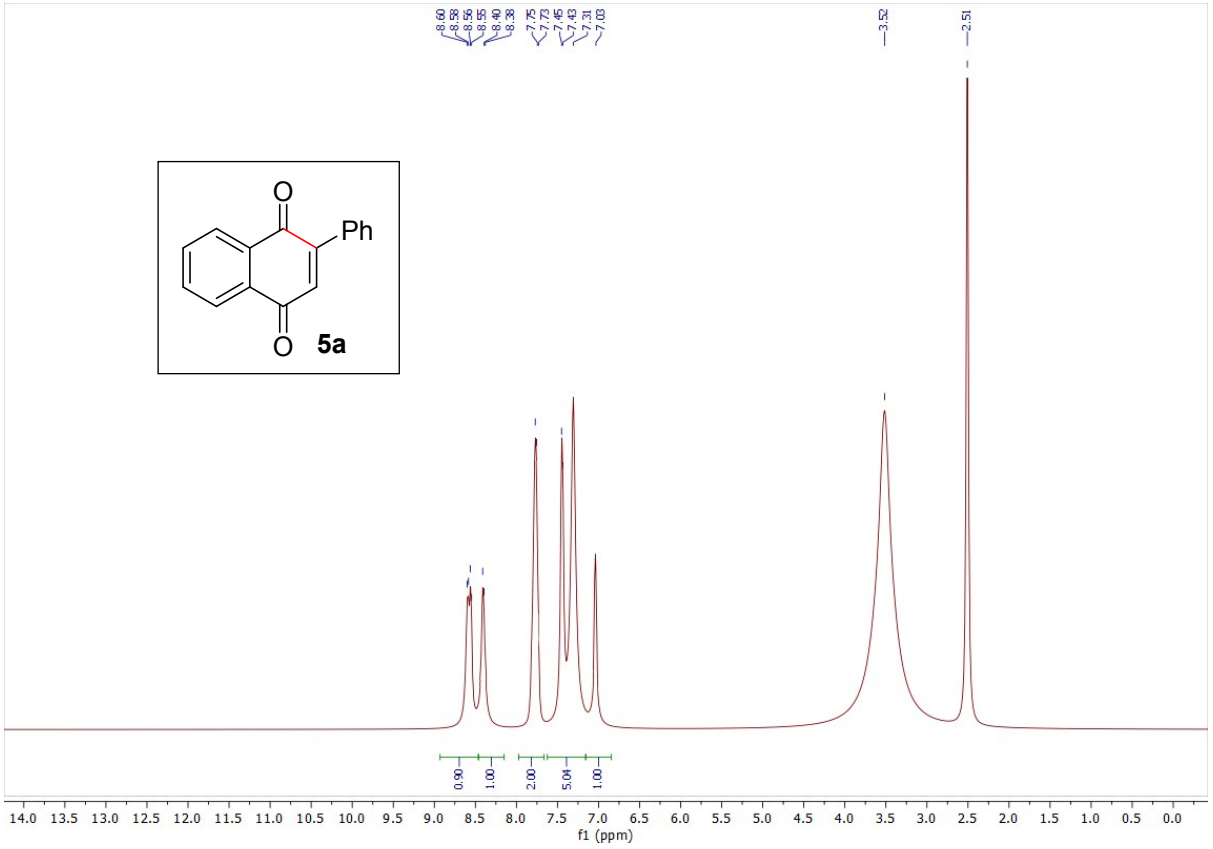
GO was synthesized from natural graphite via preoxidation/thermal expansion of graphite followed by modified Hummer's method.[1] At first step, 10 g of natural graphite flake was mixed with 3:1 volume ratio of H_2SO_4:HNO_3 for 24 h. Then 400 ml of water was added to the resultant solution for quenching the reaction. The resultant product was then filtered and washed out with water and dried under vacuum at 60 °C for 24 h. In the second step, 2 g of the product was mixed with 200 ml of H_2SO_4 and 6 g potassium permanganate for 24 h caused to a thick paste. Then, 600 ml of distilled water was added to the paste, and the reaction was terminated by addition of aqueous solution of 50 ml H_2O_2, resulting in a yellow brown mixture. Then the mixture was centrifuged and washed out three times with 10% HCl solution and then three times with distilled water. The procedure was followed by sonication for 30 minutes to produce GO stable aqueous dispersion. At the end of this process, the mixture was centrifuged for 15 min at 7000 rpm in order to remove unexfoliated graphite oxide particles. GO (1.0 g), $FeCl_3.6H_2O$ (0.036 mol) and of $FeCl_2.4H_2O$ (0.018 mol) were dissolved in 200 mL deionized water under nitrogen gas with vigorous stirring. Then, NH_3 (25%) was added into the solution until the pH of the solution reached to 10. Stirring was continued for 1 h at 60 ^oC. The color of the bulk solution turned from brown to black immediately. The magnetite GO (MGO) was separated from the solution using a magnet, washed several times with deionized water and ethanol, and dried at ambient temperature.

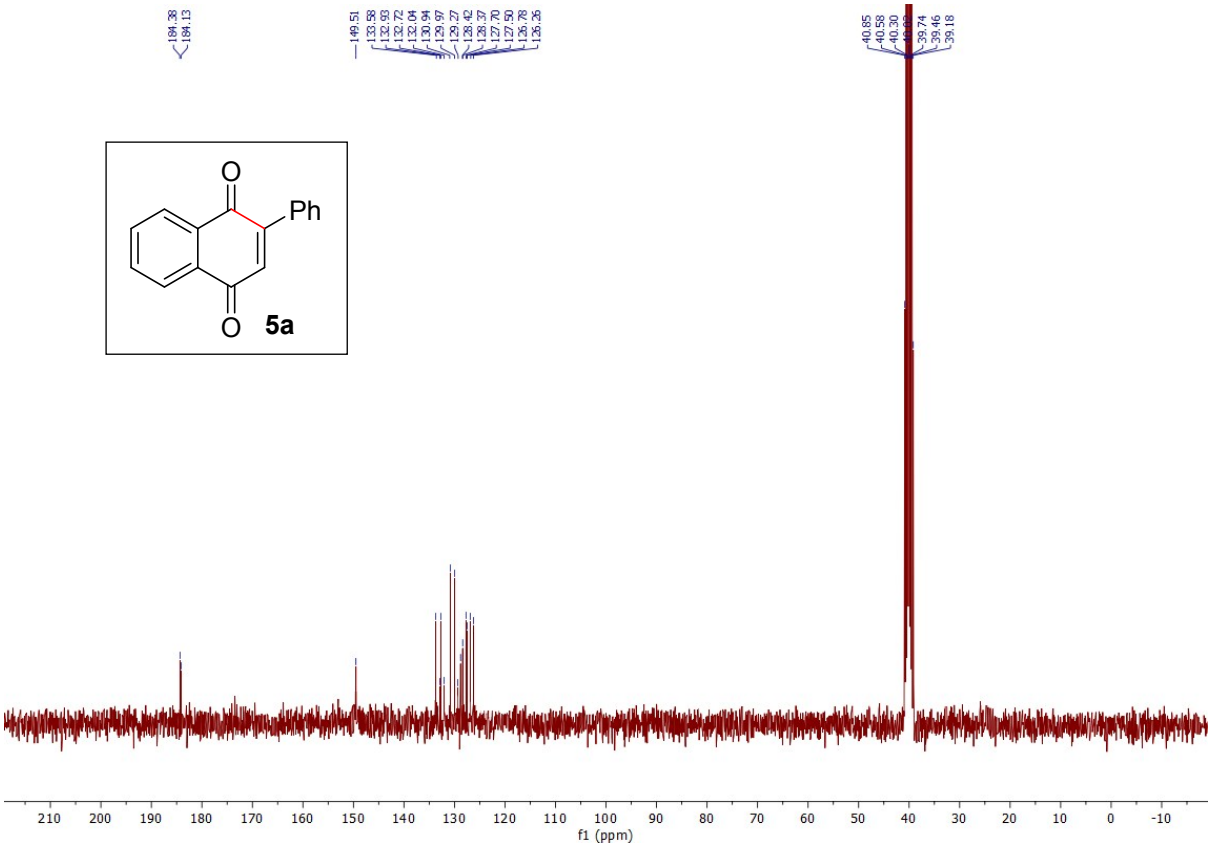
Immobilization of [IMIP]BF_4 on MGO (IMIP-MGO)

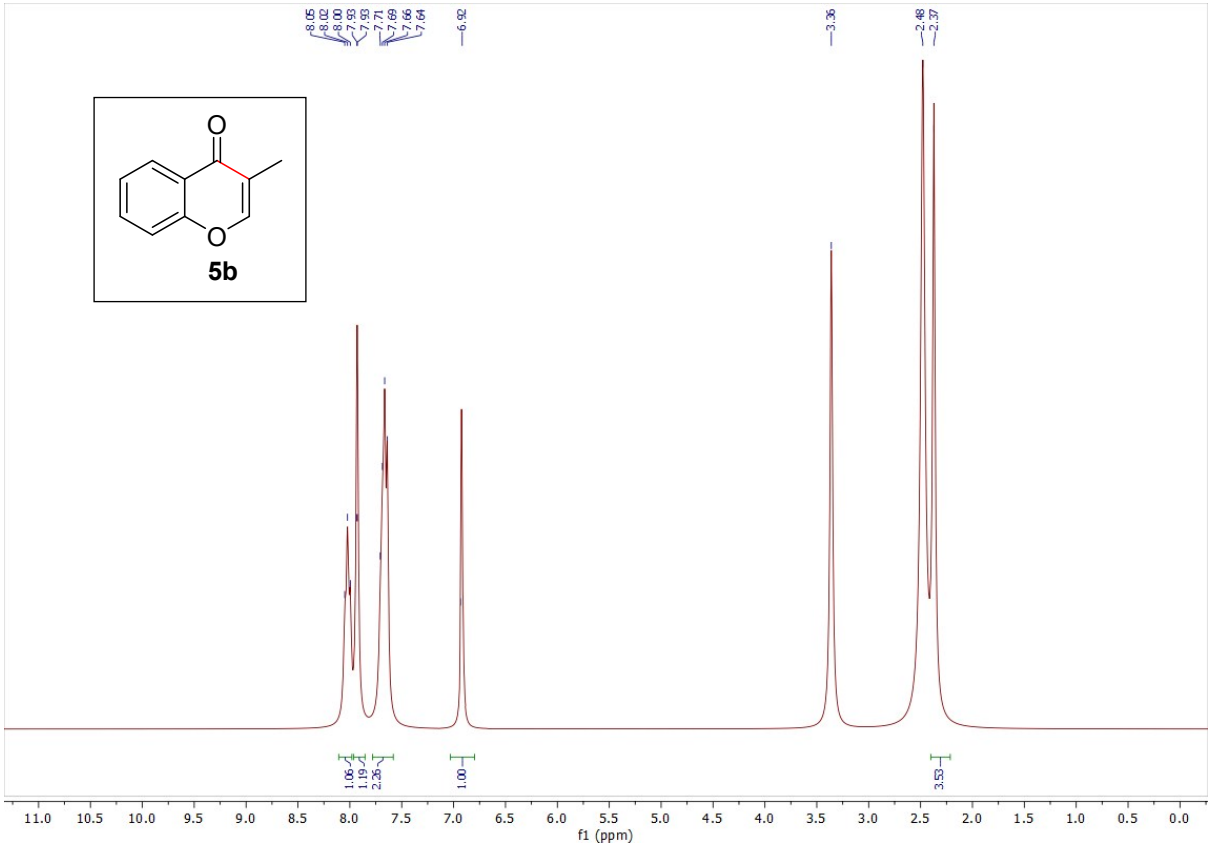
MGO (1.0 g) was added to a solution of [IMIP]BF_4 (0.1 g) in ethanol (10 mL) and slowly stirred for 2 h at room temperature. The adsorption of [IMIP]BF_4 was monitored by UV-Vis spectroscopy. Then the product was collected by an external magnet, washed by ethanol and dried at room temperature. From the UV-Vis spectroscopy data of the reaction mixture and solution of the washed catalyst, the adsorbed [IMIP]BF_4 amount was determined 0.04 g.

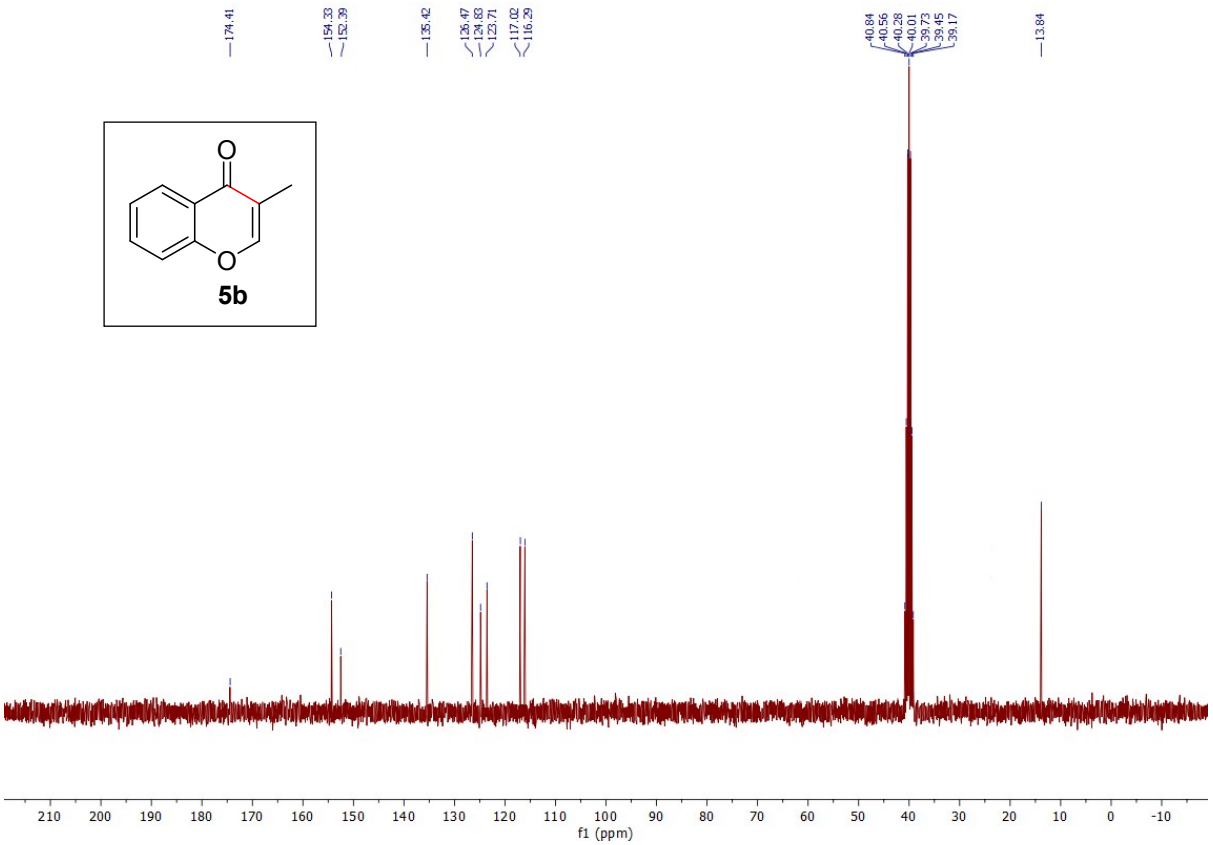


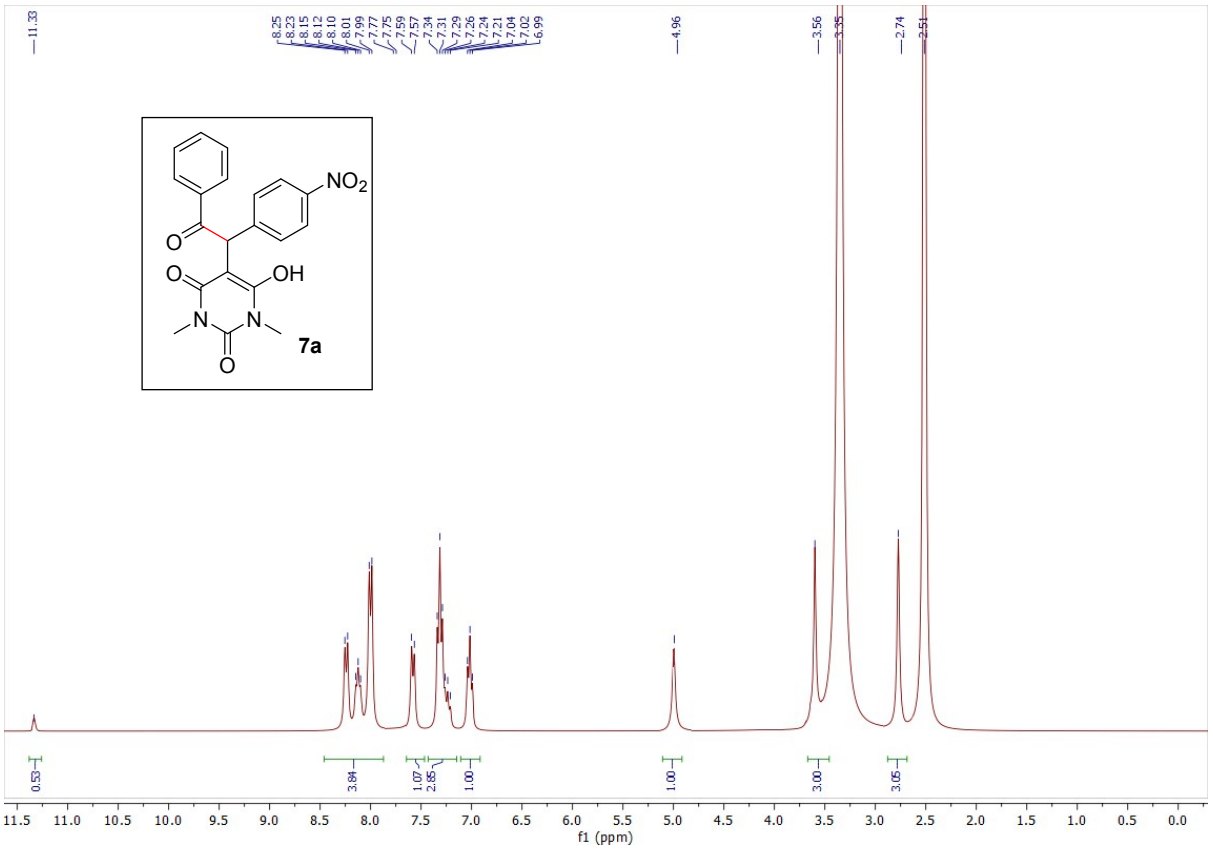

Sepahvand.22.1.1r
Cation
13C{1H} NMR
162.61
149.25
144.78
140.54
134.32
129.81
128.29
124.93
122.83
119.39
107.61
63.62
60.89
59.93
58.52
40.89
40.61
40.33
40.05
39.77
39.50
39.22
33.91
33.88
32.64
30.80
28.27
25.79
25.62
25.46
25.38
25.22
NH
BF4
HN
f1 (ppm)

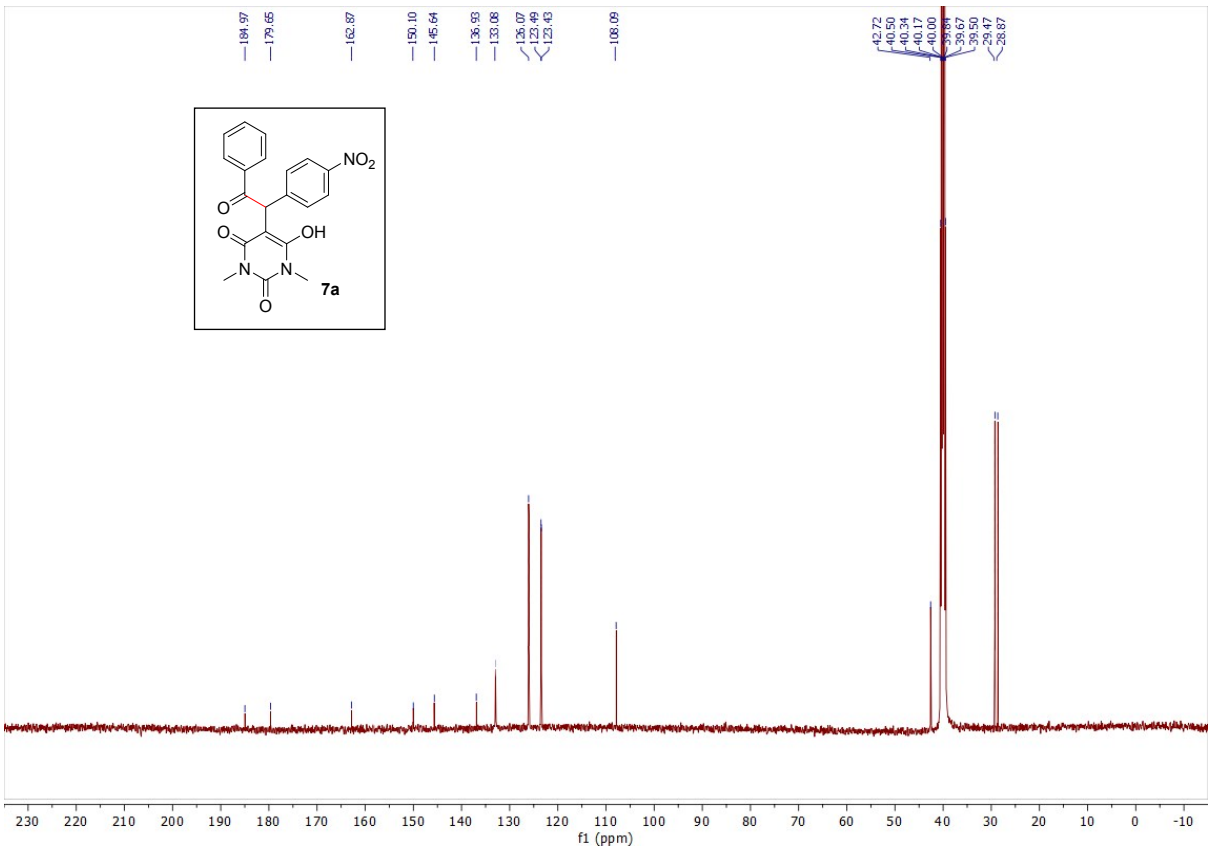

8.60
8.58
8.56
8.55
8.40
8.38
7.75
7.73
7.45
7.43
7.31
7.03
3.52
2.51
Ph
O
5a
0.90
1.00
2.00
5.04
1.00
f1 (ppm)

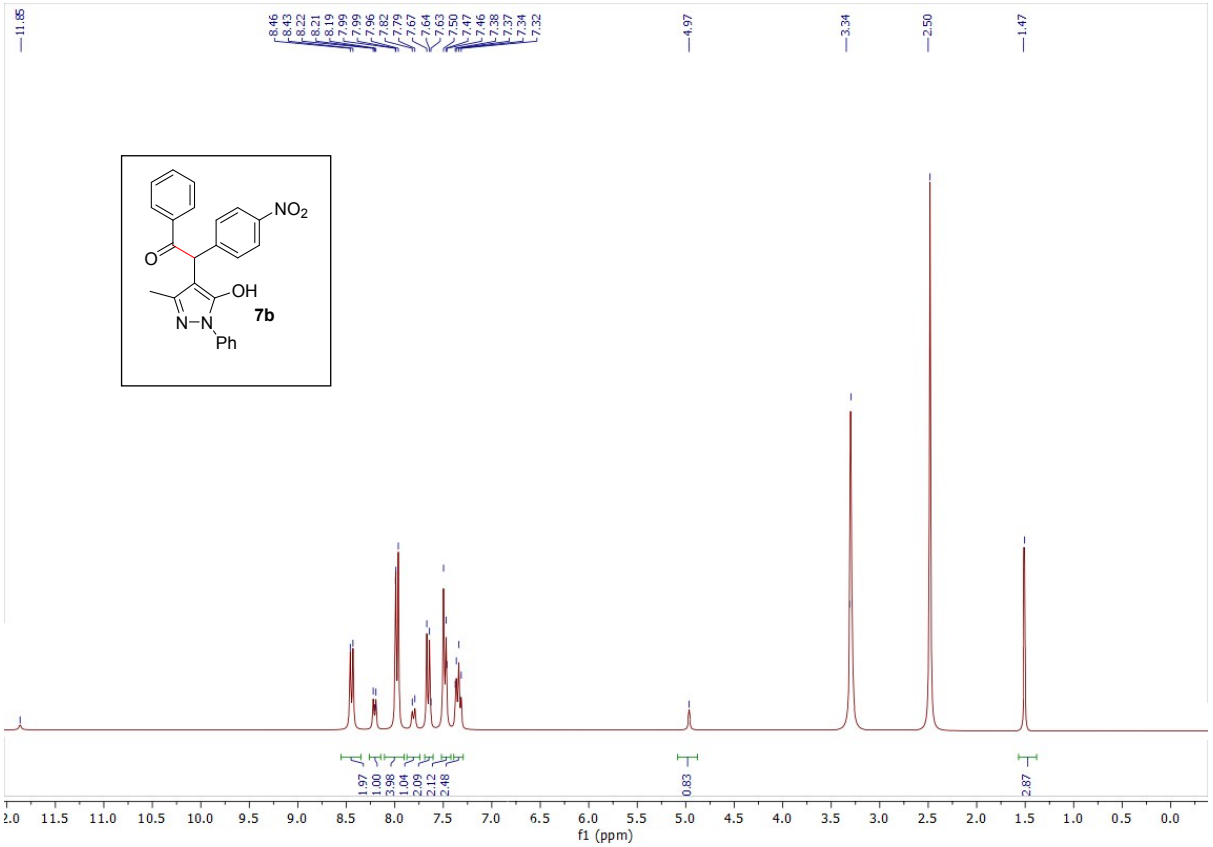

194.38
194.13
149.51
133.58
132.93
132.72
132.04
130.94
129.97
129.27
128.42
128.37
127.70
127.50
126.78
126.26
40.85
40.58
40.30
39.74
39.46
39.18
O
Ph
O
5a
210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)

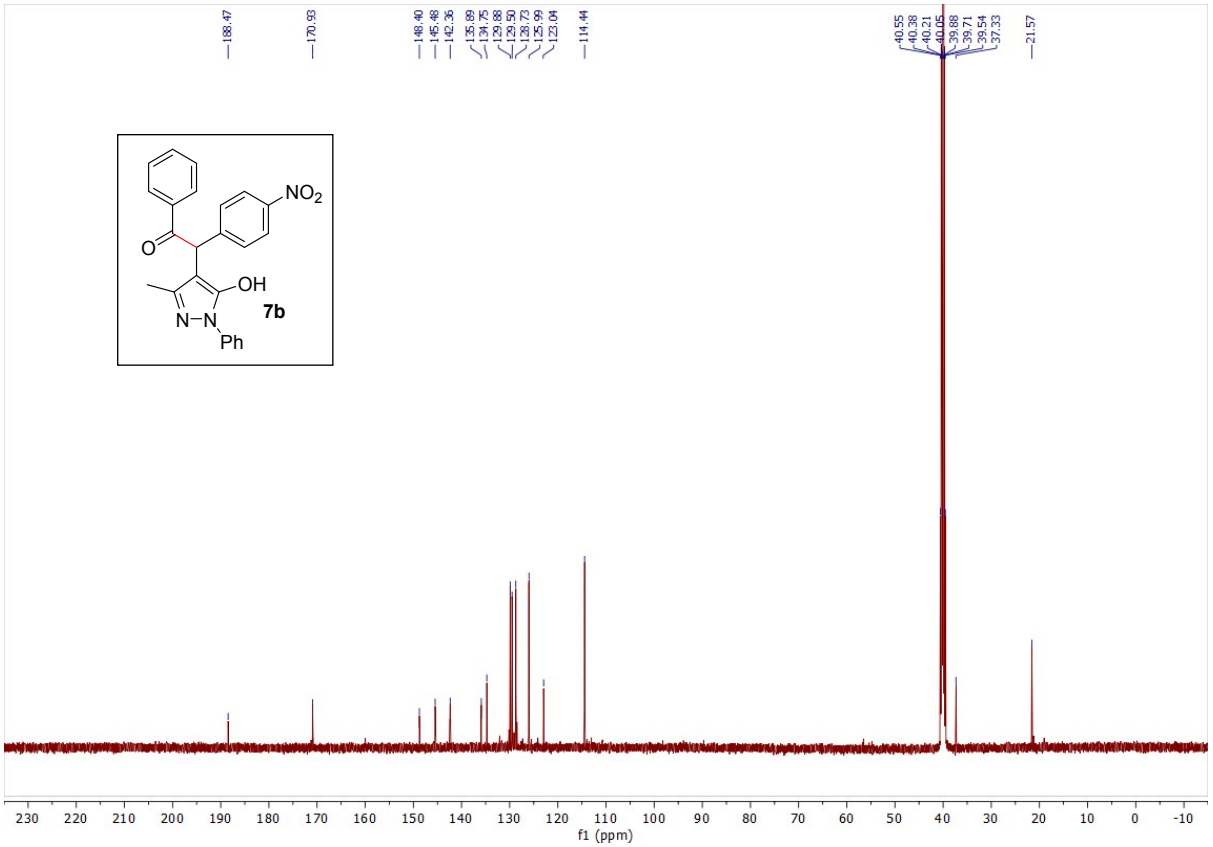

8.05
8.02
8.00
7.93
7.93
7.71
7.69
7.66
7.64
6.92
3.36
2.48
2.37
O
O
5b
1.06
1.19
2.26
1.00
3.53
11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)

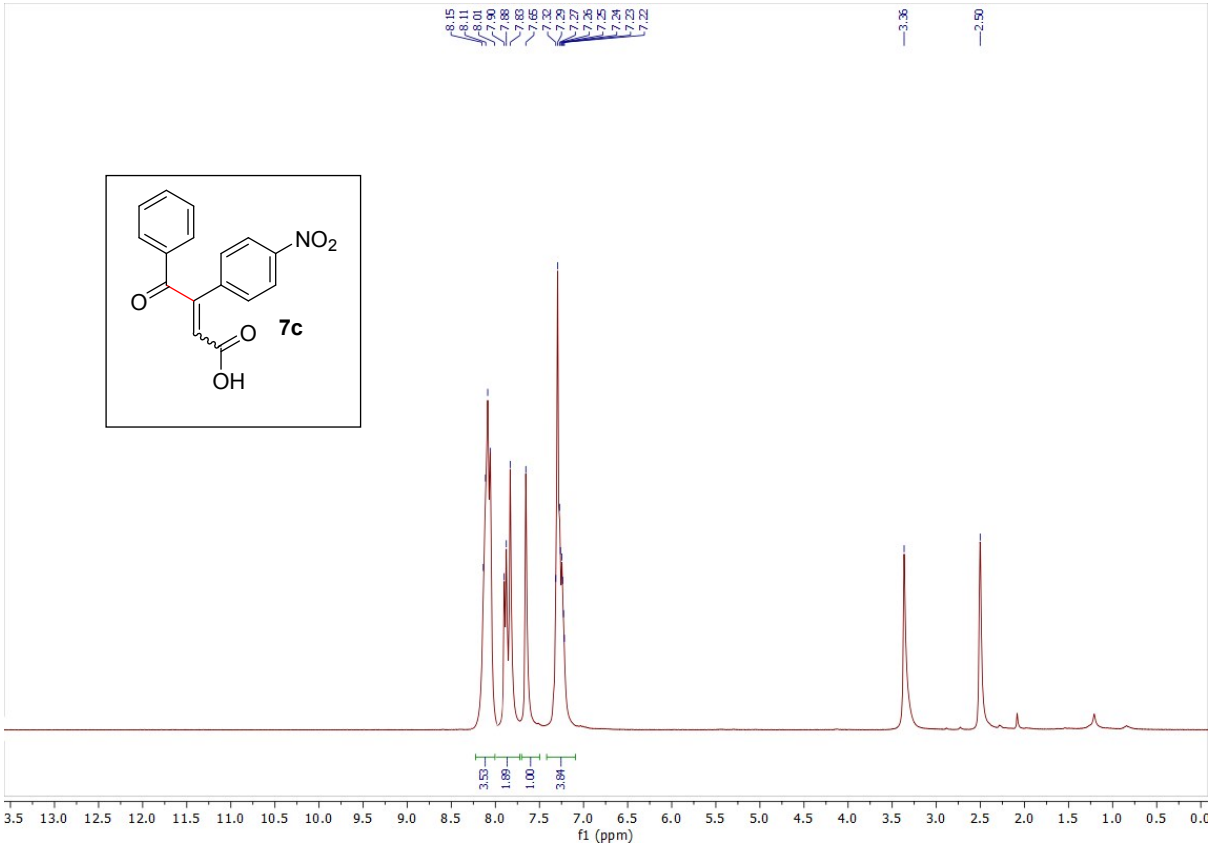

174.41
154.33
152.39
135.42
126.47
124.83
123.71
117.02
116.29
40.84
40.56
40.28
40.01
39.73
39.45
39.17
13.84
O
O
5b
f1 (ppm)

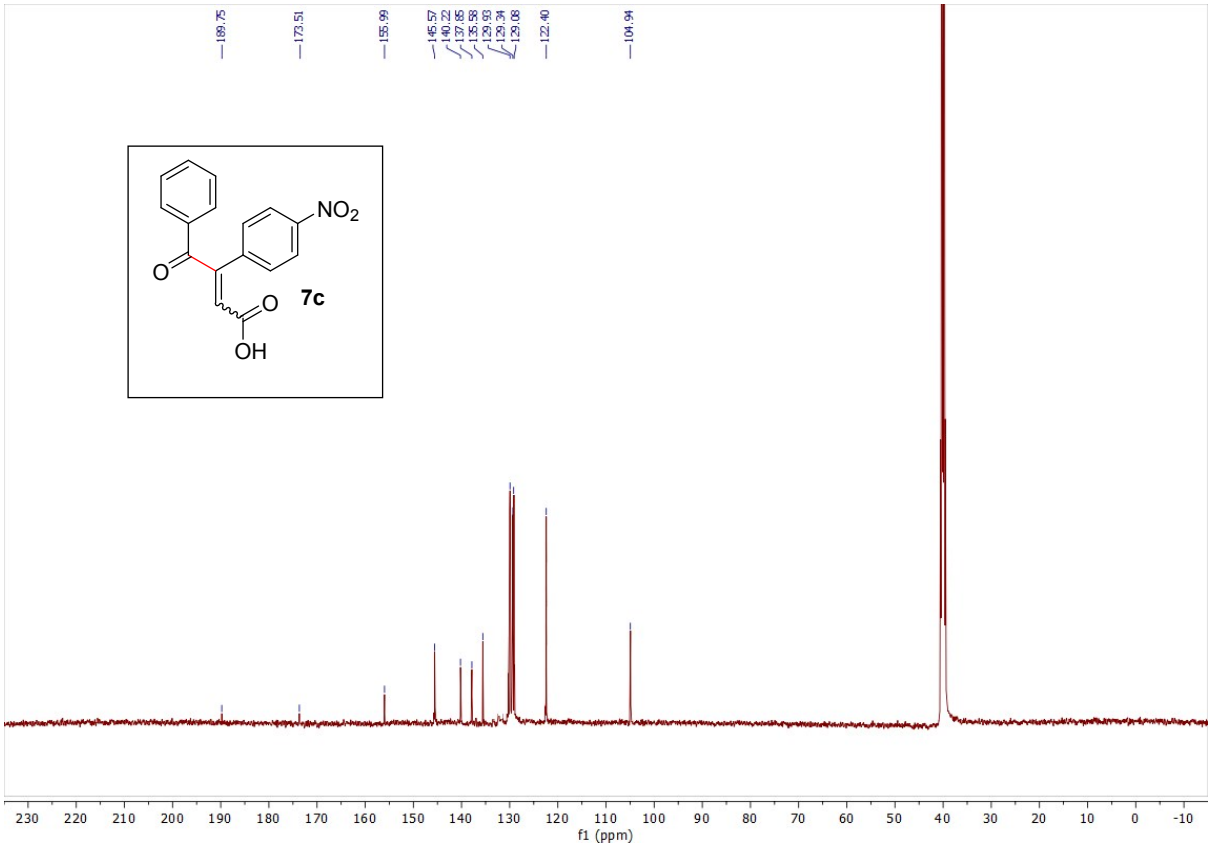

11.33
8.25
8.23
8.15
8.12
8.10
8.01
7.99
7.77
7.75
7.59
7.57
7.34
7.31
7.29
7.26
7.24
7.21
7.04
7.02
6.99
4.96
3.56
3.35
2.74
2.51
NO2
O
O
OH
N
N
O
7a
0.53
3.84
1.07
2.85
1.00
1.00
3.00
3.05
f1 (ppm)


184.97
179.65
162.87
150.10
145.64
136.93
133.08
126.07
123.49
123.43
108.09
42.72
40.50
40.34
40.17
40.00
39.84
39.67
39.50
39.47
28.87
NO2
O
O
OH
N
N
O
7a
230 220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


11.85
8.46
8.45
8.22
8.21
8.19
7.99
7.98
7.96
7.82
7.79
7.67
7.64
7.63
7.50
7.47
7.46
7.38
7.37
7.34
7.32
4.97
3.34
2.50
1.47
NO2
O
OH
N–N
Ph
7b
1.97
1.00
3.98
1.04
2.09
2.12
2.48
0.83
2.87
2.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)


188.47
170.93
148.40
145.48
142.36
135.89
134.75
129.88
129.53
128.73
125.99
123.04
114.44
40.55
40.38
40.21
40.05
39.88
39.71
39.54
37.33
21.57
NO2
O
OH
N–N
Ph
7b
230 220 210 200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0 -10
f1 (ppm)


8.15
8.11
8.01
7.90
7.88
7.83
7.65
7.32
7.29
7.27
7.26
7.25
7.24
7.23
7.22
3.36
2.50
NO2
O
O
OH
7c
3.53
1.89
1.00
3.84
3.5 13.0 12.5 12.0 11.5 11.0 10.5 10.0 9.5 9.0 8.5 8.0 7.5 7.0 6.5 6.0 5.5 5.0 4.5 4.0 3.5 3.0 2.5 2.0 1.5 1.0 0.5 0.0
f1 (ppm)



References

1. J. Brugnerotto, J. Lizardi, F. M. Goycoolea, W. Argüelles-Monal, J. Desbrieres and M. Rinaudo, *Polymer*, 2001, **42**, 3569-3580.