2-(4-chlorobenzylidene)malononitrile (3a): Colorless solid; m.p. (^{0}C): 161-163; ^{1}H-NMR (400 MHz, $CDCl_3$): δ 7.85 (d, 2H), 7.73 (s, 1H, HC=N), 7.52 (d, 2H); ^{13}C-NMR (100 MHz, $CDCl_3$): δ 83, 112, 113, 129, 130, 131, 141, 158.

Ethyl (E)-3-(4-Chlorophenyl)-2-cyano-2-propenoate (3b): White crystalline solid, 88 - 90 °C; ^{1}H-NMR (400 MHz, $CDCl_3$): δ 1.40 (t, 3H), 4.40 (q, 2H), 7.59 (d, 2H), 7.95 (d, 2H), 8.20 (s, 1H) ppm; EI-MS: m/z (%) = 235 [M+], 207, 190, 127, 75, 50.

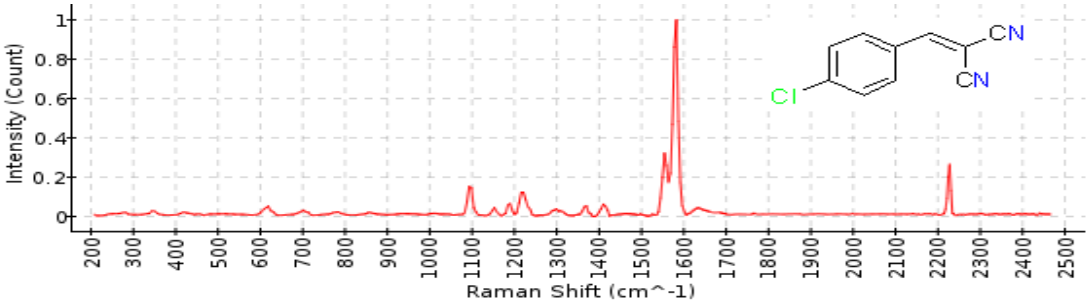
3-(4-Chlorobenzylidene)pentane-2,4-dione (3c): white solid; m.p. 167 - 169^{0}C; ^{1}H-NMR (300 MHz, $CDCl_3$): δ 7.40 (s, 1H), 7.39 -7.24 (m, 4H), 2.42 (s, 3H), 2.27 (s, 3H).

Diethyl 2-(4-chlorobenzylidene)malonate (3d): light Yellow oil, ^{1}H-NMR (300 MHz, $CDCl_3$): δ 1.31- 1.25 (2t, 6H), 4.31-4.4 (2q, 4H), 7.45-7.30 (m, 4H, Ph), 7.7 (s, 1H, imine); ^{13}C NMR (100 MHz, $CDCl_3$): 13.7, 13.8, 61.4, 61.7, 125.4, 129.0, 130.3, 130.4, 132.9, 140.0, 166.3, 163.8. MS (IE): Calc. for $[M]^+$ $C_{14}H_{15}ClO_4$: 282.07; $[M+H]^+$. = 283 (100%).

2-(4-Chlorobenzylidene)-5,5-dimethylcyclohexane-1,3-dione (3f): Pale yellow; m.p. 126–128 °C; ^{1}H-NMR (300 MHz, DMSO-d_6) δ 7.49–7.22 (m, 4H, C_6H_4), 6.07 (s, 1H, CH), 2.38, 2.21 (2s, 4H, $2CH_2$), 1.08, 1.05 (2s, 6H, $2CH_3$); ^{13}C-NMR (DMSO-d_6, 75 MHz) δ 166.5, 164.2, 128.3, 126.5, 123.2, 121.3, 108.8, 103.3, 50.6, 36.8, 24.5; EIMS: m/z 262 $[M]^+$; Calcd. for $C_{15}H_{15}ClO_2$ (262.73): C, 68.57; H, 5.75%. Found: C, 68.39; H, 6.02%.

5-(4-Chloro-benzylidene)-2,2-dimethyl-[1, 3]dioxane-4,6-dione (3h): Colorless solid; m.p. 154 – 156^{0}C; ^{1}H-NMR (300 MHz, $CDCl_3$, ppm): δ 8.37 (s, 1H), 8.03 (d, 2H), 7.46 (d, 2H), 1.81 (s, 6H); Elemental analysis: found C 58.70, H 4.25%, calculated for $C_{13}H_{11}O_4Cl$; C 58.64, H 4.13%.

Spectral information:



υ(C-Cl)	550 - 800 cm^{-1}	strong	υ(CC) aromatic ring chain vibrations	1580, 1600 cm^{-1}	strong
υ(C=C)	1500 - 1900 cm^{-1}	strong	υ(C≅N)	2220 - 2255 cm^{-1}	medium

Figure S1: Raman spectra of 2-(4-chlorobenzylidene)malononitrile (3a).

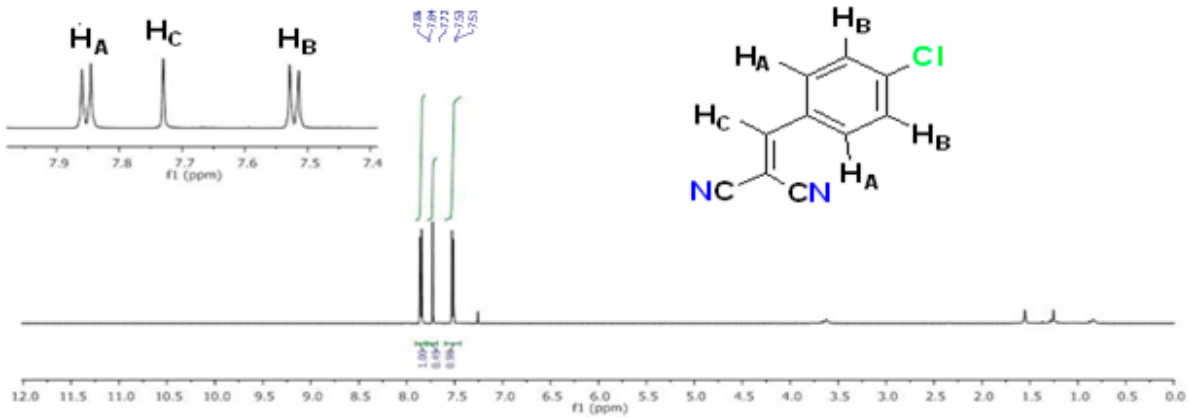


Figure S2: PMR spectra of 2-(4-chlorobenzylidene)malononitrile (3a) in $CDCl_3$.

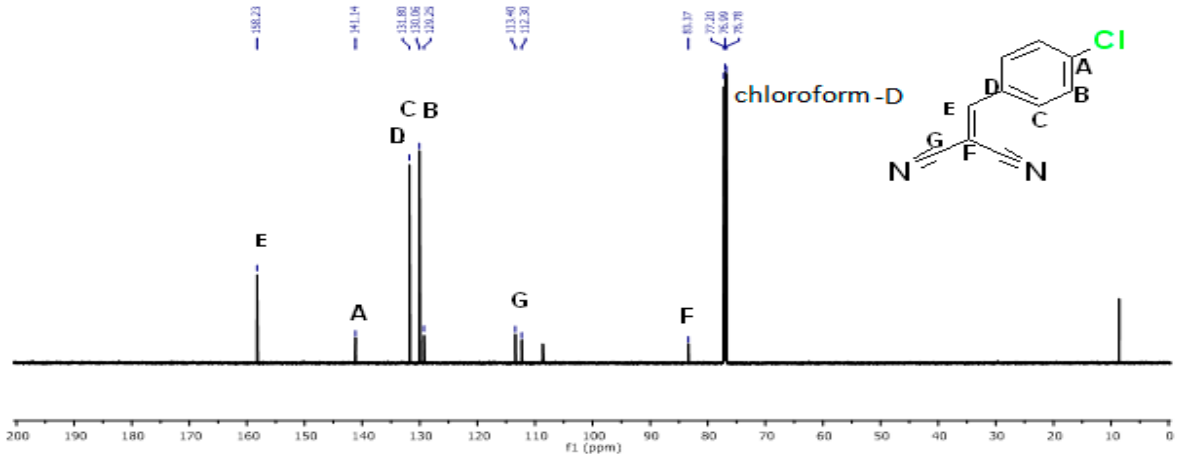


Figure S3: CMR spectra of 2-(4-chlorobenzylidene)malononitrile (3a) in $CDCl_3$.

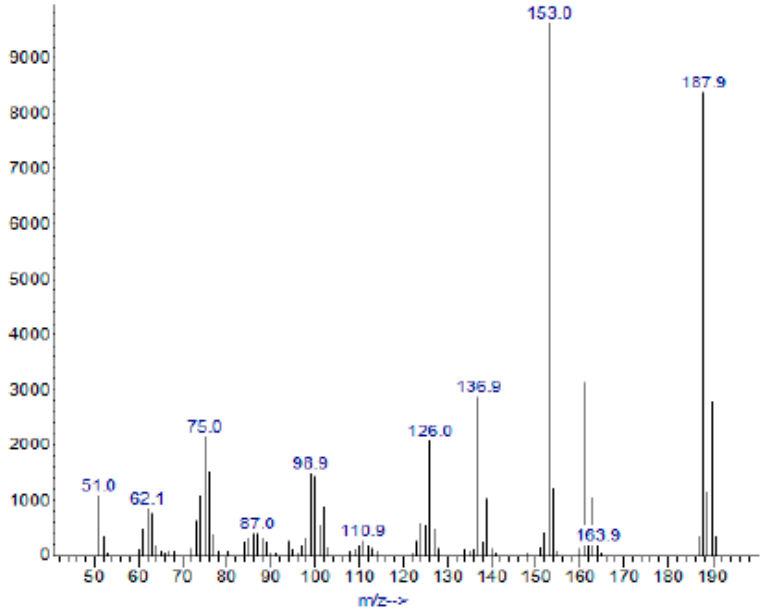


Figure S4: Mass spectra of 2-(4-chlorobenzylidene)malononitrile (3a).

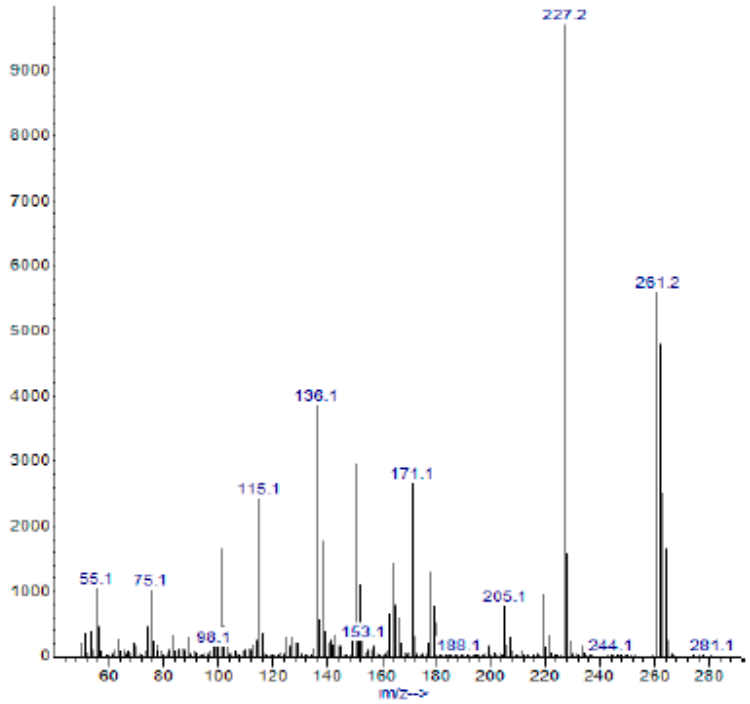


Figure S5: Mass spectra of 2-(4-Chlorobenzylidene)-5,5-dimethylcyclohexane-1,3-dione (3f)

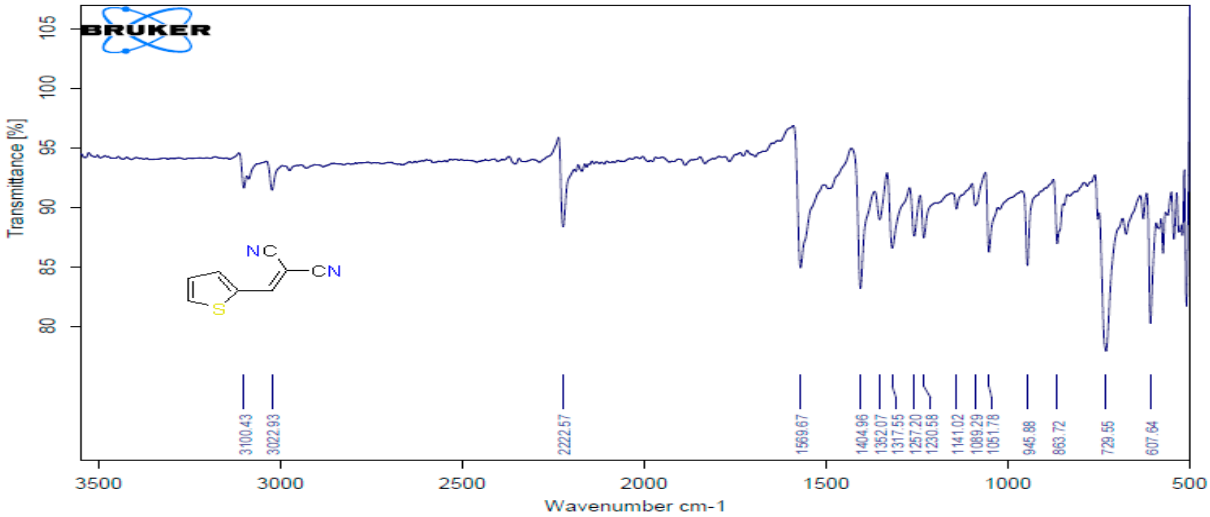
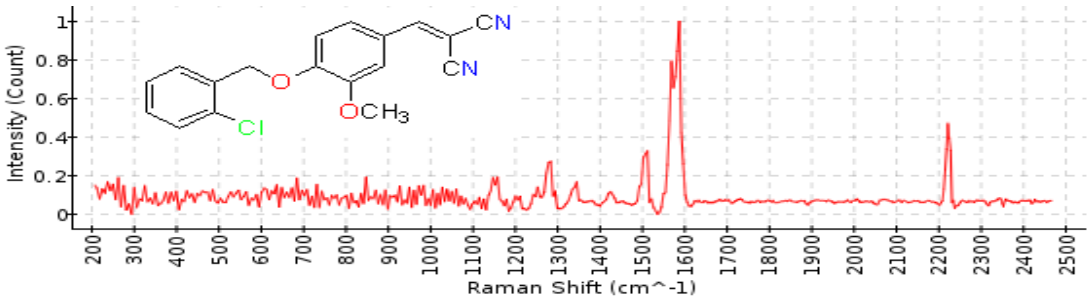


Figure S6: FTIR spectra of 2-(thiophen-2-ylmethylene)malononitrile (3k)



υ(C-O-C) asym	1060 - 1150 cm^{-1}	weak	υ(C-O-C)	800 -970 cm^{-1}	medium
υ(C=C)	1500 - 1900 cm^{-1}	strong	υ(CC) aromatic ring chain vibrations	1580, 1600 cm^{-1}	strong
υ(C≅N)	2220 - 2255 cm^{-1}	medium			

Figure S7: Raman spectra of 2-(4-((2-chlorobenzyl)oxy)-3-methoxybenzylidene)malononitrile (3l).

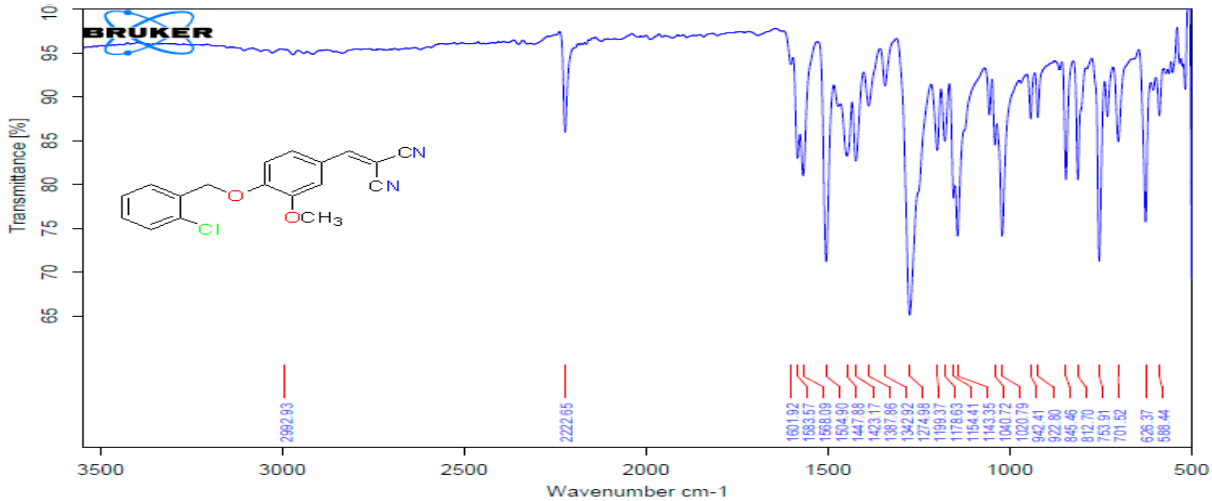
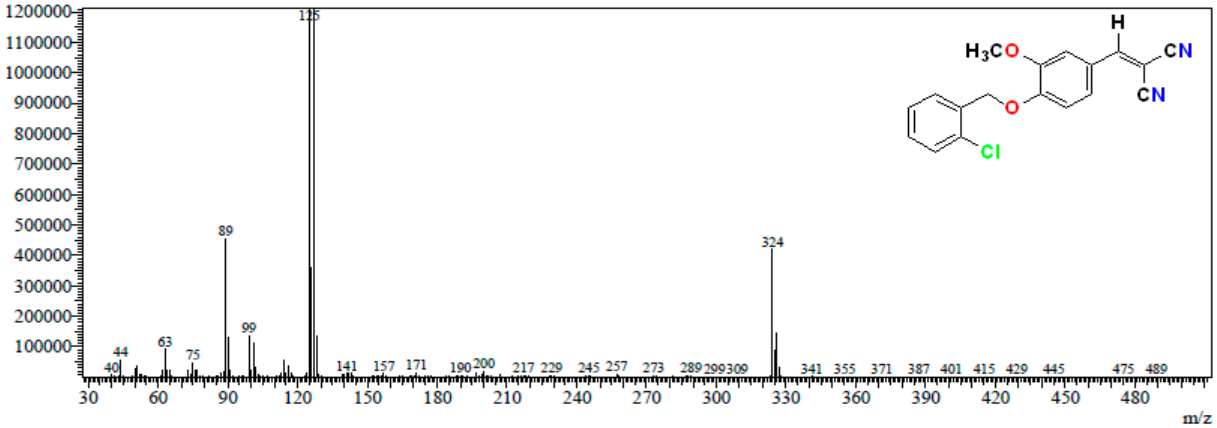
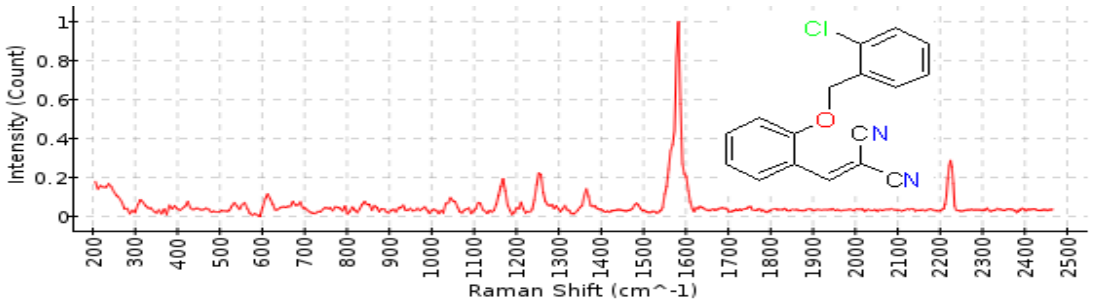


Figure S8: FTIR spectra of 2-(4-((2-chlorobenzyl)oxy)-3-methoxybenzylidene)malononitrile (3l).



Retension time (min)	Base peak (m/z)	Molecular ion peak (m/z)
21.975	125	324, 326 (1:3)

Figure S9: Mass spectra of 2-(4-((2-chlorobenzyl)oxy)-3-methoxybenzylidene)malononitrile (3l)



υ(C-Cl)	550 - 800 cm^{-1}	strong	υ(C-O-C)	800 -970 cm^{-1}	medium
υ(C-O-C) asym	1060 - 1150 cm^{-1}	weak	υ(CC) aromatic ring chain vibrations	1580, 1600 cm^{-1}	strong
υ(C=C)	1500 - 1900 cm^{-1}	strong	υ(C≅N)	2220 - 2255 cm^{-1}	medium

Figure S10: Raman spectra of 2-(2-((2-chlorobenzyl)oxy)benzylidene)malononitrile (3m).

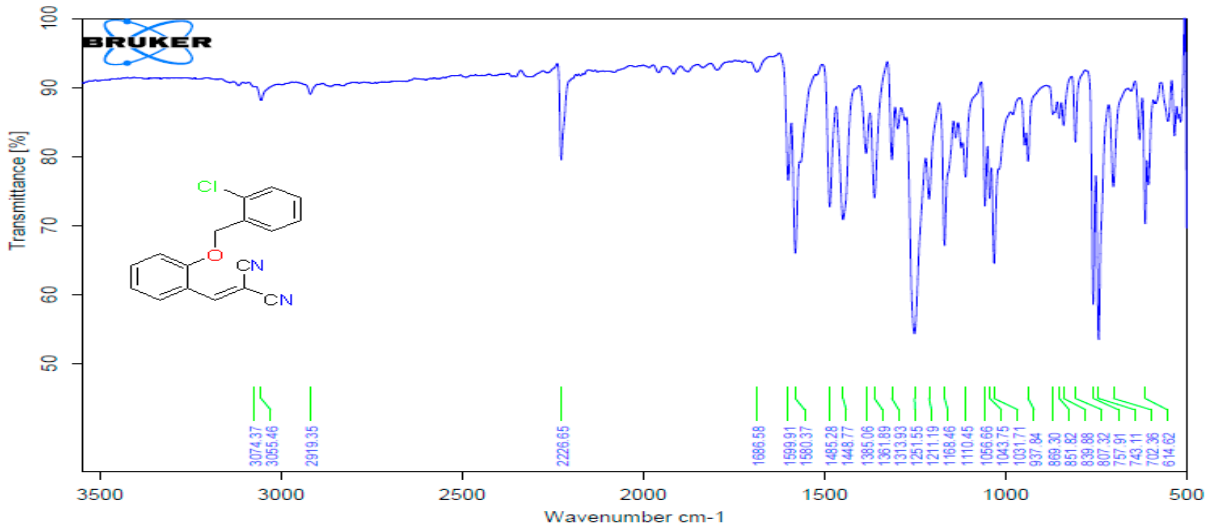
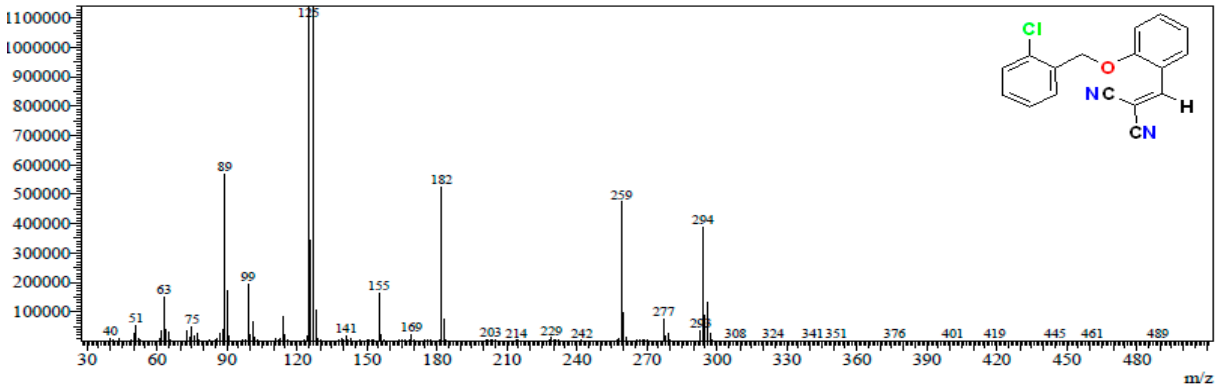
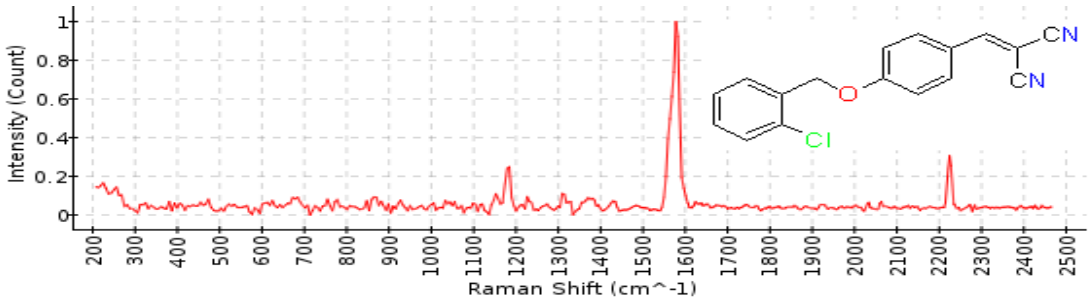


Figure S11: FTIR spectra of 2-(2-((2-chlorobenzyl)oxy)benzylidene)malononitrile (3m).



Retension time (min)	Base peak (m/z)	Molecular ion peak (m/z)
20.185	125	294, 296 (1:3)

Figure S12: Mass spectra of 2-(2-((2-chlorobenzyl)oxy)benzylidene)malononitrile (3m)



υ(C-Cl)	550 - 800 cm^{-1}	strong	υ(C-O-C)	800 -970 cm^{-1}	medium
υ(C-O-C) asym	1060 - 1150 cm^{-1}	weak	υ(CC) aromatic ring chain vibrations	1580, 1600 cm^{-1}	strong
υ(C≅N)	2220 - 2255 cm^{-1}	medium	υ(C=C)	1500 - 1900 cm^{-1}	strong

Figure S13: Raman spectra of 2-(4-((2-chlorobenzyl)oxy)benzylidene)malononitrile (3n).

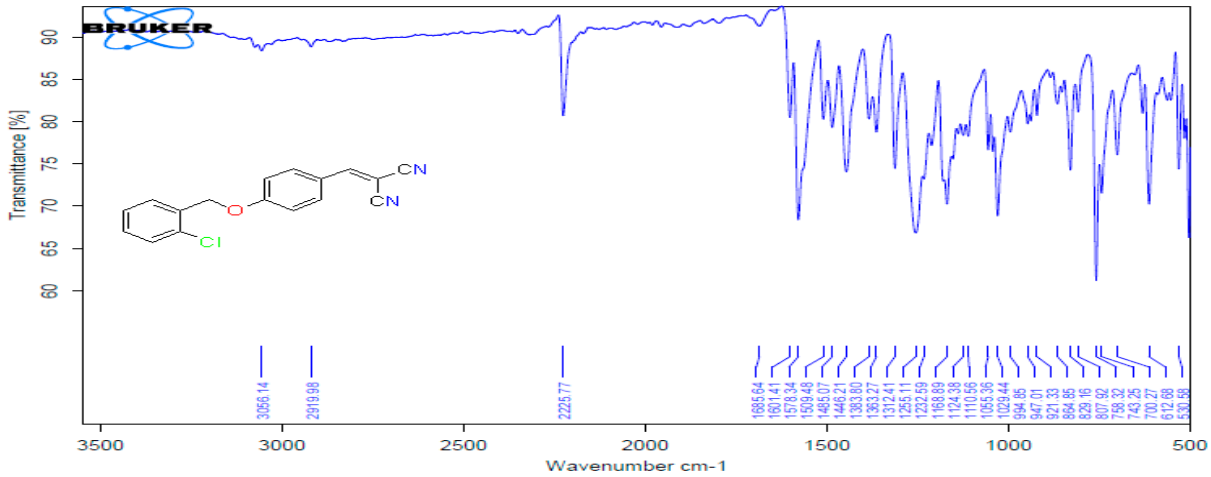
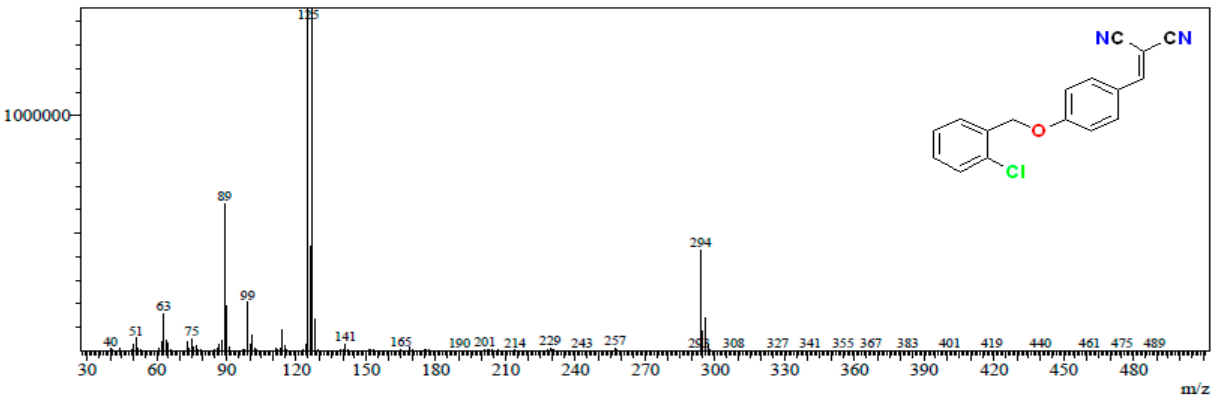
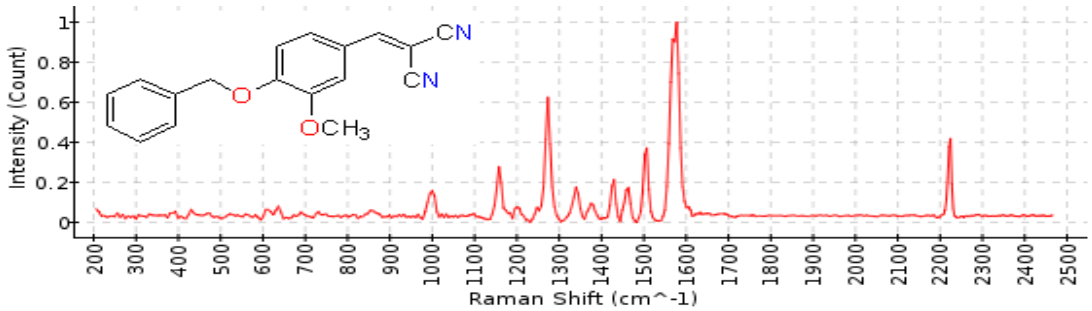


Figure S14: FTIR spectra of 2-(4-((2-chlorobenzyl)oxy)benzylidene)malononitrile (3n).



Retension time (min)	Base peak (m/z)	Molecular ion peak (m/z)
21.365	125	294, 296

Figure S15: Mass spectra of 2-(4-((2-chlorobenzyl)oxy)benzylidene)malononitrile (3n)



υ(C-O-C) asym	1060 - 1150 cm^{-1}	weak	υ(C-O-C)	800 -970 cm^{-1}	medium
υ(C=C)	1500 - 1900 cm^{-1}	strong	υ(CC) aromatic ring chain vibrations	1580, 1600 cm^{-1}	strong
υ(C≅N)	2220 - 2255 cm^{-1}	medium			

Figure S16: Raman spectra of 2-(4-(benzyloxy)-3-methoxybenzylidene)malononitrile (3o).

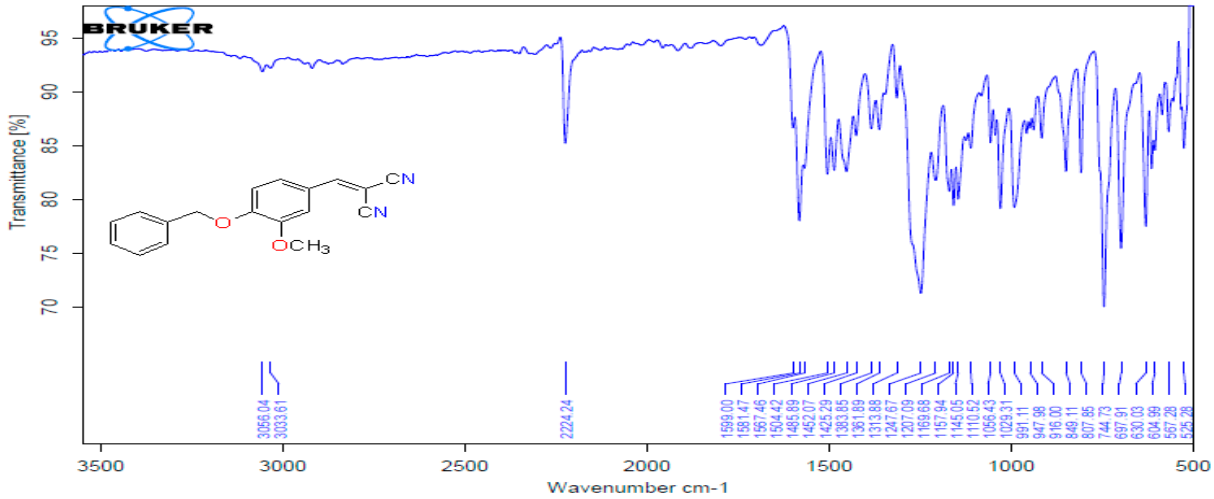
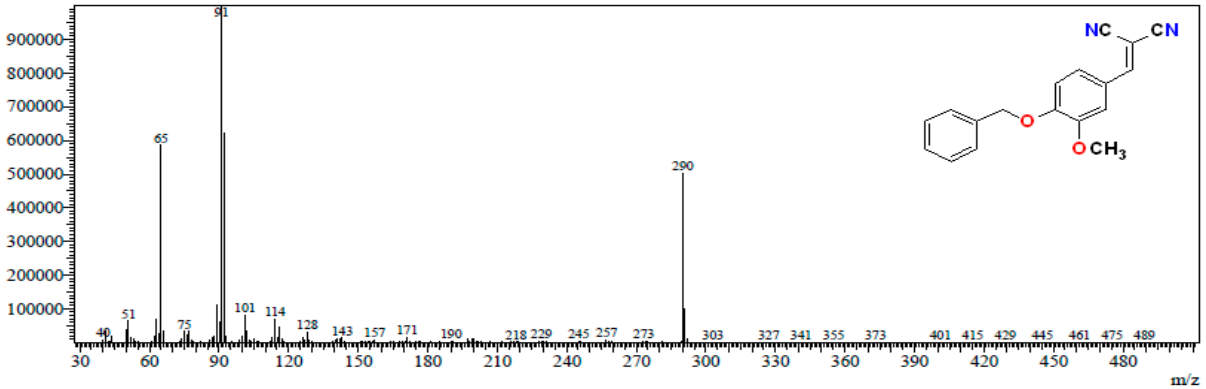
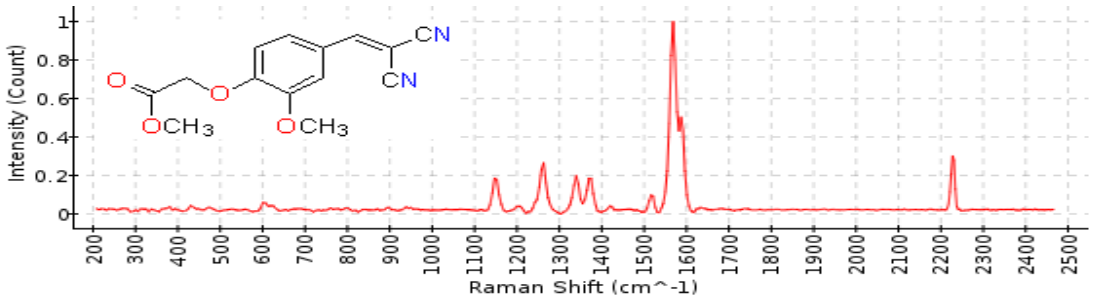


Figure S17: FTIR spectra of 2-(4-(benzyloxy)-3-methoxybenzylidene)malononitrile (3o).



Retension time (min)	Base peak (m/z)	Molecular ion peak (m/z)
20.980	91	290

Figure S18: Mass spectra of 2-(4-(benzyloxy)-3-methoxybenzylidene)malononitrile (3o)



υ(C-O-C) asym	1060 - 1150 cm^{-1}	weak	υ(C-O-C)	800 -970 cm^{-1}	medium
υ(C=O)	1680 - 1820 cm^{-1}	medium	υ(CC) aromatic ring chain vibrations	1580, 1600 cm^{-1}	strong
υ(C=C)	1500 - 1900 cm^{-1}	strong	υ(C≅N)	2220 - 2255 cm^{-1}	medium

Figure S19: Raman spectra of methyl 2-(4-(2,2-dicyanovinyl)-2-methoxyphenoxy)acetate (3p).

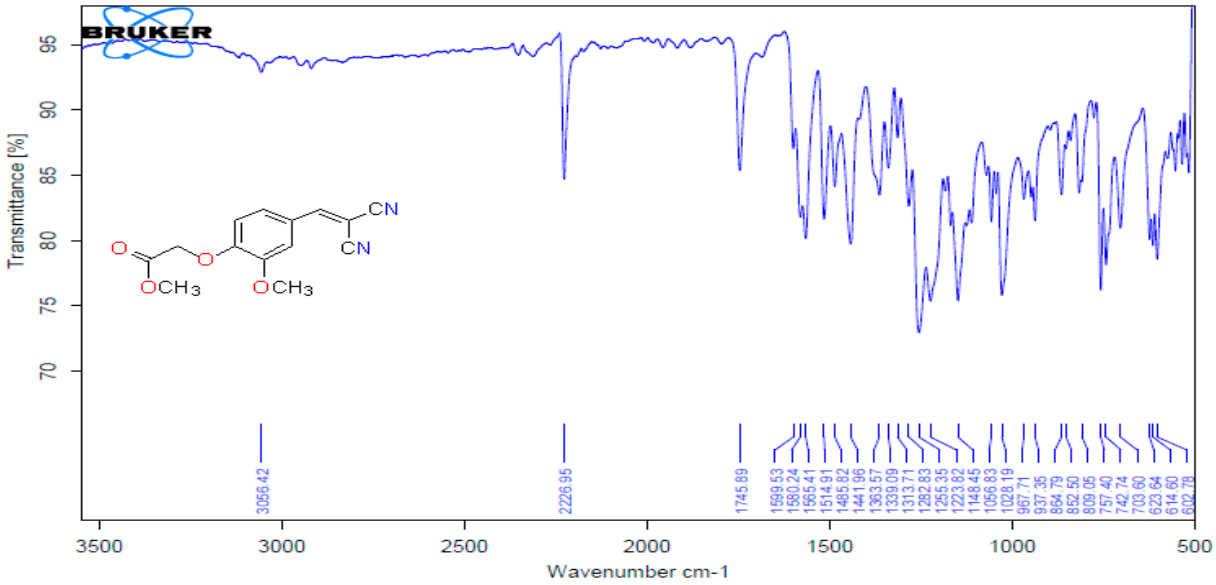
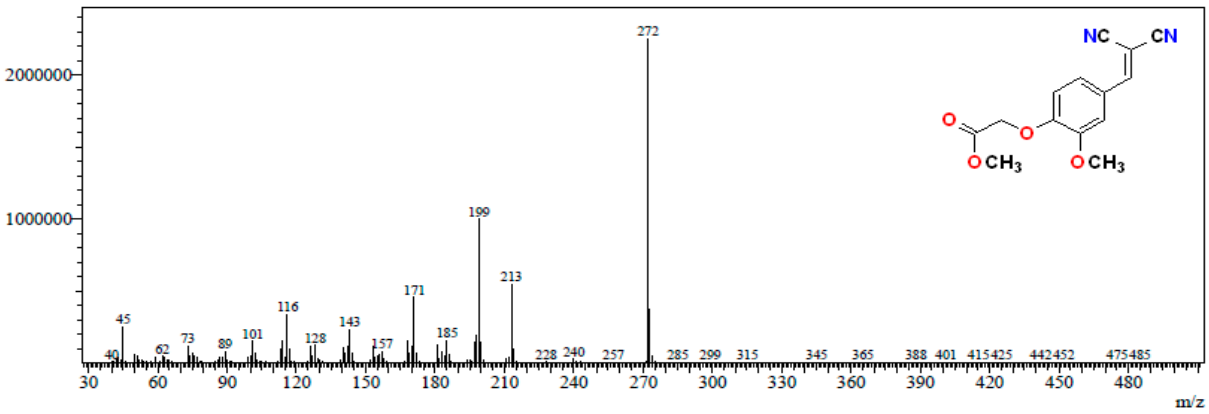
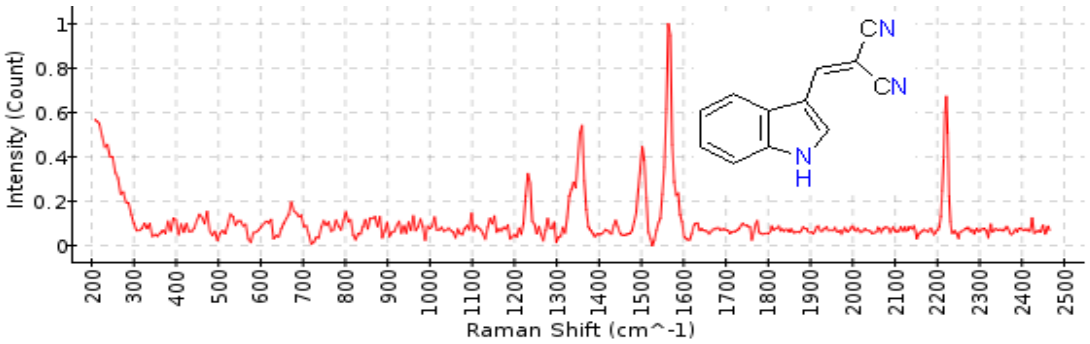


Figure S20: FTIR spectra of methyl 2-(4-(2,2-dicyanovinyl)-2-methoxyphenoxy)acetate (3p).



Retension time (min)	Base peak (m/z)	Molecular ion peak (m/z)
19.125	272	272

Figure S21: Mass spectra of methyl 2-(4-(2,2-dicyanovinyl)-2-methoxyphenoxy)acetate (3p)



υ(C=C)	1500 - 1900 cm^{-1}	strong	υ(CC) aromatic ring chain vibrations	1580, 1600 cm^{-1}	strong
υ(C≅N)	2220 - 2255 cm^{-1}	medium			

Figure S22: Raman spectra of 2-((1H-indol-3-yl)methylene)malononitrile (3q).

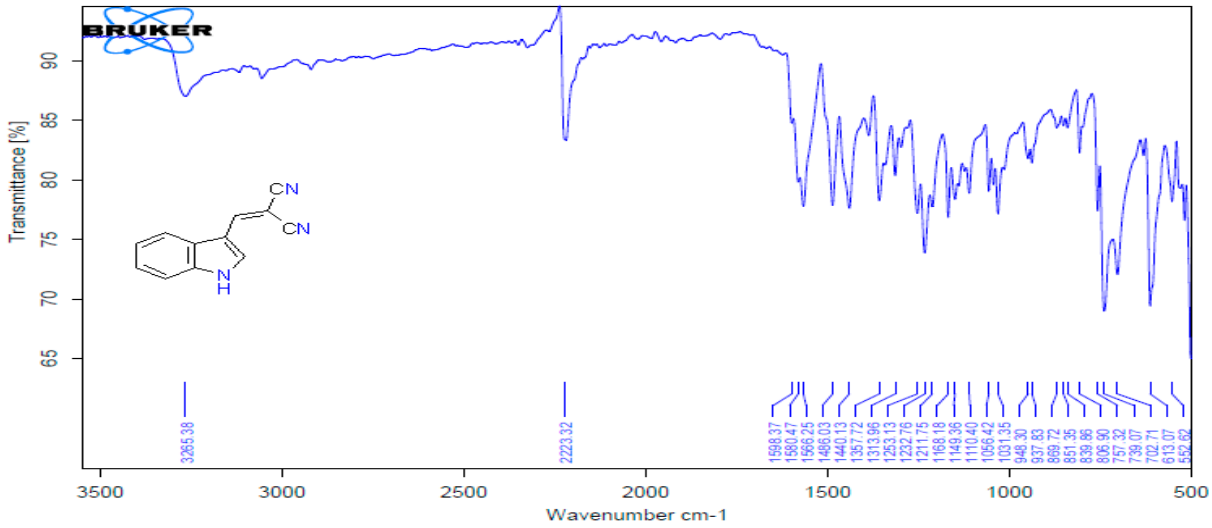
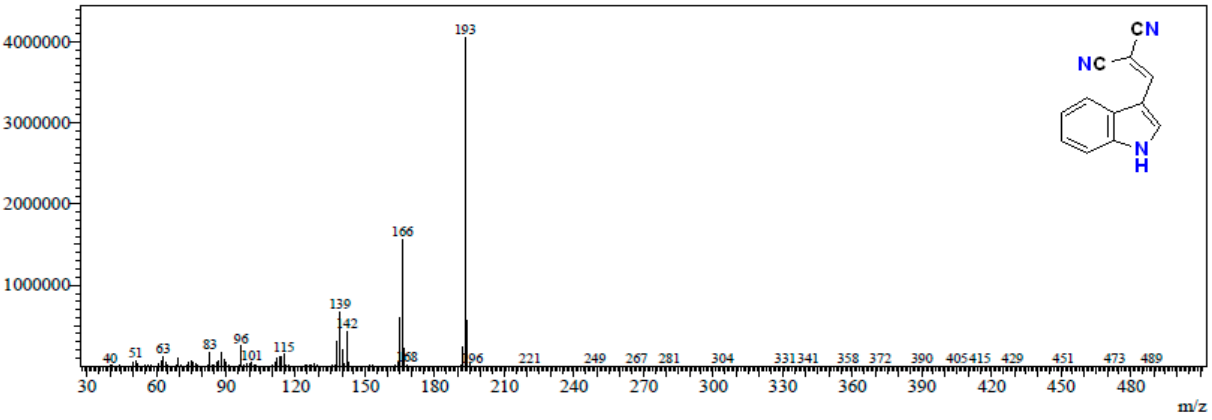
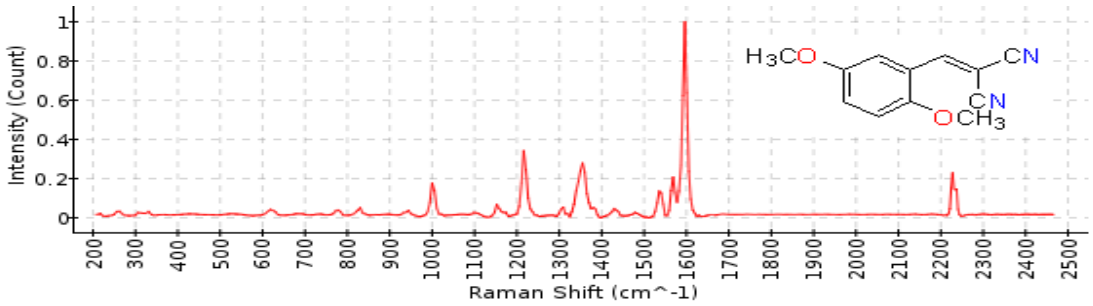


Figure S23: FTIR spectra of 2-((1H-indol-3-yl)methylene)malononitrile (3q).



Retension time (min)	Base peak (m/z)	Molecular ion peak (m/z)
19.880	193	193

Figure S24: Mass spectra of 2-((1H-indol-3-yl)methylene)malononitrile (3q)



υ(C-O-C) asym	1060 - 1150 cm^{-1}	weak	υ(C-O-C)	800 -970 cm^{-1}	medium
υ(C=C)	1500 - 1900 cm^{-1}	strong	υ(CC) aromatic ring chain vibrations	1580, 1600 cm^{-1}	strong
υ(C≅N)	2220 - 2255 cm^{-1}	medium			

Figure S25: Raman spectra of 2-(2,5-dimethoxybenzylidene)malononitrile (3r).

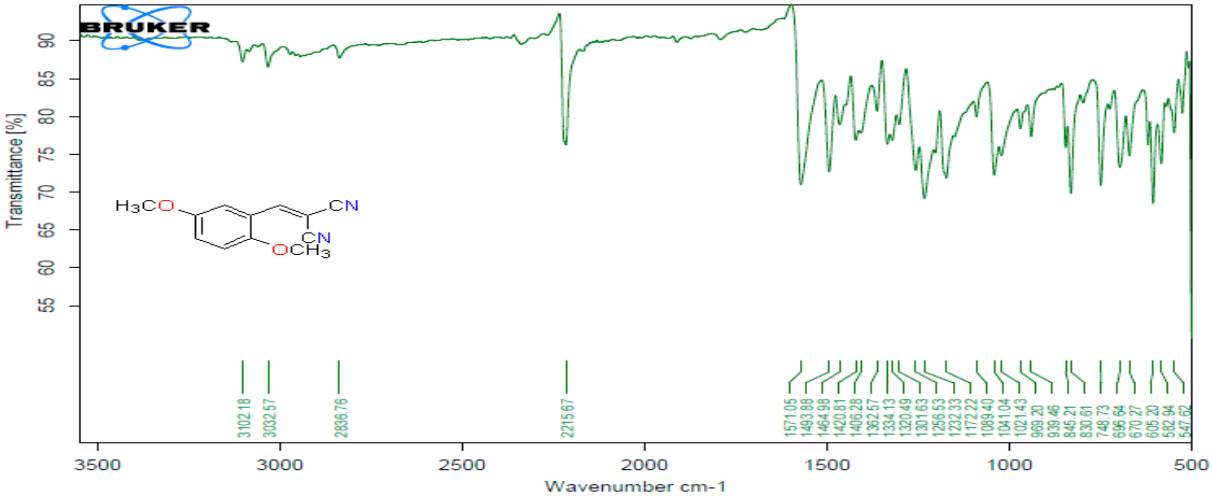
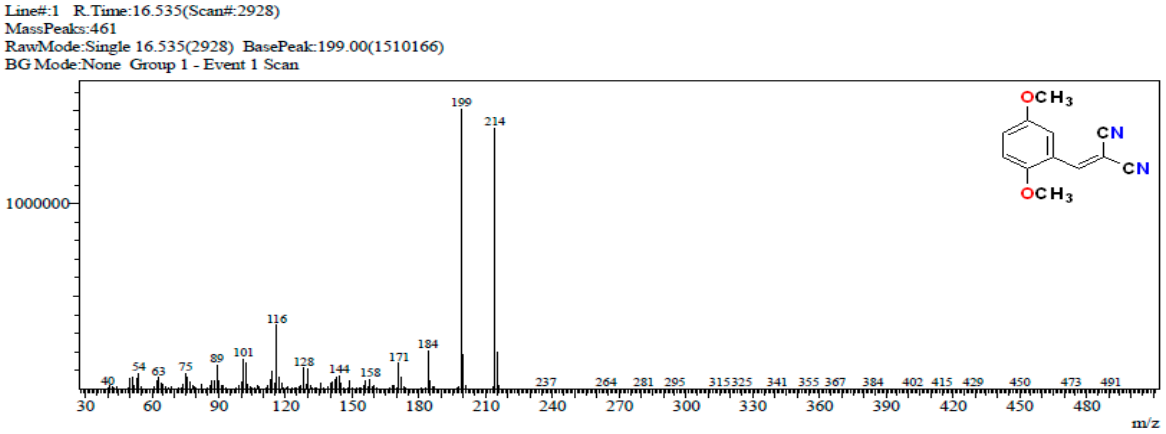


Figure S26: FTIR spectra of 2-(2,5-dimethoxybenzylidene)malononitrile (3r).



Retension time (min)	Base peak (m/z)	Molecular ion peak (m/z)
16.535	199	214

Figure S27: Mass spectra of 2-(2,5-dimethoxybenzylidene)malononitrile (3r)