Supplementary Information

An efficient synthesis, Spectroscopic Characterization and Optical Nonlinearity Response of Novel Salicylaldehyde Thiosemicarbazone Derivatives

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Table S1: Natural bond orbital (NBO) analysis of investigated compound (**1**).

Donor (i)	Type	Acceptor (j)	Type	E(2)[a] [kcal/mol]	E(j)-E(i)[b] [a.u.]	F(i,j)[c] [a.u.]
C25-C27	π	C28-C30	π*	26.69	0.3	0.08
C28-C30	π	C24-C26	π*	24.23	0.31	0.078
C24-C26	π	C25-C27	π*	23.79	0.29	0.076
C4-C5	π	C2-C3	π*	22.85	0.30	0.074
C4-C5	π	C1-C6	π*	21.61	0.30	0.072
C24-C26	π	C28-C30	π*	19.35	0.29	0.068
C25-C27	π	C24-C26	π*	18.55	0.31	0.068
C24-C26	π	N21-C22	π*	10.28	0.32	0.053
N21-C22	π	C24-C26	π*	4.87	0.37	0.041
C22-H23	∂	N19-N21	∂*	9.06	0.92	0.082
C4-C5	∂	C3-C4	∂*	4.27	1.30	0.067
C6-H11	∂	C1-C2	∂*	3.97	1.11	0.059
C25-C27	∂	C22-C24	∂*	2.98	1.21	0.054
C1-C6	∂	C5-H10	∂*	2.51	1.13	0.048
C12-H13	∂	C2-C3	∂*	1.97	1.10	0.042
C2-H8	∂	C2-C3	∂*	1.01	1.12	0.03
N15-H16	∂	C17-N19	∂*	0.99	1.09	0.03

C17-N19	∂	N19-H20	∂*	0.52	1.21	0.023
C12-H14	∂	C3-C12	∂*	0.51	0.95	0.02
N15	LP(1)	C17-S18	∂*	49.93	0.30	0.112
N19	LP(1)	C17-S18	∂*	34.05	0.31	0.093
O33	LP(2)	C25-C27	π*	31.89	0.37	0.103
S18	LP(2)	C17-N19	∂*	15.37	0.60	0.087
Br35	LP(3)	C28-C30	π*	10.01	0.32	0.055
N21	LP(1)	N19-H20	∂*	9.41	0.74	0.075
N15	LP(1)	C12-H14	∂*	7.71	0.66	0.068
O33	LP(1)	C25-C27	∂*	6.38	1.20	0.078
N15	LP(1)	C12-H13	∂*	3.08	0.66	0.043
N15	LP(1)	C17-S18	π*	1.21	0.53	0.024
N15	LP(1)	C3-C12	∂*	0.52	0.73	0.019

Table S2: Natural bond orbital (NBO) analysis of investigated compound (**2**).

Donor (i)	**Type**	**Acceptor (j)**	**Type**	**E(2)[a] [kcal/mol]**	**E(j)–E(i)[b] [a.u.]**	**F(i,j)[c] [a.u.]**
C24-C26	π	C27-C29	π*	26.68	0.3	0.08
C3-C4	π	C1-C2	π*	25.66	0.29	0.078
C27-C29	π	C23-C25	π*	24.17	0.31	0.078
C5-C6	π	C3-C4	π*	24.02	0.3	0.076
C23-C25	π	C24-C26	π*	23.73	0.29	0.076
C1-C2	π	C5-C6	π*	22.52	0.32	0.075
C5-C6	π	C1-C2	π*	21.36	0.29	0.07
C1-C2	π	C3-C4	π*	20.93	0.32	0.073
C3-C4	π	C5-C6	π*	20.64	0.3	0.07
C23-C25	π	C27-C29	π*	19.36	0.29	0.068
C24-C26	π	C23-C25	π*	18.54	0.31	0.068
C27-C29	π	C24-C26	π*	17.19	0.3	0.066
C23-C25	π	N20-C21	π*	10.37	0.32	0.053
N20-C21	π	C23-C25	π*	4.97	0.37	0.042
N20-C21	π	N20-C21	π*	1.09	0.39	0.019
C24-C26	π	C24-C26	π*	0.54	0.3	0.011
C21-H22	σ	N18-N20	σ*	9.10	0.92	0.082
C16-S17	σ	C16-S17	σ*	7.91	0.37	0.053
C1-C2	σ	C2-C3	σ*	5.73	1.31	0.078
C23-C25	σ	C27-Br34	σ*	5.15	0.83	0.058
C23-C25	σ	C25-C27	σ*	5.02	1.3	0.072
C23-C24	σ	C24-C26	σ*	4.77	1.28	0.07
C3-C4	σ	C2-F35	σ*	4.12	1	0.057
C5-H9	σ	C1-C6	σ*	3.98	1.11	0.059
C27-Br34	σ	C23-C25	σ*	3.51	1.23	0.059
C27-Br34	σ	C26-C29	σ*	3.15	1.24	0.056

C21-C23	σ	C24-C26	σ*	3.01	1.24	0.054
C24-C26	σ	C21-C23	σ*	2.97	1.21	0.054
C11-N14	σ	N14-C16	σ*	1.84	1.22	0.043
C4-C5	σ	C5-H9	σ*	1.27	1.13	0.034
C26-H30	σ	C26-C29	σ*	1.02	1.12	0.03
C6-H10	σ	C5-C6	σ*	0.75	1.11	0.026
C11-H12	σ	C3-C11	σ*	0.66	0.94	0.022
C27-Br34	σ	C25-H28	σ*	0.58	1.07	0.022
C16-S17	σ	N14-C16	σ*	0.57	0.81	0.019
C16-N18	σ	N18-H19	σ*	0.53	1.21	0.023
C2-F35	σ	C1-C2	σ*	0.51	1.64	0.026
N14-H15	σ	C16-S17	π*	5.24	0.91	0.063
C16-S17	σ	C16-S17	π*	4.16	0.6	0.046
N18-H19	σ	C16-S17	π*	3.85	0.94	0.055
C11-H13	σ	C3-C4	π*	2.44	0.54	0.035
C21-H22	σ	C23-C25	π*	1.76	0.53	0.03
C11-N14	σ	C3-C4	π*	1.57	0.79	0.034
C23-C25	σ	N20-C21	π*	0.63	0.76	0.02
N14	LP(1)	C16-S17	π*	31.93	0.53	0.024
N18	LP(1)	C16-S17	π*	31.14	0.54	0.047
N18	LP(1)	N20-C21	π*	19.14	0.33	0.093
O32	LP(2)	C24-C26	π*	10.03	0.37	0.103
Br34	LP(3)	C27-C29	π*	4.64	0.32	0.055
F35	LP(3)	C1-C2	π*	1.22	0.46	0.091
N14	LP(1)	C16-S17	σ*	48.81	0.3	0.111
N18	LP(1)	C16-S17	σ*	34.31	0.31	0.093
S17	LP(2)	C16-N18	σ*	15.38	0.61	0.088
N20	LP(1)	N18-H19	σ*	9.49	0.74	0.076
F35	LP(2)	C2-C3	σ*	5.94	1.01	0.069
Br34	LP(2)	C27-C29	σ*	3.59	0.88	0.05
F35	LP(1)	C1-C2	σ*	1.13	1.64	0.039
S17	LP(2)	C3-C11	σ*	0.97	0.62	0.023
S17	LP(1)	C16-S17	σ*	0.88	0.71	0.025

Table S3: Natural bond orbital (NBO) analysis of investigated compound (**3**).

Donor (i)	**Type**	**Acceptor (j)**	**Type**	**E(2)[a] [kcal/mol]**	**E(j)–E(i)[b] [a.u.]**	**F(i,j)[c] [a.u.]**
C1-C6	π	C4-C5	π*	25.42	0.29	0.078
C2-C3	π	C1-C6	π*	24.46	0.3	0.076
C4-C5	π	C2-C3	π*	23.92	0.32	0.078
C2-C3	π	C4-C5	π*	19.94	0.29	0.068
C4-C5	π	C1-C6	π*	19.73	0.31	0.07
C1-C6	π	C2-C3	π*	19.35	0.3	0.069

C23-C25	π	N20-C21	π*	9.87	0.32	0.052
N20-C21	π	C23-C25	π*	4.71	0.37	0.041
N20-C21	π	N20-C21	π*	1.07	0.39	0.019
C16-S17	π	C16-S17	π*	0.86	0.88	0.025
C4-C5	π	C4-C5	π*	0.51	0.3	0.011
C21-H22	σ	N18-N20	σ*	9.06	0.92	0.082
C16-S17	σ	C16-S17	σ*	7.89	0.38	0.054
C26-C29	σ	C27-Br34	σ*	5.45	0.84	0.06
C23-C25	σ	C25-C27	σ*	5.01	1.3	0.072
C2-H8	σ	C3-C4	σ*	4.92	1.1	0.066
C23-C25	σ	C23-C24	σ*	4.24	1.27	0.066
C6-H10	σ	C4-C5	σ*	4.06	1.11	0.06
C2-H8	σ	C1-C6	σ*	3.85	1.11	0.058
C6-H10	σ	C1-C2	σ*	3.79	1.12	0.058
N14-C16	σ	N18-N20	σ*	3.29	1.26	0.058
C23-C24	σ	C21-C23	σ*	2.82	1.19	0.052
C3-C11	σ	C3-C4	σ*	2.52	1.23	0.05
C25-C27	σ	C25-H28	σ*	1.85	1.15	0.041
C23-C24	σ	O32-H33	σ*	1.62	1.12	0.038
C5-C6	σ	C6-H10	σ*	1.25	1.14	0.034
C23-C25	σ	C25-H28	σ*	1.14	1.12	0.032
C26-H30	σ	C26-C29	σ*	1.02	1.12	0.03
C11-N14	σ	C3-C11	σ*	0.71	1.2	0.026
C2-C3	σ	C11-N14	σ*	0.56	1.07	0.022
C5-F35	σ	C4-C5	σ*	0.55	1.65	0.027
C16-N18	σ	N18-H19	σ*	0.54	1.21	0.023
N14-H15	σ	C16-S17	π*	5.15	0.91	0.062
C16-S17	σ	C16-S17	π*	4.23	0.6	0.046
C11-H12	σ	C2-C3	π*	4.18	0.54	0.046
N18-H19	σ	C16-S17	π*	3.81	0.93	0.055
C21-H22	σ	C23-C25	π*	1.8	0.54	0.03
C11-N14	σ	C2-C3	π*	1.15	0.8	0.03
C23-C25	σ	N20-C21	π*	0.66	0.76	0.021
N14	LP(1)	C16-S17	σ*	46.77	0.31	0.11
N18	LP(1)	C16-S17	σ*	33.68	0.31	0.093
S17	LP(2)	C16-N18	σ*	15.35	0.61	0.088
N20	LP(1)	C21-C23	σ*	12.26	0.85	0.092
N20	LP(1)	N18-H19	σ*	9.47	0.74	0.076
N14	LP(1)	C11-H13	σ*	7.91	0.66	0.069
N20	LP(1)	C21-H22	σ*	5.48	0.78	0.059
Br34	LP(2)	C27-C29	σ*	3.65	0.88	0.051
N14	LP(1)	C11-H12	σ*	2.61	0.67	0.04
Br34	LP(1)	C25-C27	σ*	1.66	1.58	0.046

S17	LP(1)	C16-S17	σ*	0.9	0.71	0.025
S17	LP(2)	C3-C11	σ*	0.9	0.62	0.022
N14	LP(1)	C3-C11	σ*	0.71	0.73	0.022
C27	LP(1)	C23-C25	π*	70.69	0.17	0.115
C27	LP(1)	C26-C29	π*	70.66	0.17	0.115
C24	LP*(1)	C26-C29	π*	64.49	0.15	0.109
C24	LP*(1)	C23-C25	π*	63.55	0.15	0.108
N18	LP(1)	N20-C21	π*	30.79	0.33	0.093
F35	LP(3)	C4-C5	π*	20.41	0.46	0.093
N14	LP(1)	C16-S17	π*	1.38	0.53	0.025

Table S4: Natural bond orbital (NBO) analysis of investigated compound (**4**).

Donor (i)	**Type**	**Acceptor (j)**	**Type**	**E(2)[a] [kcal/mol]**	**E(j)-E(i)[b] [a.u.]**	**F(i,j)[c] [a.u.]**
C23-C25	π	C26-C28	π*	26.75	0.30	0.081
C1-C6	π	C2-C3	π*	22.84	0.32	0.076
C2-C3	π	C1-C6	π*	20.56	0.29	0.07
C22-C24	π	C26-C28	π*	19.28	0.29	0.068
C23-C25	π	C22-C24	π*	18.68	0.31	0.068
C26-C28	π	C23-C25	π*	17.21	0.30	0.066
N19-C20	π	C22-C24	π*	4.58	0.37	0.04
N19-C20	π	N19-C20	π*	1.09	0.39	0.019
C20-H21	∂	N17-N19	∂*	9.03	0.92	0.082
C5-H35	∂	C3-C4	∂*	3.99	1.11	0.06
C23-C25	∂	C20-C22	∂*	3.01	1.21	0.054
C20-C22	∂	C24-C26	∂*	2.97	1.25	0.054
C1-C6	∂	C1-H7	∂*	1.28	1.14	0.034
C25-H29	∂	C25-C28	∂*	1.02	1.12	0.03
N13-H14	∂	C15-N17	∂*	0.99	1.09	0.03
C6-F34	∂	C1-C6	∂*	0.52	1.65	0.026
C6-F34	∂	C5-C6	∂*	0.50	1.64	0.026
N13	LP(1)	C15-S16	∂*	50.52	0.29	0.112
N17	LP(1)	N19-C20	π*	30.92	0.33	0.093
F34	LP(3)	C1-C6	π*	20.26	0.46	0.093
S16	LP(2)	C15-N17	∂*	15.34	0.61	0.088
N19	LP(1)	N17-H18	∂*	9.48	0.74	0.076
N19	LP(1)	C20-H21	∂*	5.48	0.78	0.059
N17	LP(1)	C15-S16	π*	4.14	0.55	0.045
Br33	LP(2)	C26-C28	∂*	3.63	0.88	0.05
N13	LP(1)	C15-S16	π*	1.04	0.54	0.022
S16	LP(1)	C15-S16	∂*	0.84	0.7	0.024
N13	LP(1)	C3-C10	∂*	0.66	0.73	0.021

Table S5: Natural bond orbital (NBO) analysis of compound (**5**).

Donor (i)	Type	Acceptor (j)	Type	E(2)[a] [kcal/mol]	E(j)-E(i)[b] [a.u.]	F(i,j)[c] [a.u.]
C22-C24	π	C22-C24	π*	0.54	0.3	0.011
N18-C19	π	N18-C19	π*	1.01	0.39	0.018
N18-C19	π	C21-C23	π*	4.73	0.37	0.041
C21-C23	π	N18-C19	π*	10.05	0.32	0.053
C21-C23	π	N18-C19	π*	54.54	0.02	0.056
C25-C27	π	C22-C24	π*	17.25	0.3	0.066
C21-C23	π	C25-C27	π*	19.32	0.29	0.068
C22-C24	π	C21-C23	π*	18.67	0.31	0.068
C3-C4	π	C5-C6	π*	21.54	0.29	0.072
C5-C6	π	C1-C2	π*	22.06	0.3	0.072
C1-C2	π	C3-C4	π*	21.83	0.3	0.073
C1-C2	π	C5-C6	π*	22.83	0.3	0.074
C3-C4	π	C1-C2	π*	23.85	0.29	0.075
C5-C6	π	C3-C4	π*	23.5	0.3	0.076
C21-C23	π	C22-C24	π*	23.91	0.29	0.076
C25-C27	π	C21-C23	π*	24.49	0.31	0.079
C22-C24	π	C25-C27	π*	26.78	0.3	0.081
C34-H37	∂	C4-C34	∂*	0.52	0.95	0.02
C3-C9	∂	C9-N12	∂*	0.63	1	0.022
C19-C21	∂	C19-H20	∂*	0.58	1.08	0.022
C25-Br32	∂	C23-H26	∂*	0.55	1.07	0.022
C25-Br32	∂	C27-H29	∂*	0.58	1.07	0.022
C3-C9	∂	C9-H11	∂*	0.67	1.02	0.023
C4-C5	∂	C34-H35	∂*	0.59	1.1	0.023
C4-C34	∂	C34-H35	∂*	0.66	1.04	0.023
C14-N16	∂	N16-H17	∂*	0.56	1.21	0.023
C22-C24	∂	C22-O30	∂*	0.6	1.09	0.023
C24-H28	∂	C22-C24	∂*	0.59	1.1	0.023
C24-H28	∂	C27-H29	∂*	0.72	0.94	0.023
C1-H7	∂	C2-H8	∂*	0.74	0.93	0.024
C1-H7	∂	C6-H38	∂*	0.74	0.94	0.024
O30-H31	∂	C21-C22	∂*	4.73	1.32	0.071
C21-C23	∂	C23-C25	∂*	5	1.3	0.072
C2-C3	∂	C3-C4	∂*	5.25	1.3	0.074
C22-C24	∂	C21-C22	∂*	5.51	1.29	0.076
C19-H20	∂	N16-N18	∂*	8.89	0.92	0.081
S15	LP2	C14-N16	∂*	15.09	0.61	0.087
S15	LP2	N12-C14	∂*	12.58	0.65	0.082
S15	LP	N12-C14	∂*	3.38	1.16	0.057
S15	LP	C14-N16	∂*	2.92	1.12	0.052
S15	LP2	C9-H10	∂*	0.78	0.57	0.019
O30	LP2	C22-C24	π*	31.93	0.37	0.103
N16	LP	N18-C19	π*	29.79	0.32	0.091
N12	LP	C14-S15	∂*	68.04	0.23	0.117
N12	LP	C9-H11	∂*	7.42	0.66	0.067
N12	LP	C14-N16	∂*	0.66	0.7	0.02
Br32	LP3	C25-C27	π*	10.04	0.32	0.055
Br32	LP	C25-C27	∂*	1.61	1.58	0.045

Table S6: Natural bond orbital (NBO) analysis of investigated compound (**6**).

Donor (i)	Type	Acceptor (j)	Type	E(2)[a] [kcal/mol]	E(j)–E(i)[b] [a.u.]	F(i,j)[c] [a.u.]
C1-C2	π	C3-C4	π*	24.69	0.3	0.077
C1-C2	π	C5-C6	π*	21.31	0.3	0.072
C3-C4	π	C1-C2	π*	21.63	0.3	0.072
C3-C4	π	C5-C6	π*	24.27	0.3	0.077
C3-C4	π	C11-H12	∂*	1.83	0.64	0.033
C3-C4	π	C11-N14	∂*	3.37	0.61	0.044
C5-C6	π	C1-C2	π*	24.71	0.29	0.076
C5-C6	π	C3-C4	π*	22.12	0.29	0.072
N20-C21	π	N20-C21	π*	1.13	0.39	0.019
N20-C21	π	C23-C24	∂*	1.19	0.9	0.029
N20-C21	π	C23-C25	∂*	1.42	0.91	0.032
N20-C21	π	C23-C25	π*	4.77	0.37	0.041
C23-C25	π	N18-H19	∂*	0.9	0.65	0.023
C23-C25	π	N20-C21	∂*	1.34	0.88	0.033
C23-C25	π	N20-C21	π*	9.82	0.32	0.052
C23-C25	π	C21-H22	∂*	1.9	0.68	0.035
C23-C25	π	C24-C26	π*	23.76	0.29	0.076
C23-C25	π	C27-C29	π*	19.36	0.29	0.068
C24-C26	π	C23-C25	π*	18.62	0.31	0.068
C24-C26	π	C24-C26	π*	0.53	0.3	0.011
C24-C26	π	C27-C29	π*	26.68	0.3	0.08
C27-C29	π	C23-C25	π*	24.16	0.31	0.078
C27-C29	π	C24-C26	π*	17.18	0.3	0.066
C1-C2	∂	C1-C6	∂*	3.55	1.31	0.061
C1-C2	∂	C1-H7	∂*	1.19	1.12	0.033
C1-C2	∂	C2-C3	∂*	4.25	1.31	0.067
C1-C2	∂	C2-H8	∂*	1.21	1.12	0.033
C1-C2	∂	C3-C11	∂*	4.03	1.15	0.061
C1-C2	∂	C6-H10	∂*	2.6	1.12	0.048
C29-H31	∂	C26-H30	∂*	0.81	0.92	0.024
C29-H31	∂	C27-C29	∂*	1.18	1.11	0.032
O32-H33	∂	C23-C24	∂*	4.74	1.32	0.071
C35-H36	∂	C4-C5	∂*	2.48	1.11	0.047
C35-H37	∂	C4-C5	∂*	1.9	1.11	0.041
C35-H38	∂	C5-C6	∂*	4.6	1.1	0.064
Br34	LP(3)	C27-C29	π*	10.03	0.32	0.055
S17	LP(2)	C3-C11	∂*	0.91	0.63	0.022
S17	LP(2)	N14-C16	∂*	12.89	0.65	0.083
S17	LP(2)	C16-N18	∂*	15.4	0.6	0.087
O32	LP(2)	C24-C26	π*	31.97	0.37	0.104
Br34	LP(2)	C25-C27	∂*	3.57	0.88	0.05
Br34	LP(2)	C27-C29	∂*	3.63	0.88	0.05
N14	LP(1)	C11-H12	∂*	3.47	0.66	0.046
N14	LP(1)	C11-H13	∂*	7.53	0.66	0.068
N14	LP(1)	C16-S17	∂*	55.03	0.28	0.114
S17	LP(1)	C16-N18	∂*	2.97	1.11	0.052
N18	LP(1)	N20-C21	π*	31.28	0.33	0.094

N20	LP(1)	C21-C23	∂*	12.31	0.84	0.092
O32	LP(1)	C24-C26	∂*	6.4	1.2	0.078
Br34	LP(1)	C25-C27	∂*	1.65	1.58	0.046
Br34	LP(1)	C27-C29	∂*	1.62	1.58	0.045

Table S7: Natural bond orbital (NBO) analysis of investigated compound (**7**).

Donor (i)	Type	Acceptor (j)	Type	E(2)[a] [kcal/mol]	E(j)–E(i)[b] [a.u.]	F(i,j)[c] [a.u.]
C24-C26	π	C27-C29	π*	26.71	0.3	0.08
C2-C3	π	C4-C5	π*	21.89	0.3	0.073
C23-C25	π	C27-C29	π*	19.33	0.29	0.068
C24-C26	π	C23-C25	π*	18.64	0.31	0.068
C23-C25	π	N20-C21	π*	9.61	0.32	0.052
N20-C21	π	C23-C25	π*	4.62	0.37	0.04
N20-C21	π	N20-C21	π*	1.13	0.39	0.019
C21-H22	∂	N18-N20	∂*	9.01	0.92	0.082
C2-H8	∂	C3-C4	∂*	4.78	1.11	0.065
C26-C29	∂	C24-O32	∂*	3.99	1.08	0.059
C24-C26	∂	C21-C23	∂*	3.01	1.21	0.054
C21-C23	∂	C25-C27	∂*	2.97	1.25	0.054
C11-N14	∂	N14-C16	∂*	1.95	1.22	0.044
N14-H15	∂	C16-N18	∂*	1.01	1.09	0.03
C1-H7	∂	C1-C6	∂*	0.99	1.12	0.03
C11-H13	∂	C3-C11	∂*	0.51	0.95	0.02
N14	LP(1)	C16-S17	∂*	54.60	0.28	0.114
N18	LP(1)	C16-S17	∂*	39.11	0.29	0.097
O32	LP(2)	C24-C26	π*	31.96	0.37	0.104
S17	LP(2)	C16-N18	∂*	15.35	0.6	0.087
N20	LP(1)	N18-H19	∂*	9.49	0.74	0.076
N18	LP(1)	C16-S17	π*	3.38	0.56	0.041
S17	LP(1)	N14-C16	∂*	3.18	1.16	0.055
N14	LP(1)	C11-H12	∂*	2.97	0.66	0.043
Br34	LP(1)	C25-C27	∂*	1.65	1.58	0.046
N14	LP(1)	C16-S17	π*	0.81	0.55	0.02
N14	LP(1)	C3-C11	∂*	0.55	0.73	0.019

$E^{(2)}$ means energy of hyper conjugative interaction (stabilization energy in kcal/mol); Energy difference between donor & acceptor *i* & *j* NBO orbitals.; F(*i;j*) is the Fock matrix element between *i*& *j* NBO orbitals.

Table S8: Computed transition energy (eV), maximum absorption wavelengths (λ_{max}/nm), oscillator strengths (*f*), and transition studies compounds (**1-7**).

Compounds	DFT λ (*nm*)	E(*eV*)	*f*	MO contributions
1	392.94	3.16	0.002	H→L (87%), H→L+3 (2%), H→L+5 (7%)
	331.00	3.75	0.166	H-1→L (85%), H-2→L (2%), H→L+1 (8%)

	315.16	3.93	0.038	H→L+1 (52%), H→L+5 (10%), H-1→L (9%), H→L (8%), H→L+2 (8%), H→L+3 (8%)
	304.14	4.08	0.007	H→L+1 (25%), H→L+2 (66%), H→L+3 (4%), H→L+5 (3%)
	293.70	4.22	0.017	H→L+1 (10%), H→L+2 (21%), H→L+3 (50%), H→L+5 (12%), H→L (2%)
	290.75	4.26	0.062	H-2→L (80%), H-3→L (4%), H-2→L+3 (2%), H-1→L (3%), H-1→L+3 (4%)
2	388.91	3.19	0.001	H→L (86%), H→L+1 (3%), H→L+5 (8%)
	322.60	3.84	0.226	H-1→L (94%), H-2→L (2%)
	306.34	4.05	0.002	H→L (12%), H→L+1 (33%), H→L+5 (31%), H→L+2 (3%), H→L+3 (8%), H→L+4 (5%), H→L+7 (2%)
	287.81	4.31	0.061	H-2→L (75%), H-3→L (5%), H-2→L+1 (2%), H-1→L (2%), H-1→L+1 (5%), H→L+2 (4%)
	286.11	4.33	0.033	H→L+1 (14%), H→L+2 (79%), H-2→L (4%)
	277.41	4.47	0.003	H→L+1 (47%), H→L+2 (11%), H→L+3 (16%), H→L+5 (17%), H→L+4 (3%), H→L+7 (2%)
3	385.67	3.21	0.001	H→L (84%), H→L+1 (4%), H→L+3 (4%), H→L+5 (7%)
	319.02	3.89	0.232	H-1→L (94%), H-2→L (3%)
	307.32	4.03	0.002	H→L (14%), H→L+1 (40%), H→L+3 (14%), H→L+5 (22%), H→L+4 (2%), H→L+7 (2%), H→L+8 (2%)
	286.25	4.33	0.041	H-2→L (71%), H-3→L (6%), H-1→L+1 (4%), H→L+2 (9%)
	283.94	4.37	0.034	H→L+2 (89%), H-2→L (6%)
	274.57	4.52	0.004	H→L+1 (54%), H→L+3 (15%), H→L+5 (20%), H→L+4 (2%), H→L+7 (3%)
4	386.71	3.21	0.001	H→L (84%), H→L+1 (4%), H→L+4 (3%), H→L+5 (8%)
	319.57	3.88	0.216	H-1→L (94%), H-2→L (3%)
	309.10	4.01	0.002	H→L (14%), H→L+1 (44%), H→L+4 (11%), H→L+5 (24%), H→L+8 (3%)
	285.84	4.34	0.057	H-2→L (75%), H-3→L (7%), H-2→L+1 (2%), H-1→L (3%), H-1→L+1 (4%)
	281.26	4.41	0.005	H→L+2 (95%)
	275.81	4.50	0.008	H→L+1 (50%), H→L+4 (15%), H→L+5 (28%), H→L+7 (3%)
5	391.80	3.16	0.001	H→L (87%), H→L+1 (4%), H→L+5 (5%)
	328.18	3.78	0.174	H-1→L (93%), H-2→L (3%)
	312.37	3.97	0.007	H→L (11%), H→L+1 (52%), H→L+5 (13%), H→L+2 (7%), H→L+3 (7%), H→L+8 (5%)
	291.47	4.25	0.089	H-2→L (81%), H-3→L (5%), H-1→L (4%), H-1→L+1 (3%)
	278.71	4.45	0.029	H-4→L (20%), H-3→L (38%), H-5→L (5%), H-2→L (5%), H-2→L+1 (3%), H-1→L+1 (9%), H→L+1 (5%), H→L+3 (2%)
	277.59	4.47	0.009	H→L+1 (36%), H→L+2 (16%), H→L+3 (15%), H→L+5 (11%), H-4→L (4%), H-3→L (8%), H→L+4 (2%), H→L+8 (2%)
6	390.42	3.18	0.001	H→L (86%), H→L+1 (3%), H→L+3 (3%), H→L+5 (7%)
	323.60	3.83	0.196	H-1→L (95%), H-2→L (3%)
	309.03	4.01	0.001	H→L (12%), H→L+1 (37%), H→L+3 (14%), H→L+5 (25%), H→L+2 (4%), H→L+8 (3%)
	287.78	4.31	0.079	H-2→L (78%), H-7→L+1 (2%), H-3→L (5%), H-2→L+1 (2%), H-1→L (3%), H-1→L+1 (5%)

	280.56	4.42	0.017	H→L+1 (54%), H→L+2 (18%), H→L+5 (13%), H→L+3 (9%), H→L+7 (2%)
	273.20	4.54	0.043	H→L+2 (69%), H-3→L (3%), H-1→L+1 (5%), H→L+1 (3%), H→L+3 (9%), H→L+5 (6%)
7	389.53	3.18	0.001	H→L (86%), H→L+1 (3%), H→L+5 (8%)
	322.20	3.85	0.199	H-1→L (94%), H-2→L (3%)
	309.92	4.00	0.001	H→L (13%), H→L+1 (39%), H→L+2 (11%), H→L+5 (27%), H→L+4 (3%), H→L+7 (2%), H→L+8 (3%)
	286.95	4.32	0.076	H-2→L (75%), H-7→L+1 (2%), H-3→L (8%), H-2→L+1 (2%), H-1→L (3%), H-1→L+1 (5%)
	279.78	4.43	0.013	H→L+1 (54%), H→L+2 (20%), H→L+5 (17%), H→L+4 (2%), H→L+7 (3%)
	272.64	4.55	0.086	H-4→L (30%), H-3→L (40%), H-2→L (12%), H-6→L (4%), H-2→L+1 (3%), H-1→L+1 (6%)

(H = HOMO, L = LUMO, H-1 = HOMO-1, etc.)

Table S9: Calculated vibrational frequencies of investigated compound (**1**)**.**

Freq	I_{IR}	**Vibrational assignments**
3889	135	υ O-H
3564	22	υ N-H
3466	29	υ N-H
3184	20	υ (s)C-H_{Ben}
3174	26	υ(as)+(s)C-H_{Ben}
3151	15	υ(as) C-H_{BrBen}
3144	5	υ C-H
3057	9	υ (as)C-H_{CH2}
3008	24	υ (s)C-H_{CH2}
1688	18	υ C=N + υ(ρ)N-H + υ(ρ)C-H
1643	26	υ (C=C-C=C_{Ben}) + υ(ρ)C-H_{BrBen} + υ(ρ)O-H
1627	15	υ (C=C-C=C_{Ben}) + υ(δ)C-H_{BrBen}
1543	356	υ(ρ)N-H + υ C-N
1520	6	υ(ρ)C-H_{Ben} + υ C-C + υ(ρ)N-H
1515	92	υ(ρ)C-H_{BrBen} + υ C-O
1508	355	υ(ρ)N-H + υ C-N
1477	15	υ(δ)C-H_{CH2} + υ(ρ + δ)C-H_{Ben}
1473	19	υ(δ)C-H_{CH2} + υ(ρ + δ)C-H_{Ben}
1436	97	υ (C=C-C=C_{Ben}) + υ(δ)C-H_{BrBen} + υ C-O
1397	22	υ C=S + υ(ρ)C-H + υ N-N
1380	217	υ C=S + υ(ρ)N-H + υ C-N + υ (w)C-H_{CH2}
1361	336	υ C=S + υ(ρ)N-H + υ C-N + υ (w)C-H_{CH2}
1339	24	υ (C=C-C=C_{Ben}) + υ(ρ)C-H_{BrBen} + υ(ρ)O-H
1320	20	υ(ρ)C-H_{Ben}
1312	113	υ(ρ)C-H_{BrBen} + υ C-O
1283	60	υ(ρ)C-H_{BrBen} + υ(ρ)O-H
1244	369	υ(ρ)N-H + υ C-N + υ(τ)C-H_{CH2}
1227	14	υ(ρ)C-H_{Ben} + υ (w)C-H_{CH2} + υ C-C
1224	54	υ(ρ)C-H_{BrBen} + υ N-N + υ(ρ)O-H
1215	41	υ(τ)C-H_{CH2} + υ N-N + υ(ρ)N-H

1172	61	$\upsilon(\rho)$O-H + $\upsilon(\rho + \delta)$C-H_{BrBen}
1119	100	$\upsilon(\rho)$O-H + $\upsilon(\rho + \delta)$C-H_{BrBen}
1098	6	$\upsilon(\rho + \delta)$C-H_{Ben}
1070	48	υ N-N + υ C-N
1047	6	$\upsilon(\rho + \delta)$C-H_{Ben}
974	8	$\upsilon(\rho)$C-H_{CH2}
944	11	$\upsilon(\tau)$C-H_{BrBen} + $\upsilon(\gamma)$C-H_{BrBen}
922	17	$\upsilon(\tau)$C-H_{BrBen} + $\upsilon(\gamma)$C-H_{BrBen} + $\upsilon(\gamma)$C-H_{Ben}
910	16	υ C=S + $\upsilon(\tau)$C-H_{BrBen} + $\upsilon(\gamma)$C-H_{BrBen} + υ C-Br_{BrBen}
893	21	$\upsilon(\gamma)$C-H + $\upsilon(\gamma)$C-H_{BrBen} + υ C-Br_{BrBen}
864	29	υ C-Br_{BrBen} + $\upsilon(w)$C-H_{CH2} + υ C-Br_{BrBen}
808	49	$\upsilon(w)$C-H_{BrBen}
797	36	υ C=S + $\upsilon(w)$C-H_{Ben}
763	13	$\upsilon(w)$C-H_{Ben}
720	26	$\upsilon(w)$C-H_{Ben} + $\upsilon(\gamma)$C-H

Frequencies are given in cm^{-1}, υ =stretching, β=in-plane bending, γ=out-plane bending δ=scissoring, ρ =rocking, w= wagging, s =symmetric, as=asymmetric, τ=twisting, ChBen = chlorobenzene, BrBen = bromobenzene.

Table S10: Calculated vibrational frequencies of investigated compound (**2**)**.**

Freq	I_{IR}	**Vibrational assignments**
3889	135	υ O-H
3578	20	υ N-H
3466	29	υ N-H
3194	7	υ(s) C-H_{Ben}
3183	14	υ(s) + υ(as) C-H_{Ben}
3151	15	υ(as) C-H_{BrBen}
3146	5	υ C-H
3086	5	υ(as) C-H_{CH2}
3012	21	υ(s) C-H_{CH2}
1689	18	υ C=N
1671	11	υ (C=C-C=C_{FlBen})
1643	26	υ (C=C-C=C_{BrBen})
1641	16	υ (C=C-C=C_{FlBen}) + $\upsilon(\delta)$ C-H_{FlBen}
1626	14	υ (C=C-C=C_{BrBen}) + $\upsilon(\rho)$ O-H + $\upsilon(\delta)$ C-H_{BrBen}
1550	180	$\upsilon(\rho)$ N-H
1547	430	$\upsilon(\rho)$ N-H
1524	70	$\upsilon(\rho)$ + $\upsilon(\delta)$ C-H_{BrBen}
1514	106	$\upsilon(\rho)$ O-H + $\upsilon(\rho)$ C-H_{BrBen}
1465	21	$\upsilon(\delta)$ C-H_{CH2}
1435	93	$\upsilon(\rho)$ C-H + υ C-Br
1378	373	υ C=S

1355	172	υ(w) C-H_{CH2}
1313	92	υ C-O + υ(ρ) C-H_{BrBen}
1283	53	υ(ρ) O-H + υ(ρ) C-H_{BrBen}
1272	145	υ C-F
1256	463	υ(ρ) N-H
1224	43	υ(ρ) O-H
1172	64	υ(ρ) O-H + υ(δ) C-H_{BrBen}
1119	106	υ(ρ) O-H + υ(δ) C-H_{BrBen}
1071	50	υ C-N + υ N-N
910	18	υ(τ)C-H_{BrBen}
895	18	υ(γ)C-H
863	29	υ(τ) + υ(w) C-H_{FlBen}
856	19	υ(τ) + υ(w) C-H_{FlBen}
808	59	υ(w) C-H_{BrBen}
796	23	β(C=C-C=C_{BrBen}) + β(C=C-C=C_{FlBen})
765	58	υ(w) C-H_{FlBen}

Frequencies are given in cm^{-1}, *υ* =stretching, *β*=in-plane bending, γ=out-plane bending *δ*=scissoring, *ρ* =rocking, *w*= wagging, s =symmetric, as=asymmetric,*τ*=twisting, Ben=benzene ring BrBen=Bromo benzene, FlBen=Fluro Benzene.

Table S11: Calculated vibrational frequencies of investigated compound (**3**)**.**

Freq	***I_{IR}***	**Vibrational assignments**
3889	136	υ O-H
3560	20	υ N-H
3457	34	υ N-H
3195	5	υ(s) C-H_{FlBen}
3180	12	υ(s) + υ(as) C-H_{FlBen}
3147	14	υ(as) C-H_{BrBen}
3136	5	υ C-H
3052	10	υ(as) C-H_{CH2}
3009	25	υ(as) C-H_{CH2}
1695	16	υ C=N
1665	42	υ(δ) C-H_{FlBen} + υ (C=C-C=C_{FlBen})
1649	27	υ(ρ) O-H + υ (C=C-C=C_{BrBen})
1644	59	υ C-F + υ (C=C-C=C_{FlBen})
1629	14	υ(ρ) O-H
1546	307	υ(ρ) N-H
1519	156	υ(ρ) N-H + υ(ρ) O-H
1518	78	υ(ρ) N-H + υ(ρ) O-H + υ(ρ) C-H_{Ben}
1511	292	υ(ρ) N-H
1479	51	υ(δ) C-H_{CH2} + υ C-F
1438	94	υ(ρ) C-H + υ(ρ) O-H + υ (C=C-C=C_{BrBen})

1376	107	υ C=S + υ(w) C-H_{CH2}
1360	456	υ C=S + υ(w) C-H_{CH2} + υ C-N
1344	35	υ(ρ) O-H + υ C-H_{BrBen}
1313	99	υ O-H + υ(ρ) C-H_{BrBen}
1294	52	υ C-F + υ(ρ) C-H_{FlBen} + υ(w) C-H_{CH2}
1285	53	υ(ρ) O-H + υ(ρ) C-H_{BrBen}
1273	126	υ(ρ) N-H + υ C-N
1239	319	υ(ρ) N-H + υ C-N + υ(w) C-H_{CH2}
1227	45	υ(ρ) C-H + υ(ρ) O-H + υ N-N
1212	26	υ(τ)C-H_{CH2} + υ N-N
1192	4	υ(τ)C-H_{CH2} + υ N-N + υ C=S
1173	68	υ(ρ) O-H + υ(δ) C-H_{BrBen}
1143	25	υ(w) C-H_{CH2}
1120	99	υ(ρ) O-H + υ(δ) C-H_{BrBen}
1069	45	υ C-N + υ N-N
958	17	υ(ρ) C-H_{CH2}
947	16	υ(τ)C-H_{BrBen} + υ(γ)C-H
922	16	υ(τ)C-H_{BrBen} + υ(γ)C-H
914	20	υ(γ) C-H
895	24	υ(γ)C-H_{BrBen} + υ(γ)C-H
873	25	υ(γ)C-H_{FlBen}
863	36	β(C=C-C=C_{BrBen})
810	50	υ(w) C-H_{BrBen}
802	42	υ(γ)υ (C=C-C=C_{FlBen}) + υ(w) C-H_{FlBen}

Frequencies are given in cm^{-1}, υ =stretching, β=in-plane bending, γ=out-plane bending δ=scissoring, ρ =rocking, w= wagging, s =symmetric, as=asymmetric,τ=twisting, Ben=benzene ring BrBen=Bromo benzene, FlBen=Fluro Benzene.

Table S12: Calculated vibrational frequencies of investigated compound (**4**)**.**

[a]***Freq***	[a]I_{IR}	**Vibrational assignments**
3887	137	υ O-H
3562	21	υ N-H
3454	33	υ N-H
3194	4	υ (s)C-H_{FlBen}
3189	3	υ (s)C-H_{FlBen}
3159	7	υ(as) C-H_{FlBen}
3148	6	υ C-H
3145	14	υ(as) C-H_{BrBen} + υ C-H
3064	7	υ(as)C-H_{CH2}
3010	26	υ(s)C-H_{CH2}
1693	18	υ C=N + υ(ρ)N-H + υ(ρ)C-H
1668	39	υ (C=C-C=C_{Ben}) + υ(δ)C-H_{FlBen} + υ C-F_{FlBen}
1647	27	υ (C=C-C=C_{Ben}) + υ(ρ)C-H_{BrBen} + υ(ρ)O-H

1626	13	υ (C=C-C=C_{Ben}) + υ(ρ + δ)C-H_{BrBen} + υ(ρ)O-H
1546	61	υ(ρ)C-H_{FlBen} + υ C-F_{FlBen} + υ(ρ)N-H + υ C-N
1543	373	υ(ρ)N-H + υ C-N
1516	103	υ(ρ)C-H_{BrBen} + υ C-O + υ(ρ)N-H + υ C-N
1511	391	υ(ρ)N-H + υ C-N
1474	21	υ(δ)C-H_{CH2}
1436	94	υ (C=C-C=C_{Ben}) + υ(δ)C-H_{Ben} + υ C-O + υ(ρ)O-H
1385	58	υ C=S + υ(ρ)C-H
1379	134	υ C=S + υ(w)C-H_{CH2} + υ C-N
1360	393	υ C=S + υ(w)C-H_{CH2} + υ C-N
1342	29	υ (C=C-C=C_{Ben}) + υ(ρ)C-H_{BrBen} + υ(ρ)O-H
1313	110	υ(ρ)C-H_{BrBen} + υ C-O
1284	79	υ(ρ)C-H_{BrBen} + υ(ρ)O-H + υ C-F_{FlBen}
1283	88	υ C-F_{FlBen} + υ(ρ) C-H_{FlBen} + υ(w)C-H_{CH2}
1246	366	υ(ρ)N-H + υ C-N + υ(τ)C-H_{CH2}
1171	68	υ(ρ + δ)C-H_{BrBen} + υ(ρ)O-H
1156	20	υ(δ)C-H_{FlBen}
1119	101	υ(δ)C-H_{BrBen} + υ C-Br_{BrBen}
1071	50	υ C-N + υ N-N
1022	6	υ(ρ)C-H_{FlBen}
948	14	υ(τ)C-H_{BrBen} + υ(γ)C-H_{BrBen}
914	24	υ(τ)C-H_{BrBen} + υ(γ)C-H_{BrBen} + υ C-Br_{BrBen}
880	14	υ(γ)C-H + υ C-F_{FlBen} + υ C-Br_{BrBen}
846	46	υ(w)C-H_{FlBen} + υ C-F_{FlBen}
802	14	υ C=S + υ(w)C-H_{FlBen} + υ(w)C-H_{CH2} + υ C-F_{FlBen}
793	26	υ C=S + υ(ρ)N-H
628	20	υ C-Br_{BrBen}

ρ=rocking, *w*= wagging, s =symmetric, as=asymmetric, τ=twisting, ChBen = chlorobenzene, BrBen = bromobenzene, FlBen = Florobenzene.

Table S13: Calculated vibrational frequencies of investigated compound (**5**)**.**

[a]Freq	***[a]I_{IR}***	**Vibrational assignments**
3889	140	υO-H_{Brph}
3570	18	υN-H
3455	32	υ N-H
3180	26	υ (s)C-H_{Ben}
3165	17	υ(as)+(s)C-H_{Ben}
3148	14	υ(as)C-H_{Brph} + υ C-H
3142	17	υ(as)C-H_{Ben}
3111	15	υ(s)+(as)C-H_{CH3}
3084	19	υ(s)+(as)C-H_{CH3}
3062	15	υ(as)C-H_{CH2}
3024	32	υ(s)C-H_{CH3}
3021	32	υ (s)C-H_{CH2}
1689	14	υ C=N + υ(ρ)N-H + υ(ρ)C-H
1643	28	υ (C=C-C=C_{Brph}) + υ(ρ)O-H_{Brph} + υ(ρ)C-H_{Brph}
1626	15	υ (C=C-C=C_{Brph}) + υ(ρ)O-H_{Brph} + υ(δ)C-H_{Brph}

1536	474	υ C-N + υ(ρ)N-H
1529	127	υ C-N + υ(ρ)N-H
1521	18	υ (C=C-C=C_{Ben}) + υ ($\delta + \rho$)C-H_{Ben} + υ($\tau + \delta$) C-H_{CH3}
1515	105	υ(ρ)O-H_{Brph} + υ(ρ)C-H_{Brph} + υC-O_{Brph} + υ(ρ)N-H
1484	11	υ(δ)C-H_{CH2} + υ($\tau + \delta$) C-H_{CH3}
1466	12	υ (ρ)C-H_{Ben} + υ(δ)C-H_{CH2} + υ($\tau + \delta$) C-H_{CH3}
1433	96	υ (C=C-C=C_{Brph}) + υ(ρ)O-H_{Brph} + υ(δ)C-H_{Brph}
1402	18	υ (w)C-H_{CH3} + υC=S
1388	304	υC=S + υ(ρ)N-H + υ C-N
1350	210	υC=S + υ(ρ)N-H + υ (w)C-H_{CH2}
1339	31	υ(ρ)O-H_{Brph} + υ(ρ)C-H_{Brph}
1312	113	υ(ρ)C-H_{Brph} + υC-O_{Brph}
1295	29	υ (ρ)C-H_{Ben}
1281	54	υ(ρ)C-H_{Brph} + υ(ρ)O-H_{Brph}
1249	294	υ(ρ)N-H + υ (τ) C-H_{CH2} + υ C-N
1238	79	υ(ρ)N-H + υ (w)C-H_{CH2} + υ ($\delta + \rho$)C-H_{Ben}
1217	25	υ (τ)C-H_{CH2} + υ(ρ)N-H + υ(ρ)O-H_{Brph}
1171	64	υ ($\delta + \rho$)C-H_{Brph} + υ(ρ)O-H_{Brph}
1118	97	υ ($\delta + \rho$)C-H_{Brph} + υ(ρ)O-H_{Brph}
1076	8	υ ($\delta + \rho$)C-H_{Ben}
1065	56	υ C-N + υ(ρ)C-H
971	11	υ (w + τ) C-H_{Ben} + υ(ρ)N-H + υ(ρ)C-H_{CH2}
949	18	υ ($\tau + \beta$) C-H_{Brph} + υ(β)C-H
923	14	υ ($\tau + \beta$) C-H_{Brph} + υ(β)C-H + υC=S
864	29	υ(β)C-H + υ(ρ)N-H
862	9	υ(β)C-H
855	23	υ(β)C-H + υ(ρ)N-H
810	44	υ (w) C-H_{Brph}
794	38	υC=S + υ (w) C-H_{Ben}
755	23	υ (w) C-H_{Ben}

Frequencies are given in cm^{-1}, υ =stretching, β=in-plane bending, γ=out-plane bending δ =scissoring, ρ=rocking, w= wagging, s =symmetric, as=asymmetric, τ=twisting, 4-bromophenol = Brph, Ben=benzene ring.

Table S14: Calculated vibrational frequencies of investigated compound (**6**)**.**

[a]***Freq***	[a]I_{IR}	**Vibrational assignments**
3889	135	υO-H_{Brph}
3565	24	υN-H
3454	30	υ N-H
3171	21	υ (s)C-H_{Ben}
3153	17	υ(as)+(s)C-H_{Ben}
3146	14	υ(as)C-H_{Brph}
3139	5	υ C-H
3130	13	υ C-H_{Ben}
3109	18	υ(s)+(as)C-H_{CH3}
3086	14	υ(as)C-H_{CH3}

3059	10	υ(as)C-H_{CH2}
3019	28	υ (s)C-H_{CH3}
3016	23	υ(s)C-H_{CH2}
1694	19	υ C=N + $\upsilon(\rho)$N-H + $\upsilon(\rho)$C-H
1664	13	υ (C=C-C=C_{Ben}) + υ ($\delta + \rho$)C-H_{Ben}
1649	27	υ (C=C-C=C_{Brph}) + $\upsilon(\rho)$O-H_{Brph} + $\upsilon(\rho)$C-H_{Brph}
1628	14	υ (C=C-C=C_{Brph}) + υ ($\delta + \rho$)C-H_{Brph}
1545	304	υ C-N + $\upsilon(\rho)$N-H
1518	99	$\upsilon(\rho)$O-H_{Brph} + $\upsilon(\rho)$C-H_{Brph} + υC-O_{Brph} + $\upsilon(\rho)$N-H
1518	17	$\upsilon(\rho)$O-H_{Brph} + $\upsilon(\rho)$C-H_{Brph} + υC-O_{Brph} + $\upsilon(\rho)$N-H
1512	404	υ C-N + $\upsilon(\rho)$N-H
1487	15	υ ($\delta + \rho$)C-H_{Ben} + $\upsilon(\tau + \delta)$ C-H_{CH3}
1475	28	υ (δ)C-H_{CH2}
1470	9	$\upsilon(\tau + \delta)$ C-H_{CH3}
1437	94	υ (C=C-C=C_{Brph}) + $\upsilon(\rho)$O-H_{Brph} + $\upsilon(\delta)$C-H_{Brph}
1402	18	$\upsilon(\rho)$C-H + υ N-N
1380	129	υ C=S + $\upsilon(\rho)$N-H + υ(w)C-H_{CH2}
1361	421	υ C=S + $\upsilon(\rho)$N-H + υ(w)C-H_{CH2} + υ C-N
1344	27	υ (C=C-C=C_{Brph}) + $\upsilon(\rho)$O-H_{Brph} + $\upsilon(\rho)$C-H_{Brph}
1341	19	υ (C=C-C=C_{Ben}) + υ (ρ)C-H_{Ben} + υ(w)C-H_{CH2}
1313	94	$\upsilon(\rho)$C-H_{Brph} + υC-O_{Brph}
1309	19	$\upsilon(\rho)$C-H_{Ben}
1285	57	$\upsilon(\rho)$O-H_{Brph} + $\upsilon(\rho)$C-H_{Brph}
1272	28	υ ($\delta + \rho$)C-H_{Ben} + $\upsilon(\rho)$N-H + υ(w)C-H_{CH2}
1242	356	$\upsilon(\rho)$N-H + $\upsilon(\tau)$ C-H_{CH2} + υ N-H
1227	67	$\upsilon(\rho)$O-H_{Brph} + υ(w)C-H_{CH2} + $\upsilon(\rho)$N-H + υ N-N
1223	28	$\upsilon(\rho)$O-H_{Brph} + υ(w)C-H_{CH2} + $\upsilon(\rho)$N-H
1183	2	$\upsilon(\rho)$C-H_{Ben} + υ(w)C-H_{CH2}
1173	66	υ ($\delta + \rho$)C-H_{Brph} + $\upsilon(\rho)$O-H_{Brph}
1119	98	υ ($\delta + \rho$)C-H_{Brph} + $\upsilon(\rho)$O-H_{Brph}
1113	6	$\upsilon(\rho)$C-H_{Ben}
1071	47	υ C-N + υ N-N + $\upsilon(\rho)$C-H
1051	5	υ (τ + w) C-H_{CH3} + $\upsilon(\beta)$ C-H_{Ben}
975	6	$\upsilon(\rho)$C-H_{CH2} + υ (τ + w) C-H_{CH3}
946	16	υ ($\tau + \beta$) C-H_{Brph} + $\upsilon(\beta)$ C-H
923	23	υ ($\tau + \beta$) C-H_{Brph} + $\upsilon(\beta)$ C-H
920	5	υ ($\tau + \beta$) C-H_{Brph} + $\upsilon(\beta)$ C-H + $\upsilon(\beta)$ C-H_{Ben}
894	20	$\upsilon(\beta)$ C-H
863	41	υ C-O_{Brph} + $\upsilon(\rho)$N-H
810	59	υ (w) C-H_{Brph} + υ (w)C-H_{Ben}
807	23	υ (w) C-H_{Brph} + υ (w)C-H_{Ben}
790	27	υ (w)C-H_{Ben} + $\upsilon(\rho)$N-H
769	10	υ (w)C-H_{Ben} + υ (w)C-H_{CH2} + $\upsilon(\rho)$N-H
630	16	υ C=S + υ C-Br

Frequencies are given in cm^{-1}, υ =stretching, β=in-plane bending, γ=out-plane bending δ =scissoring, ρ=rocking, *w*= wagging, s =symmetric, as=asymmetric, τ=twisting, 4-bromophenol = Brph, Ben=benzene ring.

Table S15: Calculated vibrational frequencies of investigated compound (**7**).

[a]*Freq*	[a]I_{IR}	**Vibrational assignments**
3887	136	υ O-H
3565	23	υ N-H
3451	31	υ N-H
3161	27	υ (s)C-H_{Ben}
3147	9	υ (as)C-H_{Ben}
3145	14	υ (as)C-H_{BrBen}
3143	14	υ (as)C-H_{Ben}
3110	14	υ (s) + (as)C-H_{CH3}
3082	14	υ (s) + (as)C-H_{CH3}
3061	8	υ(as)C-H_{CH2}
3016	35	υ(s)C-H_{CH3}
3010	25	υ(s)C-H_{CH2}
1692	20	υ C=N +υ(ρ)N-H + υ(ρ)C-H
1647	28	υ (C=C-C=C_{Ben}) + υ(ρ)C-H_{BrBen} + υ(ρ)O-H
1626	13	υ (C=C-C=C_{Ben}) + υ(ρ)C-H_{BrBen} + υ(ρ)O-H
1534	463	υ(ρ)N-H + υ C-N
1516	116	υ(ρ)C-H_{BrBen} + υ(ρ)N-H + υ(ρ)O-H
1510	184	υ(ρ)N-H + υ C-N
1474	20	υ(δ)C-H_{CH2}
1471	19	υ(τ + δ)C-H_{CH3}
1459	9	υ(τ + δ)C-H_{CH3}
1436	92	υ(δ + ρ)C-H_{BrBen} + υ(ρ)O-H
1381	223	υ C=S + υ(ρ)N-H + υ C-N + υ(ρ)C-H
1376	56	υ(w)C-H_{CH2} + υ(ρ)C-H + υ C-N
1357	289	υ C=S + υ(w)C-H_{CH2} + υ(ρ)C-H
1342	33	υ(ρ)C-H_{BrBen} + υ(ρ)C-H + υ(ρ)O-H
1325	15	υ (C=C-C=C_{Ben}) + υ(w)C-H_{CH2}
1314	20	υ(ρ)C-H_{Ben}
1313	103	υ(ρ)C-H_{Ben}
1284	60	υ(ρ)C-H_{BrBen}
1242	289	υ(τ)C-H_{CH2} + υ(ρ)N-H
1241	25	υ(w)C-H_{CH3} + υ C-C
1228	61	υ(ρ)C-H_{Ben} + υ (w)C-H_{CH2}
1171	67	υ(ρ + δ)C-H_{ChBen} + υ(ρ)O-H
1119	104	υ(ρ + δ)C-H_{ChBen} + υ(ρ)O-H
1070	46	υ C-N + υ N-N + υ (w)C-H_{CH2}
1048	11	υ(τ + w)C-H_{CH3}
948	13	υ(τ)C-H_{BrBen} + υ(γ)C-H_{BrBen} + υ C-Br_{BrBen}
921	12	υ(τ)C-H_{BrBen} + υ(γ)C-H_{BrBen} + υ C-Br_{BrBen}
913	23	υ(γ)C-H_{BrBen} + υ(τ)C-H_{BrBen} + υ C-Br_{BrBen}
823	33	υ(w)C-H_{Ben}
805	43	υ(w)C-H_{BrBen}

Frequencies are given in cm^{-1}, *υ* =stretching, *β*=in-plane bending, γ=out-plane bending *δ*=scissoring, *ρ* =rocking, *w*= wagging, s =symmetric, as=asymmetric, *τ*=twisting, ChBen = chlorobenzene, BrBen = bromobenzene.

Table S16: Computated molecular orbitals energies and their band gap for (**1-7**).

Compounds	E_{HOMO-1}	E_{LUMO+1}	ΔE	E_{HOMO-2}	E_{LUMO+2}	ΔE
1	-6.272	-0.765	5.507	-6.830	-0.502	6.328
2	-6.259	-0.745	5.514	-6.826	-0.710	6.116
3	-6.354	-0.801	5.553	-6.902	-0.753	6.149
4	-6.344	-0.824	5.52	-6.904	-0.722	6.182
5	-6.277	-0.835	5.442	-6.825	-0.454	6.371
6	-6.233	-0.754	5.479	-6.812	-0.476	6.336
7	-6.236	-0.751	5.485	-6.805	-0.410	6.395

Band gap (ΔE)= $E_{LUMO+1} - E_{HOMO-1.}$ Units are in *eV*.

Table S17: Global reactivity descriptors of studied compound (**1-7**).

Compounds	***IP***	***EA***	***X***	***η***	***μ***	***ω***	***σ***
1	5.76	1.59	3.68	2.08	-3.68	3.25	0.24
2	5.76	1.58	3.67	2.09	-3.67	3.22	0.24
3	5.84	1.62	3.73	2.11	-3.73	3.30	0.24
4	5.82	1.62	3.72	2.10	-3.72	3.30	0.24
5	5.78	1.65	3.72	2.06	-3.72	3.35	0.24
6	5.72	1.57	3.64	2.08	-3.64	3.20	0.24
7	5.71	1.55	3.63	2.08	-3.63	3.171	0.24

IP= ionization potential, *EA*= electron affinity, *X*= electro negativity, μ =chemical potential, η =global hardness, σ= global softness and ω= global electrophilicity. Units in eV

Table S18: Dipole moment (μ) and major contributing tensor (D.) of the prepared compounds (**1-7**).

Dipole Moment	1	2	3	4	5	6	7
μ_x	3.1307	2.5773	2.7209	3.3114	3.9315	2.6485	1.9746
μ_y	4.5554	5.4482	3.3246	3.8725	4.8153	5.2727	5.6294
μ_z	5.0734	4.0903	5.6257	4.6153	4.2805	4.9536	4.9313
μ_{total}	7.5028	7.2839	7.0785	6.8747	7.5476	7.7041	7.7400

Table S19: Dipole polarizability and major contributing tensor (*a.u.*) of the studied compounds.

Polarizability	1	2	3	4	5	6	7
α_{xx}	329.26	308.20	330.32	321.56	356.80	331.75	338.25
α_{yy}	366.73	355.66	367.13	367.03	384.21	392.26	391.91
α_{zz}	226.57	252.09	220.96	228.51	233.78	247.00	243.32
α_{total}	307.52	305.32	306.14	305.70	324.93	323.67	324.49

Table S20: Hyperpolarizabilities β_{total} and major contributing tensor (*a.u.*) of the prepared compounds.

Hyperpolarizability	1	2	3	4	5	6	7
β_{xxx}	-287.75	-344.79	-290.15	-181.55	-180.81	-207.20	-145.61
β_{xxy}	-269.03	-261.52	-258.44	-365.97	-198.95	-269.33	-336.95
β_{xyy}	-3.16	4.98	-170.54	15.22	-87.62	-75.16	-1.53
β_{yyy}	164.51	137.07	14.96	-32.76	153.95	88.66	88.22
β_{xxz}	-92.52	-140.09	-106.84	-99.17	-122.35	-132.87	-100.99
β_{yyz}	-15.05	-114.73	76.45	-80.44	-97.29	59.60	-40.11
β_{xzz}	3.41	-52.27	19.27	-13.49	37.63	30.52	-51.93
β_{yzz}	56.42	113.11	-0.99	44.28	57.40	43.46	71.10
β_{zzz}	72.10	9.16	92.50	86.48	24.57	51.76	103.47
β_{total}	293.64	462.82	508.41	408.22	302.44	387.60	269.44

Table S21: The second hyperpolarizabilities **<γ>** and major **γ** tensor (*a.u.*) of the prepared compounds.

2nd Hyper pol.	1	2	3	4	5	6	7
γ_X	8.96×10^4	8.29×10^4	1.01×10^4	8.23×10^4	9.53×10^4	8.40×10^4	8.55×10^4
γ_Y	5.08×10^4	4.73×10^4	8.44×10^4	5.42×10^4	6.07×10^4	5.71×10^4	6.06×10^4
γ_Z	1.42×10^4	1.48×10^4	5.32×10^4	1.34×10^4	1.31×10^4	1.60×10^4	1.43×10^4
$<\gamma>$	1.54×10^5	1.45×10^5	1.51×10^5	1.50×10^5	1.69×10^5	1.57×10^5	1.60×10^5

.

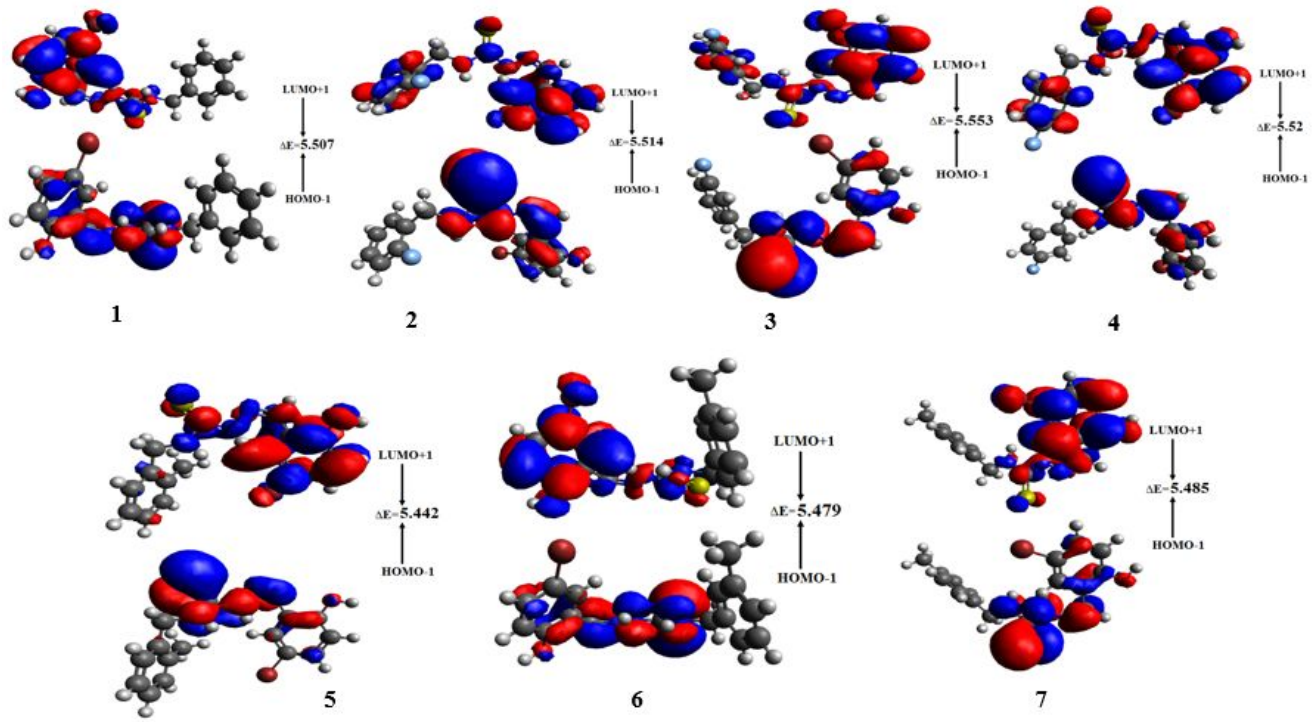


Figure S1: Frontier molecular orbitals (HOMO-1/LUMI+1) of (**1-7**).

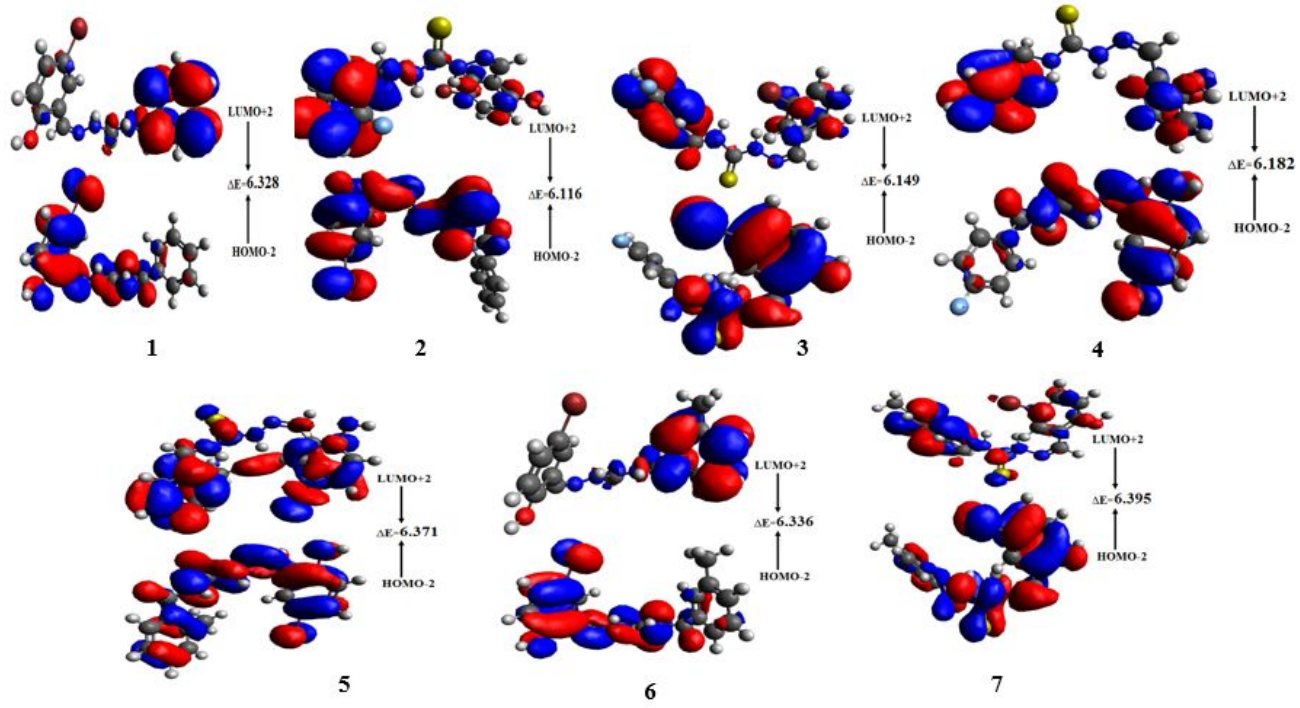


Figure S2: Frontier molecular orbitals (HOMO-2/LUMI+2) of (**1-7**).

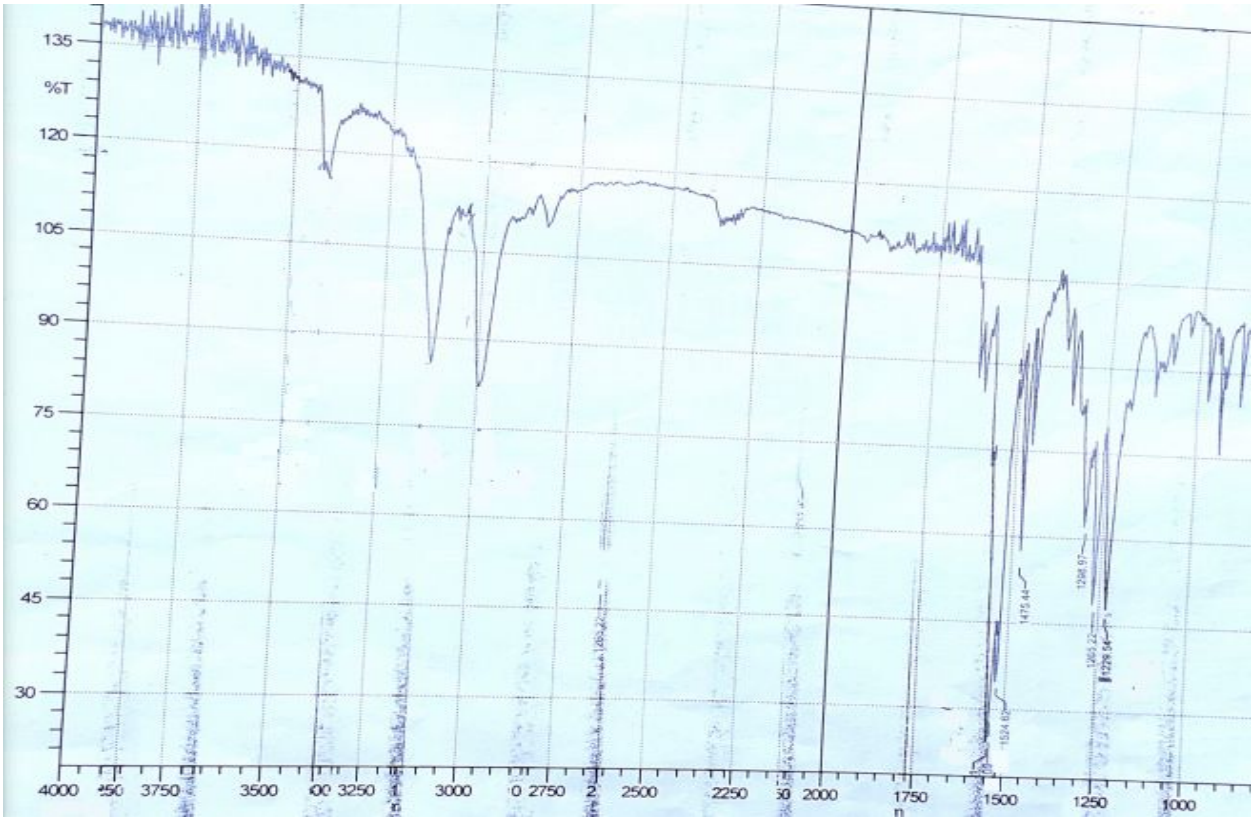
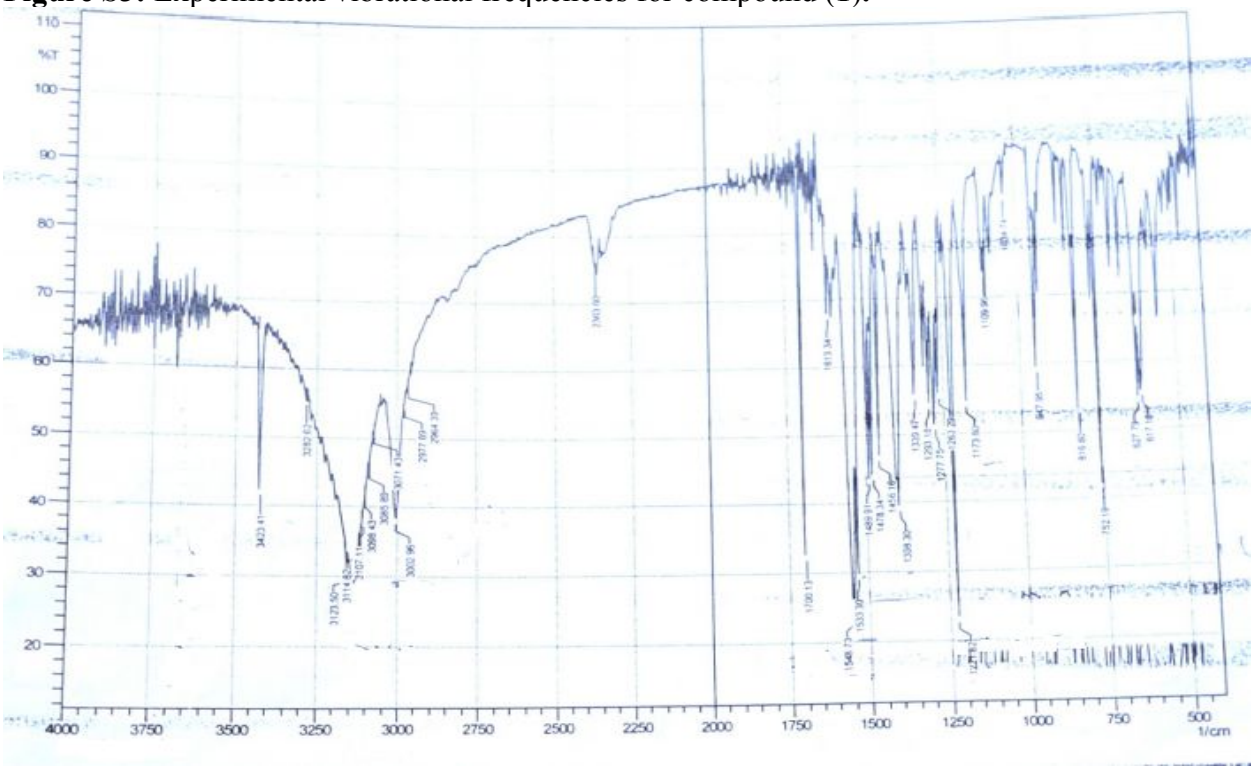


Figure S3: Experimental vibrational frequencies for compound (**1**)**.**



. Figure S4: Experimental vibrational frequencies for compound (**2**).

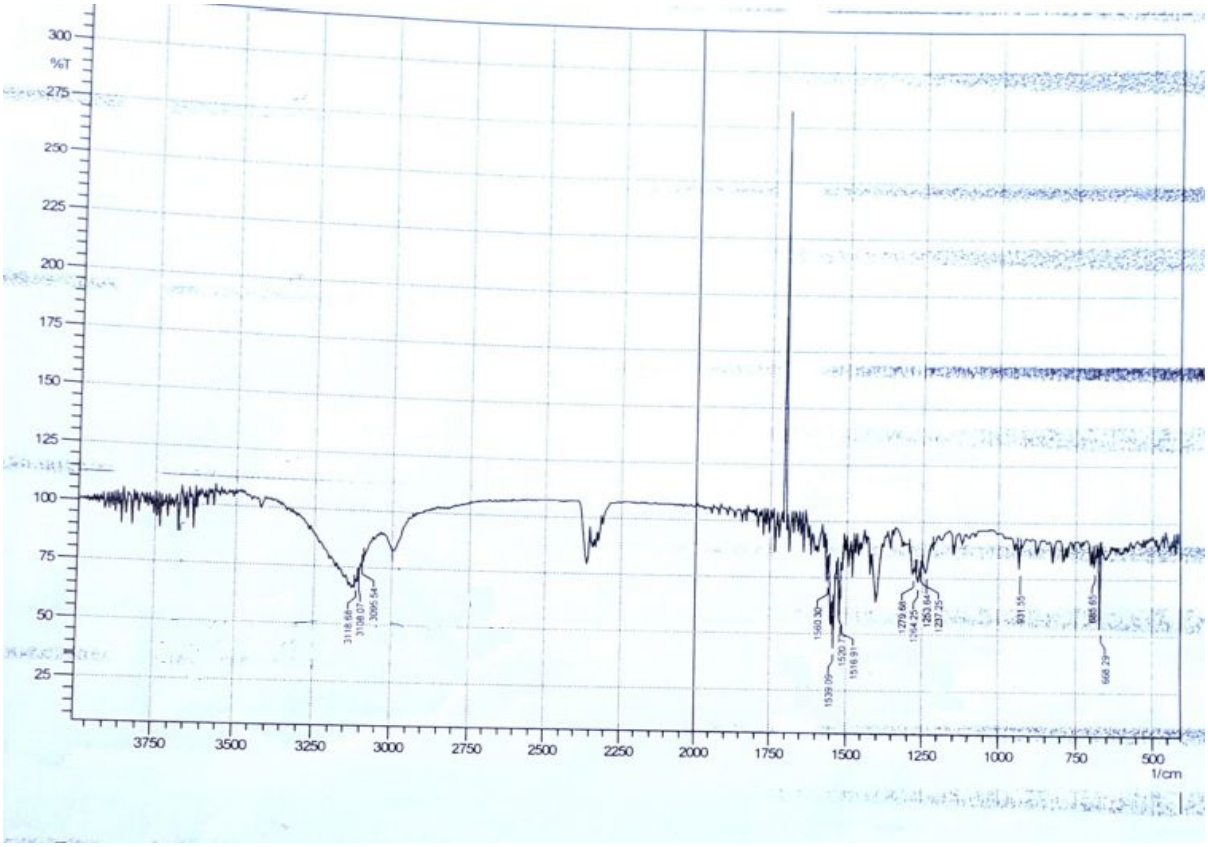


Figure S5: Experimental vibrational frequencies for compound (**3**).

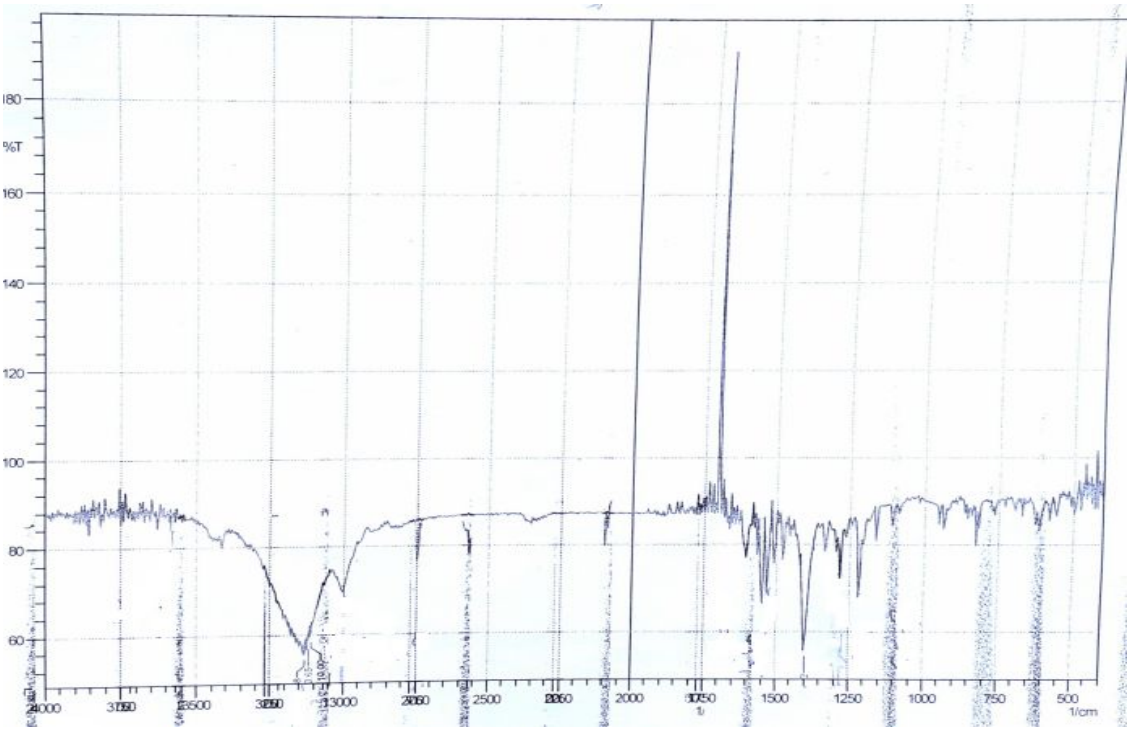


Figure S6: Experimental vibrational frequencies for compound (**4**).

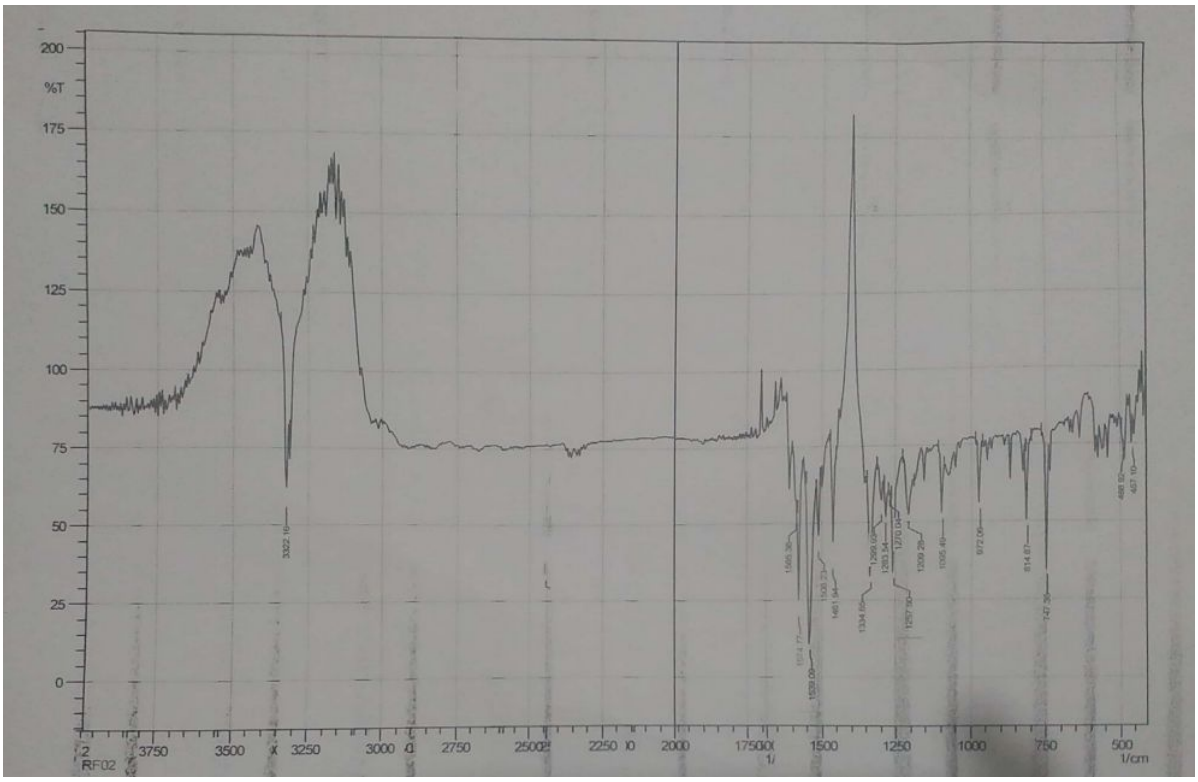


Figure S7: Experimental vibrational frequencies for compound (**5**).

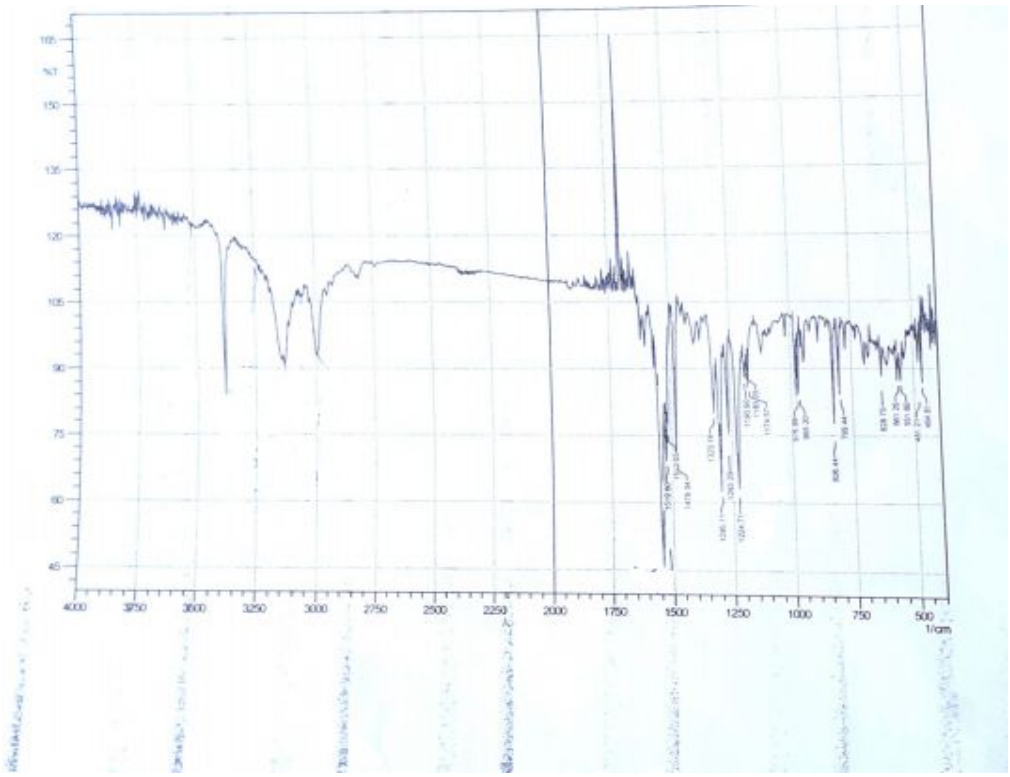


Figure S8: Experimental vibrational frequencies for compound (**6**).

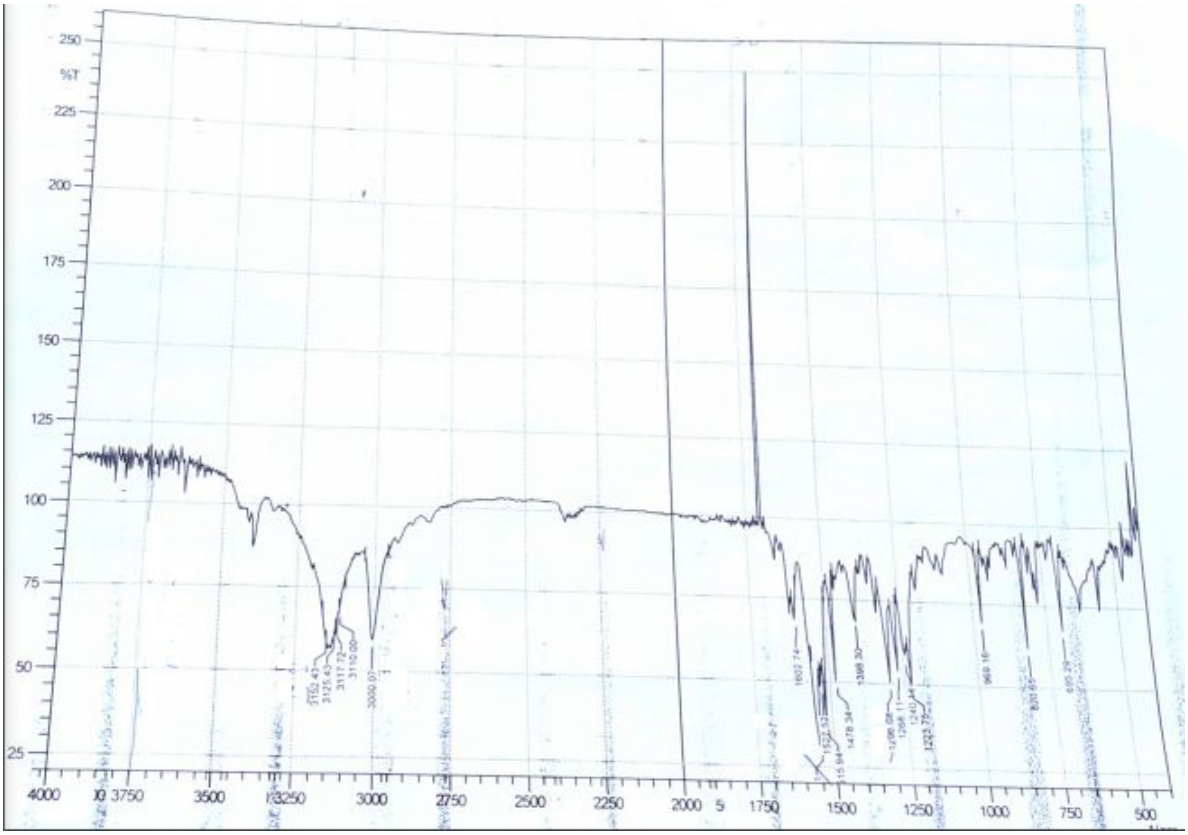


Figure S9: Experimental vibrational frequencies for compound (**7**).

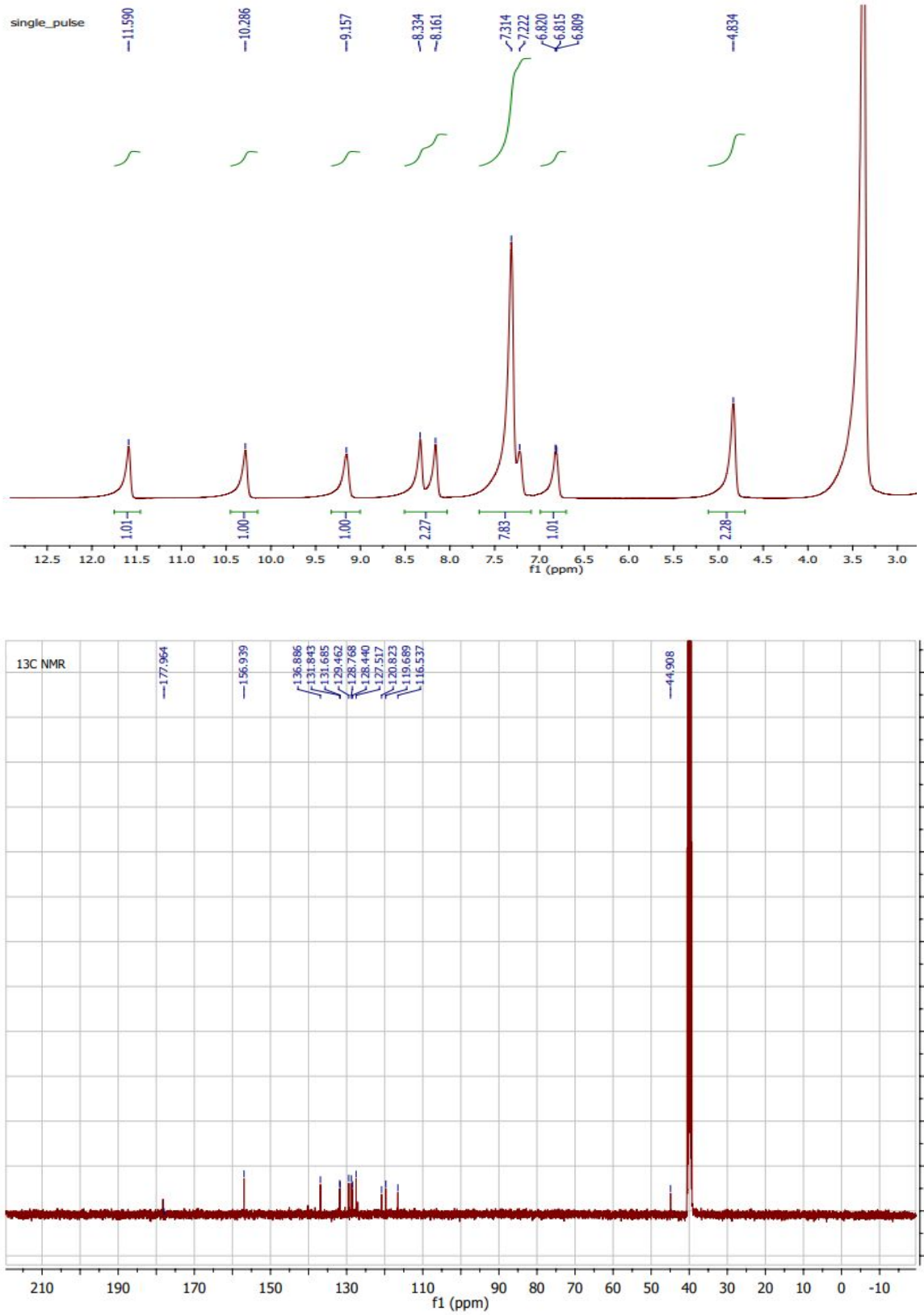


Figure S10: ^{1}H and ^{13}C NMR data of the compound (**1**).

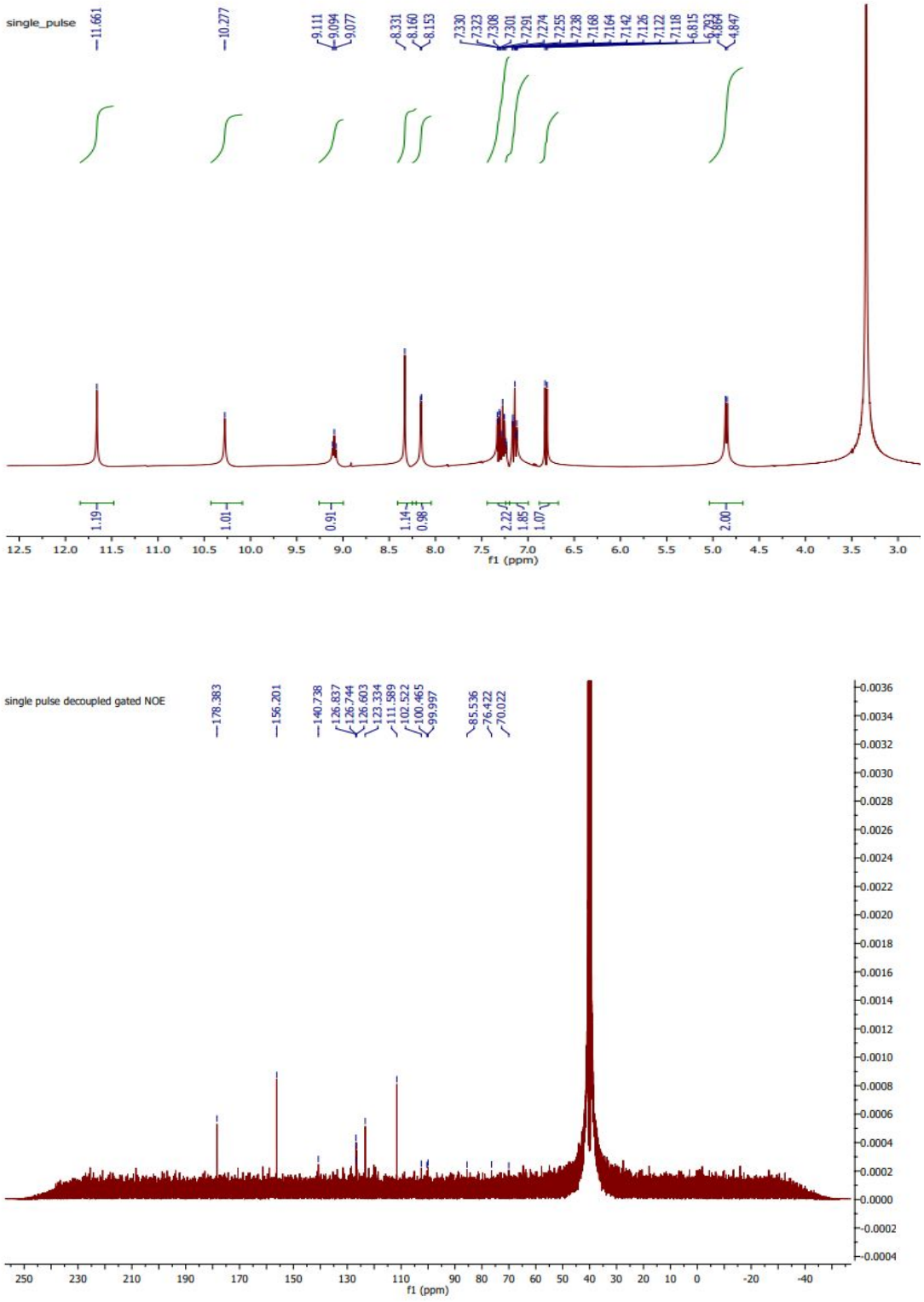


Figure S11: ^{1}H and ^{13}C NMR data of the compound (**2**).

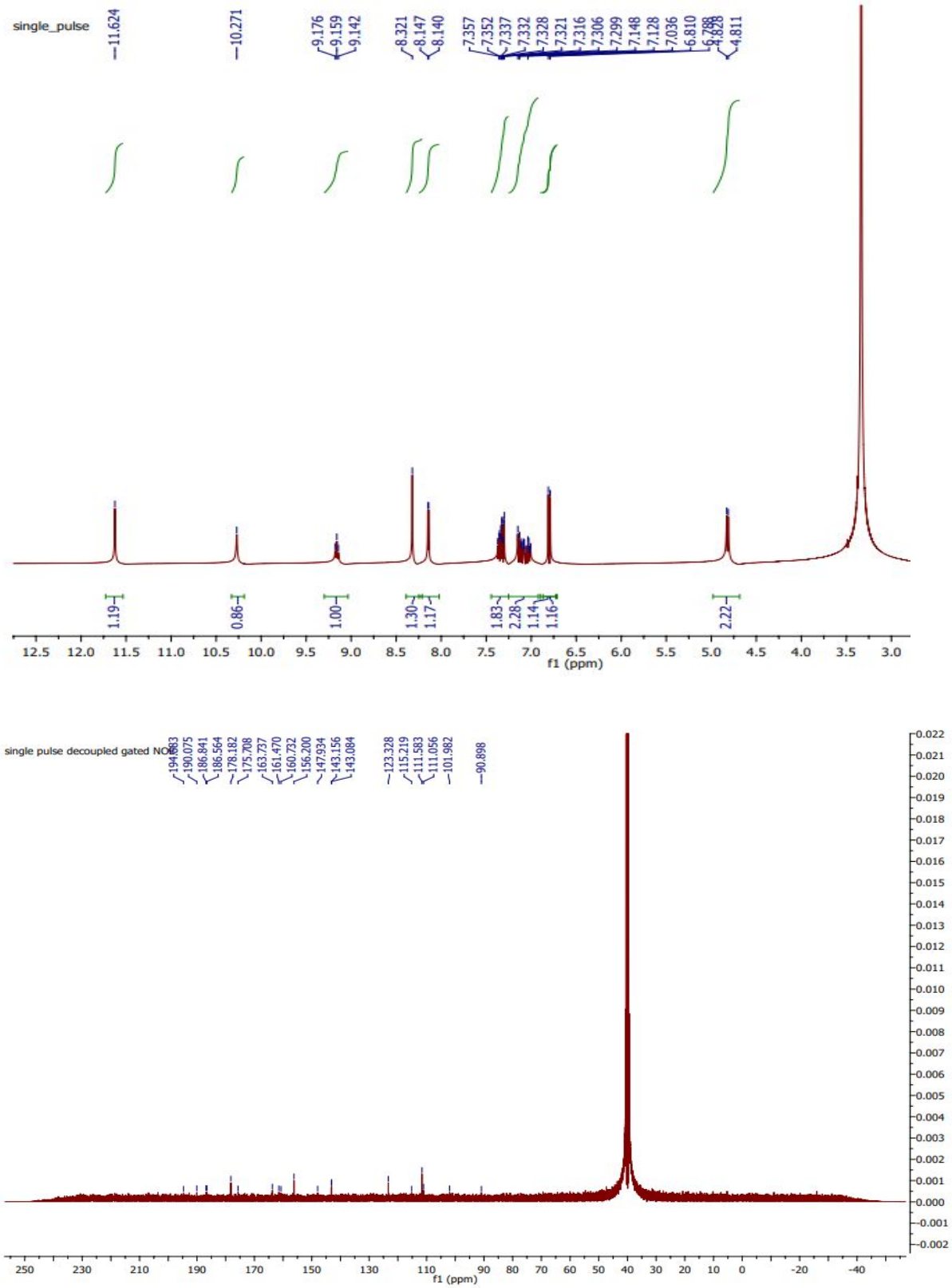


Figure S12: ^{1}H and ^{13}C NMR data of the compound (**3**).

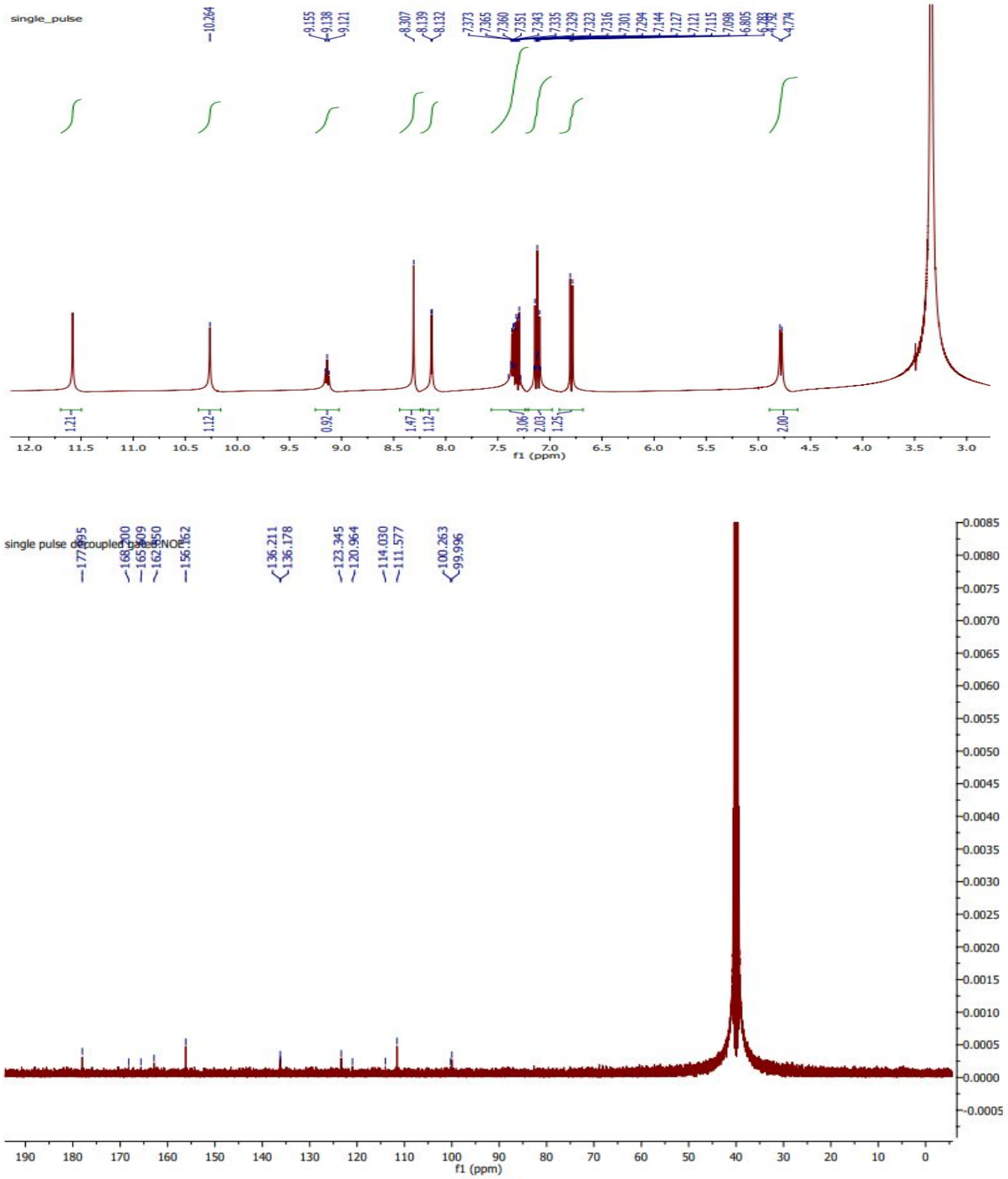


Figure S13: ^{1}H and ^{13}C NMR data of the compound (**4**).

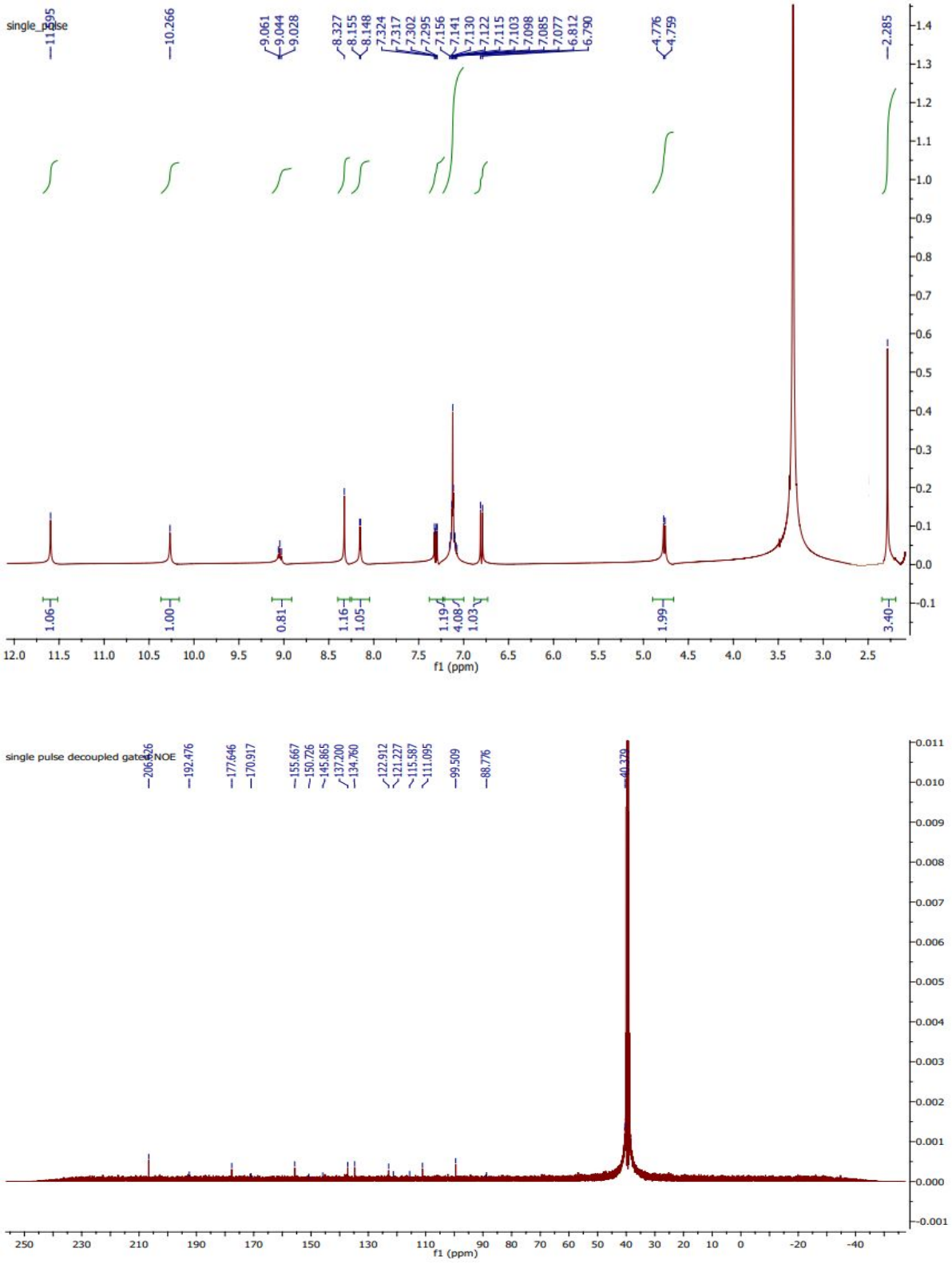


Figure S14: ^{1}H and ^{13}C NMR data of the compound (**5**).

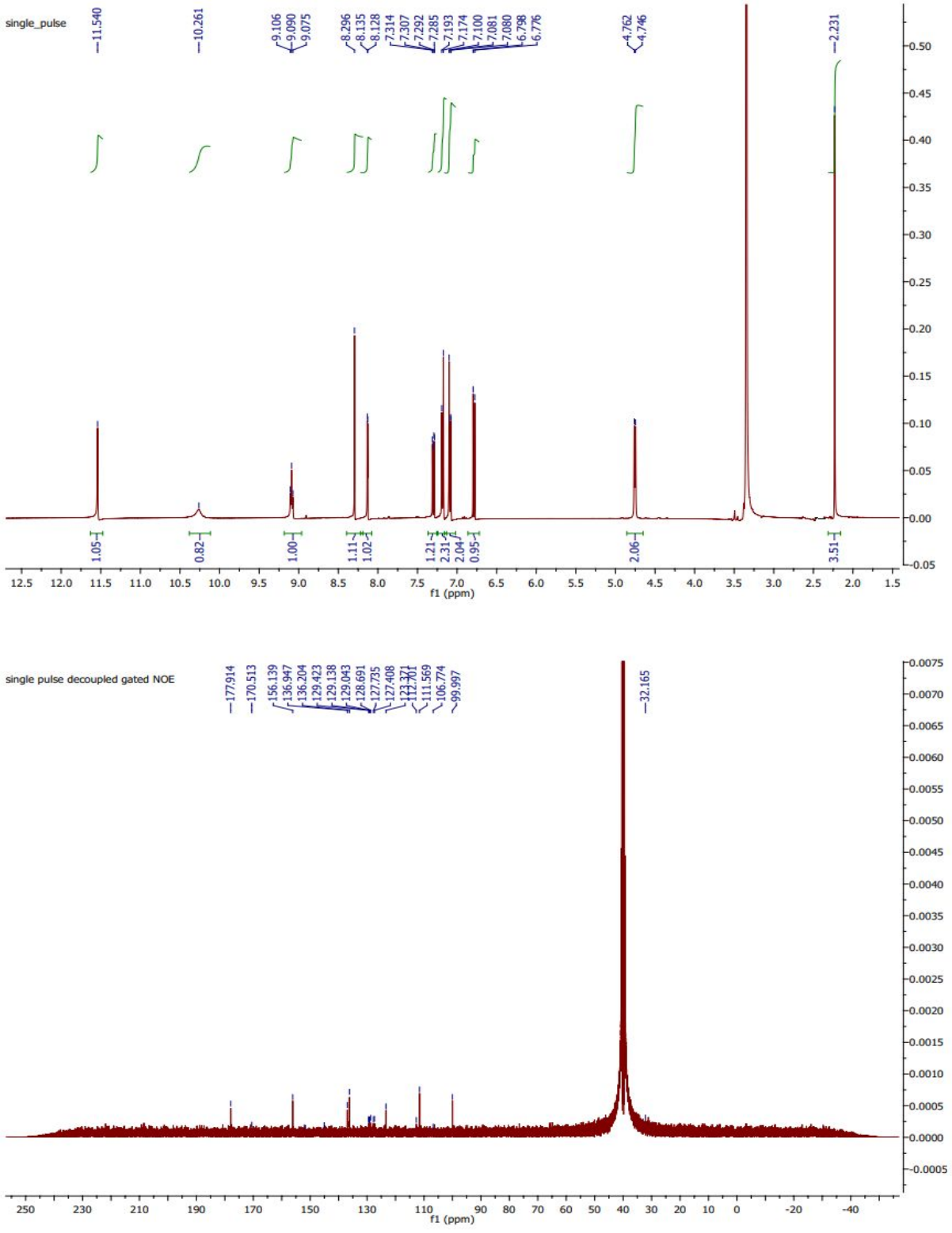


Figure S15: ^{1}H and ^{13}C NMR data of the compound (**6**).

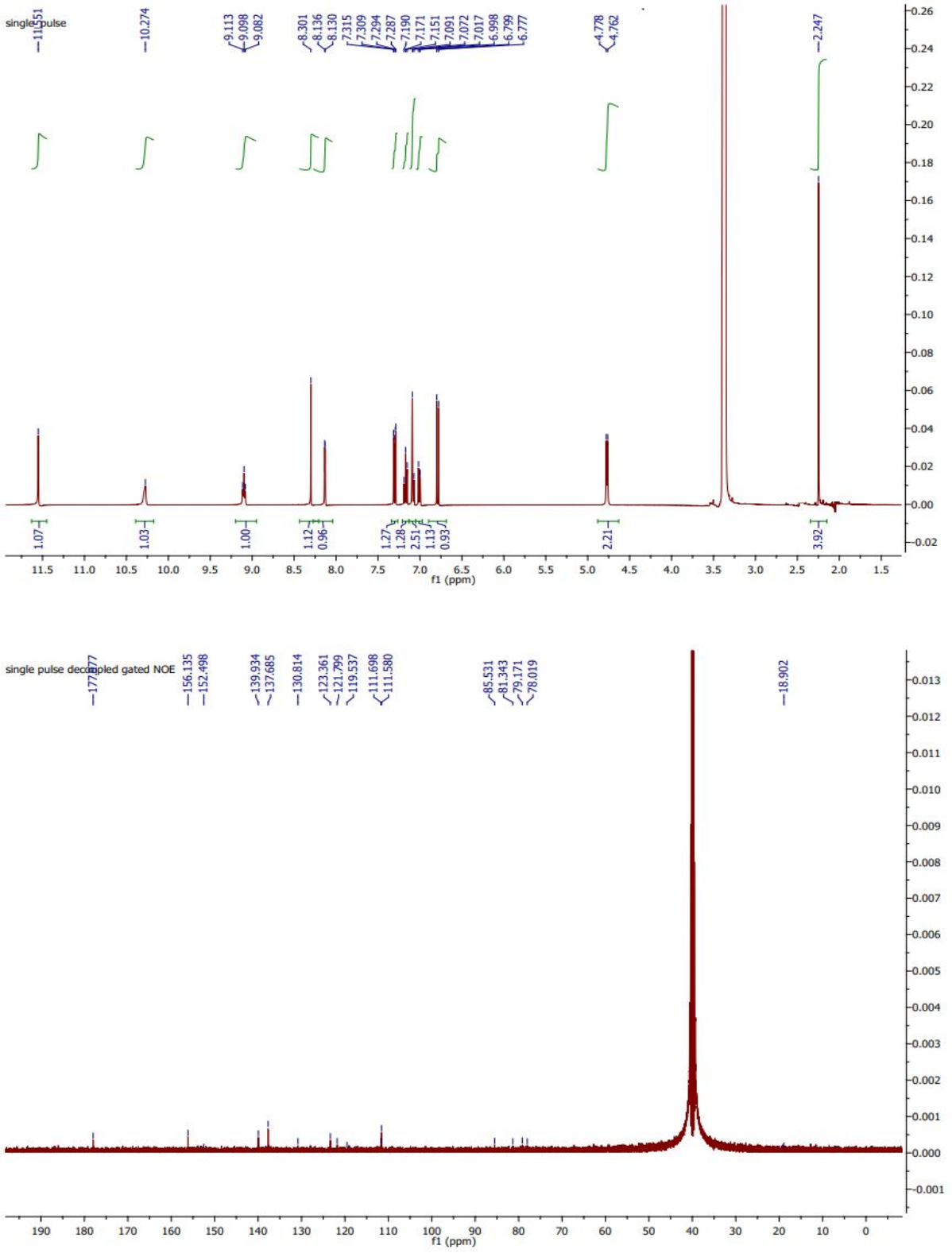


Figure S16: ^{1}H and ^{13}C NMR data of the compound (**7**).