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Supporting Information

Synthesis and characterization of In(III) S-thiobezoylethioglycolate complexes and their catalytic applications in CO_2 fixation and multicomponent synthetic reactions. †

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The NMR spectra of following synthesised indium complexes

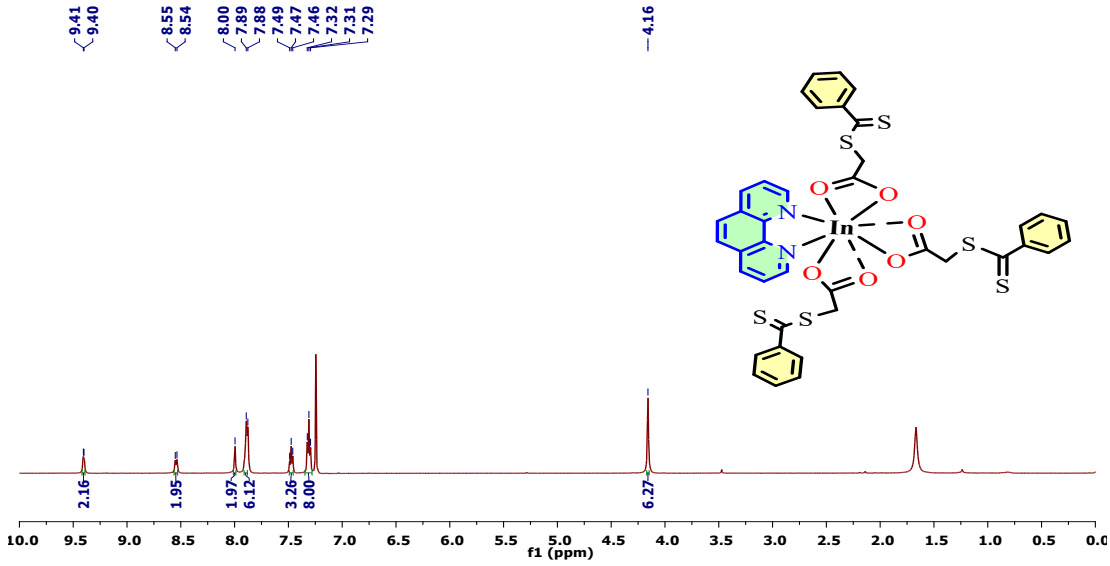


Figure S1 ^{1}H NMR spectra of complex 1 [In(1,10phen.)(Stbtg)$_3$]

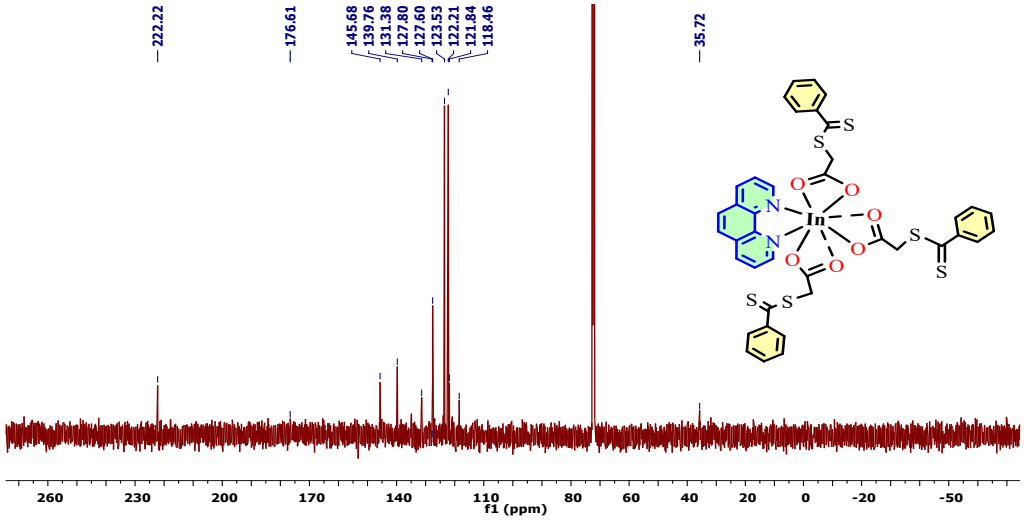


Figure S2 ^{13}C NMR spectra of complex 1 [In(1,10phen.)(Stbtg)$_3$]

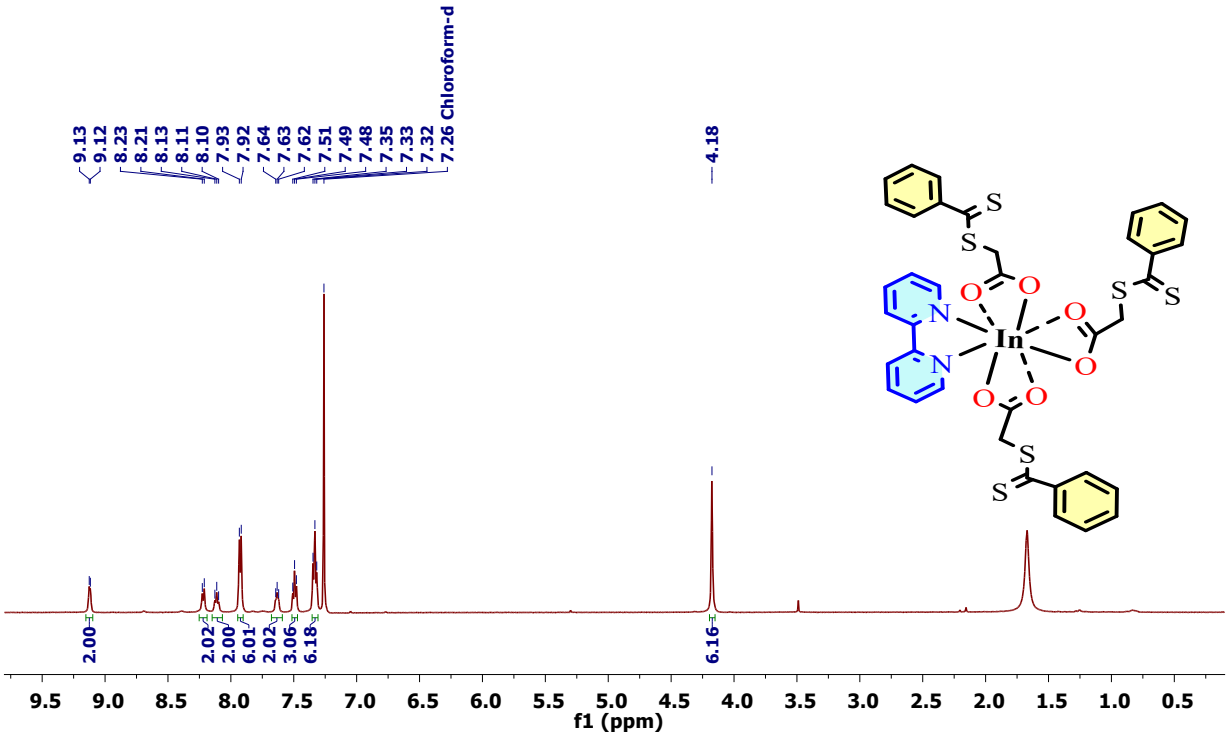


Figure-S3 ^{1}H NMR spectra of Complex 2, [In(2,2bipyridyl)(stbtg)$_3$]

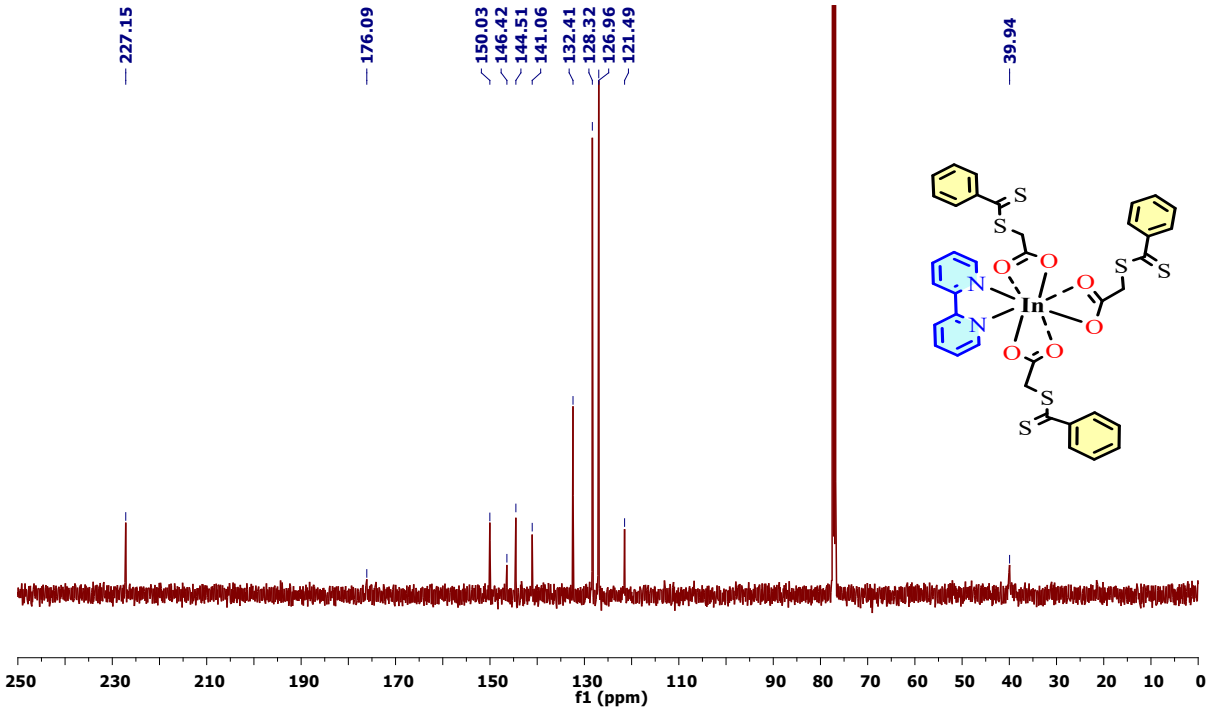


Figure S4.^{13}C NMR spectra of Complex 2, [In(2,2bipyridyl)(Stbtg)$_3$]

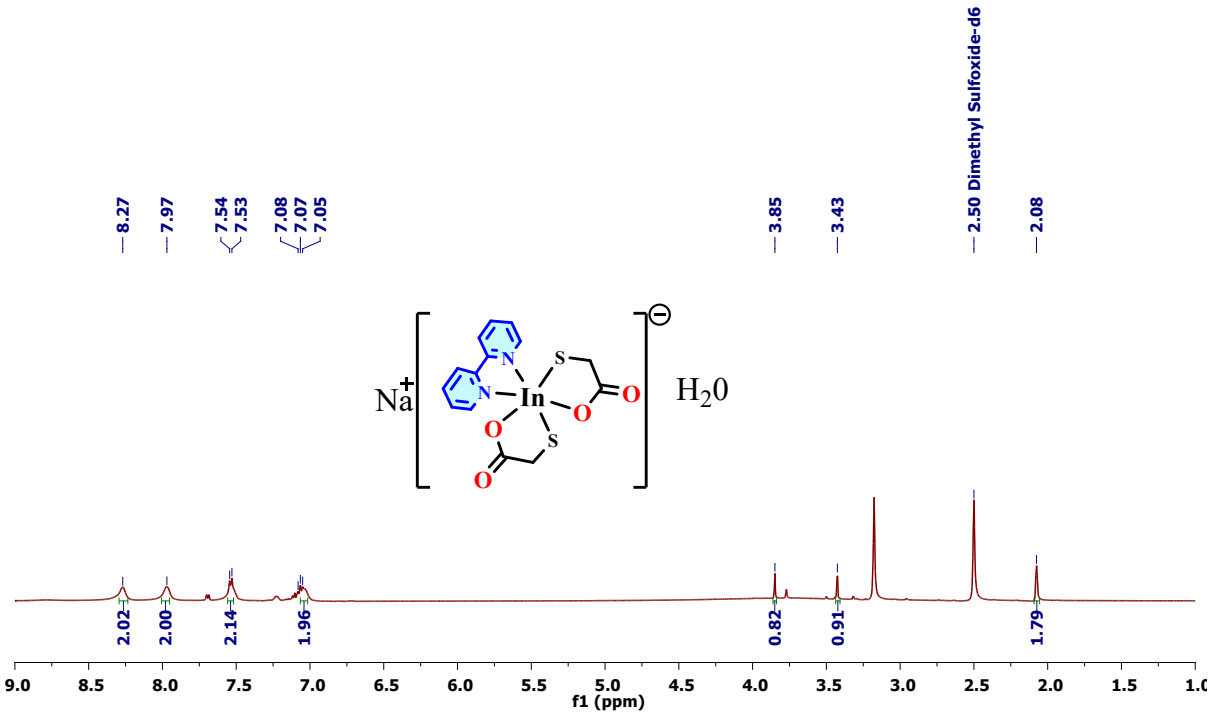


Figure S5. ^{1}H NMR spectra of Complex 3, Na [In (2,2'-bipyridyl)$(SCH_2COO)_{2}]$. (H_2O)

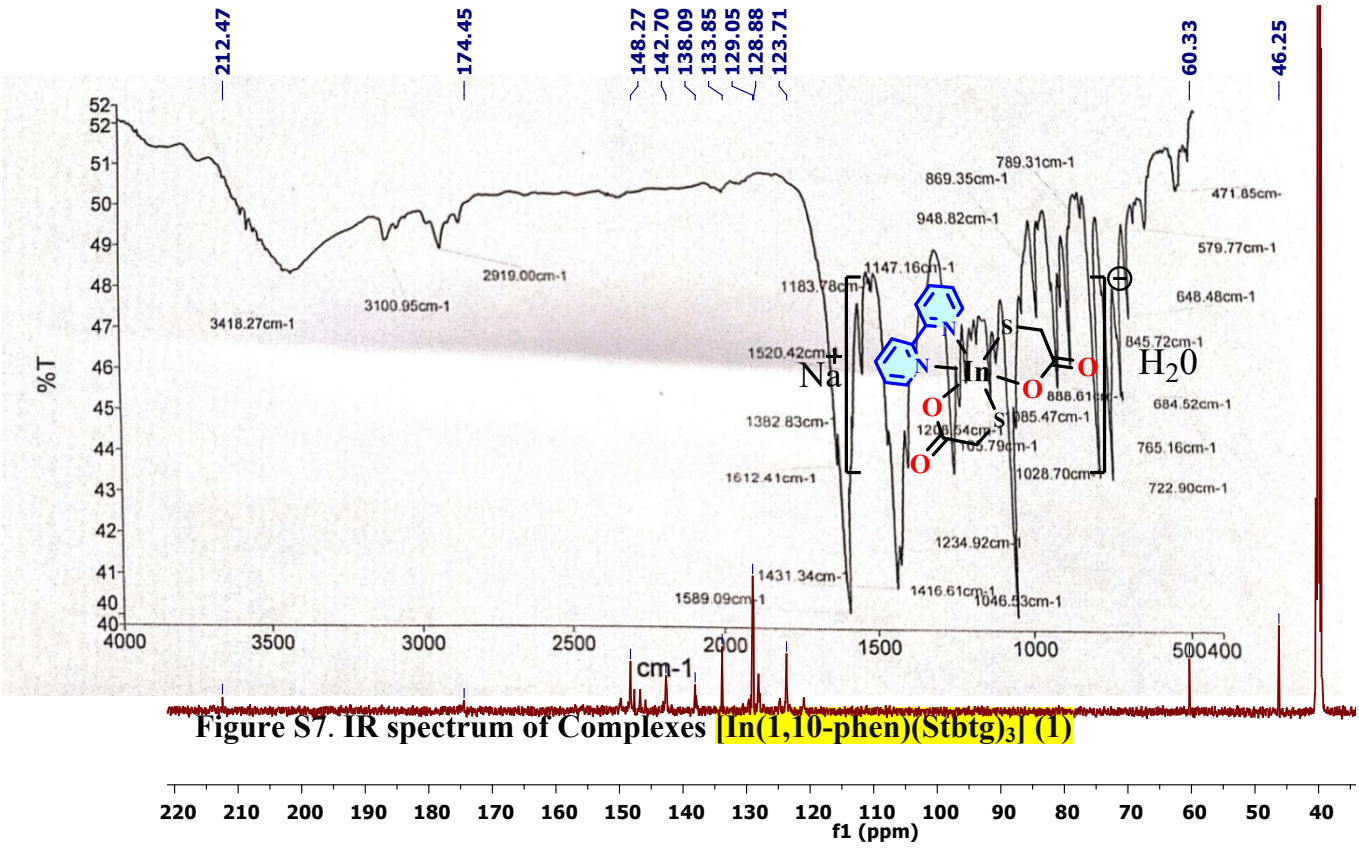


Figure S7. IR spectrum of Complexes [In(1,10-phen)$(Stbtg)_3$] (1)

Figure S6^{13}C NMR spectrum of Complex 3, Na [In (2,2'-bipyridyl)$(SCH_2COO)_{2}]$. (H_2O)

IR Spectra of the complexes

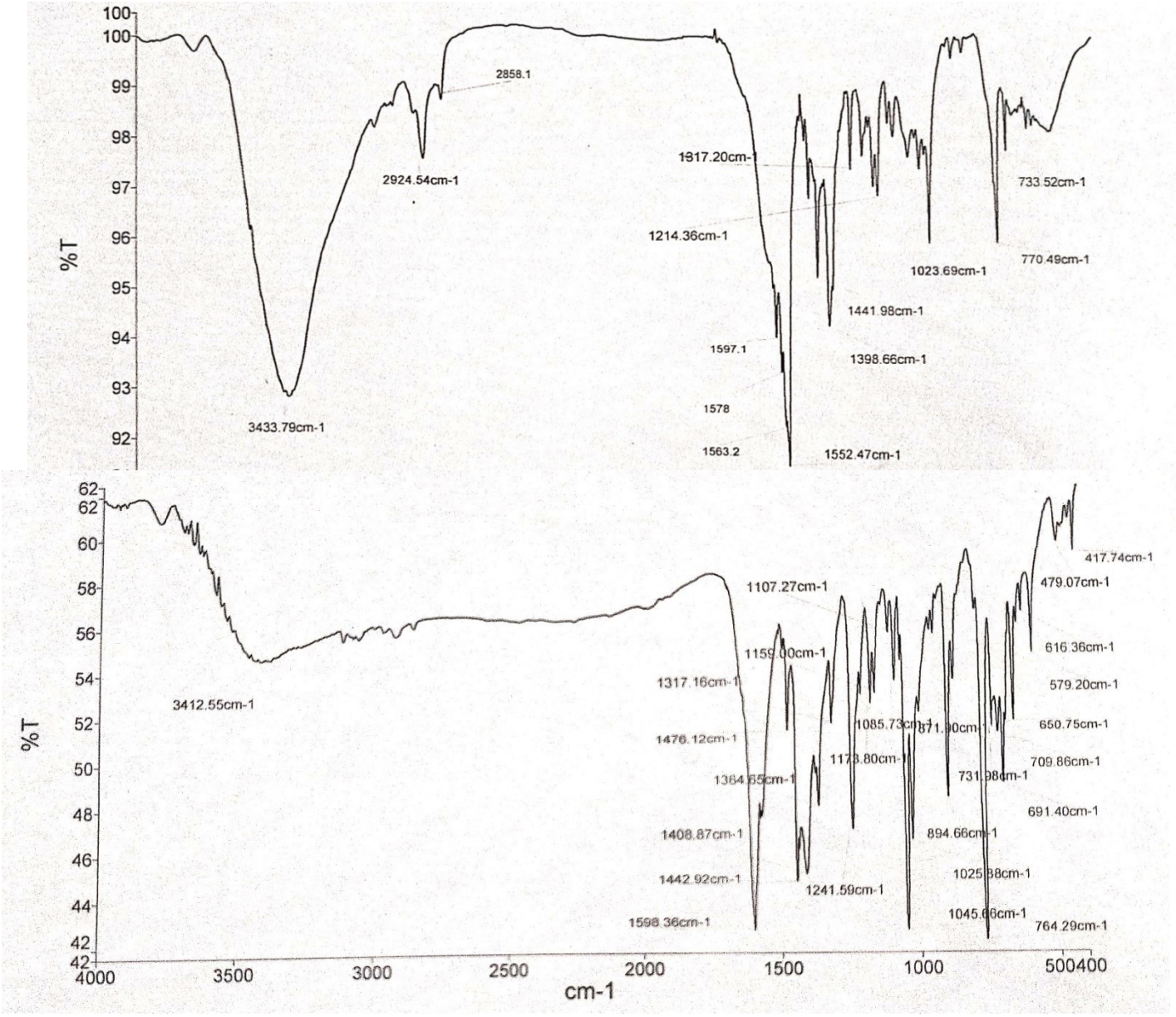


Figure S8. IR spectrum of Complexes [In(2,2′-bipy)(Stbtg)$_3$] (2)

Single Crystal X-ray diffraction

Crystals with a particular shape were used to gather X-ray intensity data using a Rigaku XtaLAB Synergy Dualflex diffractometer equipped with a HyPix 3000 detector and monochromated MoKα radiation (λ = 0.71073 Å). Data reduction, scaling, and absorption corrections were performed with the *CrysAlis Pro* software.[1,2] The structure was solved using the SHELXT program with Intrinsic Phasing and refined by least-squares procedures applying the SHELXL-2018[1] software package through the OLEX2 suite.[3] While hydrogen atom positions were geometrically calculated and refined using the riding model. Crystallographic and structure refinement data of the complex is shown in Table S1.

Identification code	Complex 1	Complex 2	Complex 3
CCDC No.	2334707	2405392	2405398
Empirical formula	$C_{39}H_{29}InN_2O_6S_6$	$C_{37}H_{29}InN_2O_6S_6$	$C_{14}H_{14}N_2O_5NaS_2In$
Formula weight	928.82	904.80	492.20
Temperature/K	293	293	293
Crystal system	monoclinic	orthorhombic	monoclinic
Space group	Pn	$Pca2_1$	$P2_1/c$
a/Å	10.9278(2)	28.2437(6)	15.7350(3)
b/Å	9.4920(2)	17.5038(3)	7.94580(10)
c/Å	19.0268(4)	14.8073(3)	14.7384(3)
α/°	90	90	90
β/°	97.401(2)	90	110.678(2)

γ/°	90	90	90
Volume/Å^3	1957.14(7)	7320.3(2)	1723.99(6)
Z	2	4	4
ρ_{calc}g/cm^3	1.576	1.642	1.896
μ/mm^{-1}	0.973	1.038	1.666
F(000)	940.0	3664.0	976.0
Radiation	Mo *K*α (λ = 0.71073)	Mo *K*α	Mo*K*α
2θ range /°	4.804 to 54.136	4.616 to 54.128	5.57 to 54.112
Index ranges	-13 ≤ h ≤ 13, -11 ≤ k ≤ 12, -24 ≤ l ≤ 23	-35 ≤ h ≤ 35, -20 ≤ k ≤ 22, -18 ≤ l ≤ 18	-20 ≤ h ≤ 20, -9 ≤ k ≤ 10, -18 ≤ l ≤ 18
Reflections collected	23670	54672	36996
Independent reflections	7385 [R_{int} = 0.0460, R_{sigma} = 0.0569]	14672 [R_{int} = 0.0623, R_{sigma} = 0.0763]	3676 [R_{int} = 0.0529, R_{sigma} = 0.0288]
Data/restraints/parameters	7385/38/488	14672/1/937	3676/0/230
Goodness-of-fit on F^2	1.037	1.042	1.065
Final R indexes [I>=2σ (I)]	R_1 = 0.0420, wR_2 = 0.0604	R_1 = 0.0570, wR_2 = 0.1231	R_1 = 0.0273, wR_2 = 0.0621
Final R indexes [all data]	R_1 = 0.0533, wR_2 = 0.0663	R_1 = 0.0978, wR_2 = 0.1436	R_1 = 0.0333, wR_2 = 0.0666
Largest diff.peak/hole /eÅ^{-3}	0.48/-0.31	1.90/-0.61	0.36/-0.56
Flack parameter	0.03(2)	0.32(2)	

Table S1. Crystal structure information and refinement parameters:

Second Order Perturbation Theory Analysis:

Table S2. For Complexes [In(1,10-phen)(Stbtg)$_3$] (1)

Donor NBO (i)	Acceptor NBO (j)	E(2) (kcal/mol)	E(j)-E(i) (a.u.)	F(i,j) (a.u.)
LP (2) O 8	**LP*(1)In 1**	**7.29**	**0.30**	**0.070**
LP (2) O 8	**LP*(2)In 1**	**8.25**	**0.49**	**0.057**
LP (2) O 8	**LP*(3)In 1**	**13.32**	**0.52**	**0.075**
LP (2) O 10	**LP*(1)In 1**	**19.87**	**0.35**	**0.081**
LP (2) O 10	**LP*(2)In 1**	**29.67**	**0.54**	**0.113**

LP (2) O 11	LP*(1)In 1	20.94	0.31	0.079
LP (2) O 11	LP*(4)In 1	21.19	0.54	0.096
LP (2) O 12	LP*(1)In 1	18.93	0.33	0.077
LP (2) O 12	LP*(3)In 1	21.64	0.56	0.098
LP (2) O 14	LP*(1)In 1	20.32	0.34	0.081
LP (2) O14	LP*(2)In 1	24.42	0.53	0.102
LP (2) O14	LP*(3)In 1	3.09	0.57	0.037
LP (2) O16	LP*(1)In 1	18.32	0.31	0.074
LP (2) O16	LP*(2)In 1	3.48	0.50	0.037
LP (2) O16	LP*(3)In 1	16.01	0.54	0.083
LP (2) O16	LP*(4) In 1	4.08	0.54	0.042
LP (1) N9	LP*(1) In 1	22.50	0.35	0.087
LP (1) N9	LP*(3) In 1	10.65	0.58	0.070
LP (1) N9	LP*(4) In 1	20.56	0.58	0.098
LP (1) N15	LP*(1)In 1	21.95	0.35	0.086
LP (1) N15	LP*(3)In 1	10.00	0.57	0.068
LP (1) N15	LP*(4)In 1	20.60	0.58	0.098

Table S3. For Complex [In(2,2′-bipy)(Stbtg)$_3$] (2)

Donor NBO (i)	Acceptor NBO (j)	E(2) (kcal/mol)	E(j)-E(i) (a.u.)	F(i,j) (a.u.)
LP (2) O 8	LP*(1)In 1	16.97	0.31	0.070
LP (2) O 8	LP*(3)In 1	19.54	0.53	0.091
LP (2) O 10	LP*(1)In 1	16.25	0.32	0.071
LP (2) O 10	LP*(3)In 1	17.28	0.55	0.087
LP (2) O 11	LP*(1)In 1	23.06	0.37	0.089
LP (2) O 11	LP*(2)In 1	23.26	0.55	0.101
LP (2) O 11	LP*(3)In 1	7.99	0.59	0.062
LP (2) O 12	LP*(1)In 1	20.46	0.35	0.082

LP (2) O 12	**LP*(2)In 1**	**27.94**	**0.53**	**0.109**
LP (2) O 13	**LP*(1)In 1**	**12.73**	**0.29**	**0.059**
LP (2) O 13	**LP*(2)In 1**	**10.47**	**0.48**	**0.063**
LP (2) O 13	**LP*(3)In 1**	**9.02**	**0.51**	**0.061**
LP (2) O 16	**LP*(1)In 1**	**26.21**	**0.34**	**0.091**
LP (2) O 16	**LP*(4)In 1**	**24.69**	**0.56**	**0.106**
LP (3) O 16	LP*(2)In 1	1.95	0.38	0.025
LP (1) N 9	**LP*(1)In 1**	**22.78**	**0.35**	**0.087**
LP (1) N 9	**LP*(3)In 1**	**5.88**	**0.57**	**0.052**
LP (1) N 9	**LP*(4)In 1**	**24.21**	**0.57**	**0.105**
LP (1) N 80	**LP*(1)In 1**	**20.79**	**0.35**	**0.083**
LP (1) N 80	**LP*(3)In 1**	**11.43**	**0.57**	**0.073**
LP (1) N 80	**LP*(4)In 1**	**16.18**	**0.57**	**0.086**

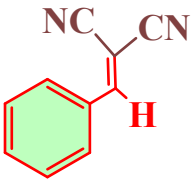
Table S4. For Complex Na[In(2,2′-bipy)($SCH_2COO)_2$].(H_2O) (3)

Donor NBO (i)	**Acceptor NBO (j)**	**E(2) (kcal/mol)**	**E(j)-E(i) (a.u.)**	**F(i,j) (a.u.)**
LP (3) S 2	**LP (1)In 1**	**115.54**	**0.22**	**0.151**
LP (3) S 2	**LP*(2)In 1**	**42.16**	**0.40**	**0.119**
LP (3) S 2	**LP*(3)In 1**	**15.89**	**0.37**	**0.072**
LP (3) S 2	**LP*(4)In 1**	**2.72**	**0.36**	**0.029**
LP (3) S 3	**LP (1)In 1**	**111.93**	**0.23**	**0.155**
LP (3) S 3	**LP*(2)In 1**	**49.67**	**0.42**	**0.131**
LP (3) S 3	**LP*(3)In 1**	**3.81**	**0.39**	**0.036**
LP (3) S 3	**LP*(4)In 1**	**2.00**	**0.38**	**0.026**
LP (2) O 4	**LP (1)In 1**	**11.75**	**0.49**	**0.078**
LP (2) O 4	**LP*(2)In 1**	**10.93**	**0.68**	**0.080**

LP (2) O 4	LP*(3)In 1	24.52	0.65	0.115
LP (2) O 4	LP*(4)In 1	8.21	0.64	0.066
LP (2) O 5	LP (1)In 1	19.19	0.53	0.104
LP (2) O 5	LP*(4)In 1	39.74	0.67	0.149
LP (1) N 7	LP (1)In 1	12.16	0.39	0.070
LP (1) N 7	LP*(2)In 1	13.11	0.57	0.080
LP (1) N 7	LP*(3)In 1	24.58	0.55	0.106
LP (1) N 21	LP (1)In 1	19.85	0.36	0.087
LP (1) N 21	LP*(3)In 1	10.82	0.52	0.068
LP (1) N 21	LP*(4)In 1	18.48	0.51	0.087
LP (2) O 37	LP*(1)Na 36	4.17	1.01	0.059
LP (1) O 12	LP*(1)Na 36	5.57	1.10	0.071
. LP (1) O 12	LP*(2)Na 36	6.39	0.98	0.071
LP (1) O 12	LP*(3)Na 36	11.48	0.92	0.092
LP (1) O 12	LP*(4)Na 36	2.49	0.92	0.043
LP (2) O 37	LP*(2)Na 36	6.09	0.89	0.066
LP (2) O 37	LP*(4)Na 36	15.17	0.82	0.101
LP (2) O 40	LP*(1)Na 36	9.30	0.96	0.086
LP (2) O 40	LP*(2)Na 36	5.77	0.84	0.063
LP (2) O 40	LP*(3)Na 36	12.21	0.78	0.088
LP (2) O 43	LP*(1)Na 36	11.92	0.95	0.096
LP (2) O 43	LP*(2)Na 36	4.89	0.83	0.058
LP (2) O 41	LP*(1)Na 36	11.29	0.96	0.094
LP (2) O 41	LP*(2)Na 36	11.52	0.84	0.088
LP (2) O 42	LP*(1)Na 36	10.32	1.03	0.093
LP (2) O 42	LP*(2)Na 36	11.52	0.91	0.092
LP*(1)Na 44	LP*(1)Na 45	17.81	0.04	0.076

Data S4-NMR (^{1}H and ^{13}C) spectra of 2 component Knoevenagel Condensation Products:

4a. 2-benzylidenemalononitrile[4]



M. F. $C_{11}H_8N_2$ (168.20),Yield: (0.156 g, 93%).White powder. ^{1}H NMR (500 MHz, $CDCl_3$, ppm) δ 7.54 (t, *J* =7.5Hz, 2H), 7.64 (t, *J* =8Hz, 2H), 7.78 (s, 1H), 2,46 (d,2H); ^{13}C NMR (125 MHz, $CDCl_3$, ppm) δ160.29, 134.99, 131.34, 131.10, 130.02, 114.07, 112.91, 83.33;

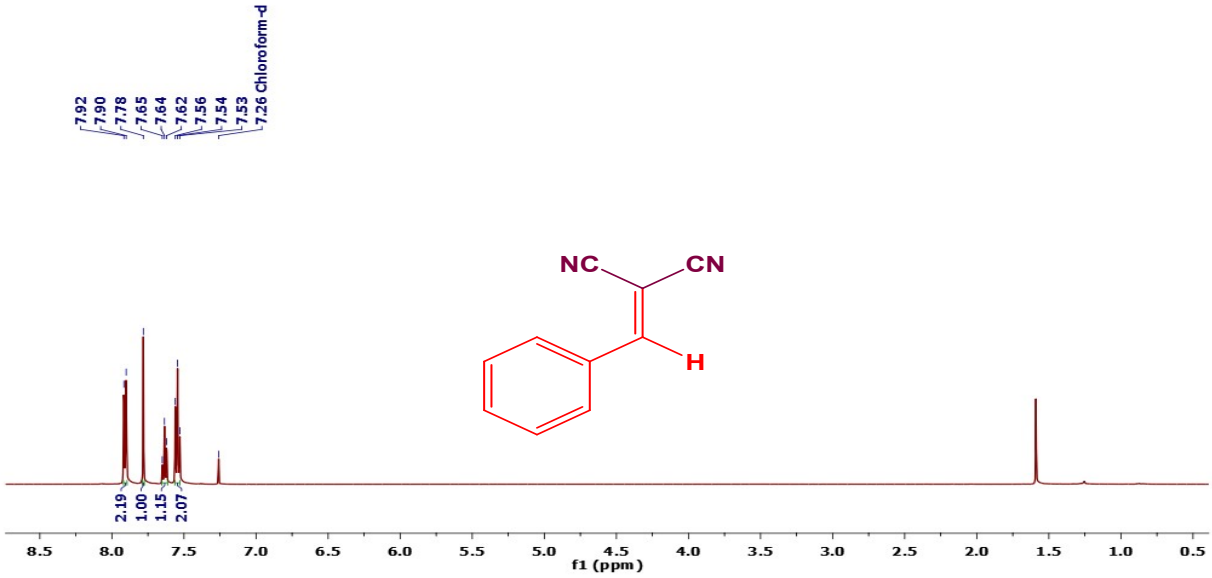


Figure S10. ^{1}H NMR spectra of 4a

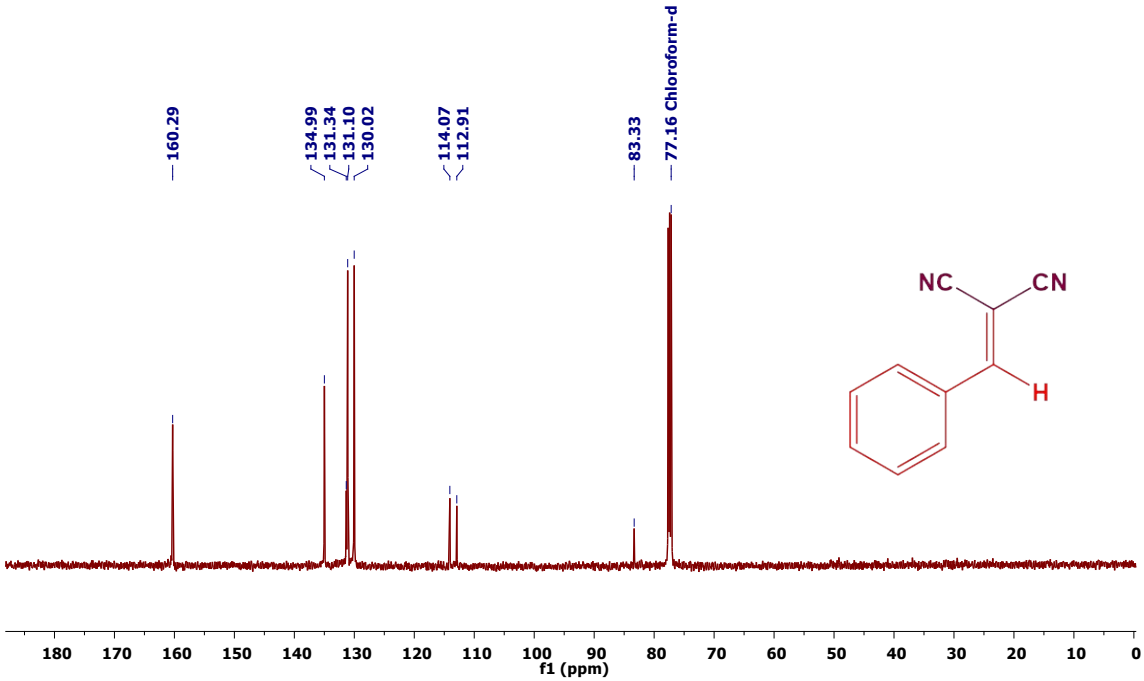
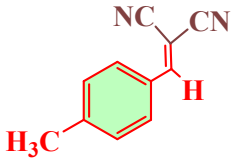


Figure S11. ^{13}CNMR spectra of 4a

4b. 2-(4-methylbenzylidene)malononitrile[4]



M. F. $C_{11}H_8N_2$ (168.20).Yield: (0.159 g, 95%).White powder. ^{1}H NMR (500 MHz, $CDCl_3$, ppm) δ 7.81 (d, *J* =8Hz, 2H), 7.72 (s, 1H), 7.34 (d, *J* = 8 Hz, 2H), 2,46 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$, ppm) δ159.81, 146.44, 130.98, 130.45, 128.57, 114.07, 112.92, 81.34, 22.05;

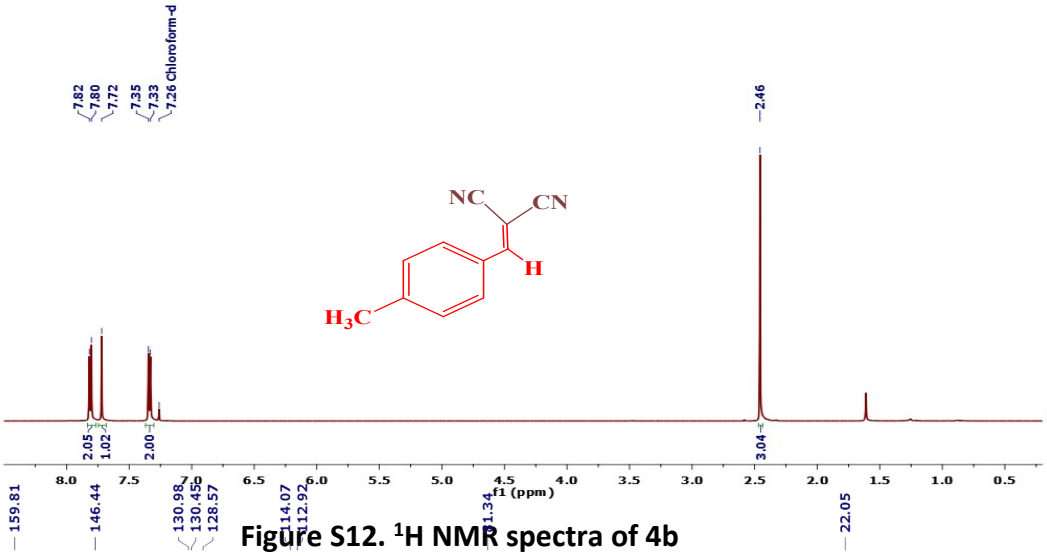


Figure S12. ^{1}H NMR spectra of 4b

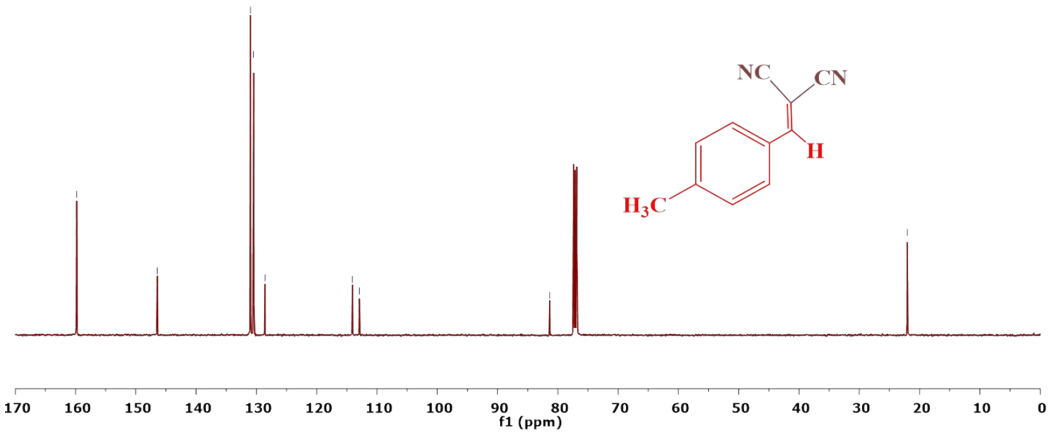
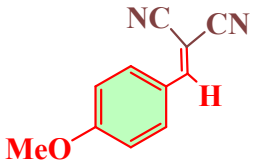


Figure S13. ^{13}C NMR spectra of 4b

4c. 2-(4-methoxybenzylidene)malononitrile[4]



M. F. $C_{11}H_8N_2O$ (184.20).Yield: (0.174 g, 95%).yellow powder. ^1H NMR (500 MHz, $CDCl_3$, ppm) δ7.91 (d, *J* = 9.5 Hz, 2H), 7.65 (s, 1H), 7.34 (d, *J* = 9.5 Hz, 2H), 3.91 (s, 3H); ^{13}C NMR (125 MHz, $CDCl_3$, ppm) δ 164.82, 158.90, 133.51, 124.12, 115.23, 114.48, 112.49, 78.71, 55.87

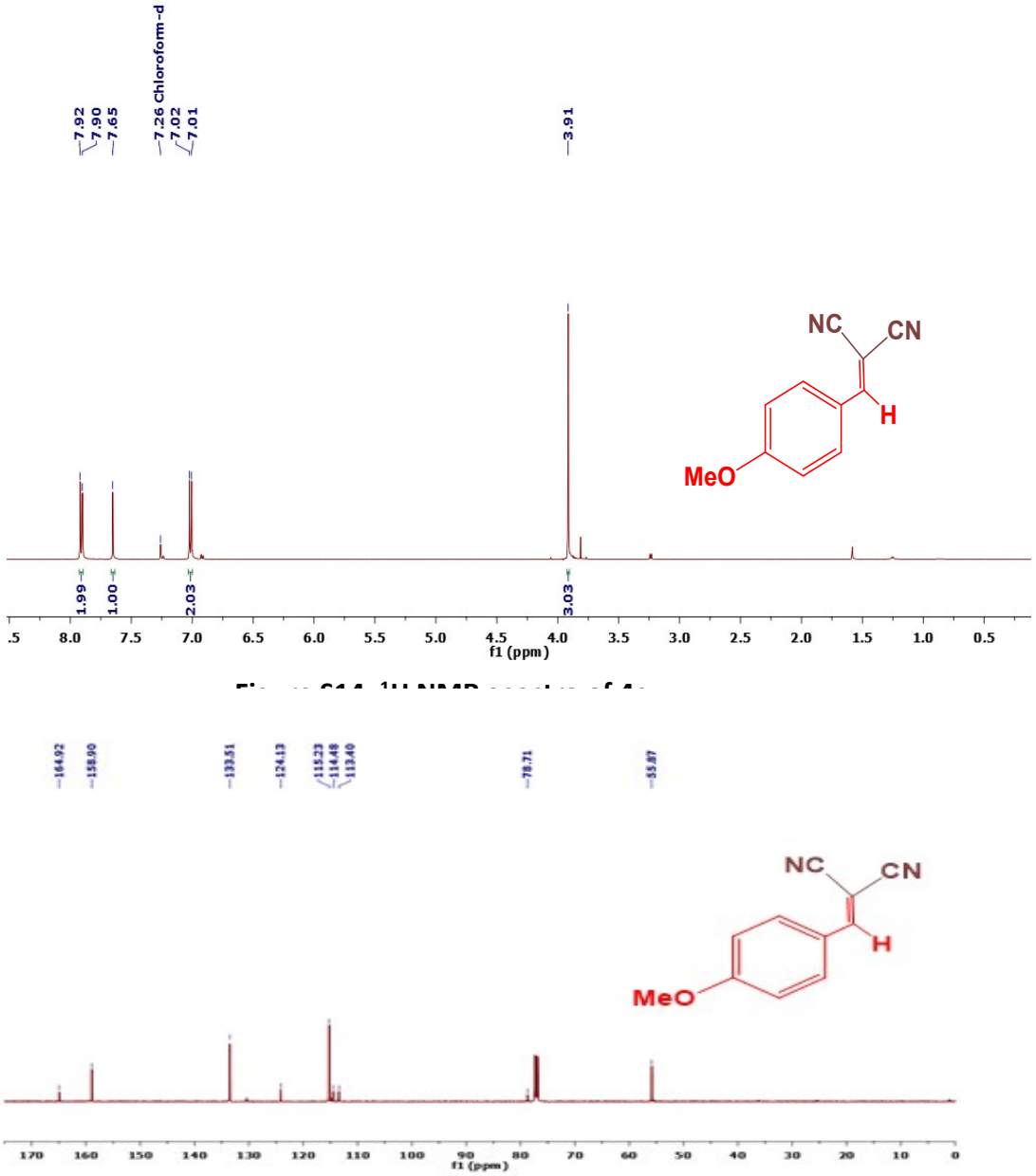
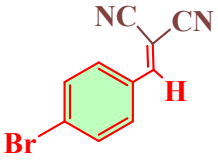


Figure S15. ^{13}C NMR spectra of 4c

4d. 2-(4-bromobenzylidene)malononitrile[5]



M.F. $C_{10}H_5N_2Br$ (233.07) Yield: (0.214g, 92%). White powder. ^1H NMR (500 MHz, $CDCl_3$, ppm) δ 8.38 (d, *J* = 9 Hz, 2H), 8.07(d, *J* = 8.5 Hz, 2H), 7.88 (s, 1H), ^{13}C NMR (125 MHz, $CDCl_3$, ppm) δ 158.42, 133.16, 131.85, 129.98, 129.75, 113.49, 112.38, 83.67

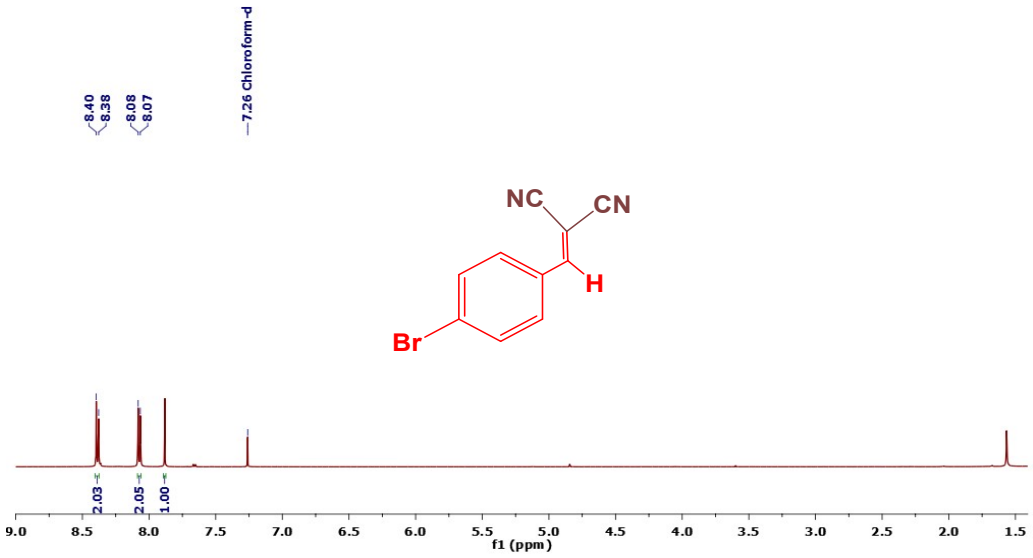


Figure S16. ^1H NMR spectra of 4d

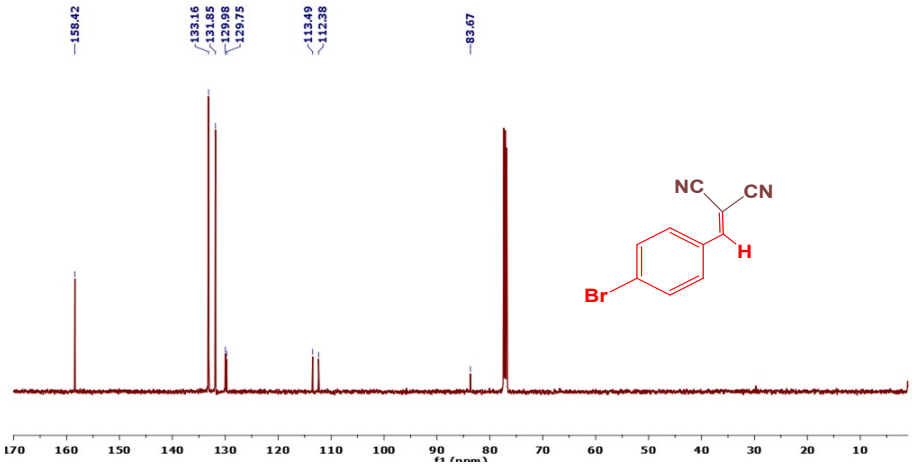
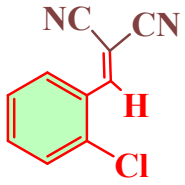


Figure S17. ^{13}C NMR spectra of 4d

4e. 2-(2-chlorobenzylidene)malononitrile[4]



M.F. $C_{10}H_5N_2Cl$ (188.61),Yield: (0.169 g, 92%). White powder. ^1H NMR (500 MHz, $CDCl_3$, ppm) δ 8.27 (s, 1H), 8.18 (d, *J* = 7.5 Hz, H), 7.55 (d, *J* = 3.5 Hz, 2H); 7.45(m, 1H), ^{13}C NMR (125 MHz, $CDCl_3$, ppm) δ 156.06, 136.41, 135.07, 130.78, 129.16, 129.16, 127. 85, 113.26, 111.95, 85.94;

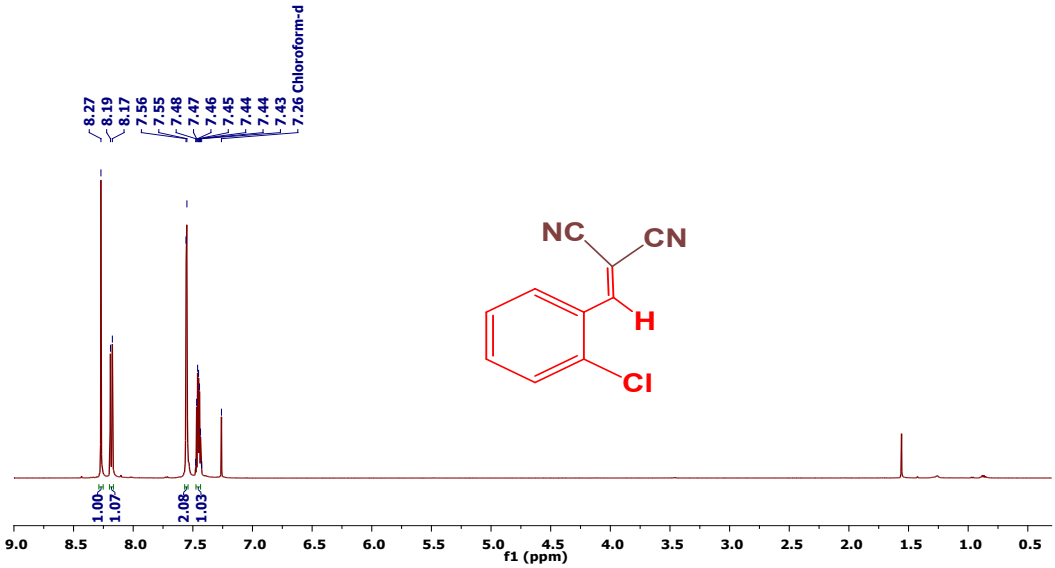


Figure S18. ^{1}H NMR spectra of 4e

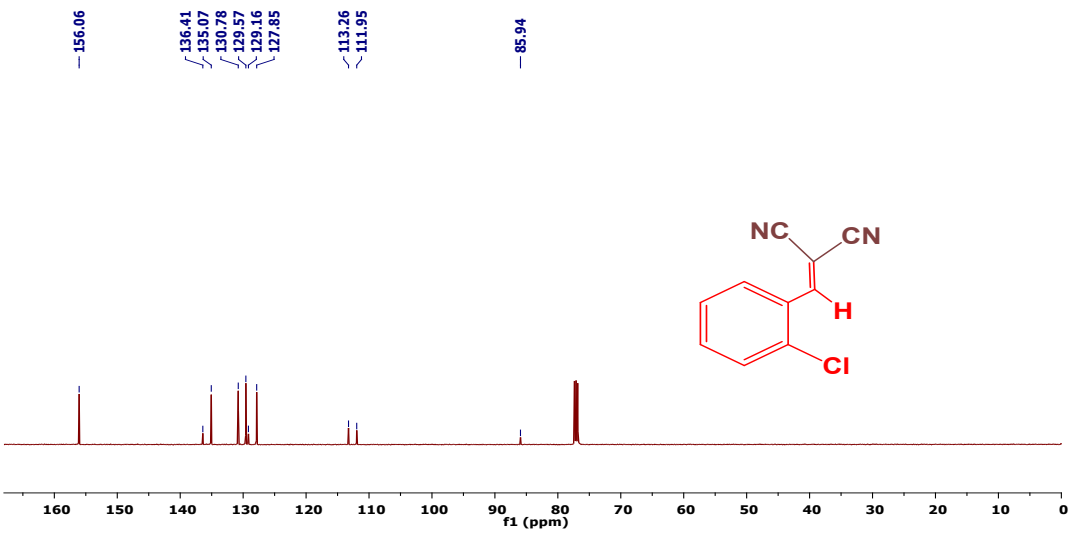
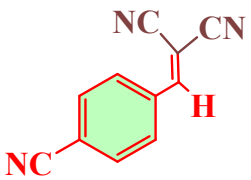


Figure S19. ^{13}C NMR spectra of 4e

4f. 2-(4-cyanobenzylidene)malononitrile[6]



M.F.$C_{11}H_5N_3$ (199.17), Yield: (0.181g, 91%). Light yellow powder. ^{1}H NMR (500 MHz, $CDCl_3$, ppm) δ 7.98 (d, *J* = 8 Hz, 2H), 7.82 (d, *J* = 8.5 Hz, 2H); 7.81 (s, 1H), ^{13}C NMR (125 MHz, $CDCl_3$, ppm) 157.36, 134.33, 133.21, 130.75, 117.37, 112.79, 111.74, 87.03;

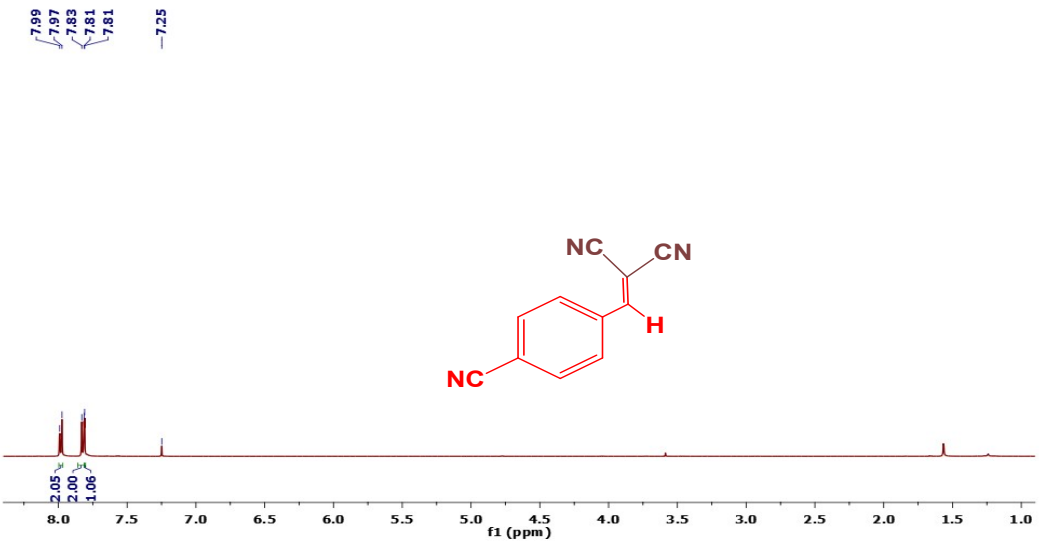


Figure S20. ^{1}H NMR spectra of 4f

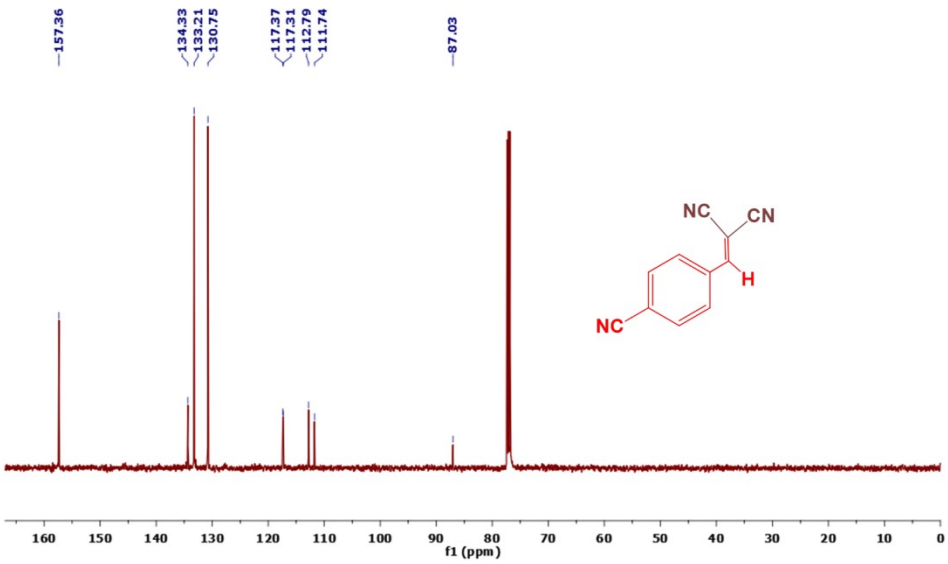
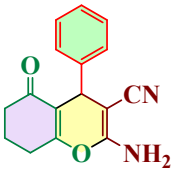


Figure S21. ^{1}H NMR spectra of 4f

The Products of Multicomponent reaction

Data S5 -NMR (^{1}H and ^{13}C) spectra of2- Amino-4H-chromeneDerivatives of cyclohexane 1,3-dione

5a. 2-amino-6,6,7,7,8,8-hexahydro-5-oxo-4-phenyl-4H-chromene-3-carbonitrile[4]



M. F. $C_{16}H_{14}N_2O_2$ (266.29), Yield: (0.239g, 90%), white powder. ^1H NMR (500 MHz, $CDCl_3$, ppm)δ 7.31 – 7.19 (m, 5H), 4.51 (s, 2H), 4.44 (s, 1H), 2.66-2.53 (m, 2H), 2.43 – 2.31 (m, 2H), 2.09 – 1.98 (m, 2H) ; ^{13}C NMR (125 MHz, $CDCl_3$, ppm) δ :195.95, 163.32, 157.48, 143.23, 128.66, 127.62, 127.24, 118.66, 115.38, 62.07, 36.87, 35.44, 27.08, 20.19

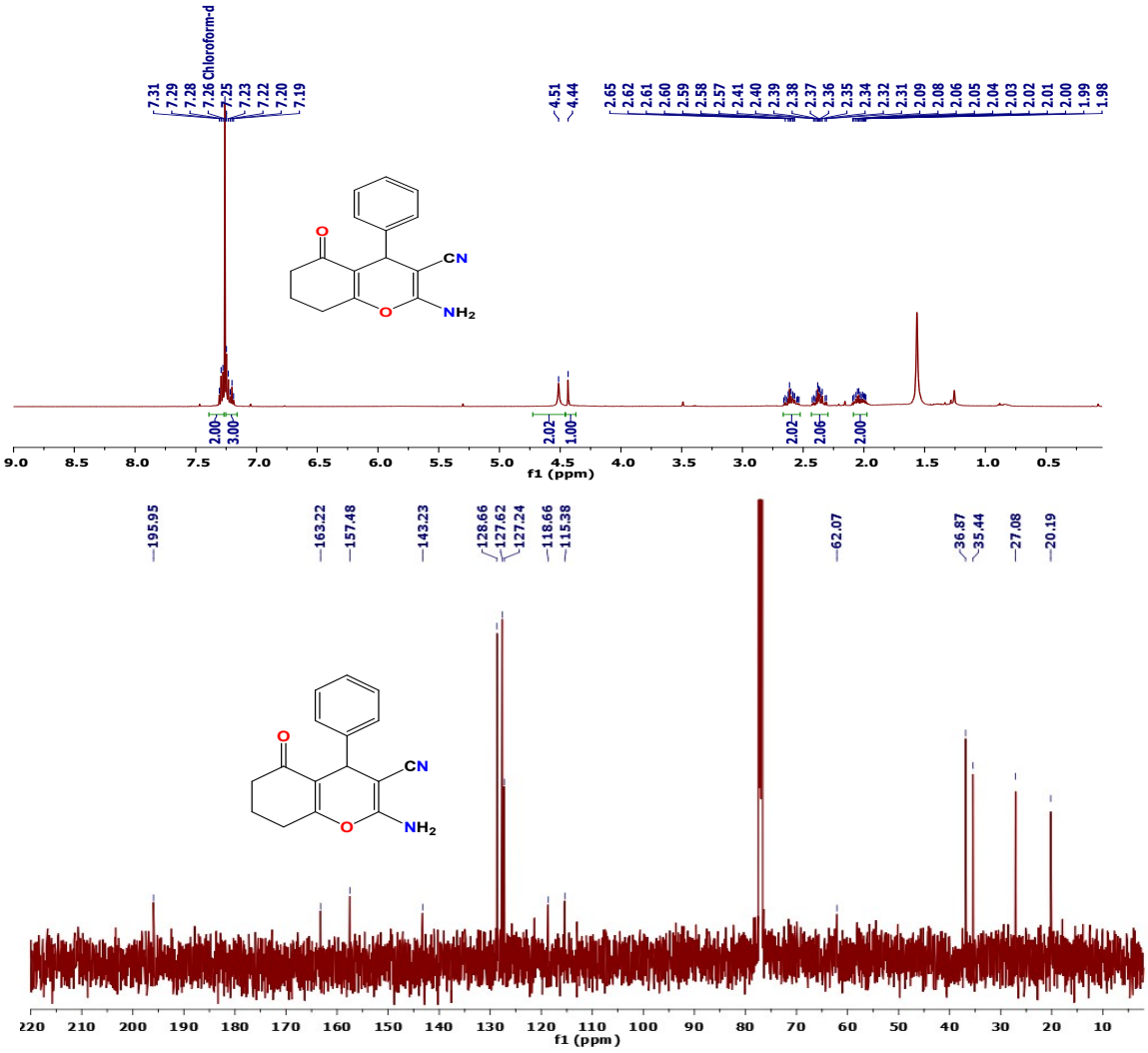
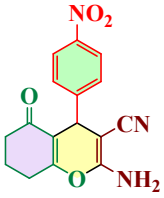


Figure S23. ^{13}C NMR Spectrum of 5a

5b. 2-amino-6,6,7,7,8,8-hexahydro-5-oxo-4-(4-nitrophenyl)-4H-chromene-3-carbonitrile[6]



M. F. $C_{16}H_{13}N_3O_4$ (311.29), Yield: (0.255 g,82%), off white powder. ^1H NMR (500 MHz, DMSO-d6, ppm) :δ 8.11 (d, *J* = 9 Hz, 2H), 7.45 (d, *J* = 9 Hz, 2H), 7.17 (s, 2H), 4.36 (s, 1H), 2.67–2.58 (m, 2H), 2.35 – 2.21 (m, 2H), 2.00 – 1.88 (m, 2H); ^{13}C NMR (125 MHz, DMSO-d6, ppm) δ

196.43, 165.66, 159.06, 152.84, 146.77, 129.10, 124.17, 119.88, 113.24, 57.42, 36.73, 36.09, 27.04, 20.25.

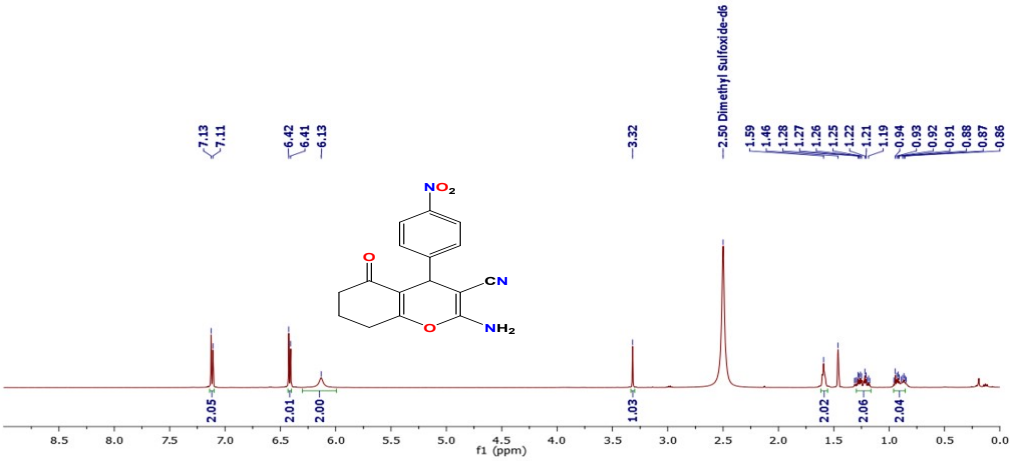


Figure S24. ^{1}H NMR Spectrum of 5b

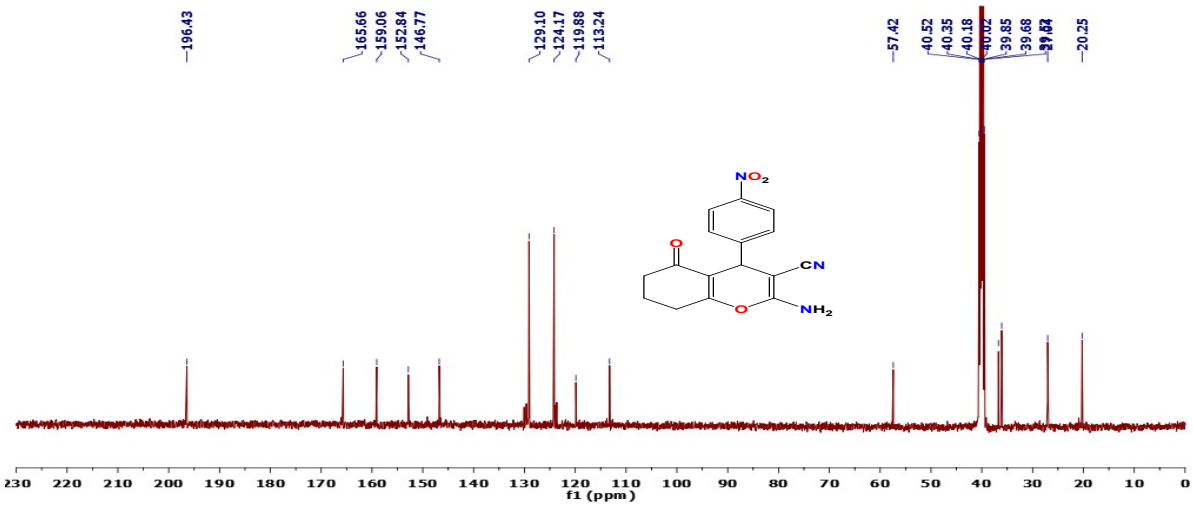
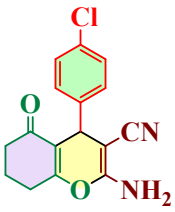


Figure S25. ^{13}C NMR Spectrum of 5b

5c. 2-amino-6,6,7,7,8,8-hexahydro-5-oxo-4-(4-chlorophenyl)-4H-chromene-3-carbonitrile[4]



M. F. $C_{16}H_{13}N_2O_3Cl$ (300), Yield: (0.264g, 88%), Pale yellow powder. 1H NMR (500 MHz, DMSO-d6, ppm) δ 7.33 (d, *J* = 9 Hz, 2H), 7.18 (d, *J* = 8Hz, 2H), 7.05 (s, 2H), 4.19 (s, 1H), ,2.64 – 2.58 (m, 2H), 2.33 – 2.21 (m, 2H), 1.98 – 1.83 (m, 2H); 13C NMR (125 MHz,

DMSO-d6, ppm) δ 196.41, 165.17, 158.98, 144.31, 131.60, 129.61, 128.79, 120.11, 113.87, 58.21, 36.80,35.51 29.99,26.99, 20.28

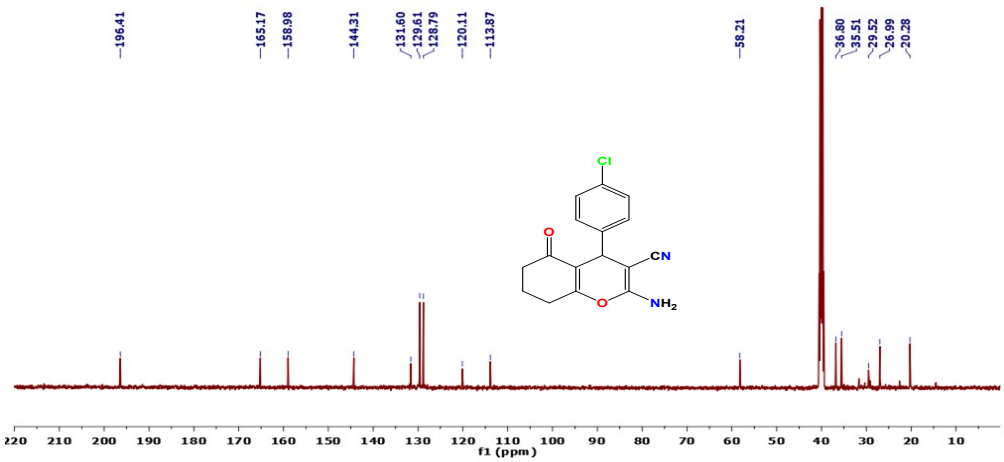
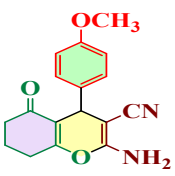


Figure S27. ^{1}H NMR Spectrum of 5c

5d. 2-amino-6,6,7,7,8,8-hexahydro-5-oxo-4-(4-methoxyphenyl)-4H-chromene-3-carbonitrile [7]



M. F. $C_{17}H_{16}N_2O_3$ (296.32), Yield: (0.281 g, 95%), pale yellow powder. ^{1}H NMR (500 MHz, CDCl3, ppm) δ 7.16 (d, *J* = 9.5 Hz, 2H), 6.82 (d, *J* = 8 Hz, 2H), 4.55 (s, 2H), 4.38 (s, 1H), 3.716(s, 3H), 2.64 – 2.51 (m, 2H), 2.40 – 2.33 (m, 2H), 2.07 – 1.94 (m, 2H); ^{13}C NMR (125 MHz, DMSO-d6, ppm) δ 196.15, 163.04, 158.69, 157.44, 135.59, 128.72, 118.72, 115.54, 114.03, 63.74, 55.30, 36.90, 34.70, 27.06, 20.20

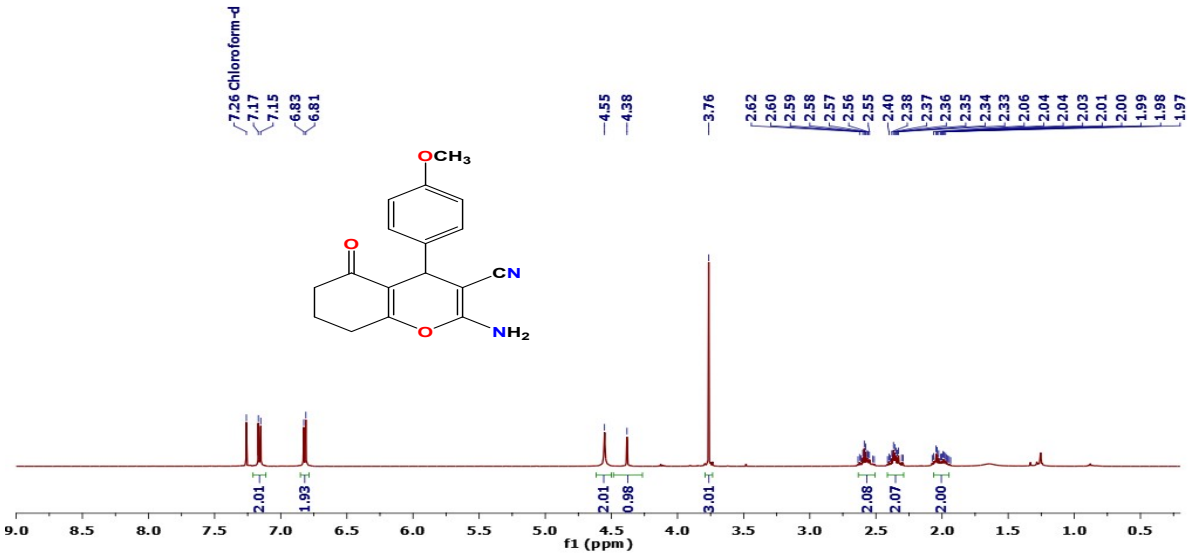


Figure S28. ^{1}H NMR Spectrum of 5d

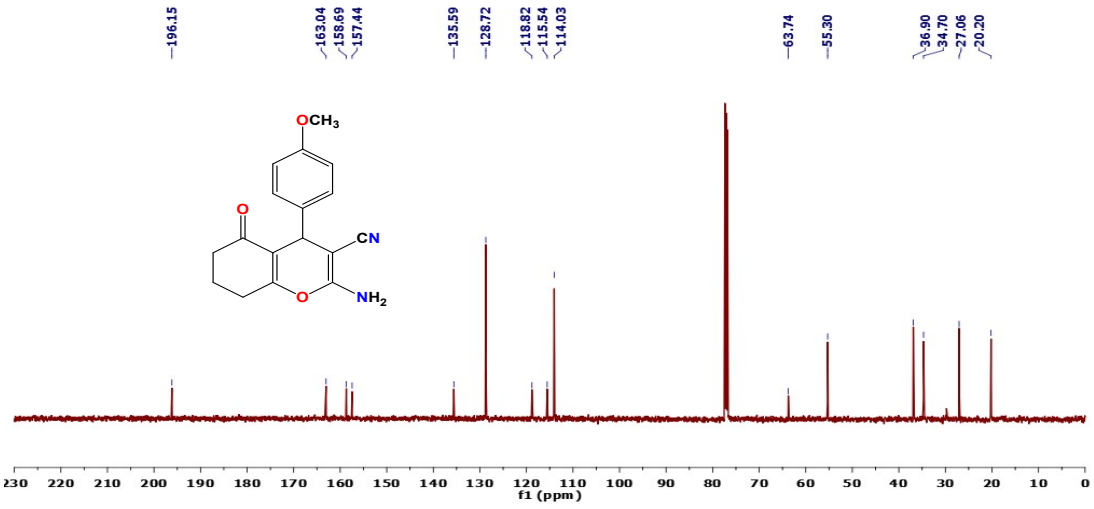
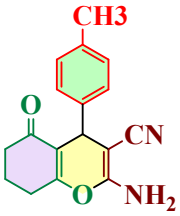


Figure S29. ^{13}C NMR Spectrum of 5d

5e. 2-amino-6,6,7,7,8,8-hexahydro-5-oxo-4-(4-methylphenyl)-4H-chromene-3-carbonitrile[4,6]



M. F. $C_{16}H_{16}N_2O_2$ (280.32), Yield: (0.266 g,95%), white powder. ^{1}H NMR (500 MHz, CDCl3, ppm) δ 7.13 (d, J = 8 Hz, 2H), 7.09 (d, *J* = 8 Hz, 2H), 4.55 (s, 2H), 4.39 (s, 1H), 2.64 – 2.52 (m, 2H), 2.39 – 2.32 (m, 2H), 2.29 (s, 3H), 2.08 – 1.95 (m, 2H); ^{13}C NMR (125 MHz, DMSO-d6, ppm) δ 196.10, 163.17, 157.47, 140.38, 136.79, 127.49, 118.80, 115.45, 63.69, 36.89, 35.06, 29.76,27.08,21.15, 20.19

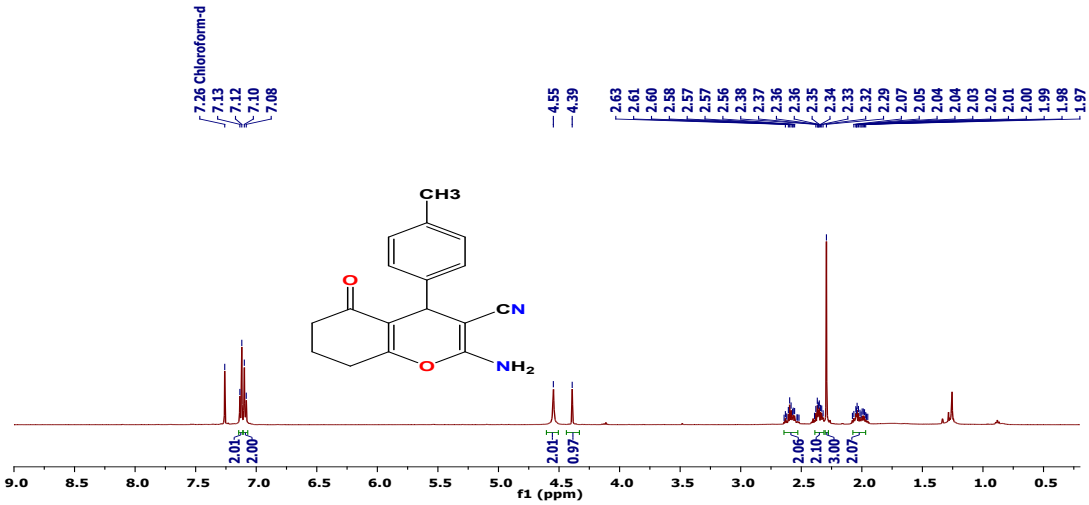


Figure S30. ^{1}H NMR Spectrum of 5e

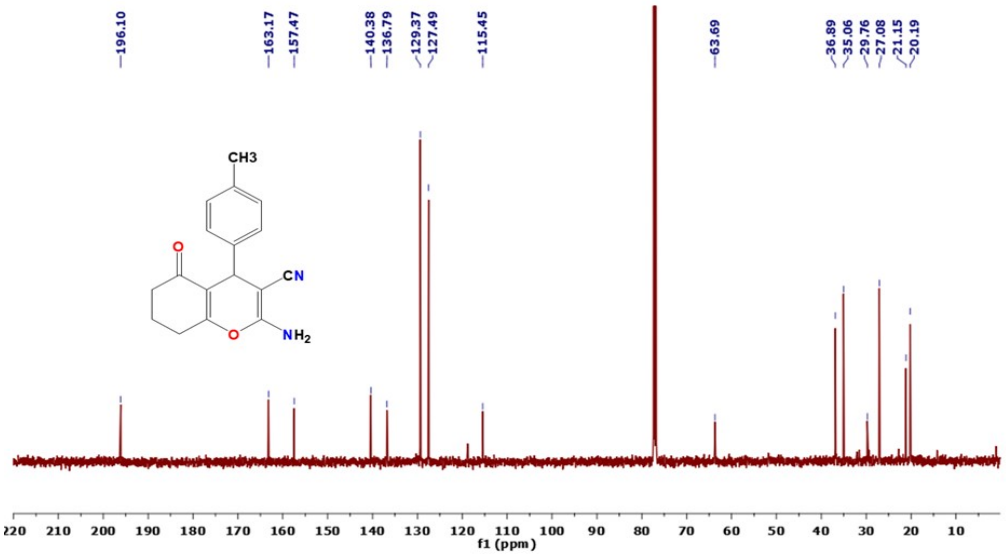
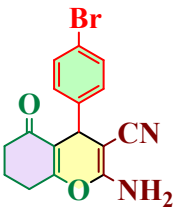


Figure S31. ^{13}C NMR Spectrum of 5e

5f. 2-amino-6,6,7,7,8,8-hexahydro-5-oxo-4-(4-Bromophenyl)-4H-chromene-3-carbonitrile[7]



M. F. $C_{16}H_{13}N_2O_3Br$ (345.19),Yield: (0.290g, 84%), Pale yellow powder. 1H NMR (500 MHz, DMSO-d6, ppm) δ 7.47 (d, *J* = 8 Hz, 2H), 7.12 (d, *J* = 8Hz, 2H), 7.03 (s, 2H), 4.18 (s, 1H), ,2.64 – 2.58 (m, 2H), 2.33

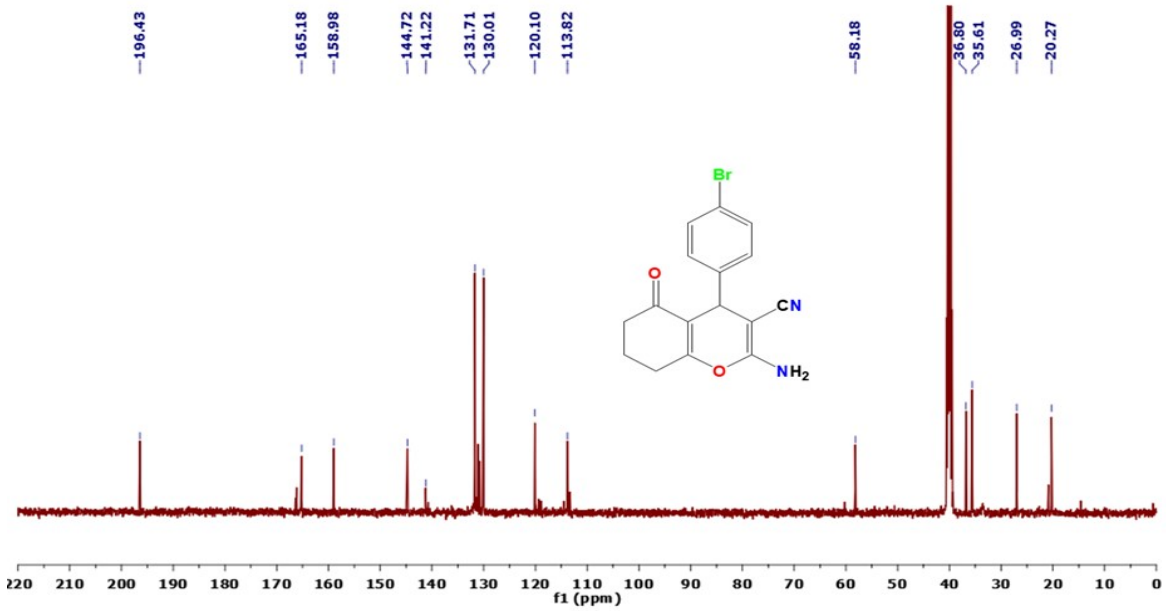


Figure S33. ^{1}H NMR Spectrum of 5f

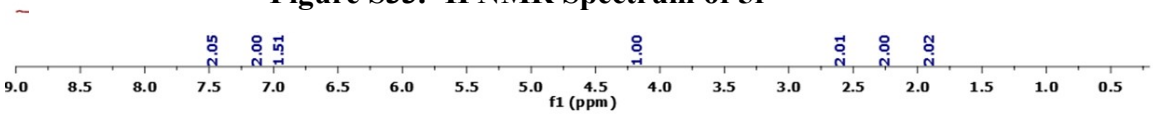
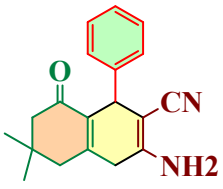


Figure S32. ^{1}H NMR Spectrum of 5f

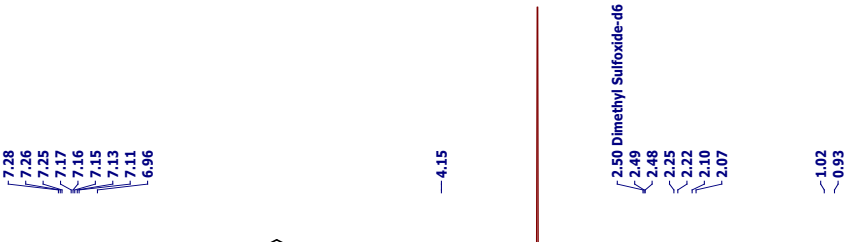
– 2.21 (m, 2H), 1.98 – 1.84 (m, 2H); 13C NMR (125 MHz, DMSO-d6, ppm) δ 196.43, 165.18, 158.98, 144.72, 141.22, 131.71, 130.01, 120.10, 113.82, 58.18, 36.80,35.61, 26.99, 20.27

Datas6- NMR (^{1}H and ^{13}C) spectra of 2- Amino-4H-chromeneDerivatives of 5,5′ dimethyl cyclohexane 1,3-dione

6a. 2-amino-6,6,8,8-tetrahydro-7,7-dimethyl-5-oxo-4-phenyl-4H-chromene-3-carbonitrile[4]



M. F. $C_{18}H_{18}N_2O_2$ (294.34). Yield: (0.264g, 90%). Pale yellow powder. ^{1}H NMR (500 MHz, DMSO-d_6, ppm) δ: 7.26 (t, *J* = 7 Hz, 2H), 7.16 (t, *J* = 7.5 Hz, 1H), 7.13 (d, *J* = 7 Hz, 2H), 6.98(s,2H),4.15 (s, 1H), 2.49(d, *J* = 4.5 Hz, 2H), 2.09 (d, *J* = 16 Hz, 1H), 2.08 (d, *J*=16Hz 1H), 102 (s, 3H), 0.93 (s, 3H); ^{13}C NMR (125 MHz, DMSO d_6, ppm) δ 196.22, 163.04, 159.02, 145.26, 128.85, 127.66, 127.10, 120.24, 113.26, 58.86, 50.50, 32.32, 28.91, 27.32.



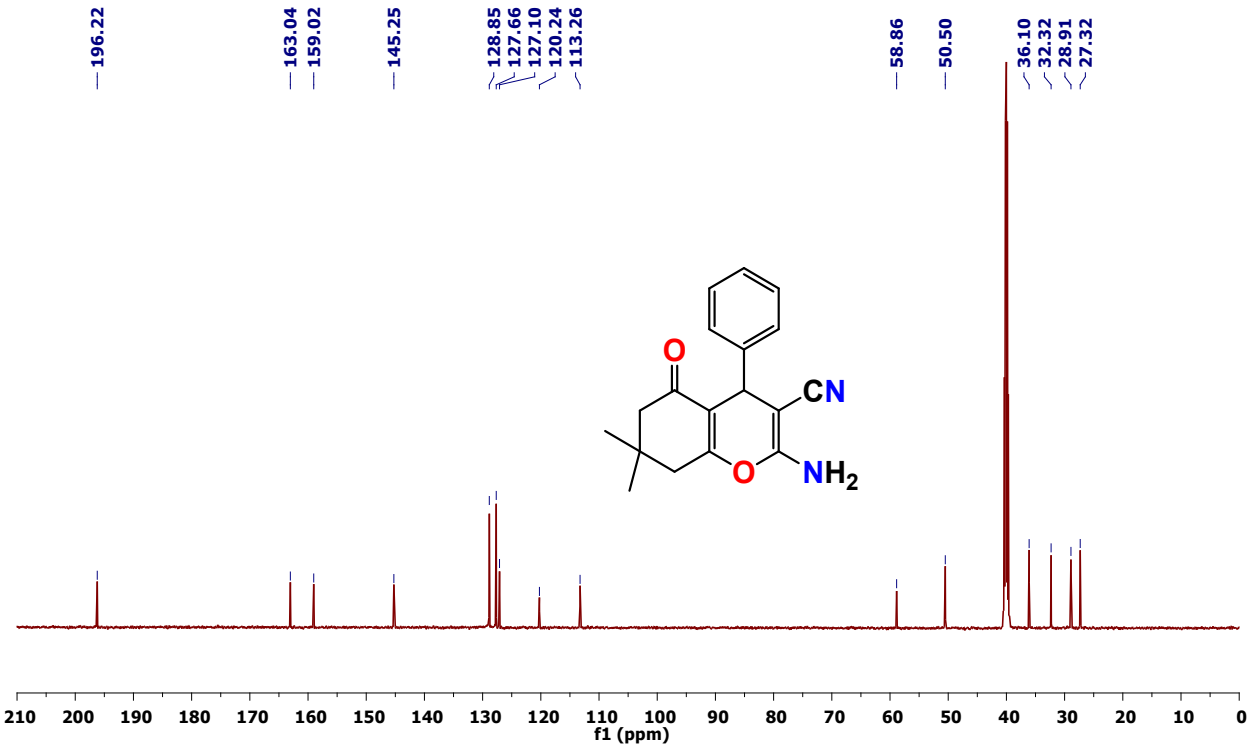
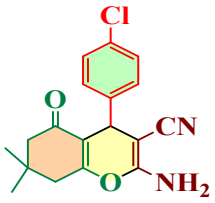
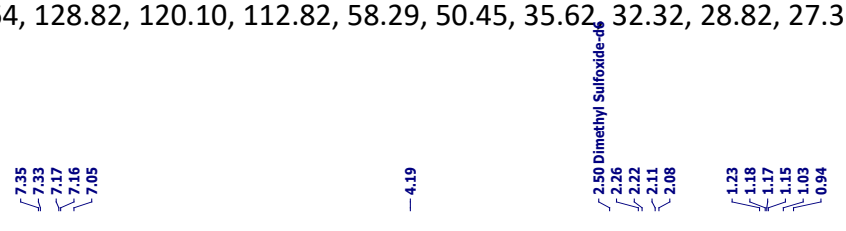


Figure S35. ^{13}C NMR Spectrum of 6a

6b.2-amino-6,6,8,8-tetrahydro-7,7-dimethyl-5-oxo-4-(4-Chlorophenyl)-4H-chromene-3-carbonitrile[4]



M.F. $C_{16}H_{13}ClN_2O_2$ (328.79), Yield: (0.270g, 82%). White powder, ^1H NMR (500 MHz, DMSO d6, ppm) δ7.34 (d, J = 8.5 Hz, 2H), 7.17 (d, *J* = 8.0 Hz, 2H), 7.05 (s, 2H), 4.19 (s, 1H), 2.24 (d, *J* = 16 Hz, 2H), 2.09 (d, *J* = 16.5 Hz, 1H), 1.24 (d, *J* = 10 Hz, 1H), 1.17 (t, *J* = 6.5 Hz, 1H), 103 (s, 3H), 0.94 (s, 3H) ^{13}C NMR (125 MHz, DMSO d_6, ppm) δ 196.30, 163.20, 159.01, 144.26, 131.64, 129.64, 128.82, 120.10, 112.82, 58.29, 50.45, 35.62, 32.32, 28.82, 27.36.



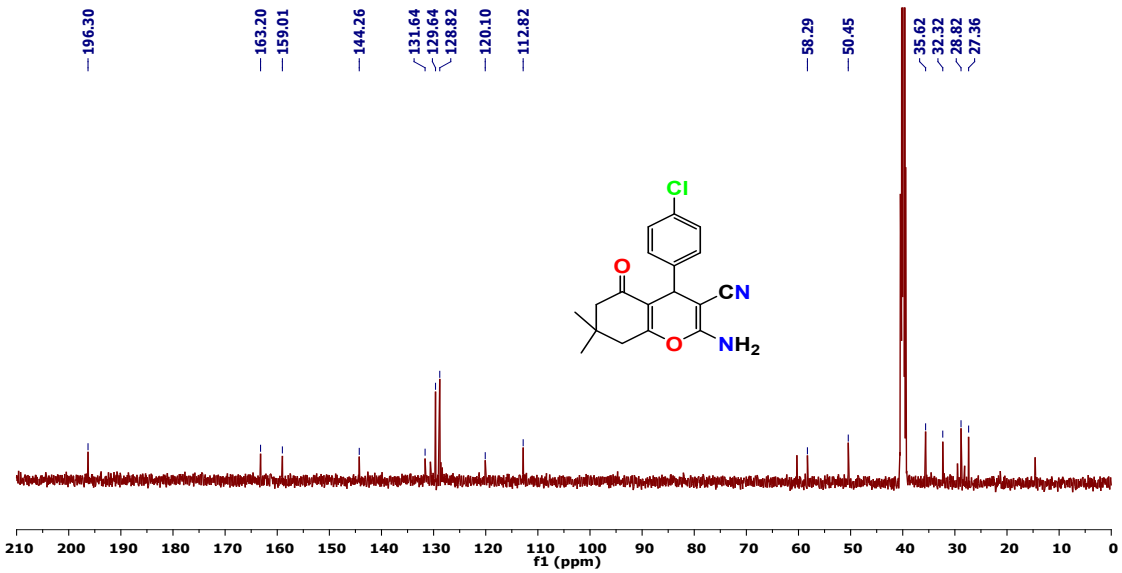
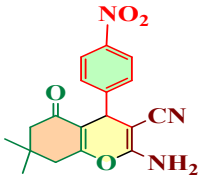
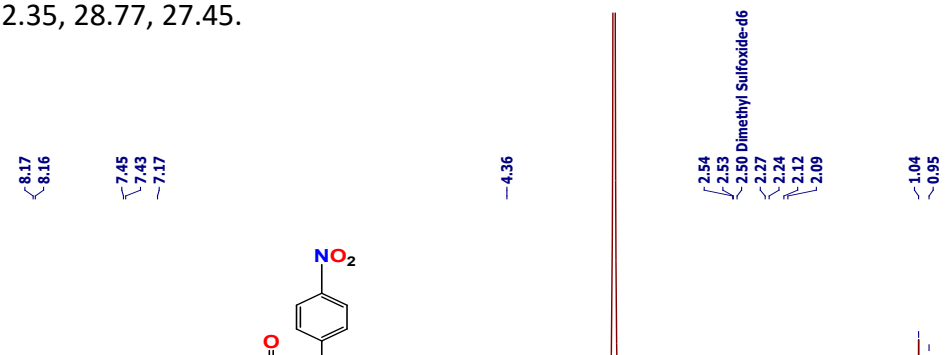


Figure S37. ^{13}C NMR Spectrum of 6b

6c.2-amino-6,6,8,8-tetrahydro-7,7-dimethyl-5-oxo-4-(4-Nitrophenyl)-4H-chromene-3-carbonitrile[7]



M. F. $C_{18}H_{17}N_3O_4$(339.35), Yield: (0.292g, 86%). Off white powder, ^{1}H NMR (500 MHz, DMSO-d6, ppm) δ8.17 (d, *J* = 9 Hz, 2H), 7.44 (d, *J* = 9 Hz, 2H), 7.17(s, 2H), 4.36 (s, 1H), 2.54 – 2.53 (m, 2H), 2.27 – 2.09 (m, 2H), 1.04 (s, 3H), 0.95 (s, 3H); 13C NMR (125 MHz, DMSO-d6, ppm) δ 196.32, 163.69, 159.10, 152.81, 146.78, 129.14, 124.21, 119.86, 112.22, 57.50, 50.37, 36.16, 32.35, 28.77, 27.45.



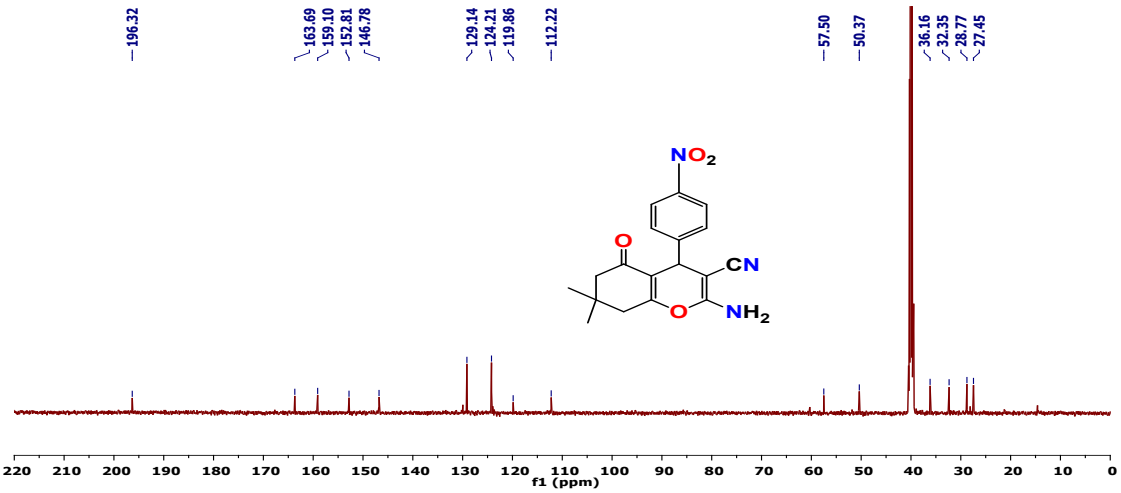
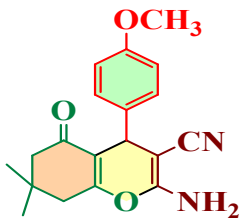
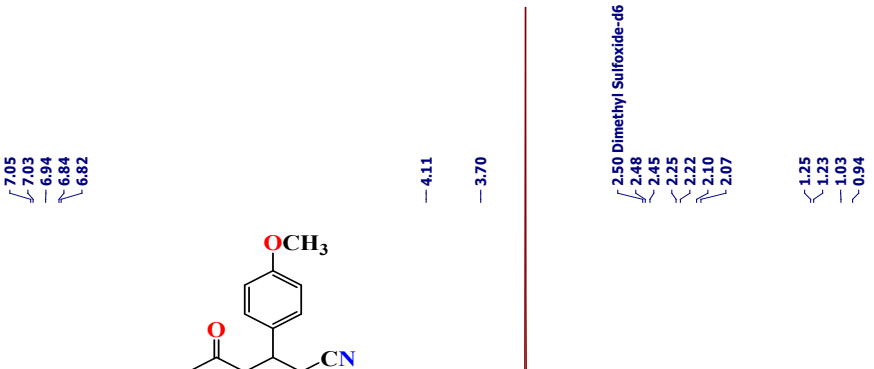


Figure S39. ^{13}C NMR Spectrum of 6c

6d. 2-amino-6,6,8,8-tetrahydro-7,7-dimethyl-5-oxo-4-(4-methoxyphenyl)-4H-chromene-3-carbonitrile[6]



M. F. $C_{19}H_{20}N_2O_3$ (324.38), Yield: (0.308 g, 95%). Off white powder, ^{1}H NMR (500 MHz, DMSO $d_6$ppm) δ: 7.04 (d, *J* = 9 Hz, 2H), 6.94 (s, 2H), 6.31 (d, *J* = 8 Hz, 2H), 4.11 (s, 1H), 3.70 (s, 3H), 2.46 (d, *J* = 17.5 Hz, 2H), 2.24 (d, *J* = 16 Hz, 1H), 2.08 (d, *J* = 16 Hz, 1H), 1.24 (t, *J* = 11 Hz, 1H), 103 (s, 3H), 0.94 (s, 3H) ^{13}C NMR (125 MHz, DMSO d_6, ppm) δ 196.22, 162.68, 158.94, 158.44, 137.36, 128.73, 120.32, 114.20, 113.51, 59.11, 55.52, 50.53 35.27, 32.30, 28.92, 27.29.



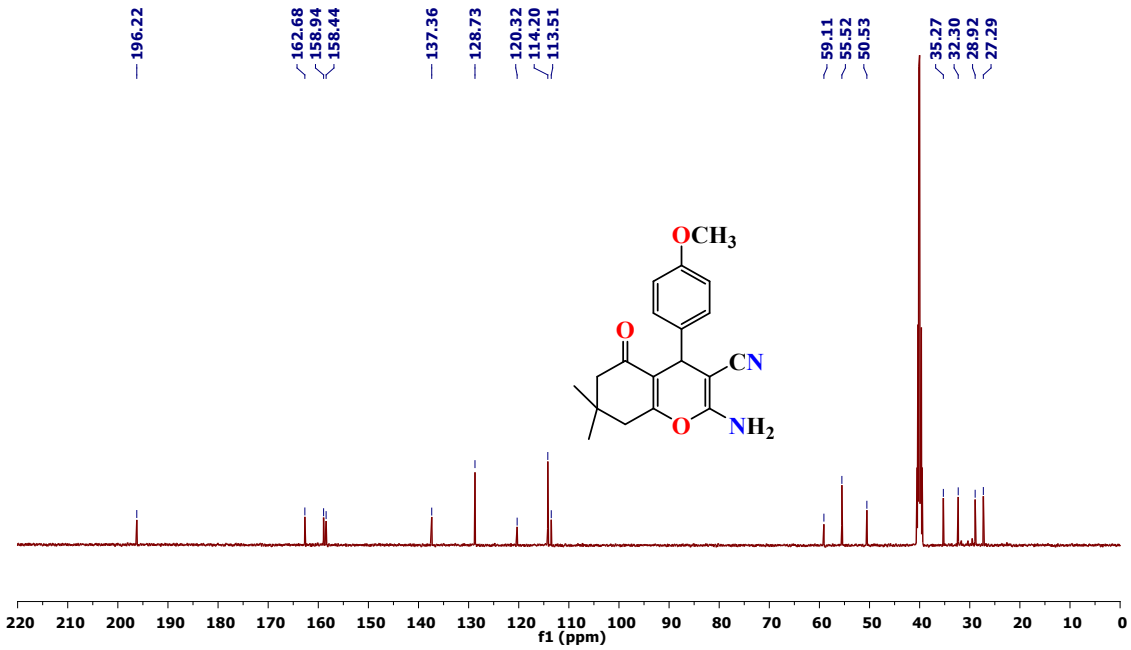
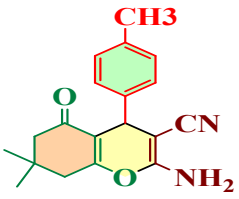


Figure S41. ^{13}C NMR Spectrum of 6d

6e. 2-amino-6,6,8,8-tetrahydro-7,7-dimethyl-5-oxo-4-(4-methylphenyl)-4H-chromene-3-carbonitrile [7]



M. F. $C_{19}H_{20}N_2O_2$ (308.38) Yield: (0.293 g, 95%). Off white powder, ^{1}H NMR (500 MHz, DMSO d_6, ppm) δ: 7.07 (d, *J* = 7.5 Hz, 2H), 7.00 (d, *J* =8 Hz, 2H), 6.94 (s, 2H), 4.11 (s, 1H), 2.49 (d, *J*=4 Hz, 2H), 2.23(s,3H), 2.25-2.22(d, *J*=16Hz, 1H) 2.08(d, *J*=16Hz, 1H) 1.02 (s, 3H), 0.94 (s, 3H); ^{13}C NMR (125 MHz, DMSO d_6, ppm) δ:196.19, 162.83, 158.96, 142.33, 136.15, 129.40 127.59, 120.27, 113.39, 58.99, 50.52, 35.70, 32.31, 28.93, 27.28, 21.10.

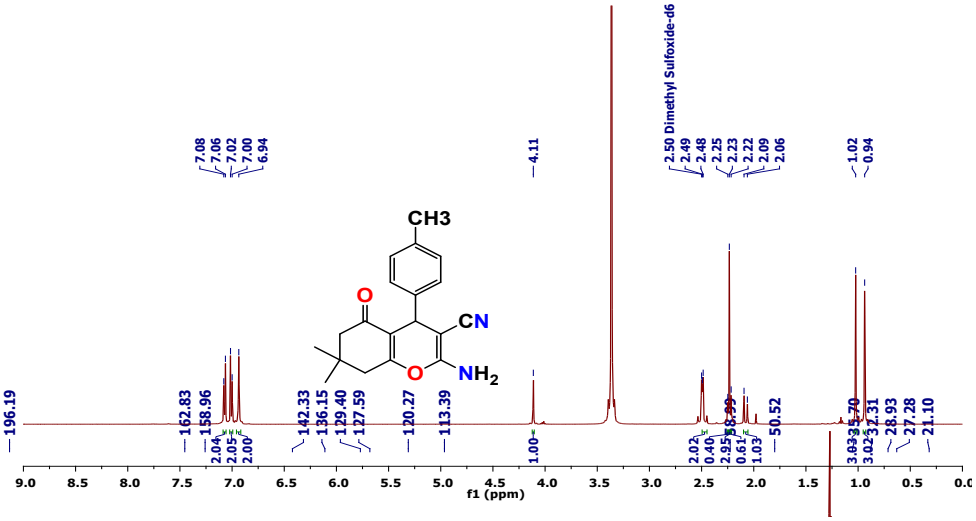


Figure S42. ^{1}H NMR Spectrum of 6e

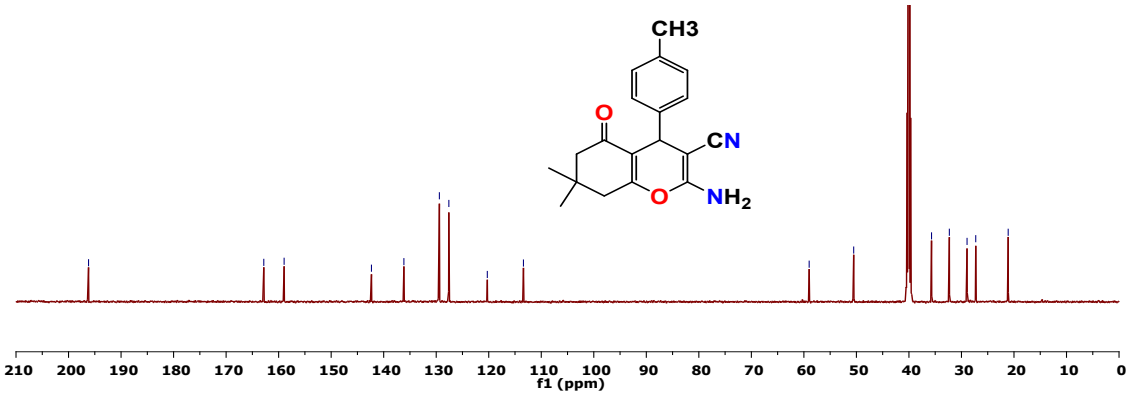
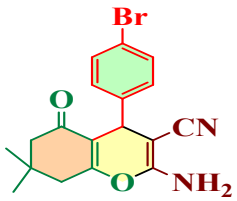
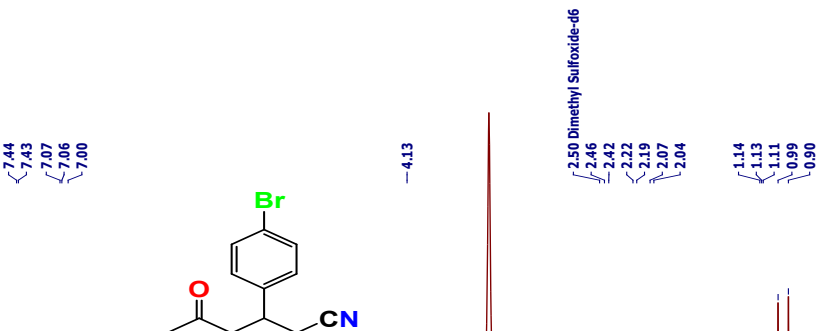


Figure S43. ^{13}C NMR Spectrum of 6e

6f. 2-amino-6,6,8,8-tetrahydro-7,7-dimethyl-5-oxo-4-(4-Chlorophenyl)-4H-chromene-3-carbonitrile[4]



M.F. $C_{18}H_{17}BrN_2O_2$ (372.25), Yield: (0.327 g, 88%). White powder, ^{1}H NMR (500 MHz, DMSO d6, ppm) δ7.43 (d, *J* = 8.0 Hz, 2H), 7.06 (d, *J* = 8.0 Hz, 2H), 7.00 (s, 2H), 4.13 (s, 1H), 2.44 (d, *J* = 18.5 Hz, 2H), 2.20 (d, *J* = 16 Hz, 1H), 2.05 (d, *J* = 16 Hz, 1H), 1.13 (t, *J* = 7 Hz, 1H), o.99 (s, 3H), 0.90 (s, 3H), ^{13}C NMR (125 MHz, DMSO d6, ppm) δ 196.27, 163.35, 163.20, 159.01, 144.69, 141.17, 131.73, 130.03, 120.13, 112.77, 58.23, 50.45, 35.71, 32.32, 28.82, 27.37.



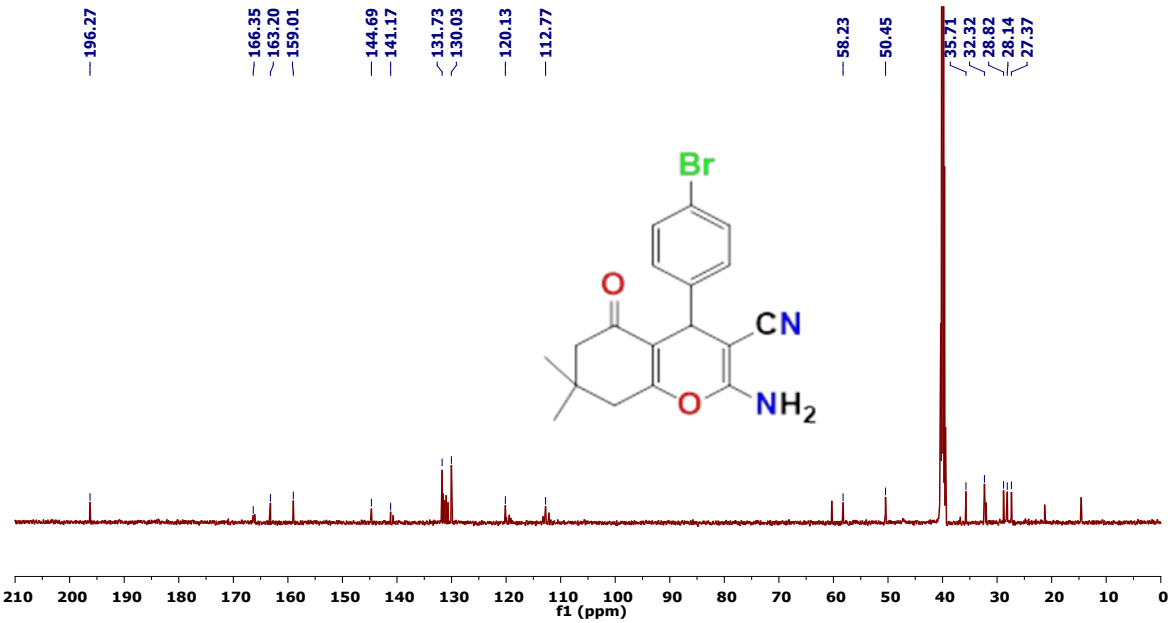
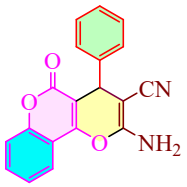


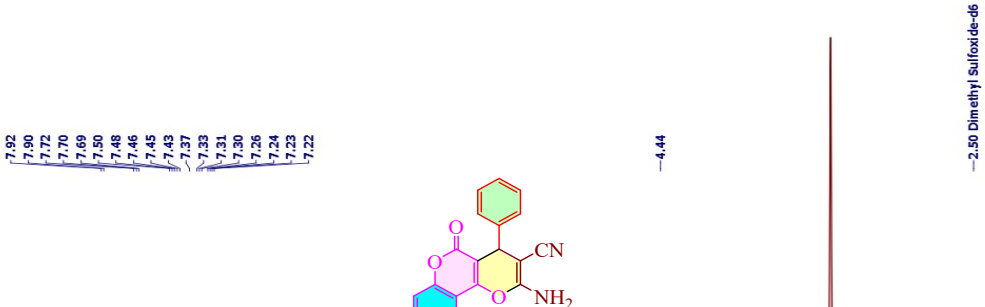
Figure S45. ^{13}C NMR Spectrum of 6f

Data S7- NMR (^{1}H and ^{13}C) spectra of 2- Amino-4chromeneDerivatives of 4-Hydroxycoumarin

7a. 2-amino-5-hydroxy-4-phenyl-4H,5H-pyrano[3,2-c]chromene-3-carbonitrile[7]



M. F. $C_{19}H_{12}N_2O_3$ (316.32), Yield: (0.284g, 90%), white powder. ^{1}H NMR (500 MHz, DMSO-d6, ppm) δ: 7.91 (d, *J* = 8 Hz, H), 7.70 (t, *J* = 8.5Hz, H), 7.48(t, *J* = 7.5Hz, H), 7.44(d, *J* = 8.5Hz, H), 7.37 (s, 2H), 7.31(t, *J* = 8Hz, 2H), 7.25(t, *J* = 7.25Hz, 2H) 4.44 (s, 1H), ^{13}C NMR (125 MHz, DMSO-d6, ppm) δ: 160.05, 158.90, 158.48, 153.65, 152.64, 135.89, 133.37, 129.25, 125.16, 123.01, 119.74, 117.05, 114.40, 113.54, 104.84, 58.84, 55.59, 36.72, 31.45



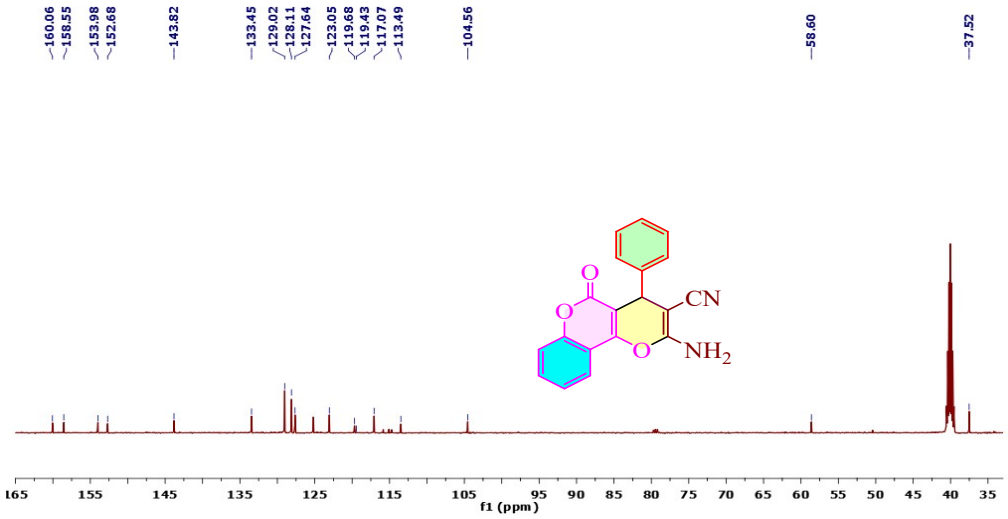
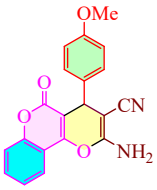


Figure S47. ^{13}C NMR Spectrum of 7a

7b.2-amino-4-(4-methoxyphenyl)-5-oxo-4H,5H-pyrano[3,2-c]chromene-3-carbonitrile[6]:



M. F. $C_{20}H_{14}N_2O_4$ (346.34), Yield: (0.325 g,94%), white powder. ^{1}H NMR (500 MHz, DMSO-d6, ppm) δ: 7.86 (d, *J* = 7.5 Hz, H), 7.66 (t, *J* = 8Hz, H), 7.44(t, *J* = 8Hz, H), 7.40(d, *J* = 8.5Hz, H), 7.28 (s, 2H), 7.13(d, *J* = 8Hz, H), 6.82(d, *J* = 8Hz, 2H) 4.39 (s, 1H), 3.72(s, 3H); ^{13}C NMR (125 MHz, DMSO-d6, ppm) δ: 160.05, 158.90, 158.48, 153.65, 152.64, 135.89, 133.37, 129.25, 125.16, 123.01, 119.74, 117.05, 114.40, 113.54, 104.84, 58.84, 55.59, 36.72,31.45

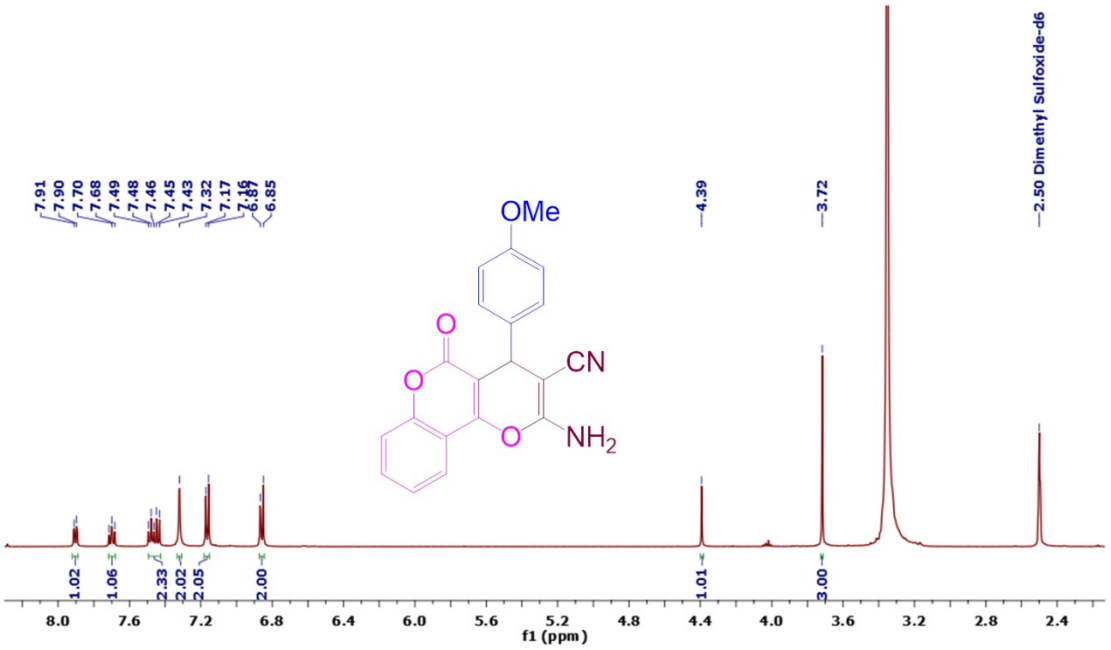


Figure S48. ^{1}H NMR Spectrum of 7b

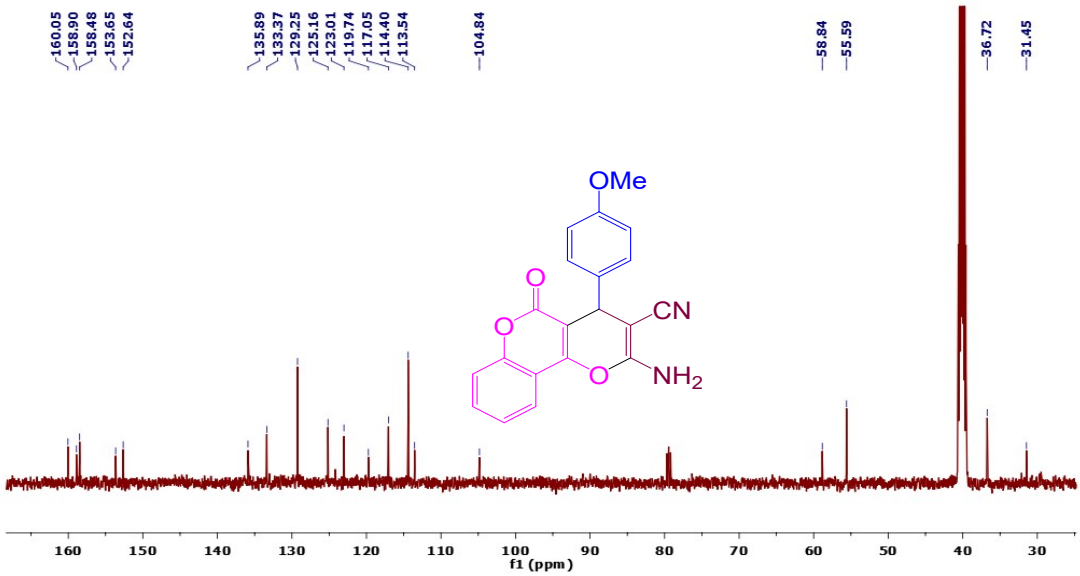
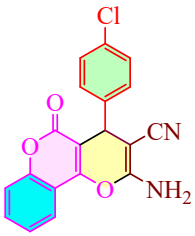


Figure S49. ^{13}C NMR Spectrum of 7b

7c. 2-amino-4-(4-chlorophenyl)-5-oxo-4H,5H-pyrano[3,2-c]chromene-3-carbonitrilearbonitrile[6]



M. F. $C_{19}H_{11}N_2O_3Cl$ (350.76), Yield: (0.308 g, 88%), reddish brown powder. . ^{1}H NMR (500 MHz, DMSO-d6, ppm) δ: 7.91 (d, *J* = 8 Hz, H), 7.71 (t, J = 8.5Hz, H), 7.48(t, *J* = 7.5Hz, H),

7.45(d, *J* = 8.5Hz, H), 7.41 (s, 2H), 7.36(d, *J* = 8Hz, 2H), 7.29(d, *J* = 9Hz, 2H) 4.48 (s, 1H), ^{13}C NMR (125 MHz, DMSO-d6, ppm) δ: 160.05, 158.53, 154.10, 152.73, 133.53, 132.26, 130.11, 128.59, 125.20, 123.09, 119.52, 117.09, 113.47, 104.05, 58.14, 36.97, 29.50

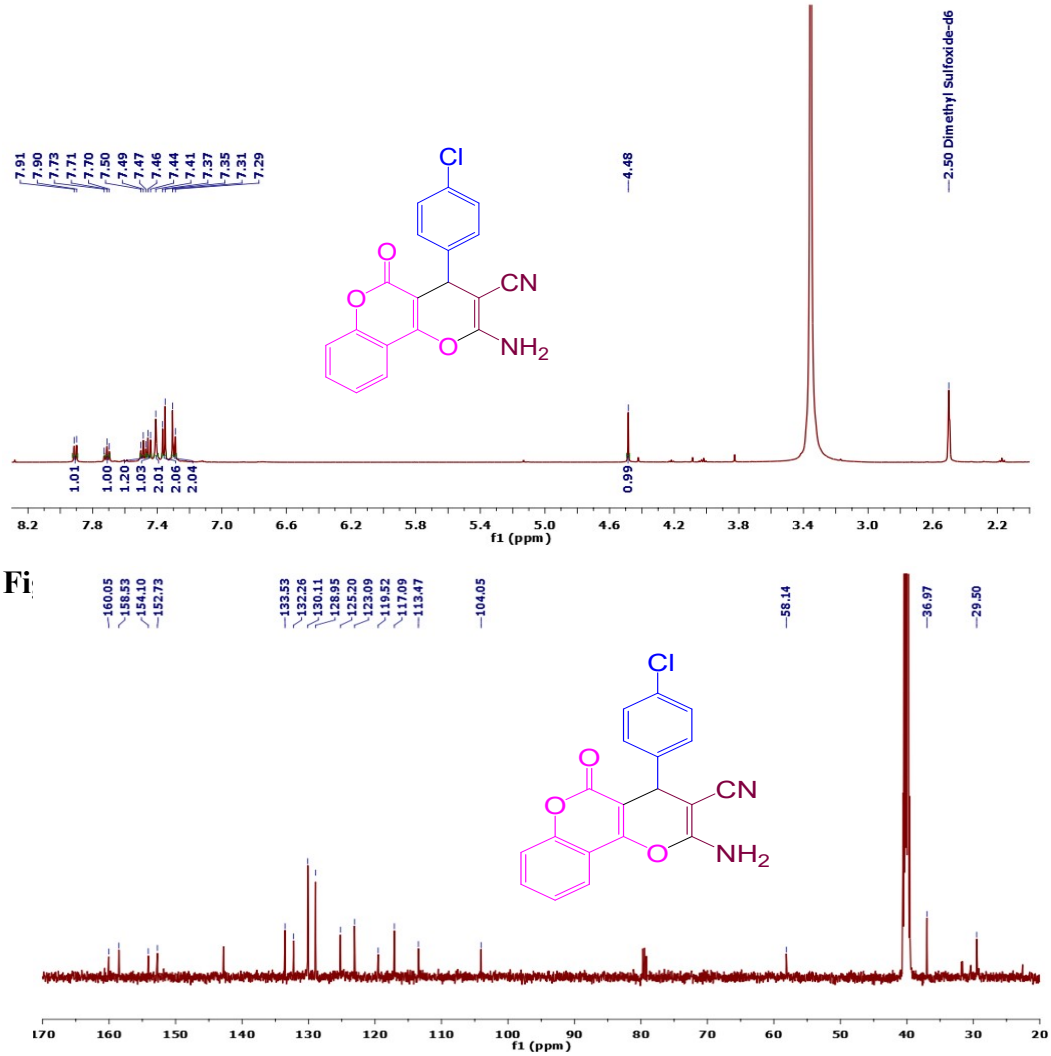
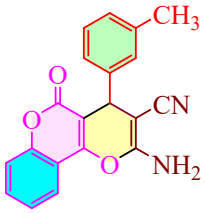


Figure S51. ^{1}H NMR Spectrum of 7c

7d. 2-amino-5-oxo-4-(m-tolyl)-4H,5H-pyrano[3,2-c]chromene-3-carbonitrile[7]



M. F. $C_{20}H_{14}N_2O_3$ (330.34), Yield: (0.304 g, 92%), reddish brown powder. ^{1}H NMR (500 MHz, DMSO-d6, ppm) δ: 7.91 (d, *J* = 10 Hz, H), 7.71 (t, *J* = 7.5Hz, H), 7.49(t, *J* = 7.5Hz, H), 7.45(d, *J* = 8Hz, H), 7.36 (s, 2H), 7.19(d, *J* = 8Hz, H), 7.04(d, *J* = 10Hz, 3H) 4.40 (s, 1H), 2.27(s, 3H); ^{13}C NMR (125 MHz, DMSO-d6, ppm) δ: 160.06,158.50, 153.94, 152.69,

143.82, 138.18, 133.91, 128.53, 128.37, 125.35, 125.19, 123.02, 119.72, 117.10, 113.55, 104.61, 58.70, 37.47, 21.54

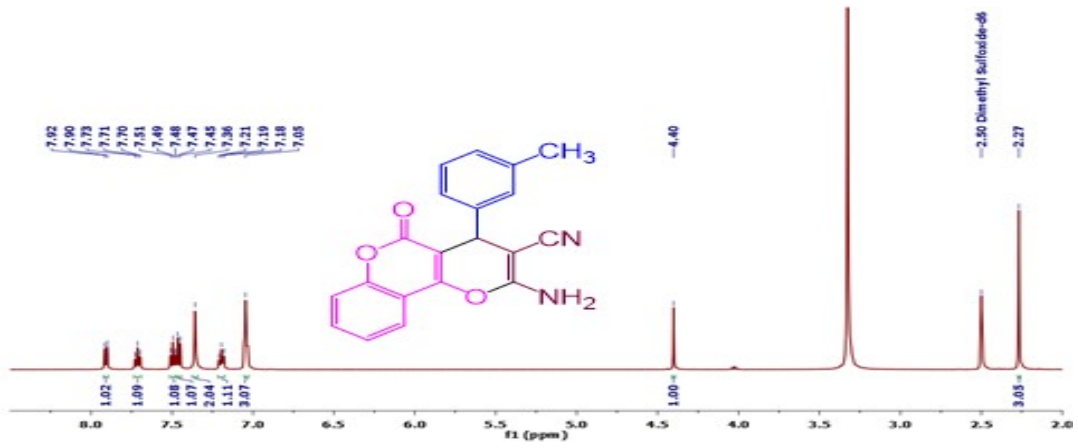


Figure S52. ^{1}H NMR Spectrum of 7d

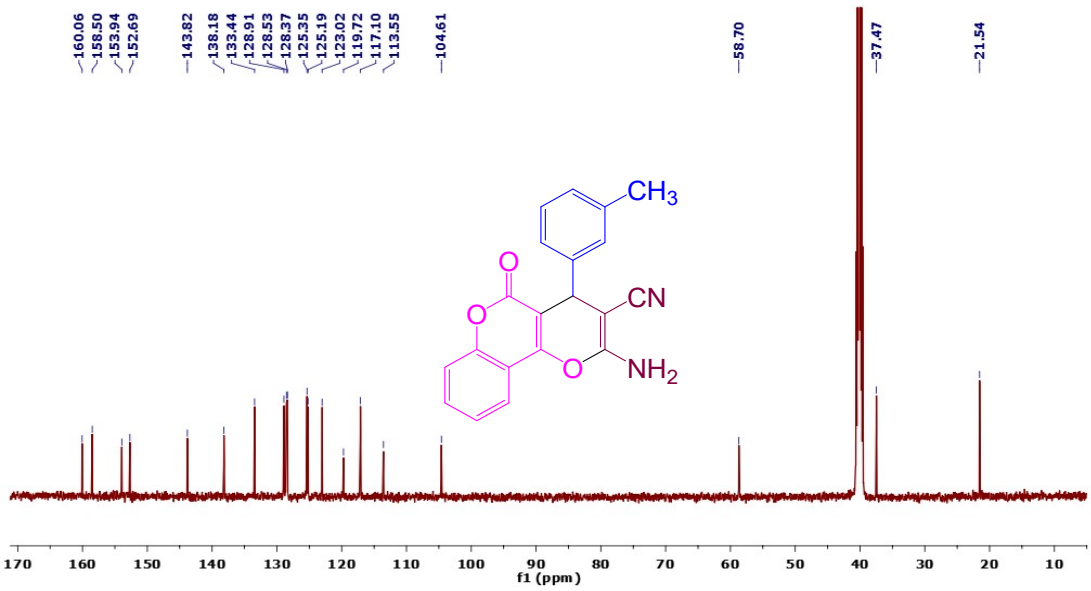
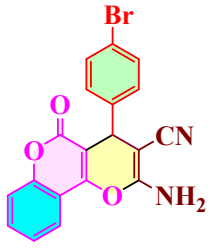


Figure S53. ^{13}C NMR Spectrum of 7d

7e. 2-amino-4-(4-bromophenyl)-5-oxo-4H,5H-pyrano[3,2-c]chromene-3-carbonitrile[6]



M. F. $C_{19}H_{11}N_2O_3Br$ (395.21), Yield: (0.340g, 86%), reddish brown powder. ^1H NMR (500 MHz, DMSO-d6, ppm) δ: 7.90 (d, *J* = 8 Hz, H), 7.72 (t, *J* = 7.5Hz, H), 7.49(t, *J* = 7.5Hz, 2H), 7.46(d, *J* = 8.5Hz, 2H), 7.43 (s, 2H), 7.24(d, *J* = 8Hz, 2H), 7.29(d, *J* = 8Hz, 2H) 4.47 (s, 1H), ^{13}C NMR (125 MHz, DMSO-d6, ppm) δ: 160.05, 158.52, 154.11, 152.74, 143.27, 133.54, 131.86, 130.51, 125.21, 123.06, 120.75, 119.52, 117.11, 113.49, 104.00, 58.09, 37.06, 29.50

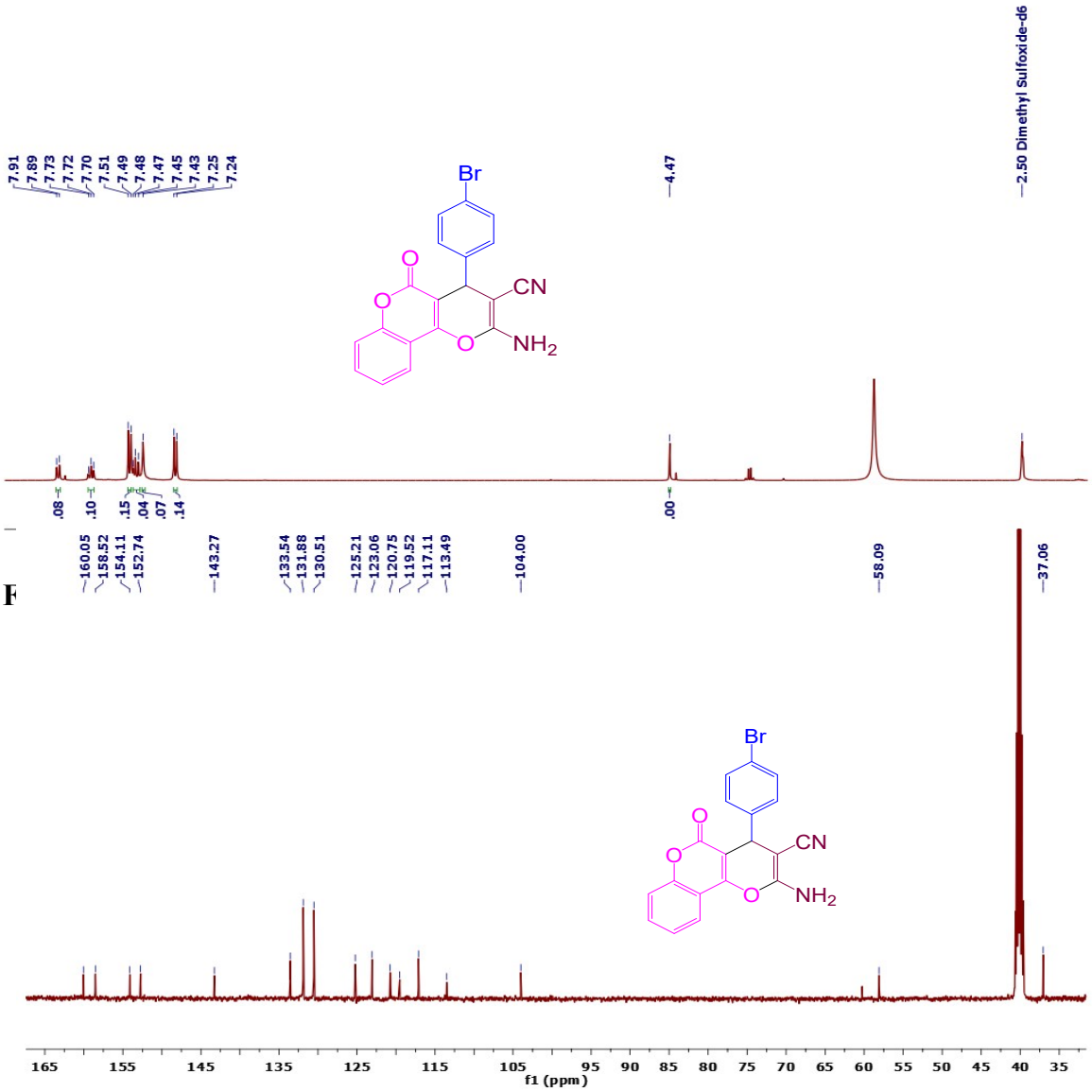
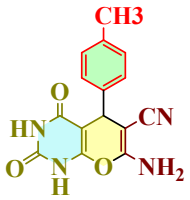


Figure S55. ^{13}C NMR Spectrum of 7e

Data S8- NMR (^1H and ^{13}C) spectra of 2- Amino4-chromeneDerivatives of of Burbuteric acidderivative

8a. 7-amino-2,4-dioxo-5-(p-tolyl)-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile [6]



M. F. $C_{15}H_{12}N_4O_3$ (296.29), Yield: (0.284g, 96%), white powder, ^{1}H NMR (500 MHz, DMSO-d6, ppm) δ: 12.03 (s, 1H), 11.11(s, 1H), 7.08(d, *J* = 2Hz, 4H), 7.06(s, 2H), 4.17(s, 1H),2.25(s, 3H); ^{13}C NMR (125 MHz, DMSO-d6, ppm) δ: 168.25, 162.95, 158.10, 152.17, 141.73, 136.31, 129.35, 127.70, 89.17, 59.63, 35.81; 21.13;

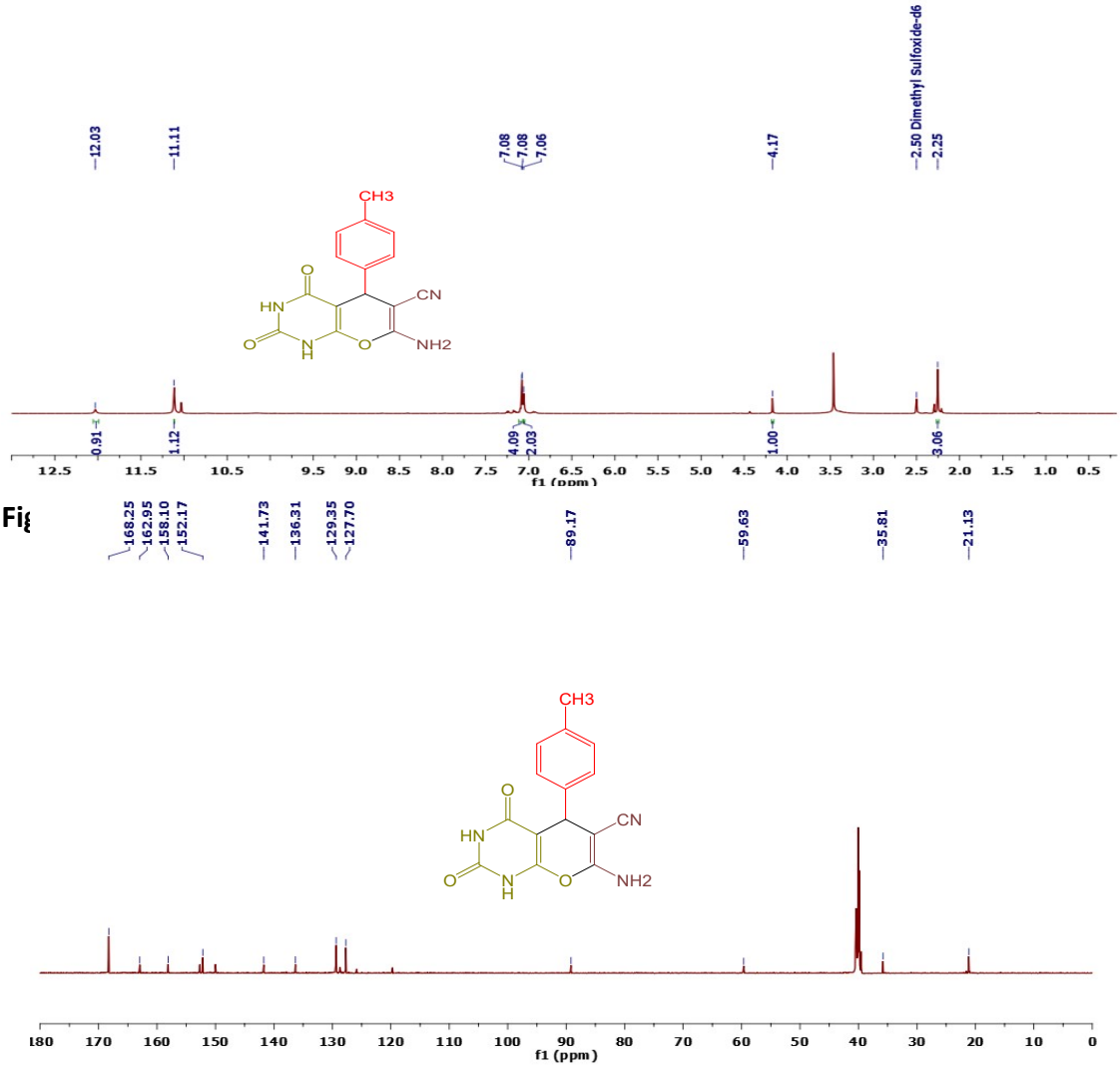
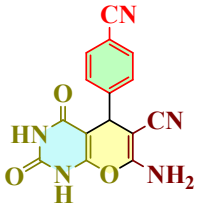


Figure S57. ^{13}C NMR Spectrum of 8a

8b.7-amino-5-(4-cyanophenyl)-2,4-dioxo-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile[7]



M. F. $C_{15}H_9N_5O_3$ (307.27), Yield: (0.251 g, 82%), white brown powder. ^{1}H NMR (500 MHz, DMSO-d6, ppm) δ: 12.11 (s, 1H), 11.09(s, 1H), 7.72 (t, *J* = 8 Hz, 2H), 7.39(d, *J* = 8Hz, 2H), 7.22(s, 2H), 4.34(s, 1H); ^{13}C NMR (125 MHz, DMSO-d6, ppm) δ: 162.99, 158.28, 153.14, 150.18, 150.00, 132.82, 129.09, 119.35, 110.10, 87.98, 58.23, 36.36;

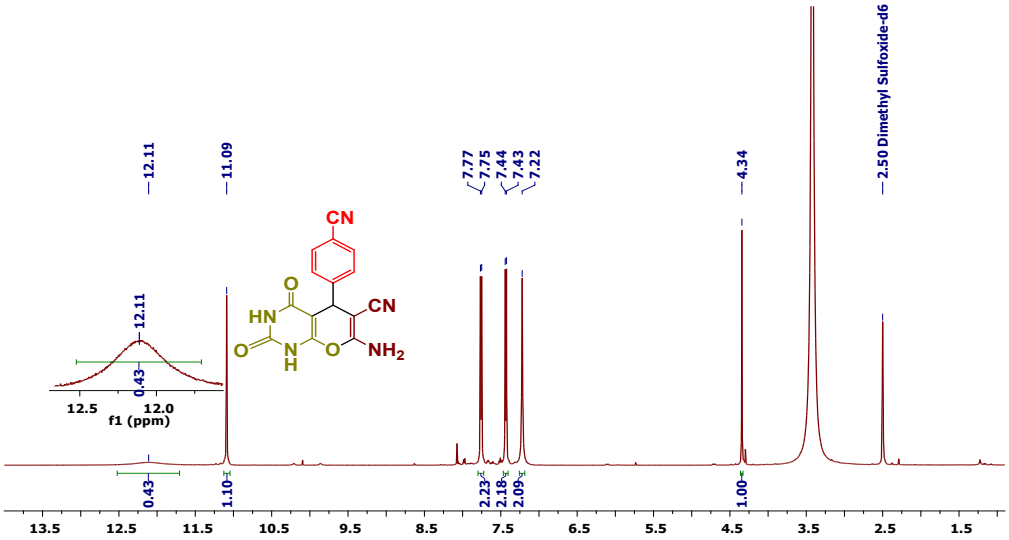


Figure S58. ^{13}H NMR Spectrum of 8b

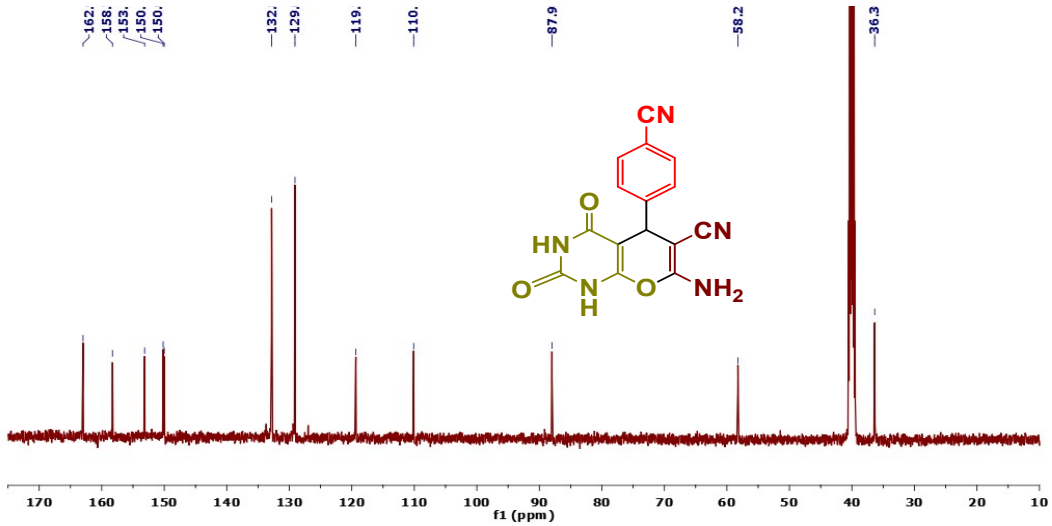
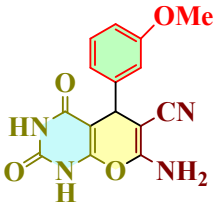


Figure S59. ^{13}C NMR Spectrum of 8b

8c. 7-amino-5-(3-methoxyphenyl)-2,4-dioxo-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile [6]



M. F. $C_{15}H_{12}N_4O_4$ (312.29), Yield: (0.296 g, 95%), white powder. ^{1}H NMR (500 MHz, DMSO-d6, ppm) δ: 12.01 (s, 1H), 11.06 (s, 1H), 7.21 (t, *J* = 8 Hz, H), 7.06 (s, 2H), 6.75(d, *J* = 10Hz, H), 6.72(d, *J* = 7.5Hz, H), 6.68(s, H), 4.20 (s, 1H), 3.72(s, 3H); ^{13}C NMR (125 MHz, DMSO-d6, ppm) δ: 162.98,159.70, 158.22, 152.82, 150.00, 146.22, 129.94, 119.92, 119.66, 114.00, 112.20, 88.92, 59.36, 55. 49, 36.10;

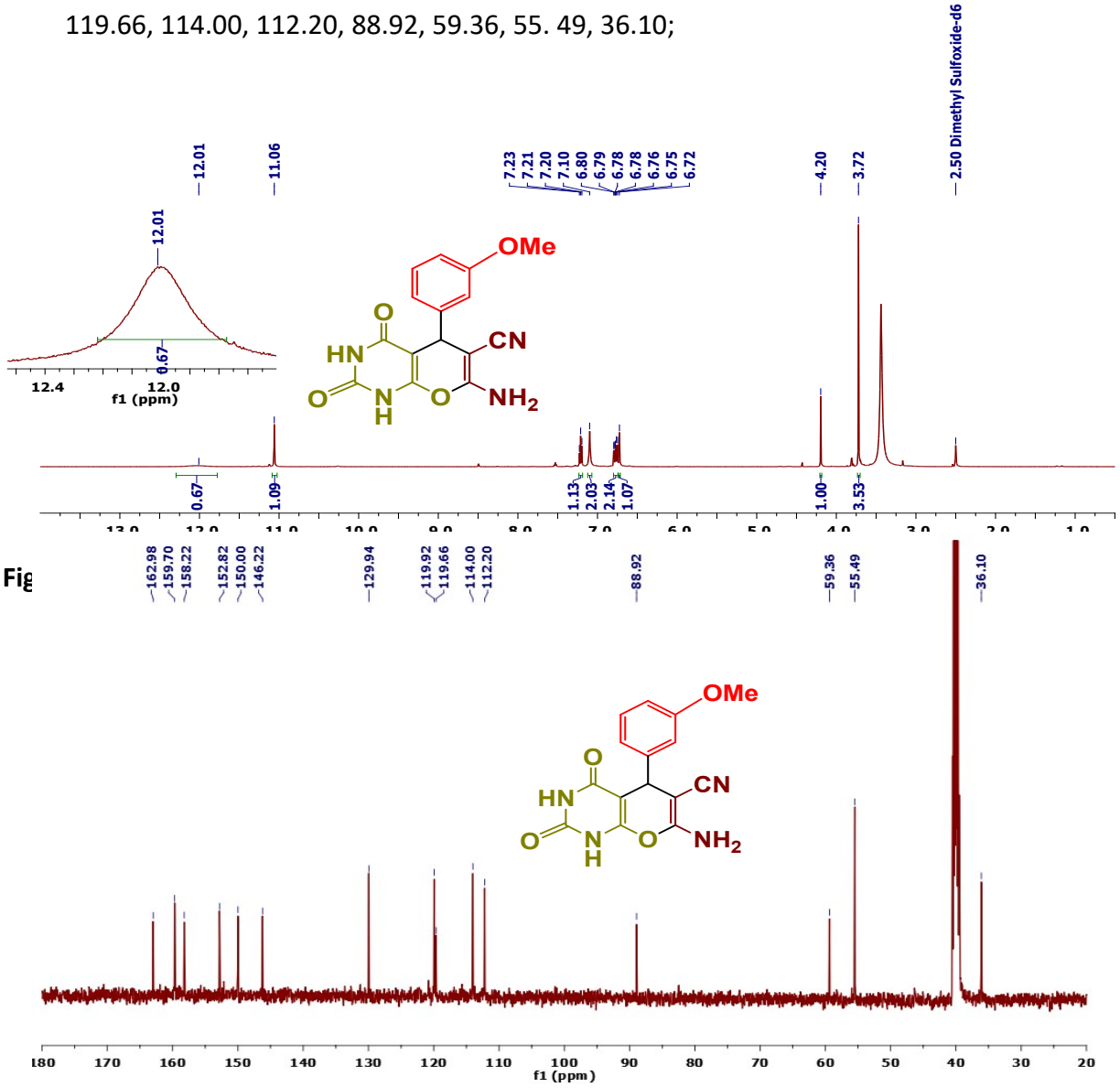
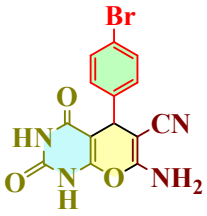


Figure S61. ^{13}C NMR Spectrum of 8c

8d. 7-amino-5-(4-bromophenyl)-2,4-dioxo-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile[6]



M. F. $C_{14}H_9BrN_4O_3$ (361.16), Yield: (0.324 g, 89%), off white powder. ^{1}H NMR (500 MHz, DMSO-d6, ppm) δ: 12.01 (s, 1H), 11.07 (s, 1H), 7.43 (t, *J* = 8 Hz, 2H), 7.14(d, *J* = 9Hz, 2H), 7.10(s, 2H), 4.23(s, 1H), ^{13}C NMR (125 MHz, DMSO-d6, ppm) δ: 162.97, 158.16, 152.87,

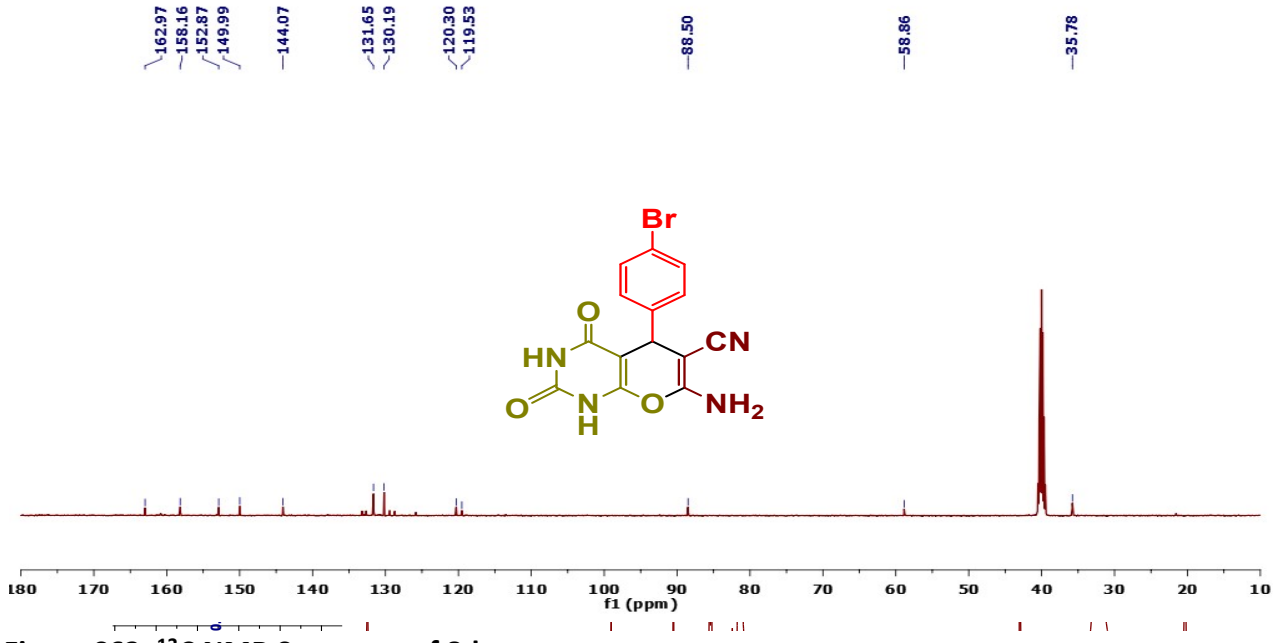


Figure S63. ^{13}C NMR Spectrum of 8d

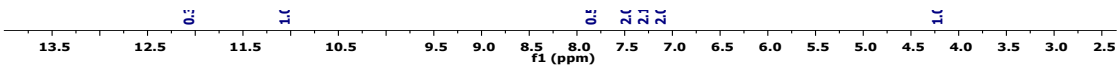
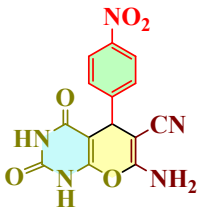


Figure S62. ^{1}H NMR Spectrum of 8d

8e. 7-amino-5-(4-nitrophenyl)-2,4-dioxo-1,3,4,5-tetrahydro-2H-pyrano[2,3-d]pyrimidine-6-carbonitrile[7]



M. F. $C_{14}H_9N_5O_5$ (327.26), Yield: (0.261g, 80%), light brown powder. ^{1}H NMR (500 MHz, DMSO-d6, ppm) δ: 12.15 (s, 1H), 11.10(s, 1H), 8.16 (s, 2H), 7.53(s, 2H), 7.25(s, 2H), 4.42(s, 1H); ^{13}C NMR (125 MHz, DMSO-d6, ppm) δ: 167.73, 163.99, 157.89, 156.97, 154.76, 153.14, 151.70, 134.09, 128.79,124..07, 92.72, 62.82, 40.94;

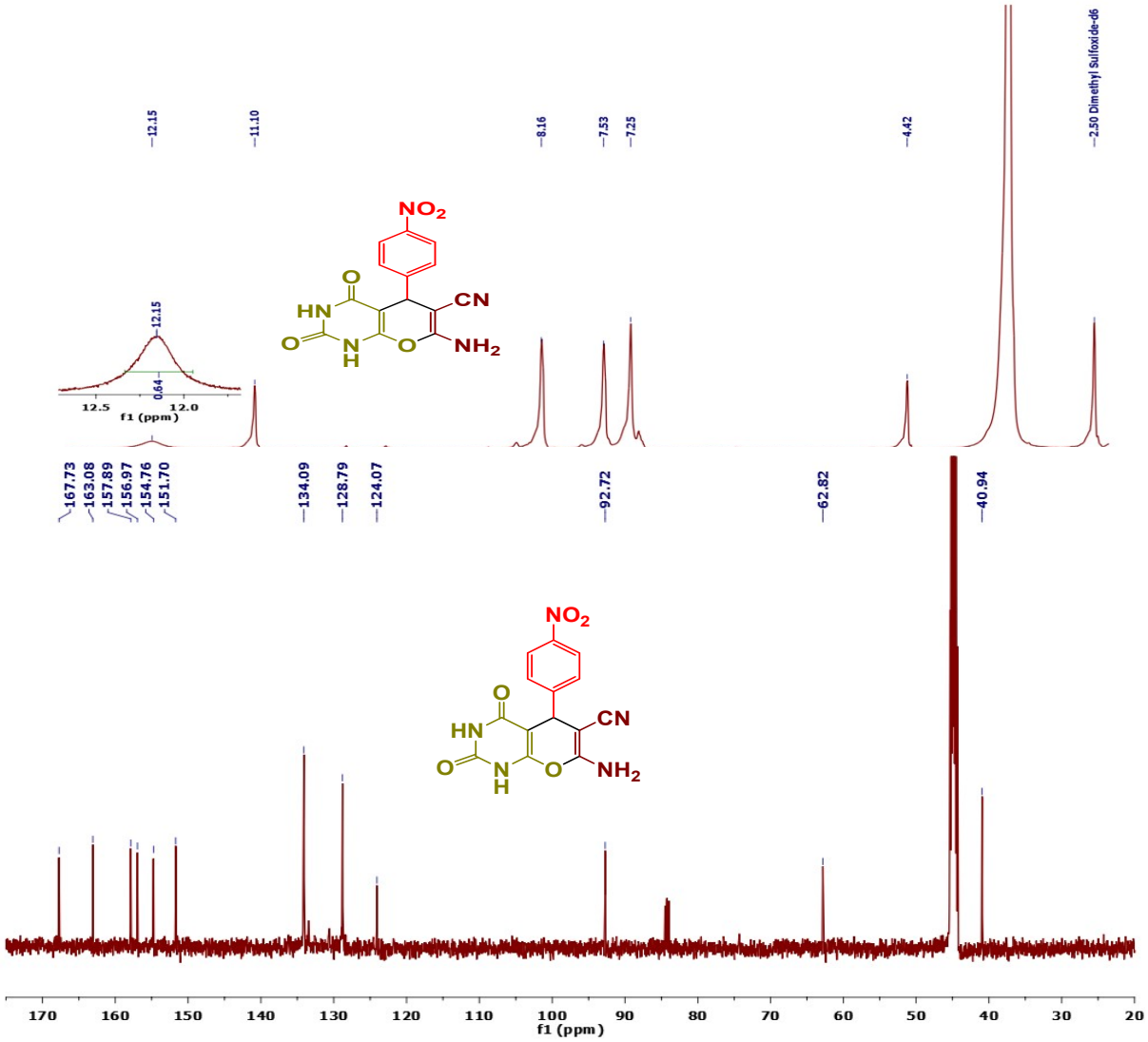
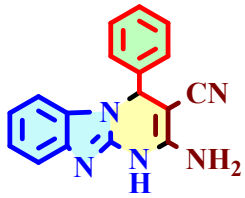


Figure S65. ^{13}C NMR Spectrum of 8e

DataS9- NMR (^{1}H and ^{13}C) spectra of Imidazopyrimidine derivatives:

9a. 2-amino-4-phenyl-1,4-dihydrobenzo[4,5]imidazo[1,2-a]pyrimidine-3-carbonitrile[4]



M. F. $C_{17}H_{13}N_5$ (287.31), Yield: (0.253 g, 88%). Light yellow powder. 1H NMR (500 MHz, DMSO-d6, ppm) δ 9.03 (s, 1H, NH), 7.68 (d, *J* = 8 Hz, 1H), 7.38 – 7.35 (m, 2H), 7.31 (dd, *J* = 7, 8 Hz, 4H), 7.16 (t, *J* = 8Hz, 1H), 7.06(t, *J* = 7 Hz, 1H), 6.87 (s, 2H), 5.28 (s, 1H); 13C NMR (125 MHz, DMSO-d6, ppm) δ 156.37, 154.13, 147.86, 134.011, 134.03, 133.29, 131.35, 128.92, 125.73, 124.20, 120.78, 117.98, 67.45, 58.60.

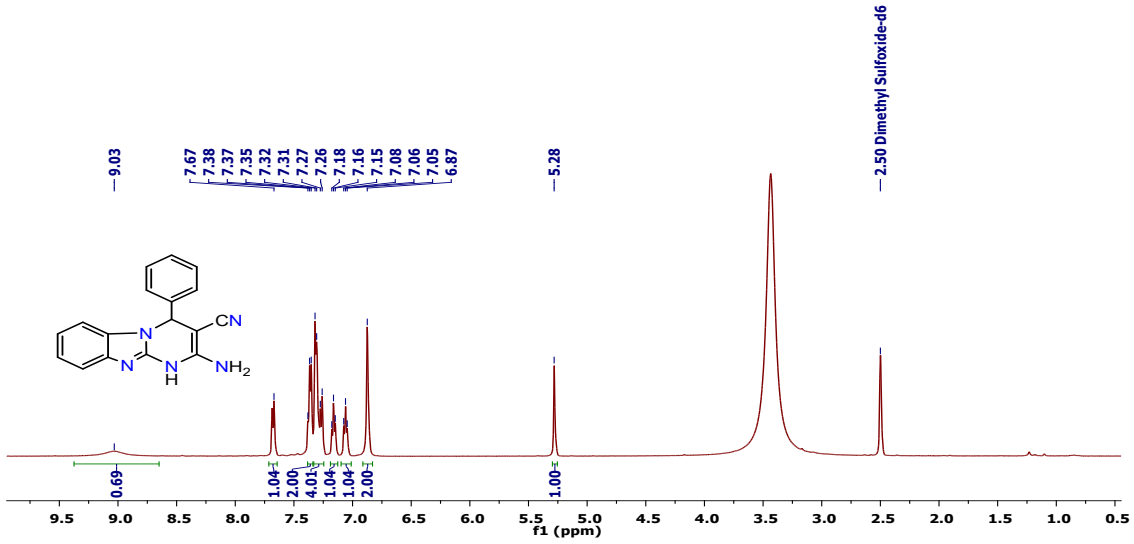


Figure S66. ^{1}H NMR Spectrum of 9a

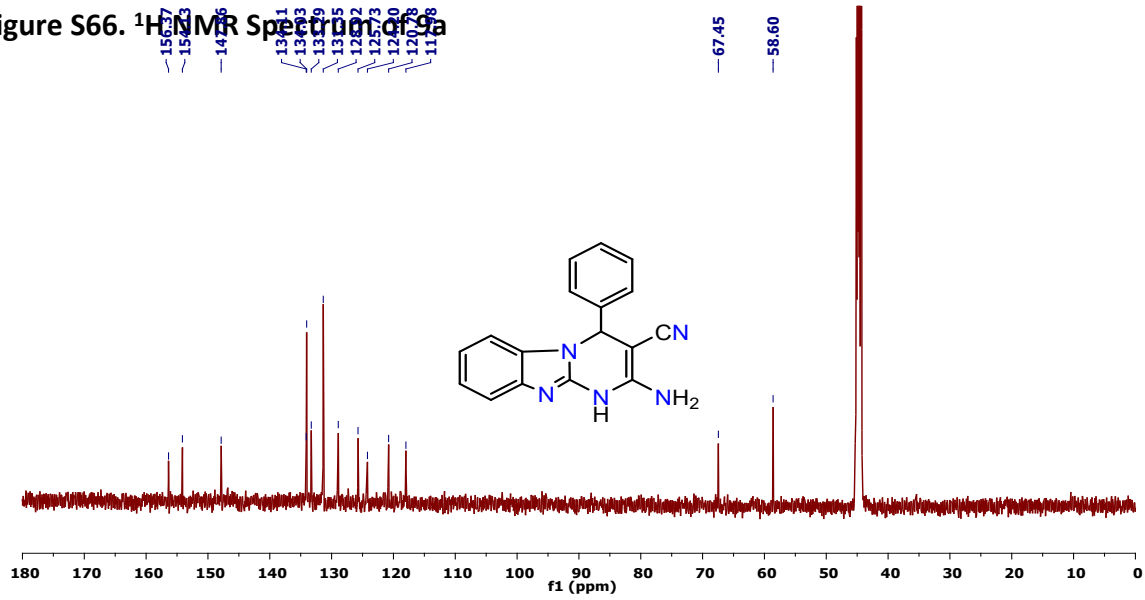
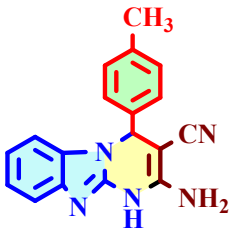


Figure S67. ^{13}C NMR Spectrum of 9a

9b. 2-amino-4-(4-methylphenyl)-1,4-dihydrobenzo[4,5]imidazo[1,2-a]pyrimidine-3 carbonitrile[5]



M. F. $C_{18}H_{15}N_5$ (301.13), Yield: (0.280g, 93%). Off white powder. 1H NMR (500 MHz, DMSOd6, ppm) δ 8.83 (s, 1H, NH), 7.65 (d, *J* = 8 Hz, 1H), 7.24 (d, *J* = 7.5 Hz, 1H), 7.19 (d, *J* = 8 Hz, 4H), 7.15 (d, *J* = 7.5 Hz, 1H),), 7.13 (d, *J* = 7.5 Hz, 1H) 7.03(t, *J* = 7.5 Hz, 1H), 6.83 (s, 2H), 5.20 (s, 1H), 2.26 (s, 3H); 13C NMR (125 MHz, DMSO-d6, ppm) δ 152.06, 149.50, 140.31, 137.70, 129.73, 126.48,123.95, 120.58, 119.60, 116.35, 113.01, 62.76, 53.58, 21.16.

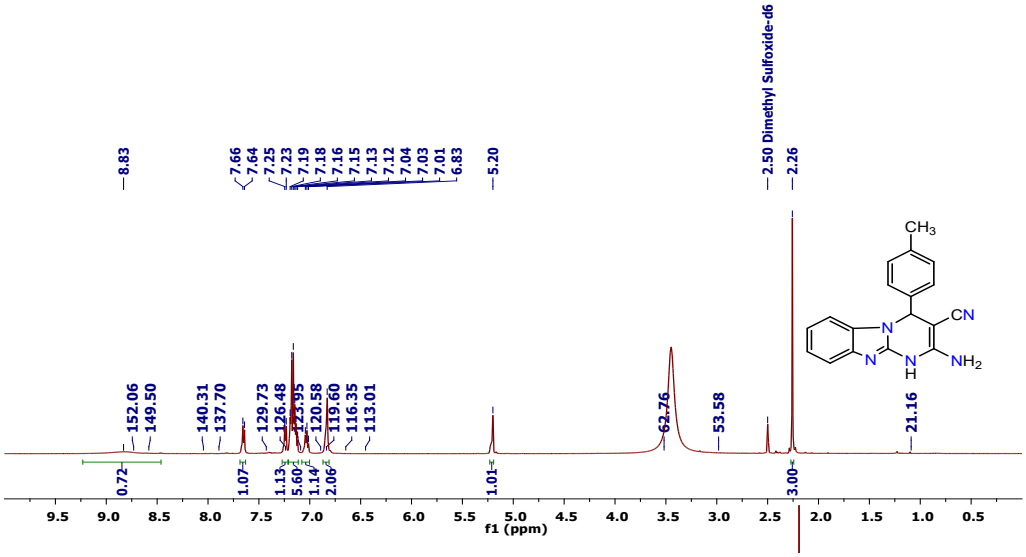


Figure S68. ^{1}H NMR Spectrum of 9b

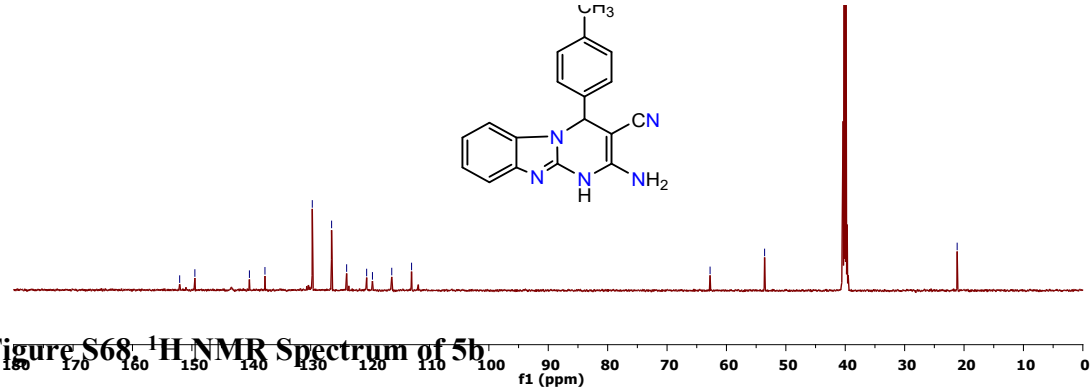
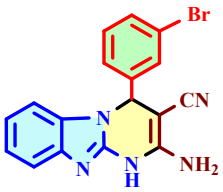


Figure S68. ^{1}H NMR Spectrum of 5b

Figure S69. ^{13}C NMR Spectrum of 9b

9c. 2-Amino-4-(3-bromophenyl)-1,4-dihydrobenzo[4,5]imidazolo[1,2-a]pyrimidine-3-carbonitrile[5]



M. F. $C_{17}H_{12}BrN_5$ (366.21), Yield: (0.311 g, 85%). yellow powder. ^{1}H NMR (500 MHz, DMSO-D_6) δ 8.83 (s, 1H), 7.66 (d, J = 8.0 Hz, 1H), 7.52 (s, 2H),7.50(d, J=8.5Hz,1H) 7.35 – 7.29 (m, 2H), 7.25 (d, J = 8.0 Hz, 1H), 7.14 (t, J = 8.0 Hz, 1H), 7.04 (t, J = 7.5 Hz, 1H), 6.93 (s, 2H), 5.31 (s, 1H):^{13}C NMR (126 MHz, DMSO-d_6) δ 151.77, 149.78, 145.96, 143.27, 131.56, 131.31, 129.54, 125.63, 124.07, 122.39, 120.75, 119.46, 116.47, 113.09, , 61.72, 53.10.

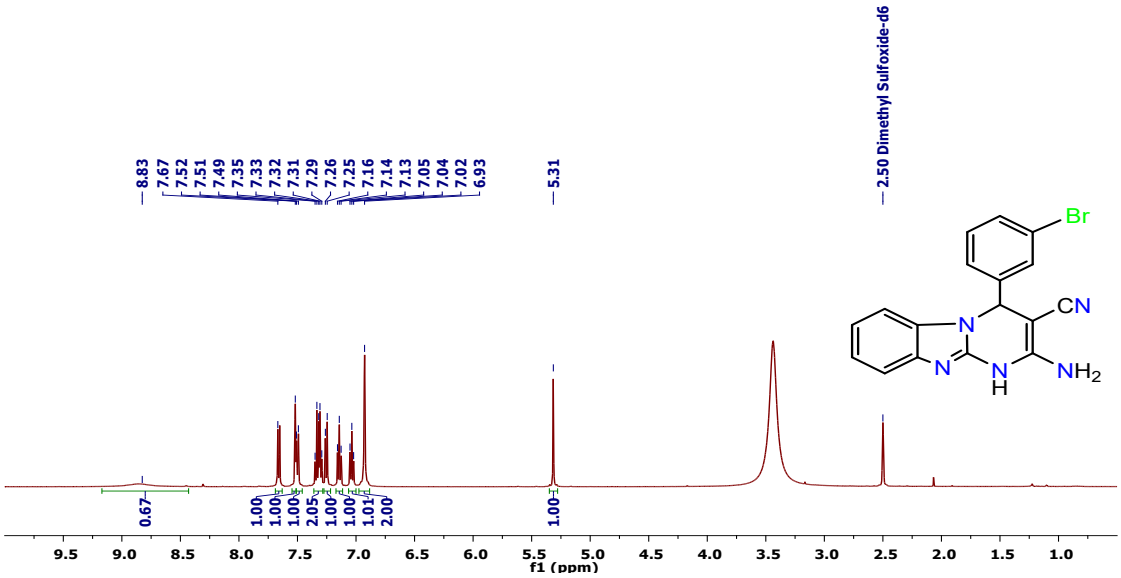


Figure S70. ^{1}H NMR Spectrum of 9c

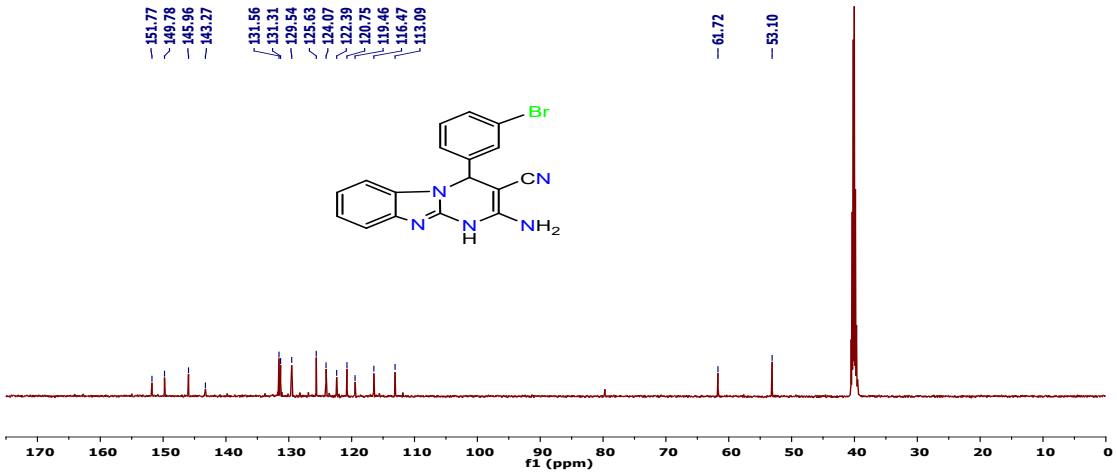
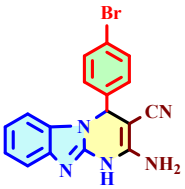


Figure S71. ^{13}C NMR Spectrum of 9c

9d. 2-Amino-4-(4-Brormophenyl)-1,4-dihydrobenzo[4,5]imidazolo[1,2-a]pyrimidine-3-carbonitrile[4]



M. F. $C_{17}H_{12}N_5Br$ (366.21), Yield: (0.314 g, 86%). yellow powder. 1H NMR (500 MHz, DMSO-d6, ppm): δ 8.60 (s, 1H, NH), 7.63 (d, *J* = 8.5 Hz, 1H), 7.55 (d, *J* = 7.5 Hz, 2H), 7.24 (dd, *J* = 8, 6.5 Hz, 3H), 7.12 (t, J = 7.5 Hz, 1H), 7.00 (d, *J* = 7.5 Hz, 1H), 6.86 (s, 2H), 5.24 (s, 1H); 13C NMR (125 Hz, DMSO-d6, ppm): δ 152.07, 149.76, 144.00, 142.73, 142.68, 132.15, 129.76, 128.80, 123.94, 121.51, 120.51, 119.54, 116.64, 112.98, 61.95, 53.17.

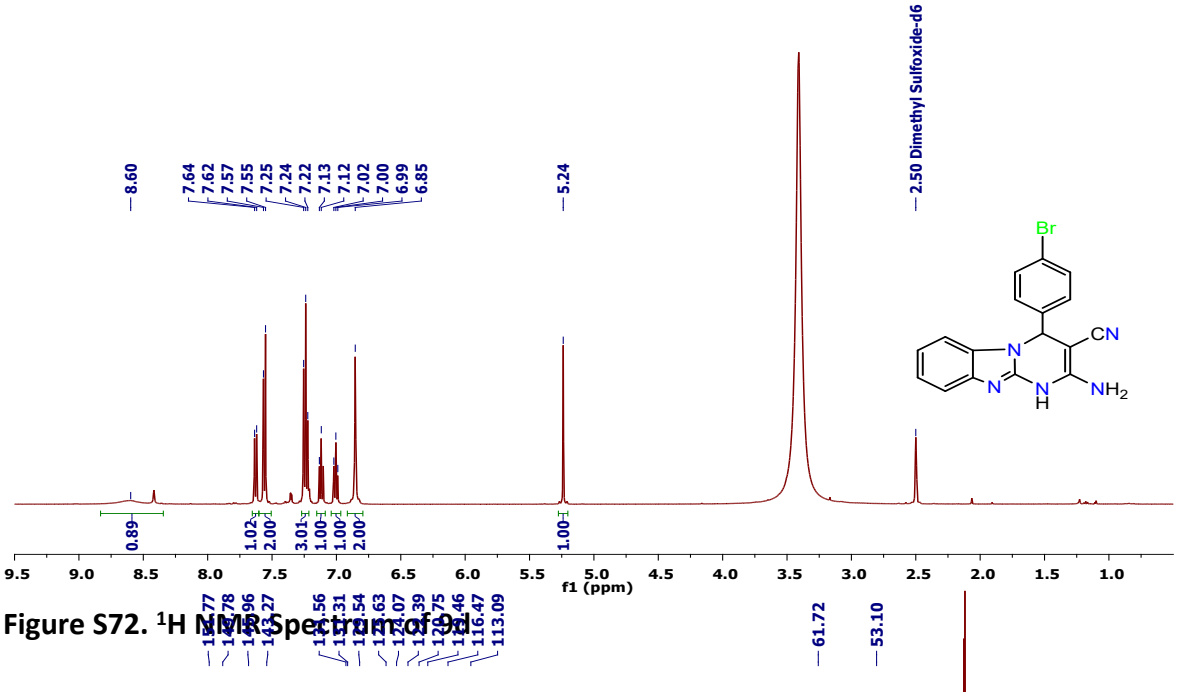
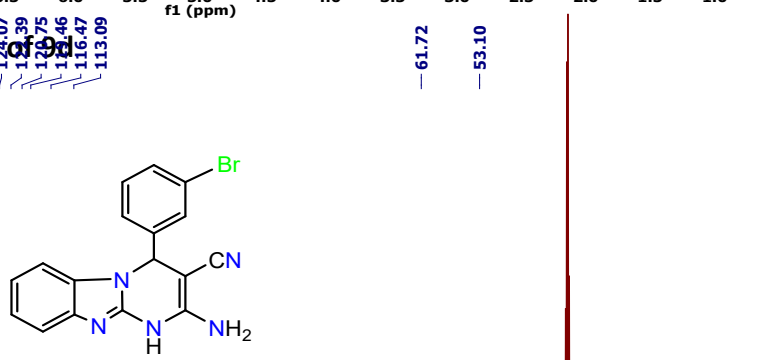
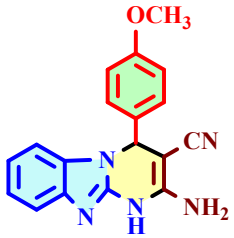


Figure S72. ^{1}H NMR Spectrum of 9d



9e. 2-amino-4-(4-methoxyphenyl)-1,4-dihydrobenzo[4,5]imidazo[1,2-a]pyrimidine-3 carbonitrile[4]



M. F. C18H15N5O (317.34), Yield: (0.288g,91%). Light yellow powder. 1H NMR (500 MHz,DMSO-d6, ppm) δ 8.53 (s, 1H, NH), 7.63 (d, *J* = 7.5 Hz, 1H), 7.20 (t, *J* = 8.0 Hz, 3H), 7.11 (t, *J* = 7.5 Hz, 1H), 6.99 (t, *J* = 8.0 Hz, 1H), 6.90 (d, *J* = 9.0 Hz, 2H), 6.79 (s, 2H), 5.15 (s, 1H), 3.71 (s, 3H); 13C NMR (125 MHz, DMSO-d6, ppm) δ 159.45, 152.24, 149.56, 144.07, 135.32, 129.80, 127.85, 123.82, 112036, 116.53, 114.55, 112.90, 79.68,62.87, 55.63, 53.31.

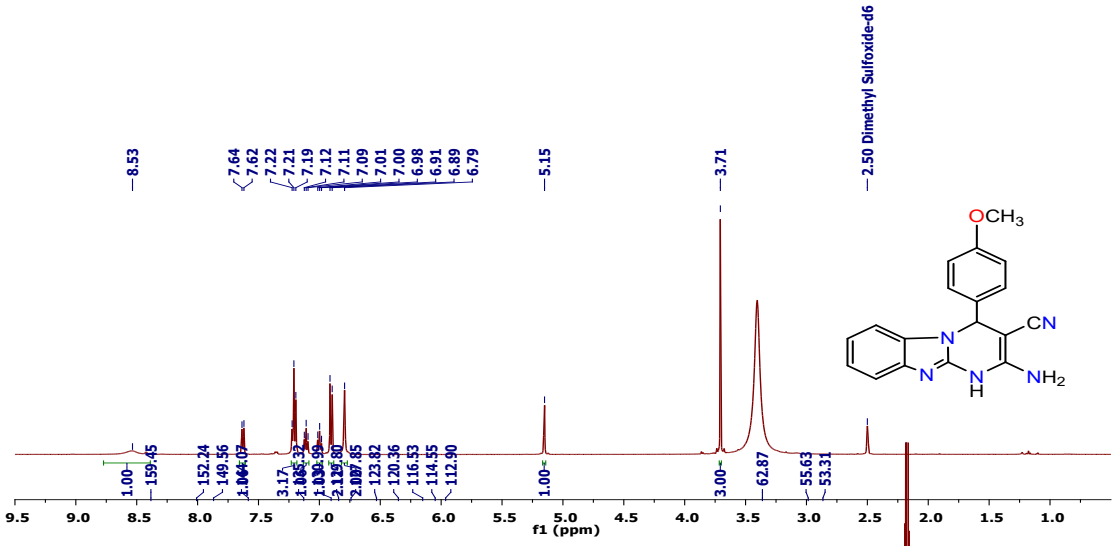
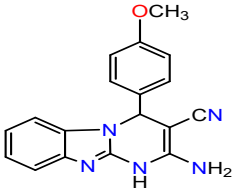
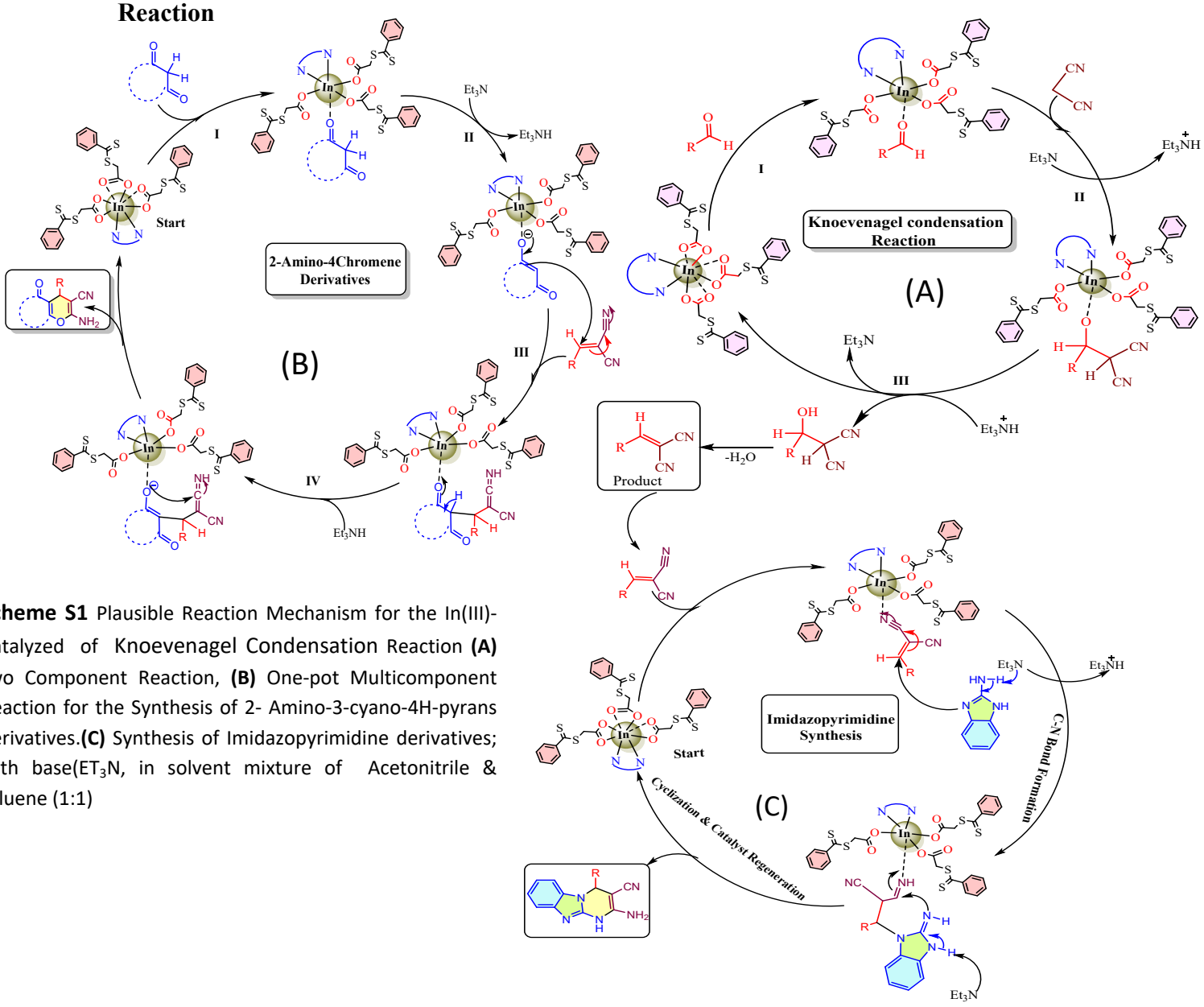


Figure S74. ^{1}H NMR Spectrum of 9e



Plausible Reaction Mechanism for the In(III)-Catalyzed One-pot Multicomponent Reaction



Scheme S1 Plausible Reaction Mechanism for the In(III)-Catalyzed of Knoevenagel Condensation Reaction **(A)** two Component Reaction, **(B)** One-pot Multicomponent Reaction for the Synthesis of 2- Amino-3-cyano-4H-pyrans derivatives.**(C)** Synthesis of Imidazopyrimidine derivatives; with base(ET_3N, in solvent mixture of Acetonitrile & toluene (1:1)

Data S10- NMR(^{1}H & ^{13}C) spectra For CO_2 conversion Reaction

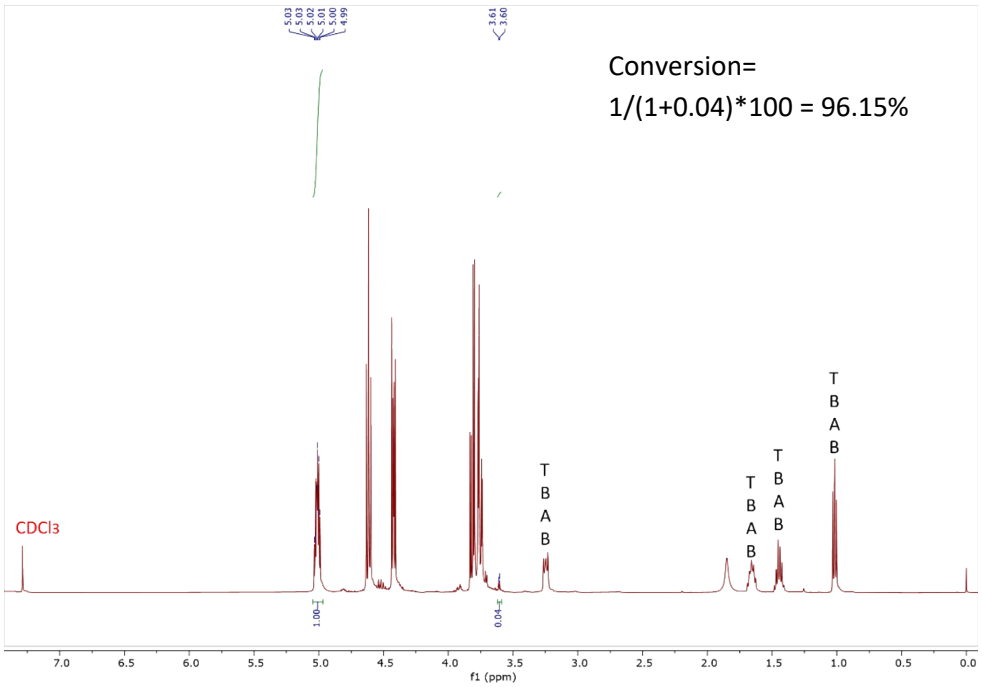


Figure S76: ^{1}H NMR spectrum of **catalyst 1**(Epichlorohydrin**) EPH** in $CDCl_3$ at 100°C

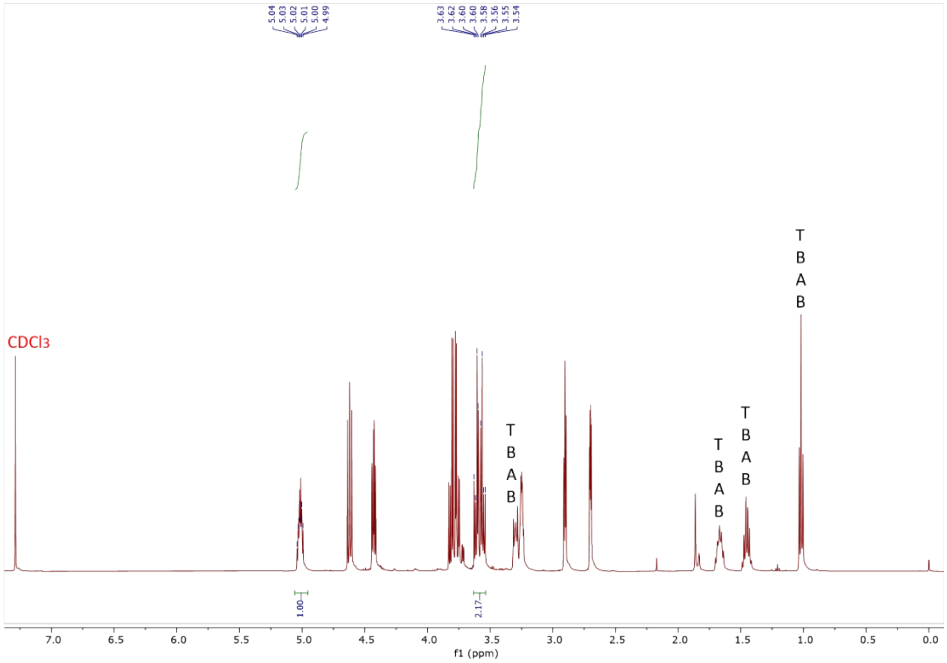


Figure S77: ^{1}H NMR spectrum of **catalyst 1** (Epichlorohydrin) **EPH** in $CDCl_3$ at 40 C

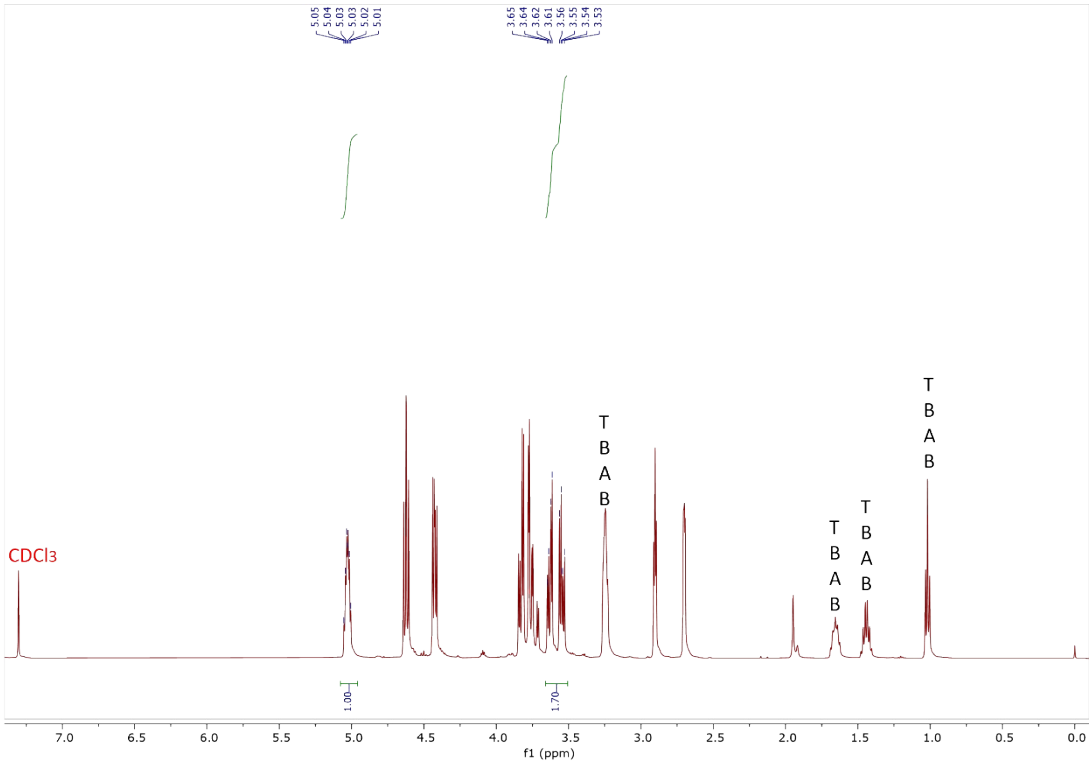
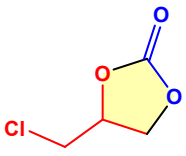


Figure S78: ^{1}H NMR spectrum of **catalyst 1** Epichlorohydrin **EPH** in $CDCl_3$ at 60 C

The residue was dissolved in Dichloromethane (5 mL) and washed with water (5 mL) and brine (5 mL × 2), dried over anhydrous sodium sulfate, filtered, and concentrated under reduced pressure. The crude product was purified by column chromatography (hexane: ethyl acetate = 1 : 1) to afford the desired cyclic carbonate

Epichlorohydrin**EPH**



M.F. $C_4H_5O_3Cl$ (136.0) (130g, 96%yield); ^{1}H NMR (500 MHz, $CDCl_3$) δ : 4.94-4.99 (m, 1H, -CH-), 4.56 (t, J = 8.0 Hz, 1H –CH_2- O(CO)O-), 4.34.3.37 (m, 1H –CH_2-O(CO)O-), 3.76-3.80 (m, 1H –CH_2-Cl), 3.67-3.71 (m, 1H –CH_2-Cl); ^{13}C NMR (126 MHz, $CDCl_3$) δ 154.55(C, -O(CO)O-), 74.64(-CH_2,) 67.06(-CH-), 44.11(-CH_2-Cl)

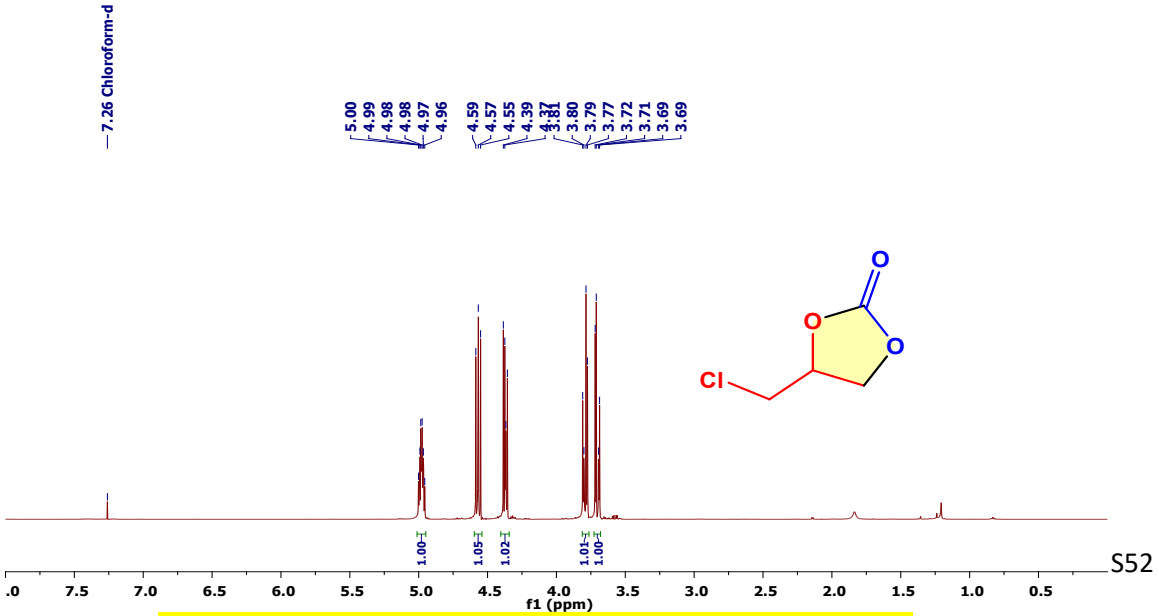
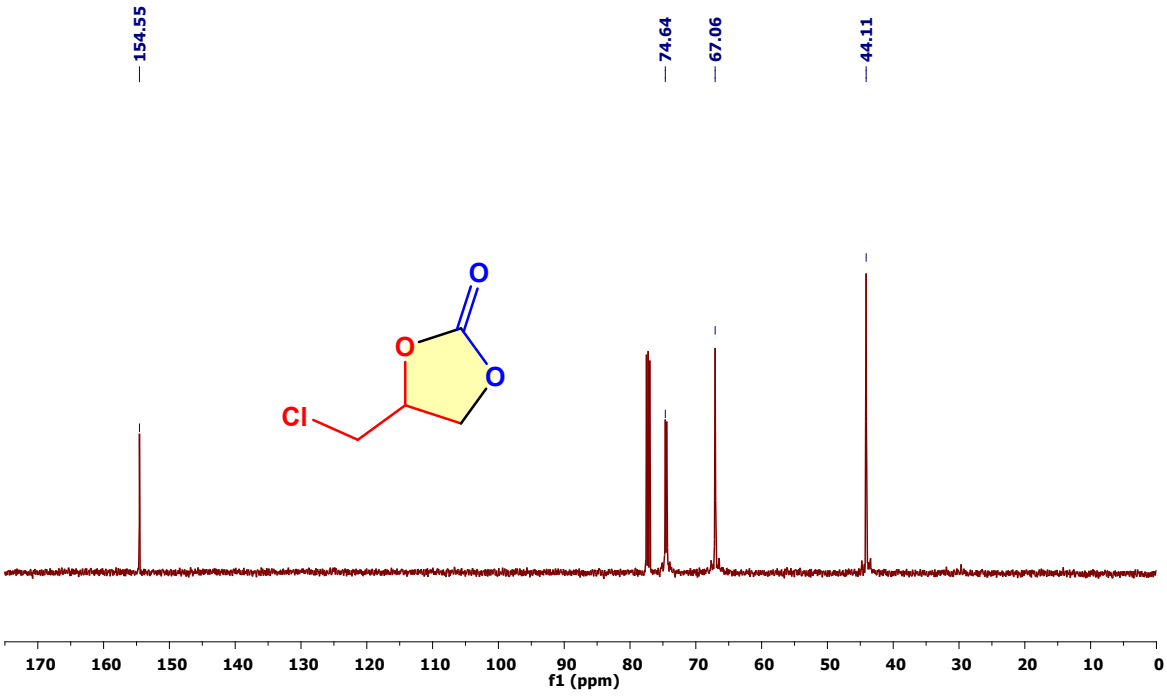


Figure S79: ^{1}H NMR spectrum of Epichlorohydrin (**EPH)** in $CDCl_3$



gure S80: ^{13}C NMR spectrum of Epichlorohydrin**EPH** in $CDCl_3$

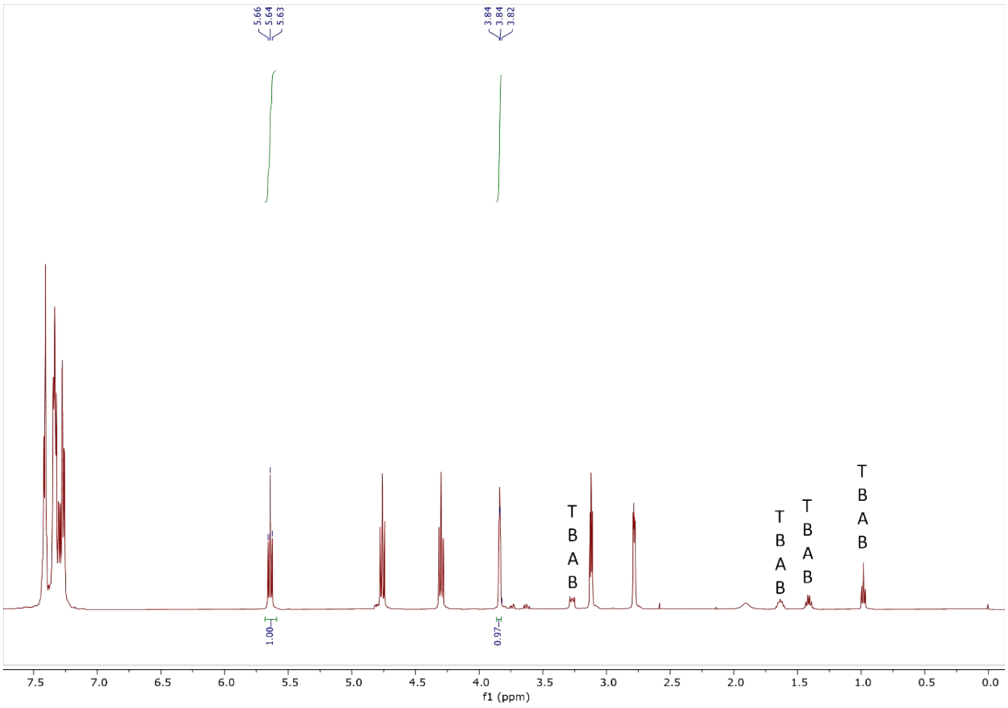
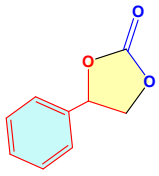


Figure S81: ^{1}H NMR spectrum of **catalyst 1 styrene oxide (STO)** in $CDCl_3$

The residue was dissolved in Dichloromethane (5mL) and washed with water (5 ml) and brine (5 mL × 2), dried over anhydrous sodium sulfate, filtered, and concentrated under reduced pressure. The crude product was purified by column chromatography (hexane: ethyl acetate = 3 : 1) to afford the desired cyclic carbonate.

Styrene oxide (STO)



M. F. $C_9H_6O_3$ (M.W 162.0) (80g, 49%yield) (^{1}H NMR (500 MHz, $CDCl_3$) δ : 7.35-746 (m, 5H, Ph), 5.67 (t, *J*=8Hz, 1H, -CH-O(CO)O-), 4.48 (t, *J* = 8.0 Hz, 1H –CH_2-O(CO)O-), 4.33 (t, *J* = 8.0 Hz, 1H –CH_2-O(CO)O-); ^{13}C NMR (126 MHz, $CDCl_3$) δ 154.99(C, -O(CO)O-), 135.91(C, Ph), 129.81(C, Ph),, 126.00(C, Ph), 71.28(-CH_2,) 78.11(-CH-).

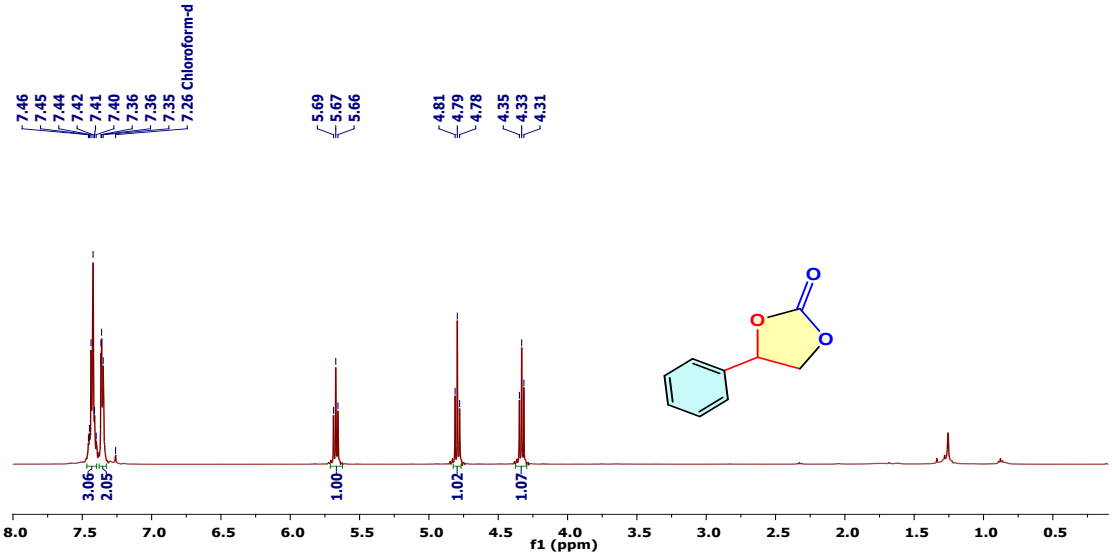


Figure S82: ^{1}H NMR spectrum of **styrene oxide (STO)** in $CDCl_3$

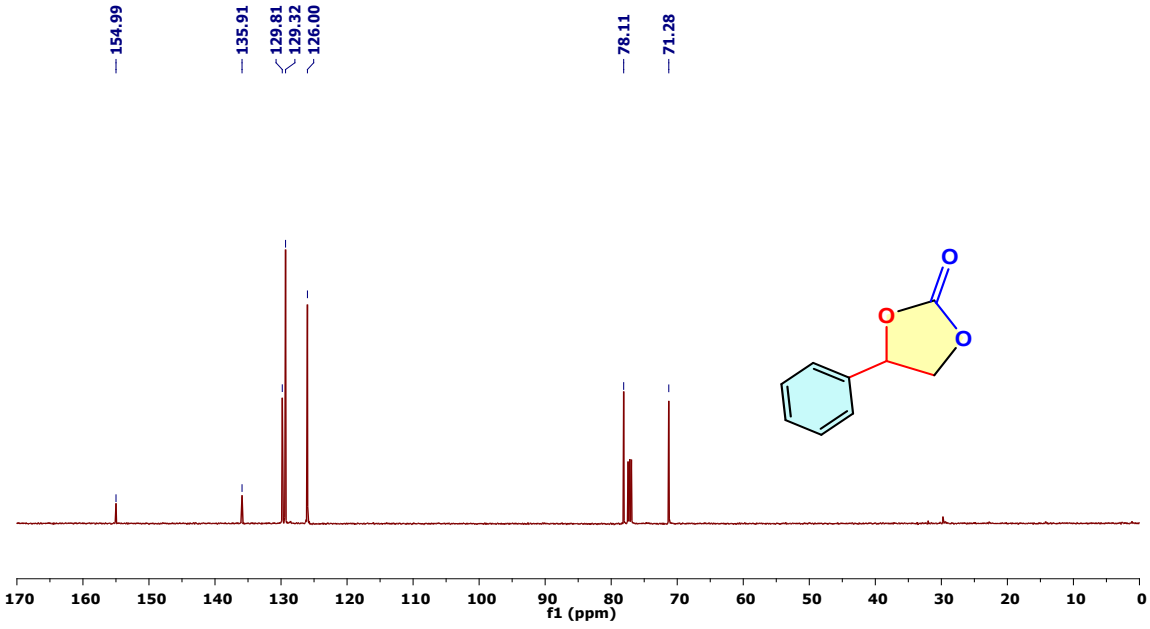


Figure S83: ^{13}C NMR spectrum of **styrene oxide (STO)** in $CDCl_3$

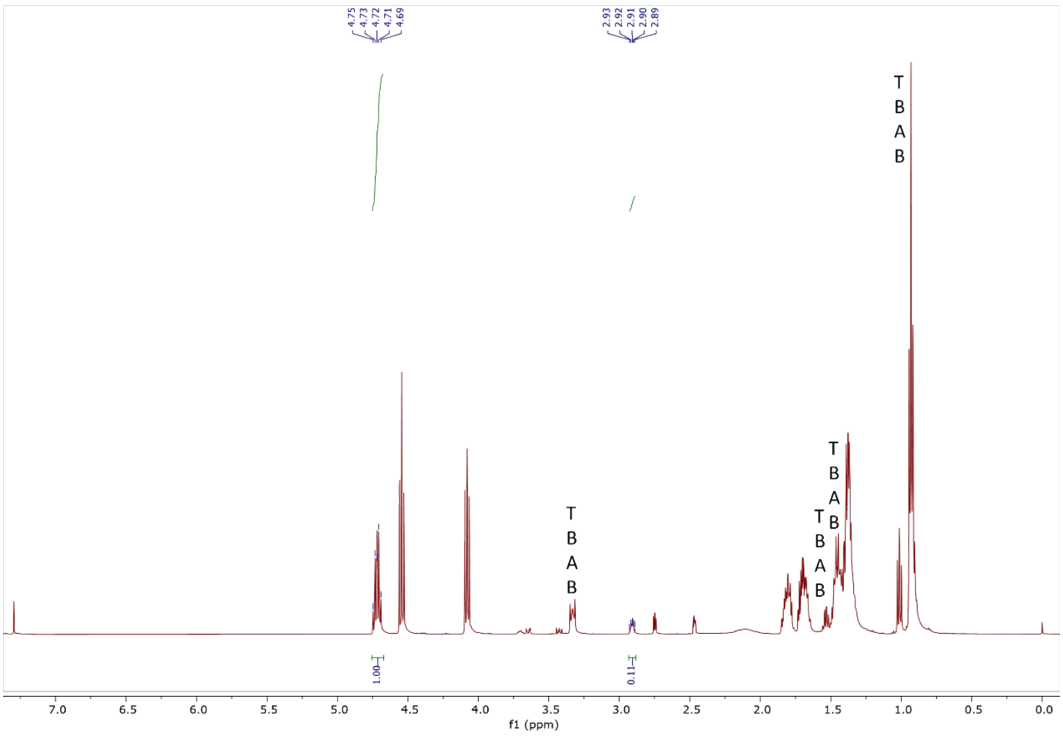
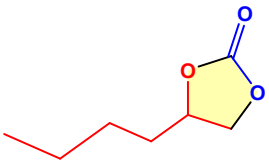


Figure S84: ^{1}H NMR spectrum of **1 Epoxy hexane (EH)** in $CDCl_3$

The residue was dissolved in Dichloromethane (5mL) and washed with water (5 ml) and brine (5 mL × 2), dried over anhydrous sodium sulfate, filtered, and concentrated under reduced pressure. The crude product was purified by column chromatography (hexane: ethyl acetate = 2 : 1) to afford the desired cyclic carbonate

Epoxy hexane(EH)



F. $C_7H_{612}O_3$ (M.W 144.1); (131g, 91% yield) ^{1}H NMR (500 MHz, $CDCl_3$) δ : 4.66 (m, , 1H,), 4.51 (t, J = 8.0 Hz, 1H –CH_2- O(CO)O-), 4.06 (t, J = 7.5 Hz, 1H –CH_2-O(CO)O-), 1.79-1.83(m, 1H, -CH_2-), 1.64-171 (m, 1H, -CH_2-), 1.31-147 (m, 4H, -CH_2-), 0.91(t, J=6.5Hz, 3H, CH_3); ^{13}C NMR (126 MHz, $CDCl_3$) δ: 155.16(C, -O(CO)O-), 69.46(C, -CH_2,) 32.62(-CH-), 29.74(-CH_2-), 26.49(-CH_2-), 22.31(-CH_2-), 13.84(-CH_3),

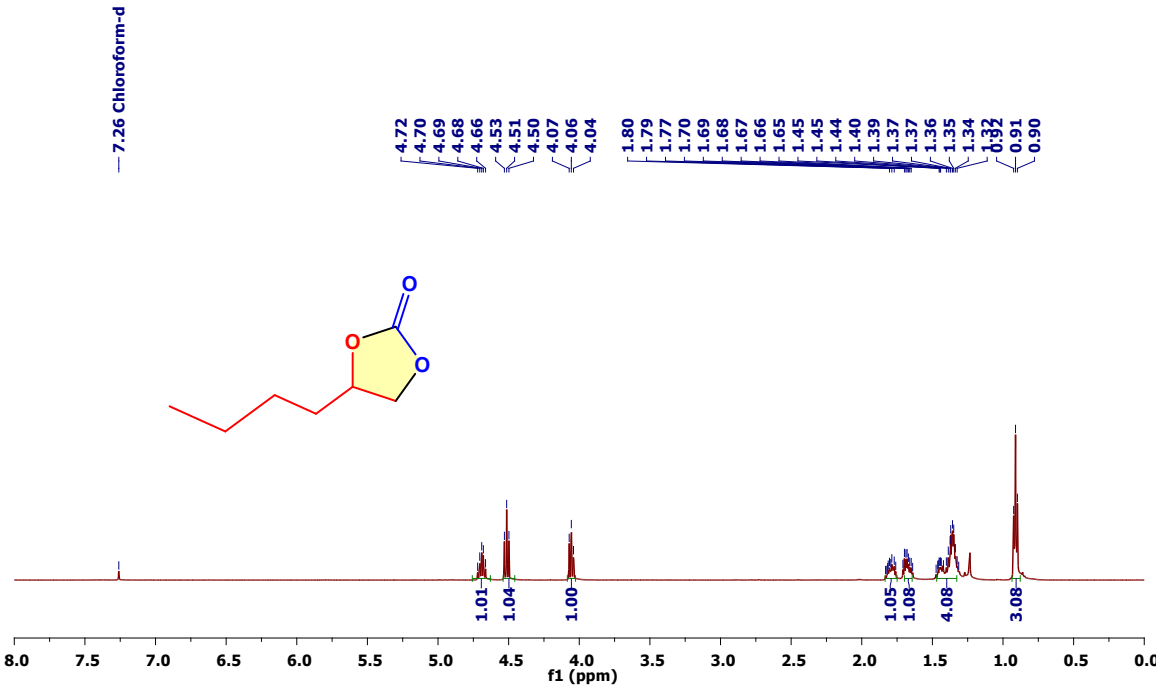


Figure S85: ^{1}H NMR spectrum of **Epoxy hexane (EH)** in $CDCl_3$

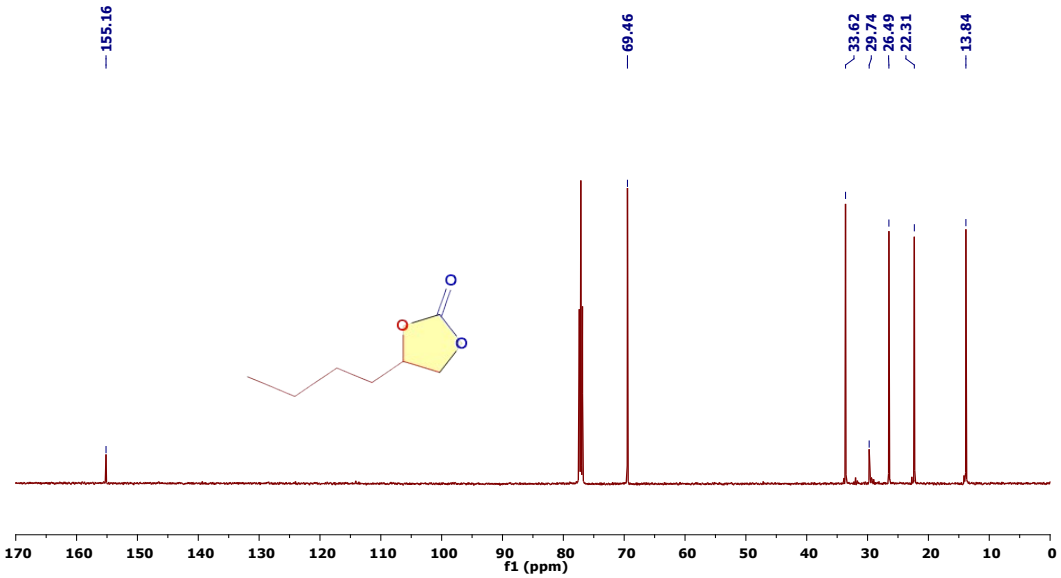
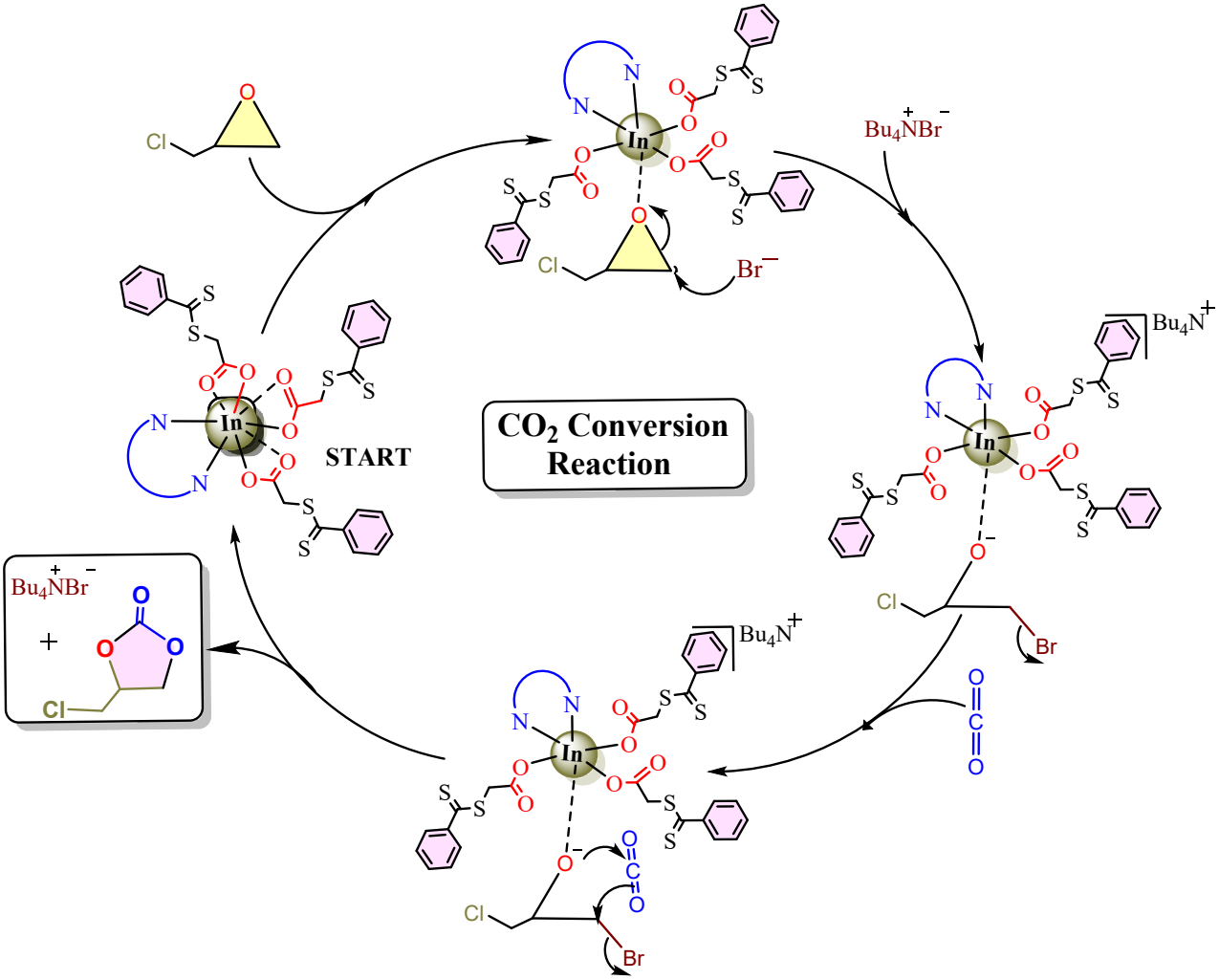


Figure S86: ^{1}H NMR spectrum of **Epoxy hexane (EH)** in $CDCl_3$

The catalyst **1** was added to TBAB as a cocatalyst to check the conversion of epoxide to cyclic carbonate using ^{1}H NMR spectra at different temperatures (100 C, 40 C and 60 C) Figure S1-3. The peak emerged around 5 ppm, depicting the formation of cyclic carbonate and the peak around 3.6

corresponds to the epichlorohydrin (EPH). The % conversion was calculated by using the integral of product and reactant peak.

Plausible Reaction Mechanism for the conversion of CO_2 into cyclic carbonate



Scheme S2. A Proposed mechanism for the coupling of epoxide and CO_2catalyzed by In/TBAB

Reference

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3 O. V. Dolomanov, L. J. Bourhis, R. J. Gildea, J. A. Howard and H. Puschmann, *J. Appl. Crystallogr.*, 2009, **42**, 339–341.
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