

Synthesis, X-Ray powder diffraction and DFT calculations of vasorelaxant active 3-(arylmethylidene)pyrrolidine-2,5-diones

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Table S1. Selected intramolecular experimental (X-ray) and computationally optimized geometrical parameters (bond lengths, Å) of **12a** and **12b** by AM1, PM3 and DFT methods.

Geometric parameters	Compound 3a				Compound 3b			
	Exp. X-ray data	AM1	PM3	DFT	Exp. X-ray data	AM1	PM3	DFT
N(3)–C(4)	1.434	1.418	1.418	1.431	1.434	1.416	1.416	1.428
C(4)–C(9)	1.371	1.414	1.414	1.399	1.371	1.415	1.415	1.400
C(8)–C(9)	1.388	1.392	1.392	1.394	1.387	1.391	1.391	1.393
C(14)–C(15)	1.468	1.450	1.450	1.459	1.468	1.450	1.450	1.460
C(17)–C(18)	1.372	1.400	1.400	1.395	1.373	1.396	1.396	1.396
O(1)–C(2)	1.208	1.236	1.236	1.216	1.207	1.236	1.236	1.216
N(3)–C(10)	1.396	1.422	1.422	1.405	1.396	1.423	1.423	1.406
C(5)–C(6)	1.378	1.392	1.392	1.394	1.378	1.391	1.391	1.393
C(10)–C(12)	1.500	1.531	1.531	1.527	1.500	1.531	1.531	1.525
C(15)–C(16)	1.387	1.409	1.409	1.409	1.387	1.409	1.409	1.410
C(18)–C(19)	1.382	1.397	1.397	1.396	1.382	1.393	1.393	1.397
O(11)–C(10)	1.204	1.233	1.233	1.211	1.204	1.233	1.233	1.212
C(2)–C(13)	1.487	1.504	1.504	1.488	1.487	1.502	1.502	1.485
C(6)–C(7)	1.360	1.392	1.392	1.395	1.360	1.396	1.396	1.393
C(12)–C(13)	1.502	1.487	1.487	1.500	1.502	1.488	1.488	1.501

C(15)–C(20)	1.400	1.401	1.401	1.410	1.400	1.401	1.401	1.410
C(19)–C(20)	1.393	1.394	1.394	1.391	1.393	1.395	1.395	1.392
N(3)–C(2)	1.403	1.426	1.426	1.417	1.404	1.427	1.427	1.420
C(4)–C(5)	1.368	1.415	1.415	1.399	1.368	1.415	1.415	1.400
C(7)–C(8)	1.376	1.392	1.392	1.395	1.376	1.396	1.396	1.393
C(13)–C(14)	1.331	1.340	1.340	1.348	1.331	1.341	1.341	1.349
C(16)–C(17)	1.375	1.390	1.390	1.390	1.374	1.391	1.391	1.391
Cl(21)–C(18)	1.758	1.697	1.697	1.754	---	---	---	---
Cl(21)–C(7)	---	---	---	---	1.758	1.699	1.699	1.758
RMSE	---	0.0236	0.0236	0.0174	---	0.0264	0.0264	0.0171
Maximum difference	---	0.047	0.047	0.035	---	0.059	0.059	0.033

Table S2. Selected intramolecular experimental (X-ray) and computationally optimized geometrical parameters (bond angles, °) of **12a** and **12b** by AM1, PM3 and DFT methods.

Geometric parameters	Compound 3a				Compound 3b			
	Exp. X-ray data	AM1	PM3	DFT	Exp. X-ray data	AM1	PM3	DFT
C(2)–N(3)–C(4)	124.4	125.3	125.3	123.8	124.4	125.3	125.3	123.8
O(1)–C(2)–N(3)	124.8	126.4	126.4	125.0	124.8	126.2	126.2	124.8
N(3)–C(4)–C(5)	119.3	121.0	121.0	119.9	119.2	121.2	121.2	120.0
C(4)–C(5)–C(6)	119.6	120.6	120.6	119.5	119.6	120.9	120.9	120.0
C(7)–C(8)–C(9)	120.1	121.0	121.0	120.5	120.2	120.4	120.4	119.5
O(11)–C(10)–C(12)	127.6	123.5	123.5	126.1	127.6	123.7	123.7	126.2
C(2)–C(13)–C(12)	107.4	107.4	107.4	108.0	107.38	107.4	107.4	108.0
C(13)–C(14)–C(15)	130.4	129.6	129.6	130.8	130.4	129.7	129.7	130.9
C(16)–C(15)–C(20)	117.6	118.5	118.5	117.6	117.6	118.5	118.5	117.9
C(18)–C(19)–C(20)	120.4	119.8	119.8	119.5	120.5	120.3	120.3	120.5
C(2)–N(3)–C(10)	112.3	109.3	109.3	112.2	112.3	109.3	109.3	112.1
O(1)–C(2)–C(13)	128.0	124.5	124.5	127.7	128.1	124.8	124.8	128.0
N(3)–C(4)–C(9)	119.8	121.2	121.2	119.8	119.8	121.0	121.0	120.1
C(5)–C(6)–C(7)	120.4	121.0	121.0	120.5	120.4	120.4	120.4	119.5
C(4)–C(9)–C(8)	119.0	120.6	120.6	119.5	118.9	120.9	120.9	120.0
N(3)–C(10)–C(12)	108.5	109.6	109.6	108.0	108.5	109.6	109.6	108.0

C(2)–C(13)–C(14)	119.9	120.7	120.7	119.1	119.9	120.7	120.7	119.2
C(14)–C(15)–C(16)	118.2	117.5	117.5	117.7	118.2	117.4	117.4	117.5
C(15)–C(16)–C(17)	122.0	121.0	121.0	121.8	122	120.7	120.7	121.3
C(15)–C(20)–C(19)	120.2	120.9	120.9	121.2	120.3	120.6	120.6	120.7
C(4)–N(3)–C(10)	123.3	125.4	125.4	123.9	123.3	125.4	125.4	124.0
N(3)–C(2)–C(13)	107.2	109.1	109.1	107.3	107.2	109.1	109.1	107.3
C(5)–C(4)–C(9)	120.9	117.9	117.9	120.3	120.9	117.8	117.8	119.9
C(6)–C(7)–C(8)	120.0	119.0	119.0	119.6	120	119.6	119.6	120.9
O(11)–C(10)–N(3)	123.9	126.9	126.9	125.9	123.9	126.7	126.7	125.8
C(10)–C(12)–C(13)	104.6	104.6	104.6	104.5	104.6	104.6	104.6	104.5
C(12)–C(13)–C(14)	132.7	132.0	132.0	132.9	132.74	131.9	131.9	132.8
C(14)–C(15)–C(20)	124.2	124.0	124.0	124.6	124.2	124.0	124.0	124.5
C(16)–C(17)–C(18)	120.1	119.6	119.6	119.0	120.1	120.1	120.1	119.9
C(17)–C(18)–C(19)	119.6	120.2	120.2	120.9	119.5	119.7	119.7	119.7
Cl(21)–C(18)–C(17)	120.2	119.8	119.8	119.6	---	---	---	---
Cl(21)–C(18)–C(19)	120.2	119.9	119.9	119.5	---	---	---	---
Cl(21)–C(7)–C(8)	---	---	---	---	120.2	120.2	120.2	119.5
Cl(21)–C(7)–C(6)	---	---	---	---	119.8	120.2	120.2	119.5
RMSE	---	1.632	1.632	0.705	---	1.593	1.593	0.686
Maximum difference	---	4.1	4.1	2.0	---	3.9	3.9	1.9

Table S3. Calculated vibrational spectra (IR) of compound **12a**.

Scaled freq. (cm ⁻¹)	Intensity	Scaled freq. (cm ⁻¹)	Intensity	Scaled freq. (cm ⁻¹)	Intensity
19	0.001	688	1.011	1243	2.134
31	0.028	711	0.112	1284	0.831
51	0.010	730	3.889	1291	4.515
53	0.110	749	3.666	1298	0.346
67	0.261	798	1.541	1316	0.488
102	0.314	804	5.966	1336	100.000
121	1.100	807	5.571	1363	0.059
152	0.133	809	0.015	1399	2.330
176	0.006	882	0.675	1420	1.159
186	0.251	887	4.354	1444	0.797
208	0.061	919	0.073	1478	14.452
268	0.132	920	0.551	1486	11.873
289	1.294	925	0.071	1548	0.562
308	0.020	936	0.617	1581	2.062
322	0.105	951	0.386	1582	11.183
376	0.031	952	0.556	1593	2.698
398	0.003	953	0.680	1641	23.580
401	0.025	980	0.199	1725	67.240
407	7.034	987	5.550	1779	23.752
427	0.022	1018	0.237	2930	0.665
479	0.210	1064	7.142	2962	0.405
498	0.650	1070	13.999	3039	0.101
512	2.941	1093	2.838	3059	0.002
568	1.414	1105	3.128	3067	0.894
572	0.329	1136	0.338	3070	2.887
602	3.848	1146	10.957	3081	6.004
607	0.270	1148	35.542	3092	0.419
619	1.092	1156	7.753	3097	0.736
644	2.994	1171	1.005	3115	0.336
672	2.338	1172	0.444	3115	0.174
681	4.189	1203	2.545	3116	0.584

Table S4. Calculated vibrational spectra (IR) of compound **12b**.

Scaled freq. (cm ⁻¹)	Intensity	Scaled freq. (cm ⁻¹)	Intensity	Scaled freq. (cm ⁻¹)	Intensity
19	0.009	704	2.454	1245	2.107
29	0.032	705	0.781	1284	0.150
45	0.024	737	2.310	1292	1.397
53	0.107	751	5.328	1295	4.339
83	0.005	792	0.046	1319	0.122
99	0.213	801	2.503	1336	100.000
124	1.006	804	6.455	1369	0.308
136	0.340	817	0.001	1396	0.656
170	0.071	886	8.581	1420	0.789
194	0.223	888	0.295	1437	2.470
245	0.173	914	0.307	1480	34.938
253	0.142	918	0.068	1484	0.427
269	0.134	926	0.294	1566	0.832
281	0.475	937	0.707	1566	0.074
337	0.054	950	0.542	1585	0.012
383	0.055	953	1.056	1592	2.430
396	0.010	964	0.158	1639	30.475
403	0.006	975	0.223	1724	64.989
416	2.951	993	4.874	1777	21.160
416	5.116	1017	0.208	2930	0.656
498	1.075	1068	13.311	2963	0.380
509	1.294	1072	2.269	3038	0.108
511	2.096	1083	6.298	3057	0.540
557	3.492	1108	2.311	3064	0.049
571	0.498	1136	0.428	3073	3.528
578	4.455	1145	44.228	3084	5.143
607	0.301	1148	1.672	3094	0.651
619	0.016	1158	7.593	3094	0.372
632	1.774	1171	2.085	3112	0.977
675	4.600	1173	0.177	3123	0.060
679	0.122	1203	2.045	3124	0.060

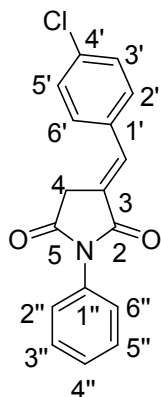


Table S5. Theoretical ^1H - and ^{13}C -NMR chemical shifts (ppm) of compound **12a**.

Atom	Calculated (ppm) DFT/B3LYP and 6-31G(d) level of theory
$\text{H}_2\text{C-4}$	3.28, 3.33 (mean = 3.31)
H-3'	7.29
H-5'	7.30
H-4''	7.33
H-2'	7.36
H-3''	7.43
H-5''	7.44
H-6'	7.44
Olefinic CH	7.46
H-2''	7.68
H-6''	7.70
$\text{H}_2\text{C-4}$	34.1
C-6''	126.1
C-2''	126.2
C-3	126.5
C-4''	127.3
C-3''	128.5
C-5''	128.6
C-6'	129.6
C-5'	130.3
C-3'	130.5
C-1'	134.3
C-1''	135.2
Olefinic CH	136.8
C-2'	137.4
C-4'	147.8
CO-2	171.4
CO-5	173.1

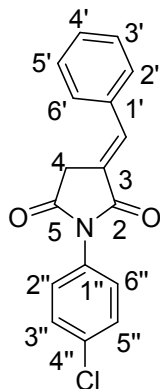


Table S6. Theoretical ^1H - and ^{13}C -NMR chemical shifts (ppm) of compound **12b**.

Atom	Calculated (ppm) DFT/B3LYP and 6-31G(d) level of theory
H_2C -4	3.34, 3.39 (mean = 3.37)
HC-3''	7.24
HC-5''	7.25
HC-4'	7.45
HC-2'	7.47
HC-3'	7.50
HC-5'	7.50
HC-6'	7.54
Olefinic-CH	7.57
HC-2''	7.72
HC-6''	7.74
H_2C -4	34.4
C-3	124.8
C-2''	126.9
C-6''	126.9
C-6'	129.0
C-3''	129.3
C-5'	129.4
C-5''	129.4
C-3'	129.7
C-4'	131.2
C-1''	133.9
C-1'	135.6
C-2'	137.1
Olefinic-CH	139.4
C-4''	143.2
CO-2	171.5
CO-5	173.5

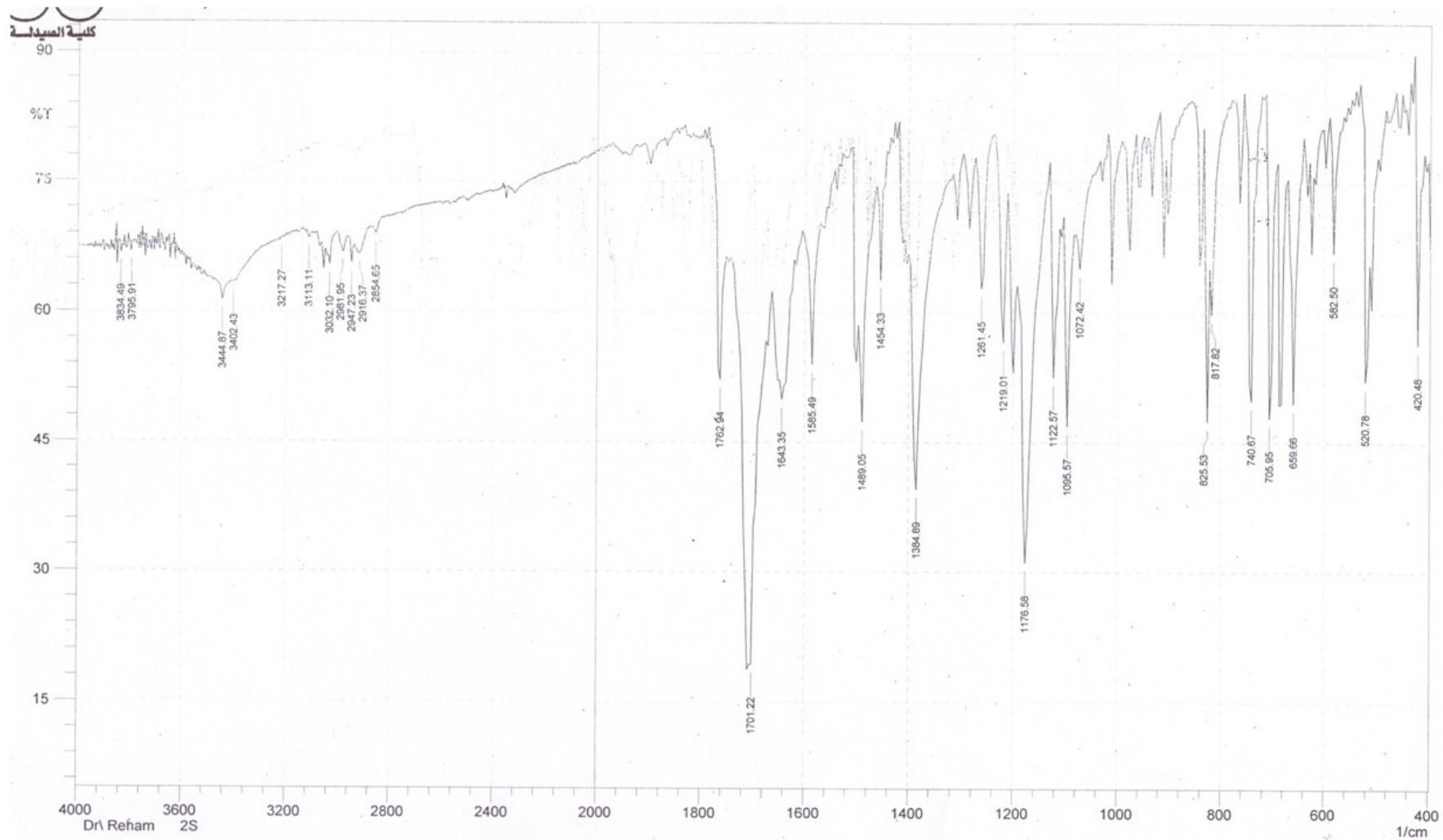


Figure S1. IR spectrum of compound 12a (KBr pellet).

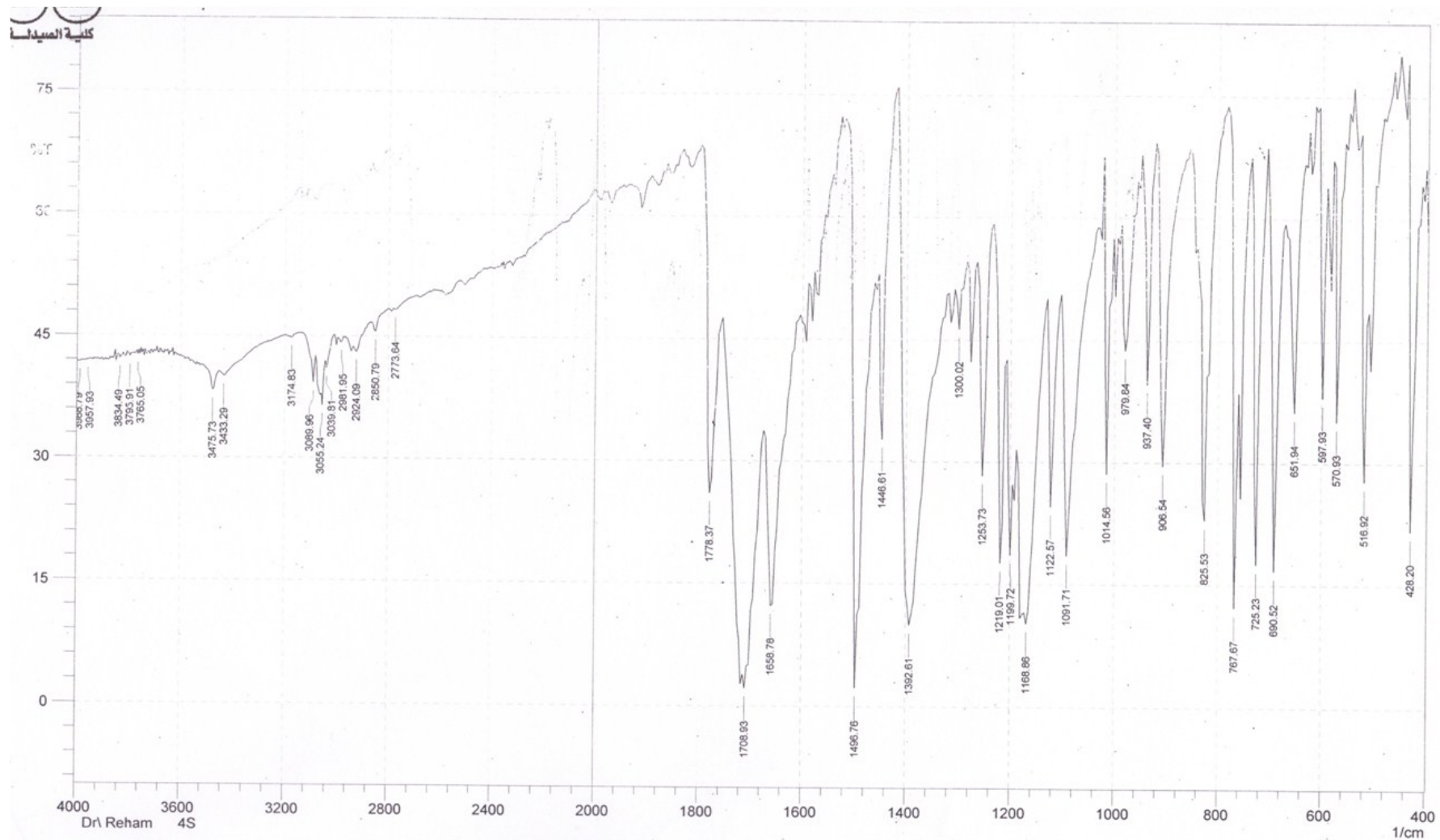


Figure S2. IR spectrum of compound **12b** (KBr pellet).

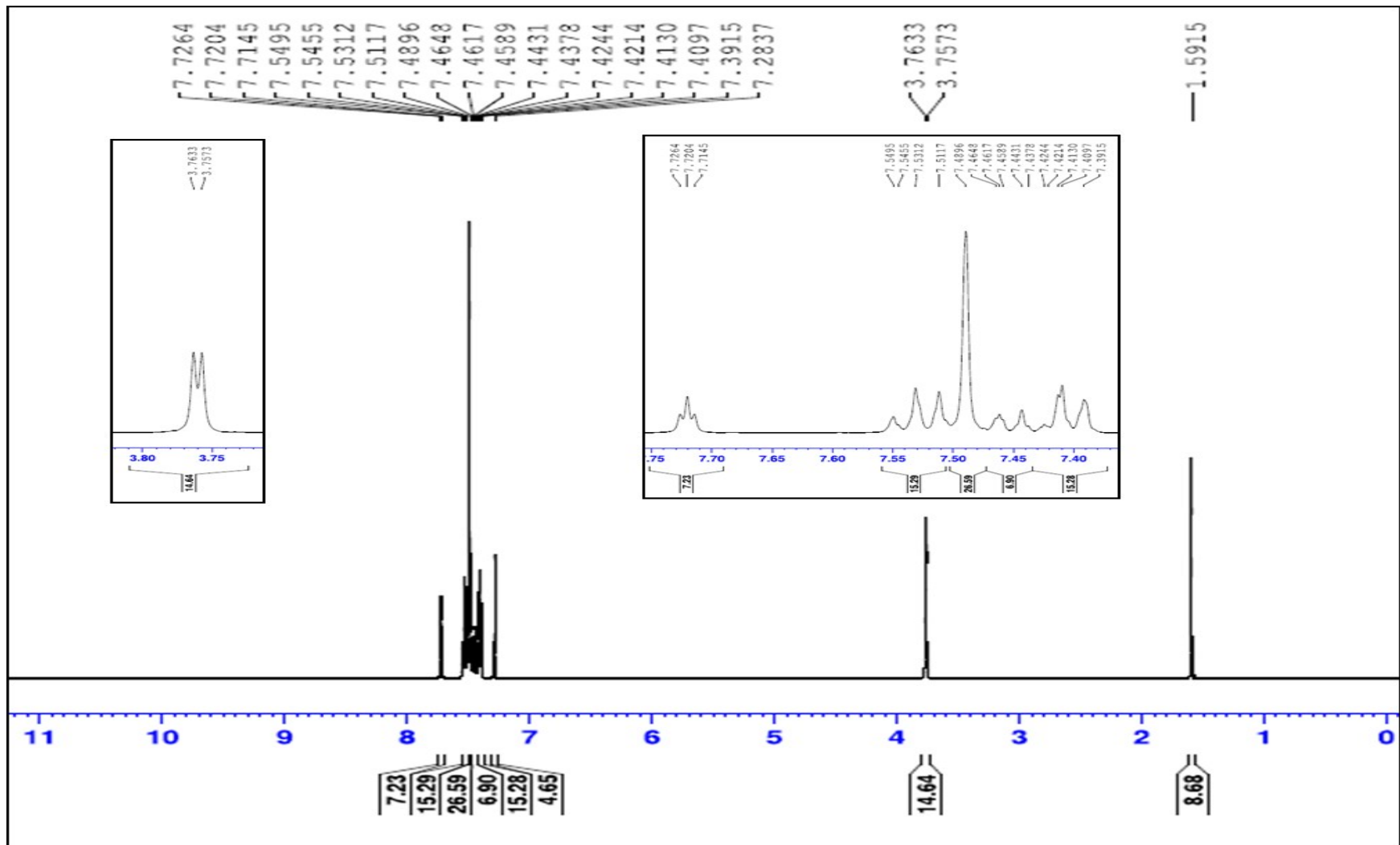


Figure S3. ¹H-NMR spectrum of compound **12a** in CDCl₃.

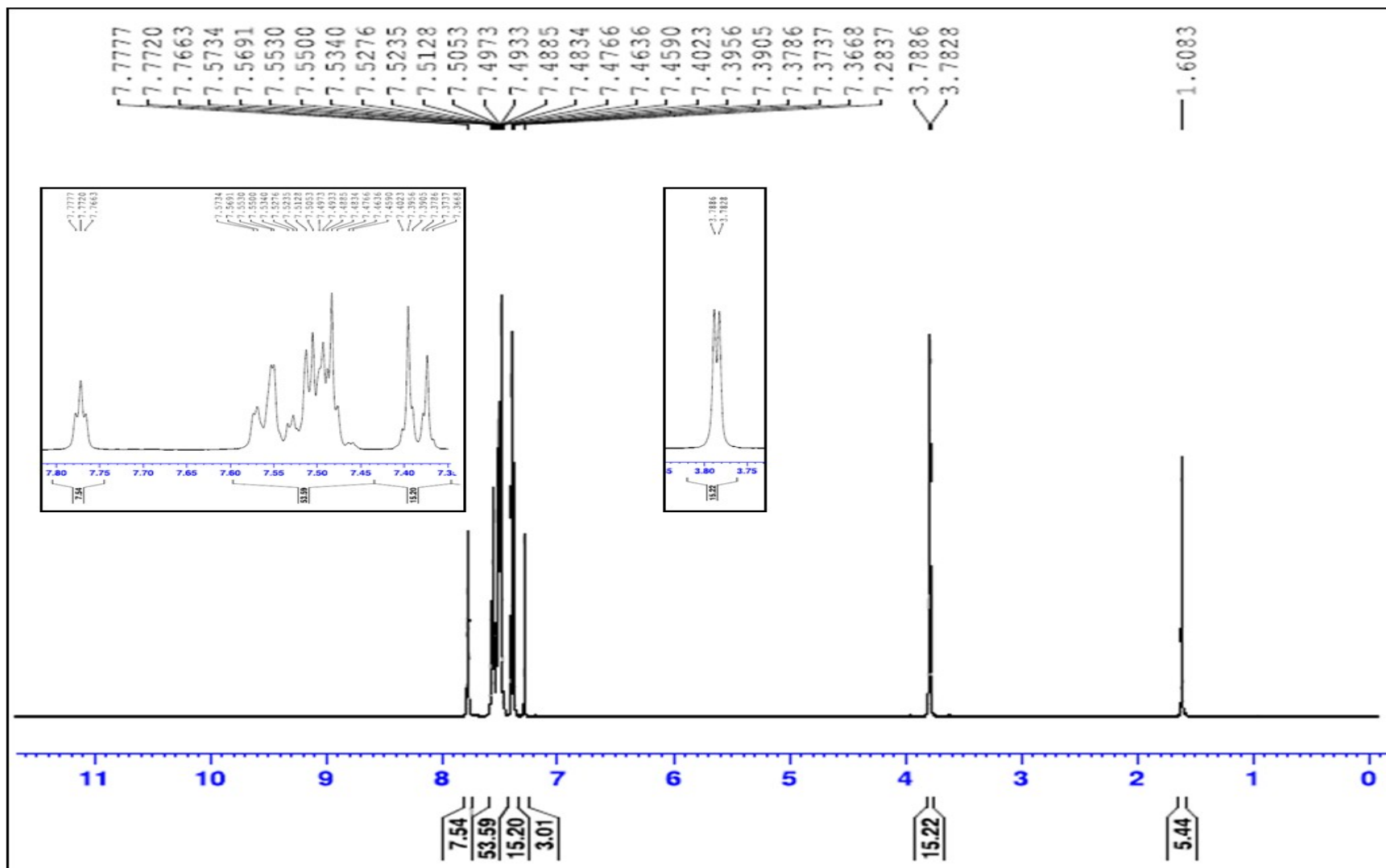


Figure S4. ¹H-NMR spectrum of compound **12b** in CDCl₃.

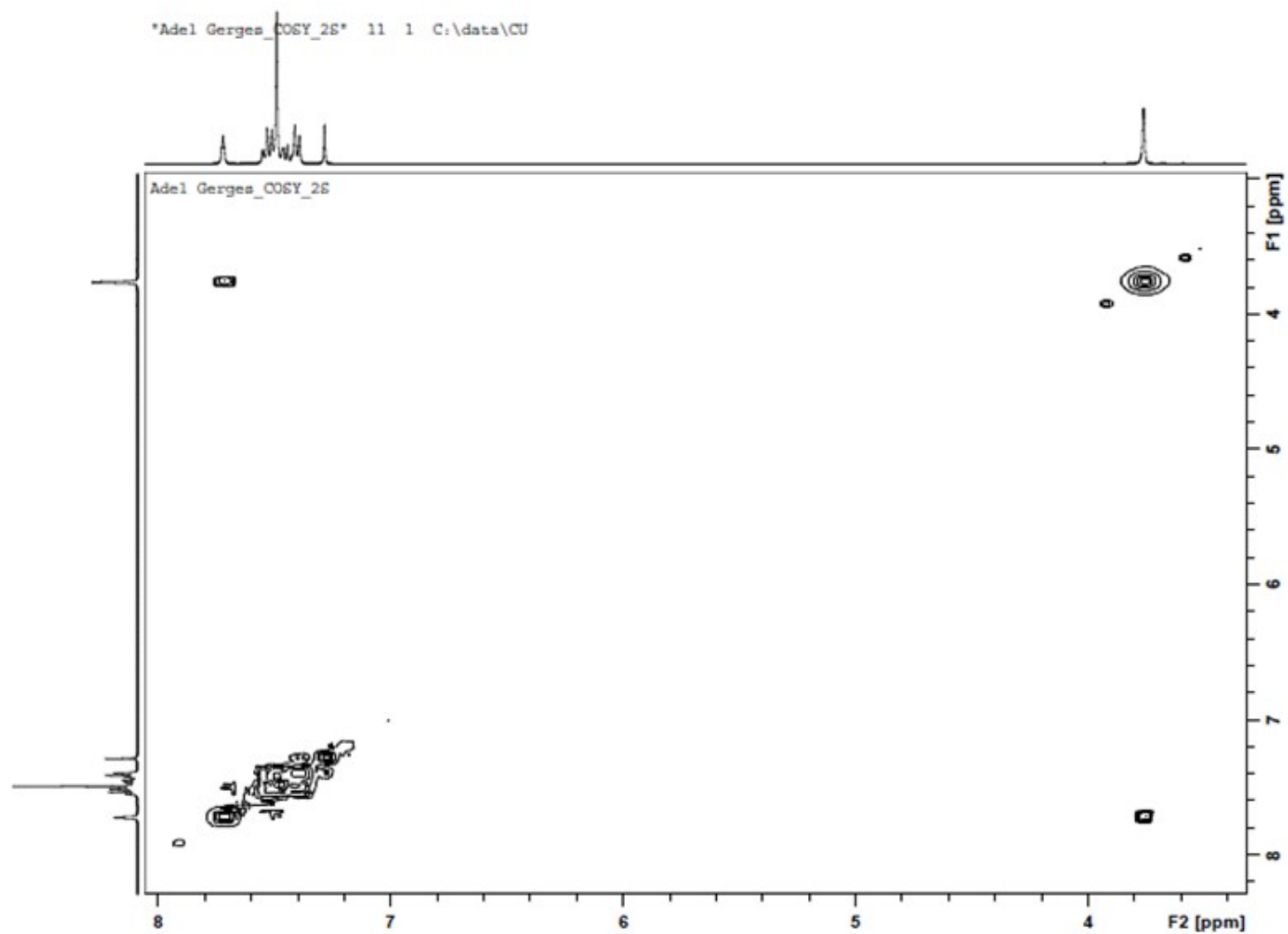


Figure S5. ^1H , ^1H -COSY spectrum of compound **12a** in CDCl_3 .

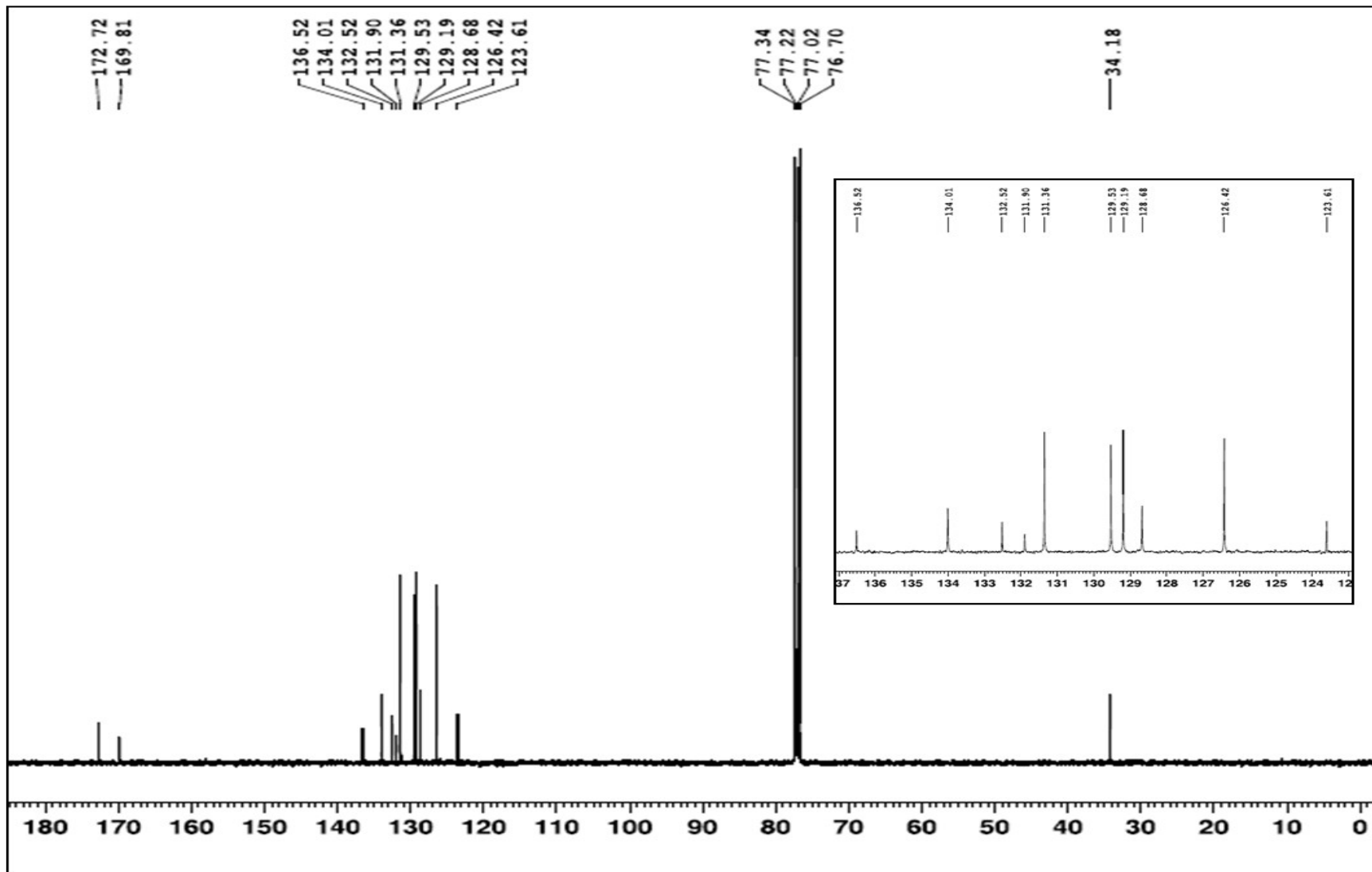


Figure S6. ^{13}C -NMR spectrum of compound **12a** in CDCl_3 .

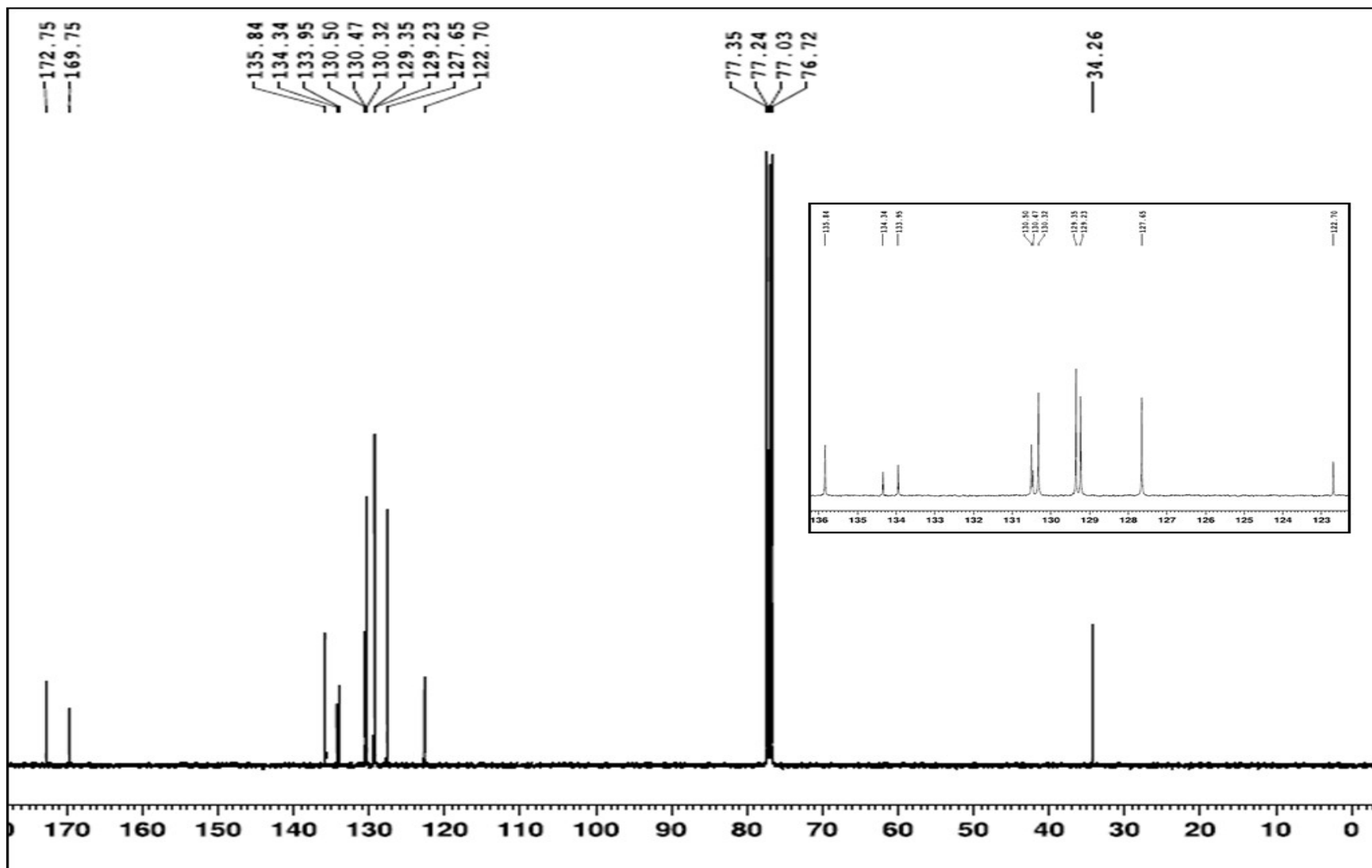


Figure S7. ^{13}C -NMR spectrum of compound **12b** in CDCl_3 .

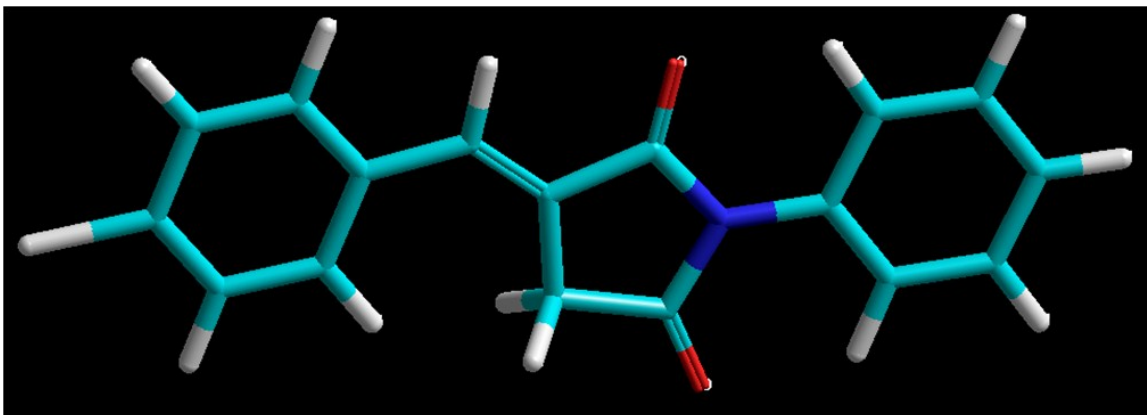


Figure S8. A projection of the optimized structure of compound **12a** by semi-empirical AM1.

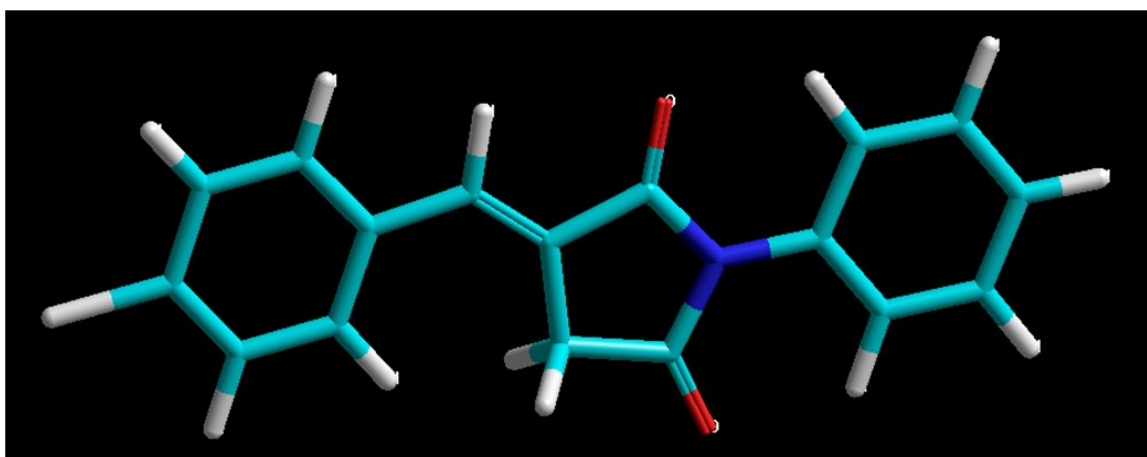


Figure S9. A projection of the optimized structure of compound **12a** by semi-empirical PM3.

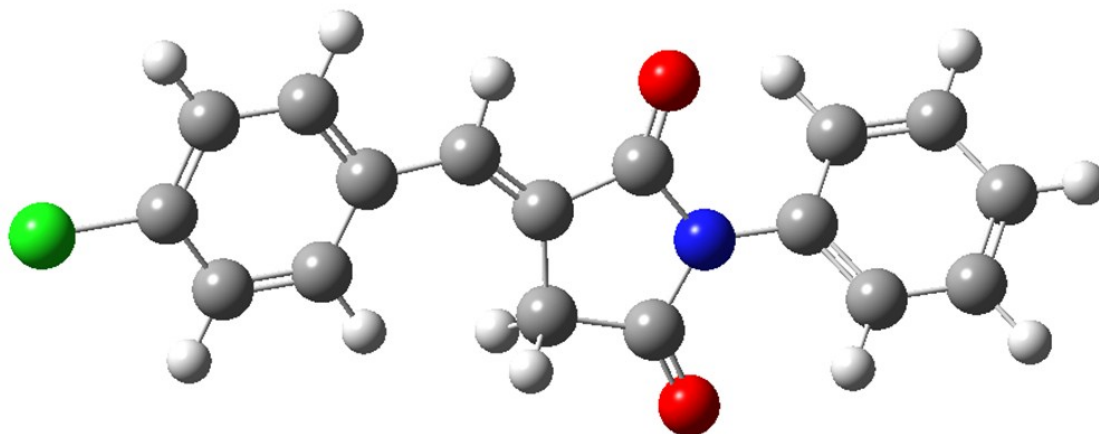


Figure S10. A projection of the optimized structure of compound **12a** by DFT/B3LYP method with 6-31G(d,p) basis set.

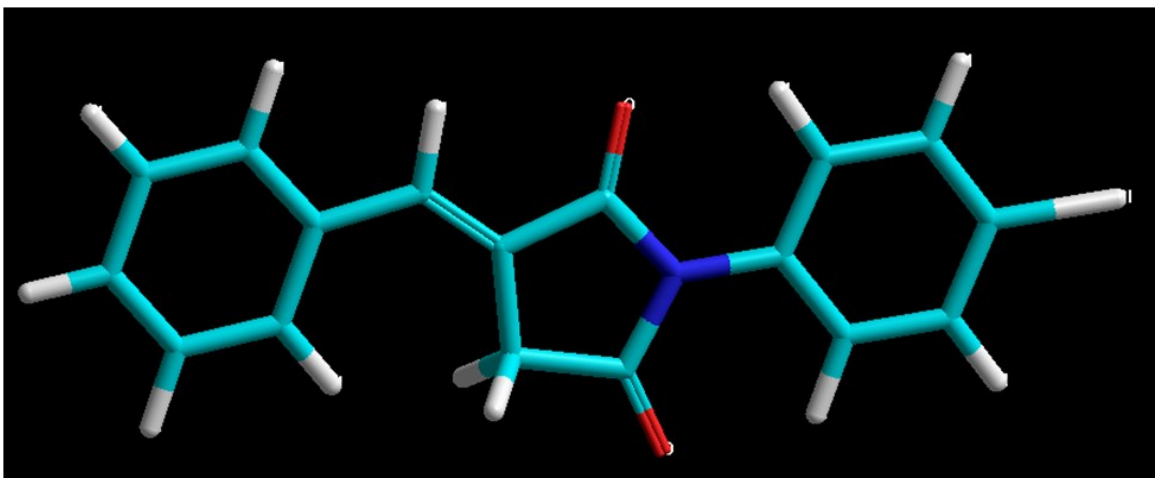


Figure S11. A projection of the optimized structure of compound **12b** by semi-empirical AM1.

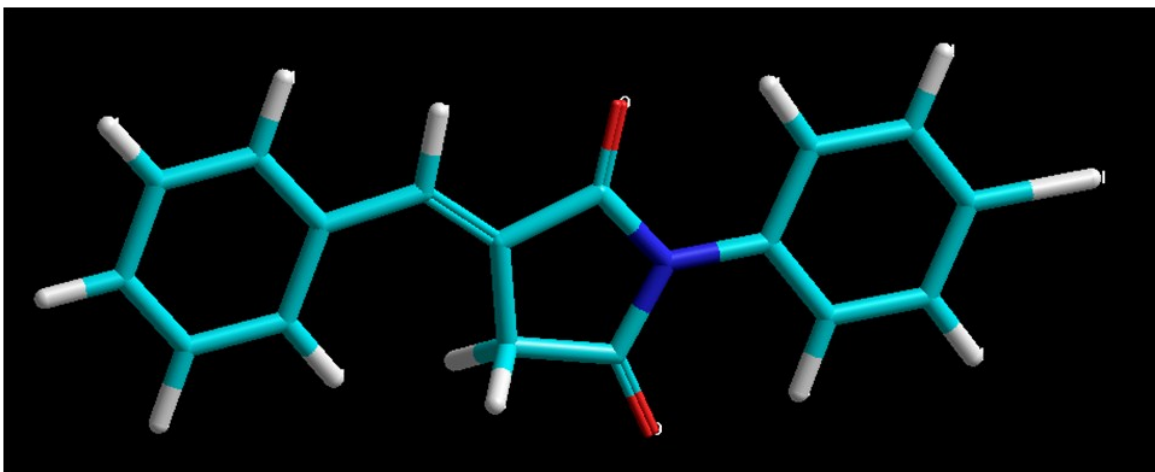


Figure S12. A projection of the optimized structure of compound **12b** by semi-empirical PM3.

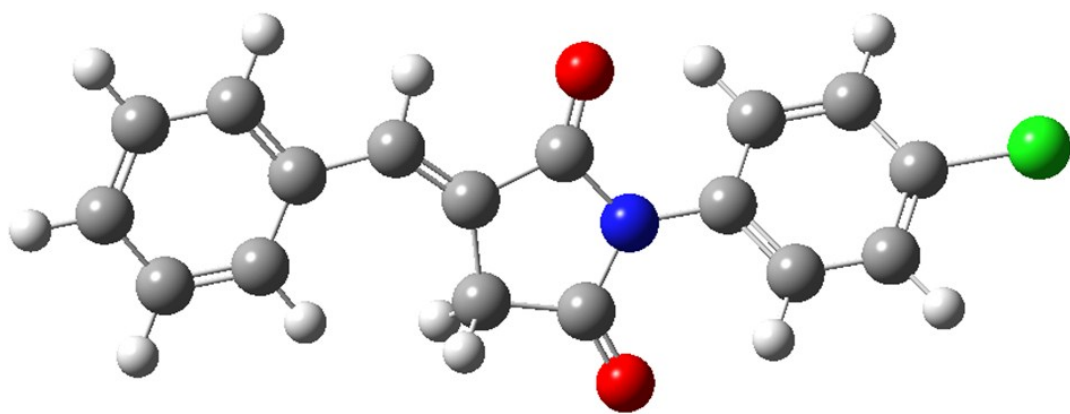


Figure S13. A projection of the optimized structure of compound **12b** by DFT/B3LYP method with 6-31G(d,p) basis set.

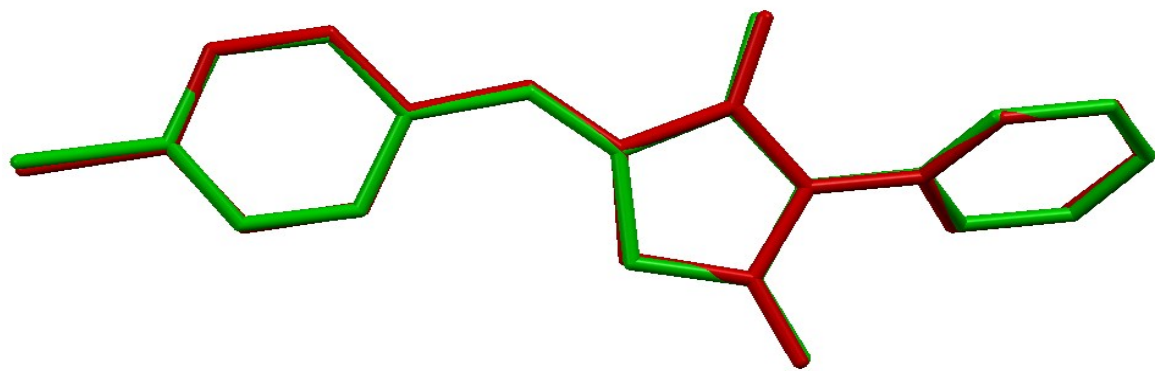


Figure S14. Molecule Overlay view of the structures of **12a**, red (X-ray structure), green (DFT).

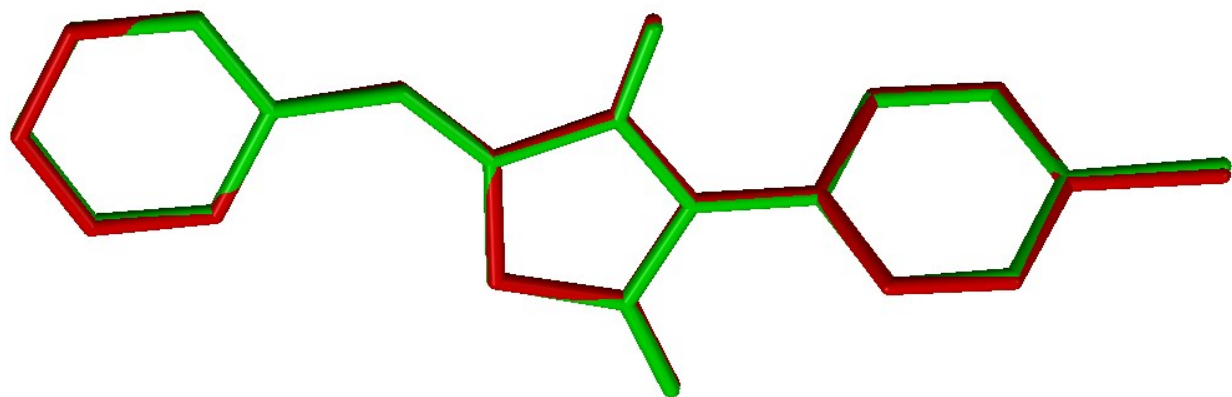


Figure S15. MoleculeOverlay view of the structures of **12b**, red (X-ray structure), green (DFT).

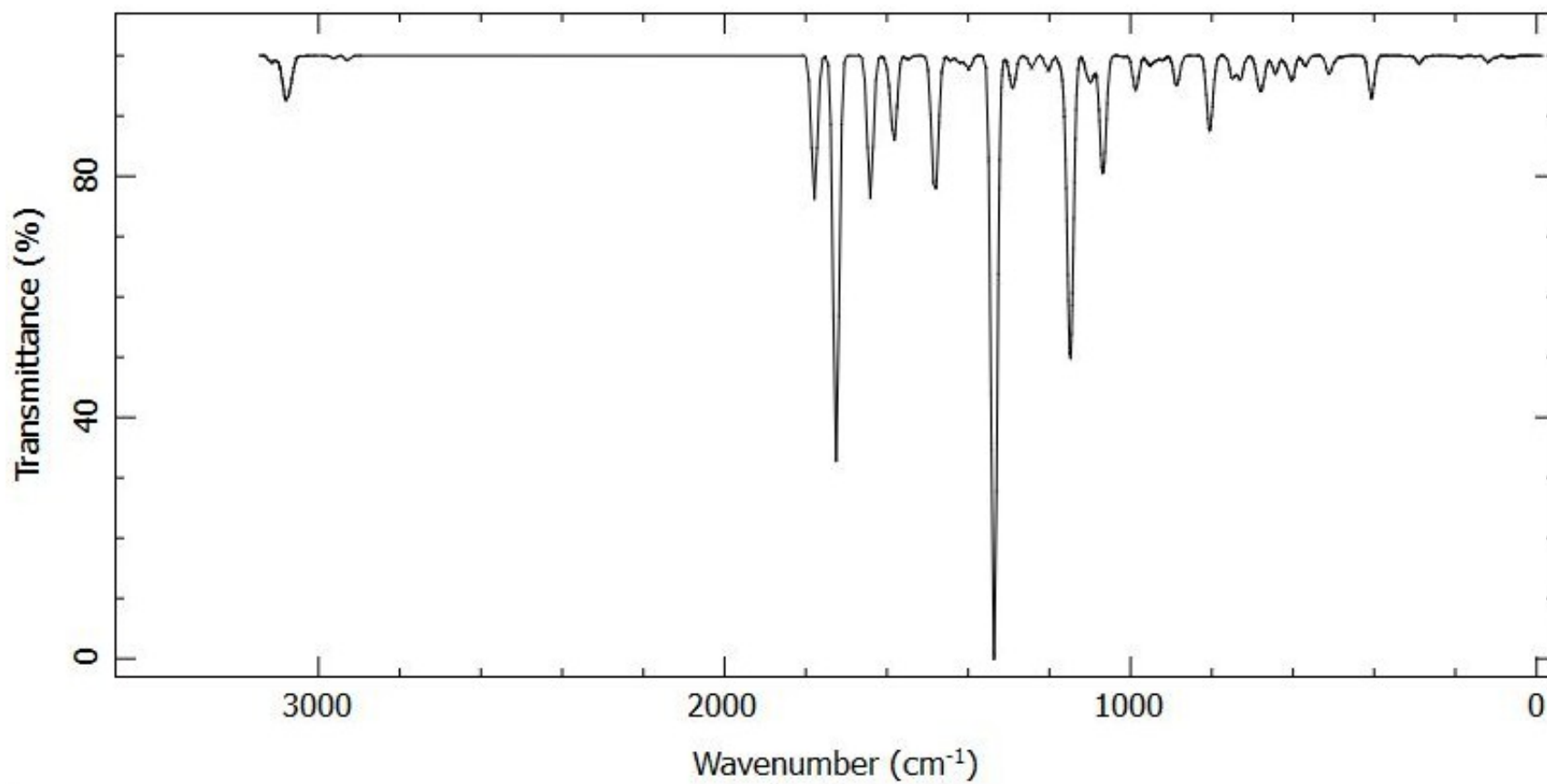


Figure S16. Simulated IR spectrum of compound **12a** computed by DFT/B3LYP method with 6-31G(d) basis set.

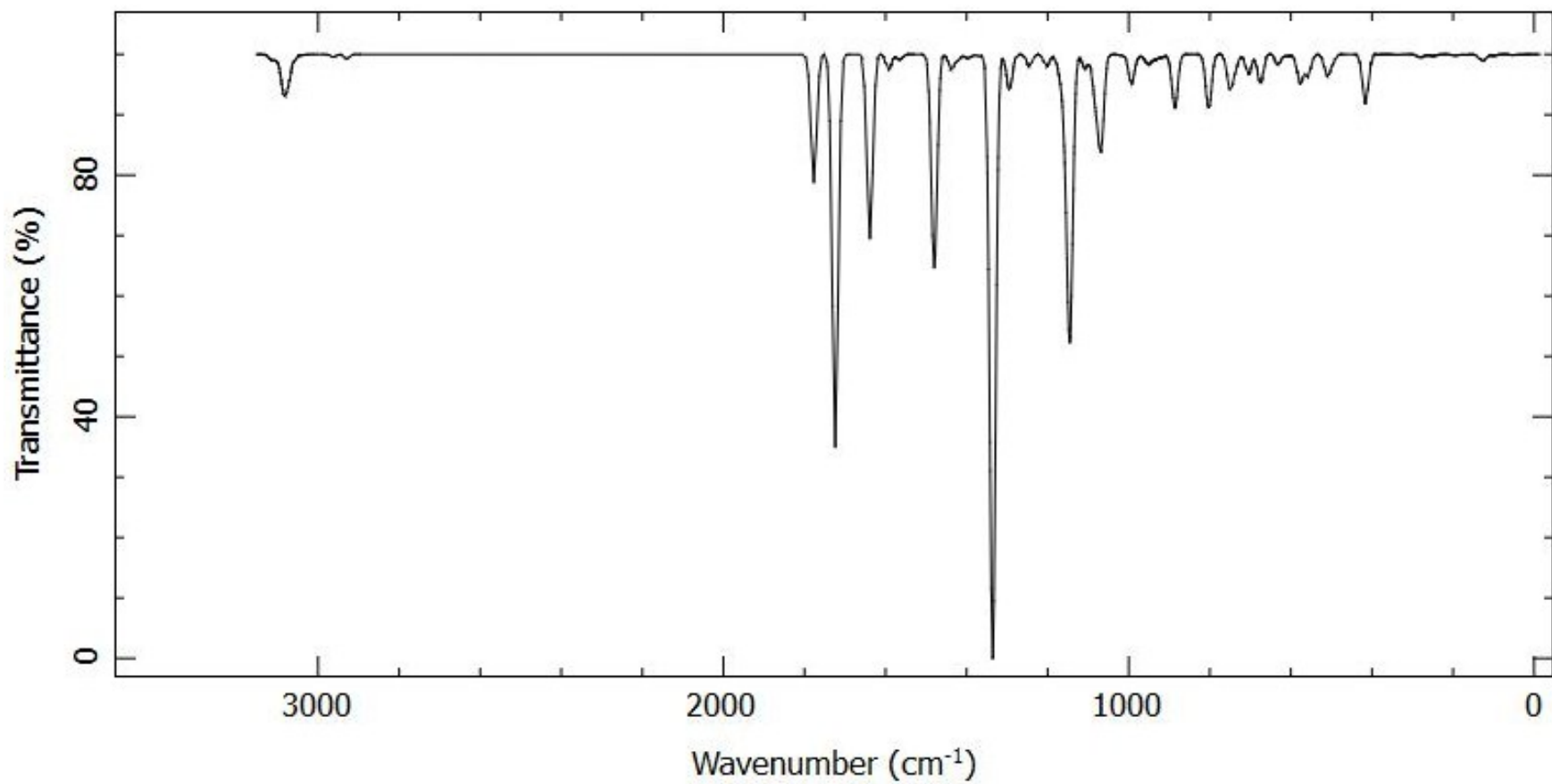


Figure S17. Simulated IR spectrum of compound **12b** computed by DFT/B3LYP method with 6-31G(d) basis set.