Electronic Supplementary Material (ESI) for CrystEngComm.

Imidazole-induced self-assembly of polyoxovanadate cluster organic framework for efficient Knoevenagel condensation under mild conditions

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Table S1 The crystallographic data and structure refinement for **1**, **2** and **3**.

	1	**2**	**3**
Formula	$C_{56}H_{98}N_{16}Ni_2O_{13}V_4$	$C_{20}H_{54}N_8Ni_2O_{38}V_{10}$	$C_{14}H_{42}N_4Ni_2O_{38}V_{10}$
M_r	1524.68	1641.53	1501.34
Crystal system	Monoclinic	Monoclinic	Monoclinic
Space group	*P*2/*c*	$P2_1/n$	*C*2/*m*
T(K)	298(2) K	298(2) K	293(2) K
a (Å)	14.6250(12)	11.8760(9)	19.4747(16)
b (Å)	16.5741(14)	12.2282(11)	9.2565(8)
c (Å)	16.2669(14)	17.4796(14)	14.0833(12)
α (deg)	90	90	90
β (deg)	107.071(3)	101.696(3)	106.177(2)
γ (deg)	90	90	90
V (Å^3)	3769.3(5)	2485.7(4)	2438.2(4)
Z	2	2	2
$D_{calc.}$(g/cm^{-3})	1.343	2.193	2.045
F(000)	1596	1640	1488
$R_1[I>2\sigma(I)]$	0.0755	0.0859	0.0498
$wR_2[I>2\sigma(I)]$	0.1970	0.2212	0.1355
R_1(all data)	0.1794	0.1332	0.0681
wR_2(all data)	0.2372	0.2439	0.1497

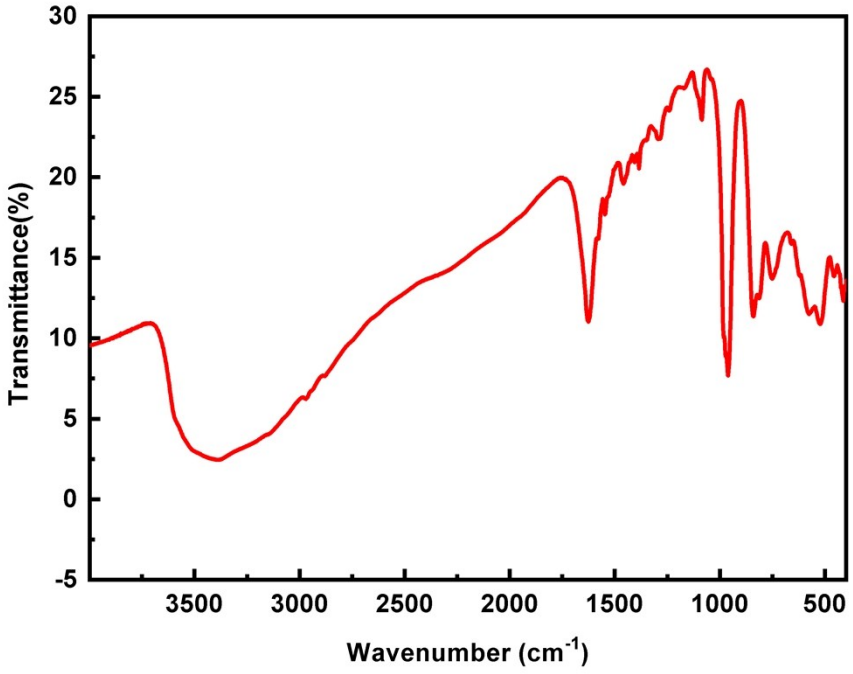

Figure S1. The FT-IR spectrum of compound **1**.

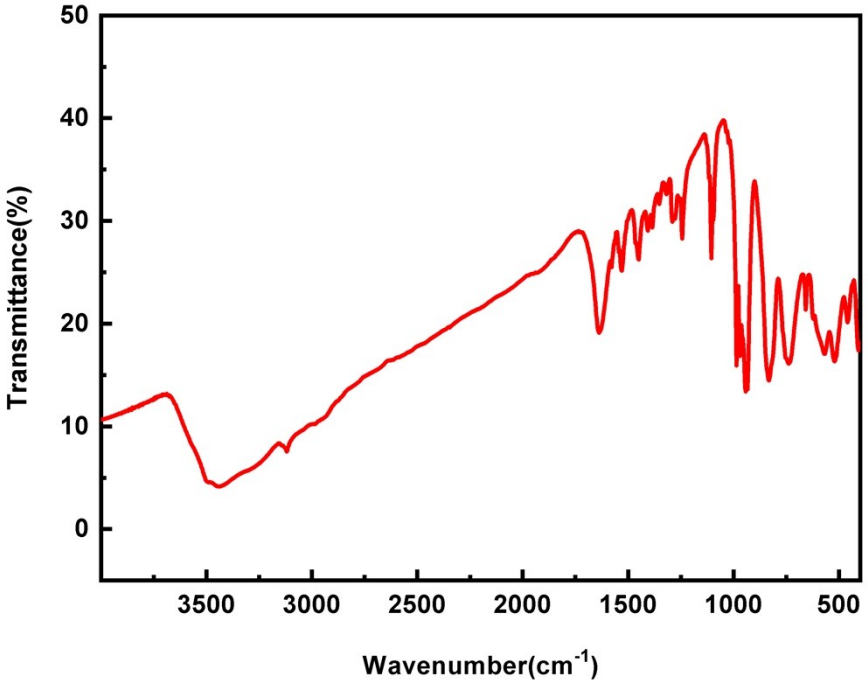

Figure S2. The FT-IR spectrum of compound **2**.

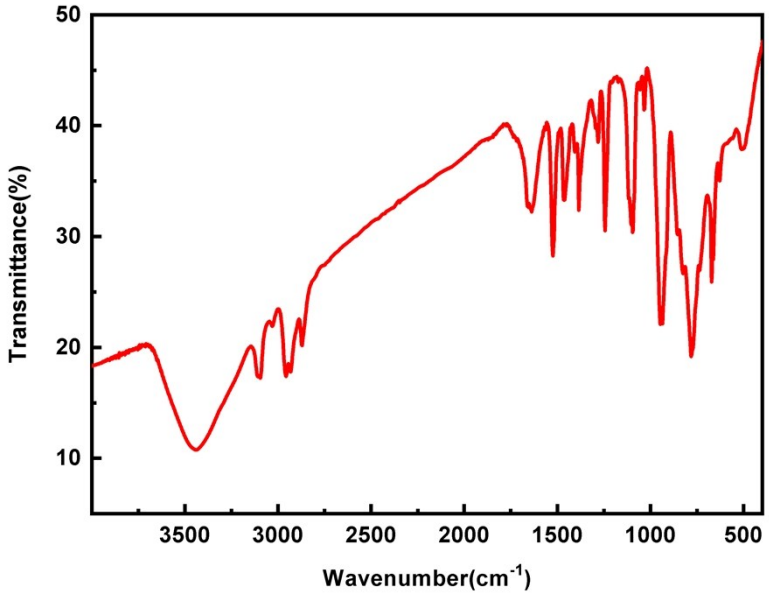

Figure S3. The FT-IR spectrum of compound **3**.

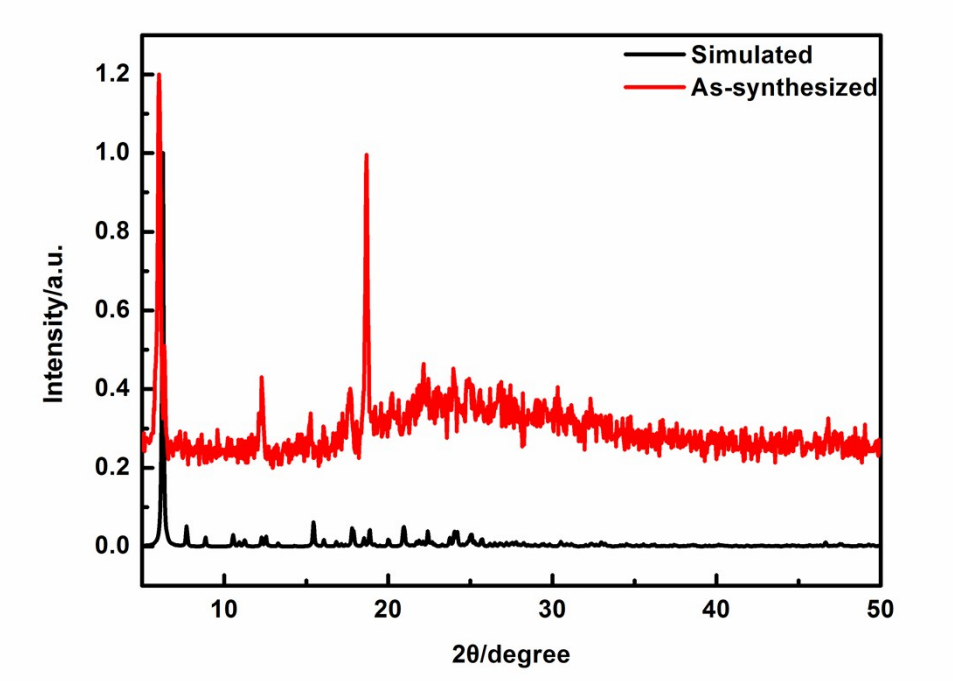

Figure S4. The simulated (black) and experimental (red) PXRD patterns of compound **1**.

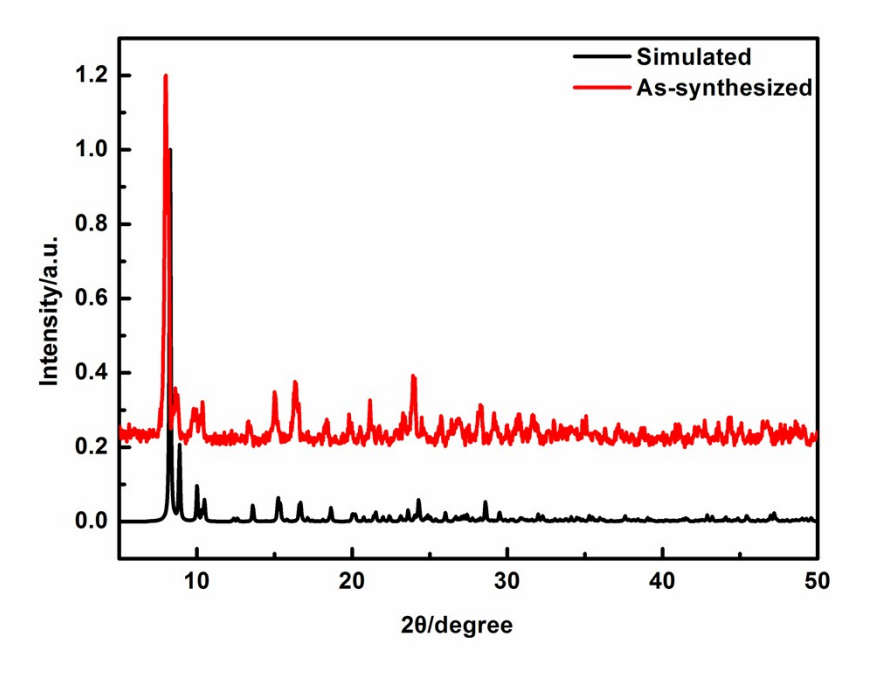

Figure S5. The simulated (black) and experimental (red) PXRD patterns of compound **2**.

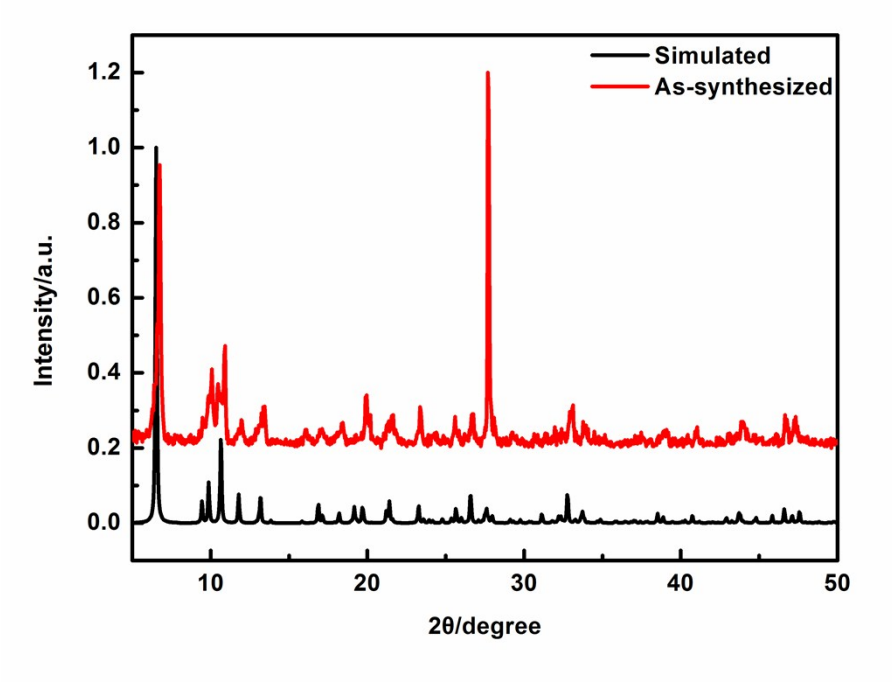

Figure S6. The simulated (black) and experimental (red) PXRD patterns of compound **3**.

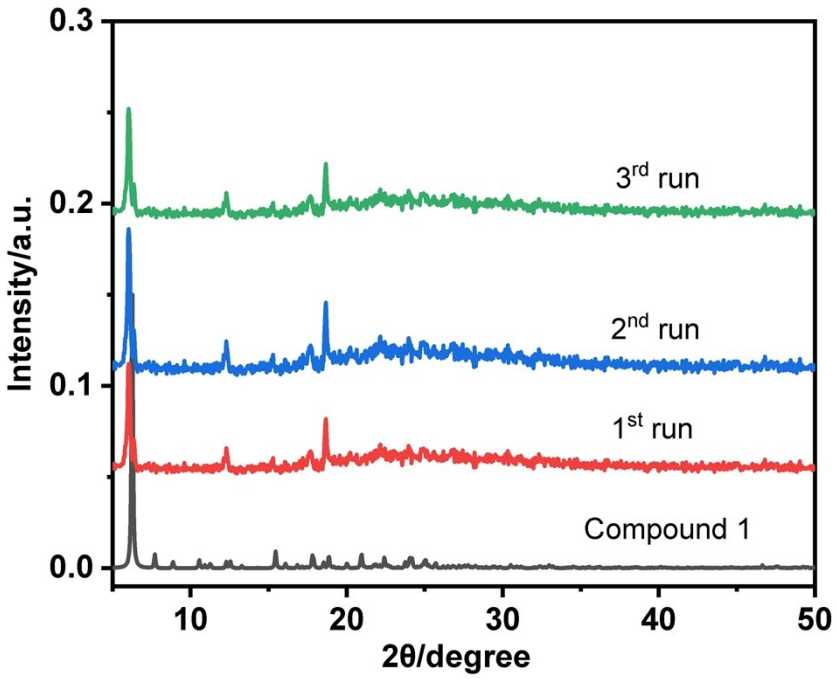

Figure S7. The PXRD patterns of compound **1** of three run cycles.

Figures S8-S21. The NMR spectra of the products in the Table 2

Entry 1. 2-benzylidenemalononitrile.

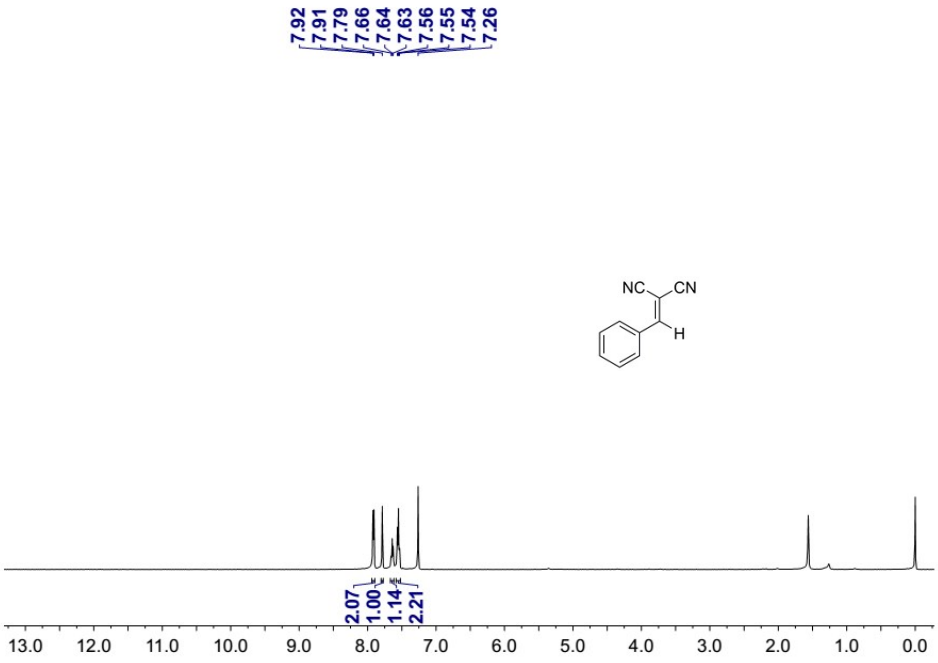

Figure S8. ^{1}H NMR spectrum of 2-benzylidenemalononitrile (entry 1) in $CDCl_3$.

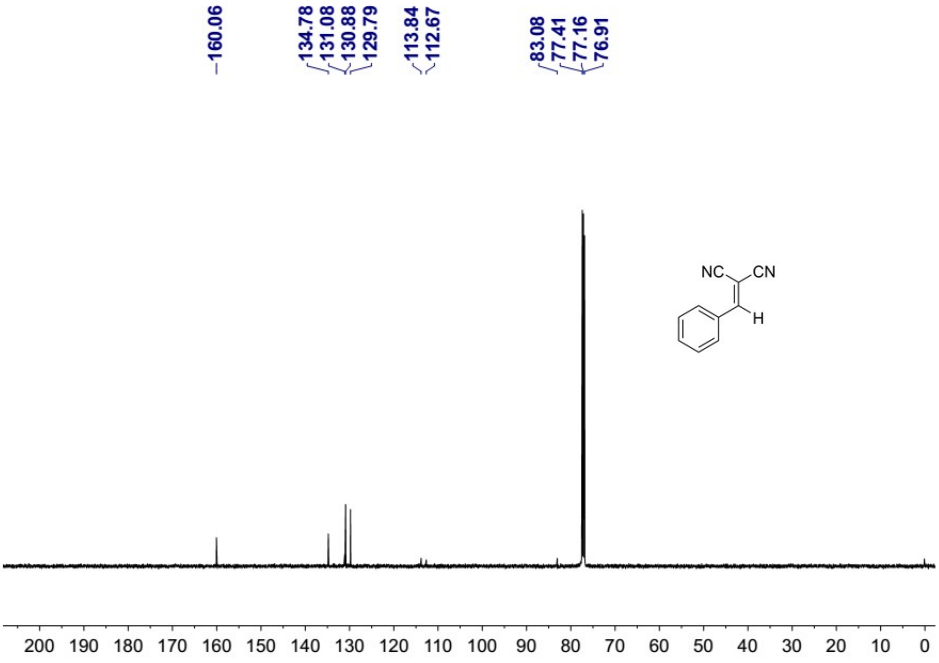

Figure S9. ^{13}C NMR spectrum of 2-benzylidenemalononitrile (entry 1) in $CDCl_3$.

Entry 2. 2-(4-nitrobenzylidene)malononitrile.

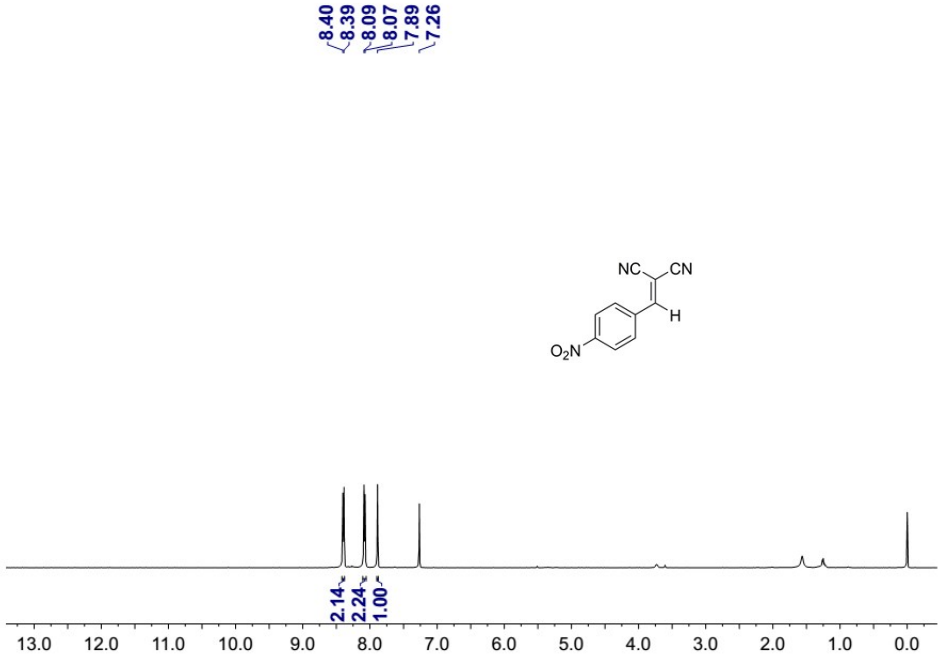

Figure S10. ^{1}H NMR spectrum of 2-(4-nitrobenzylidene)malononitrile (entry 2) in $CDCl_3$.

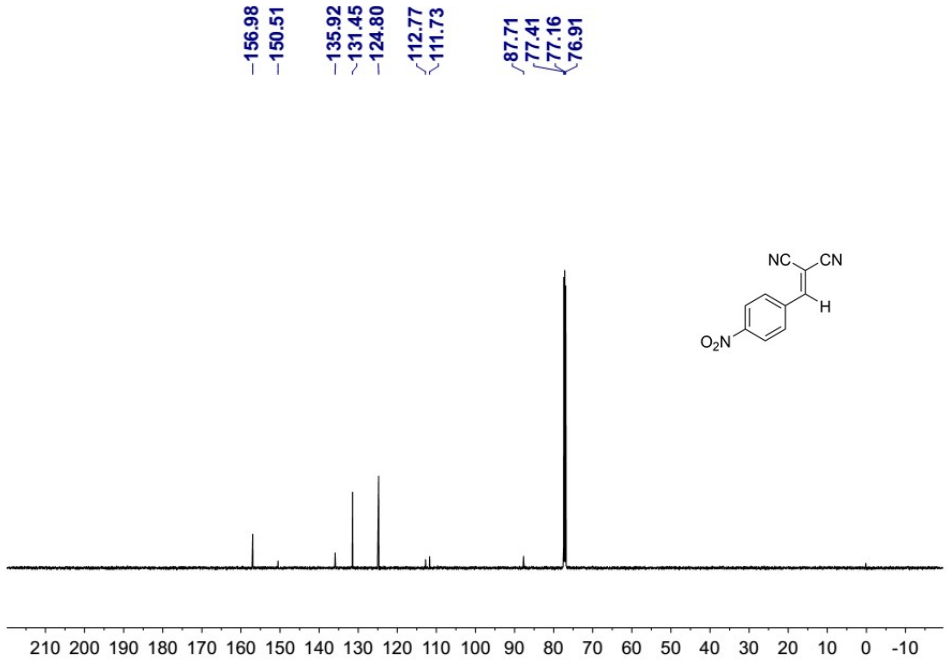

Figure S11. ^{13}C NMR spectrum of 2-(4-nitrobenzylidene)malononitrile (entry 2) in $CDCl_3$.

Entry 3. 2-(4-chlorobenzylidene)malononitrile.

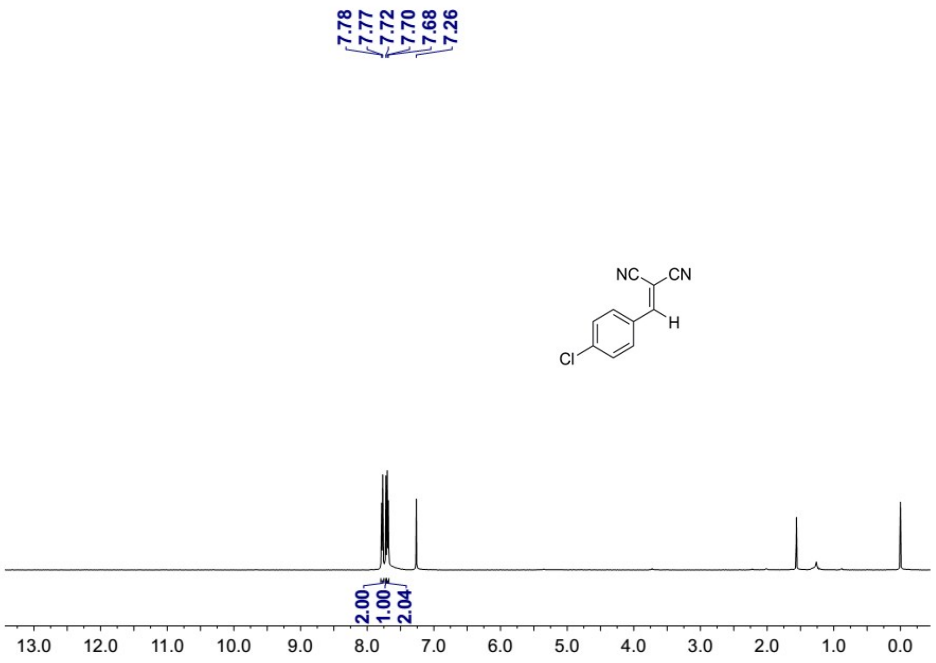

Figure S12. ^{1}H NMR spectrum of 2-(4-chlorobenzylidene)malononitrile (entry 3) in $CDCl_3$.

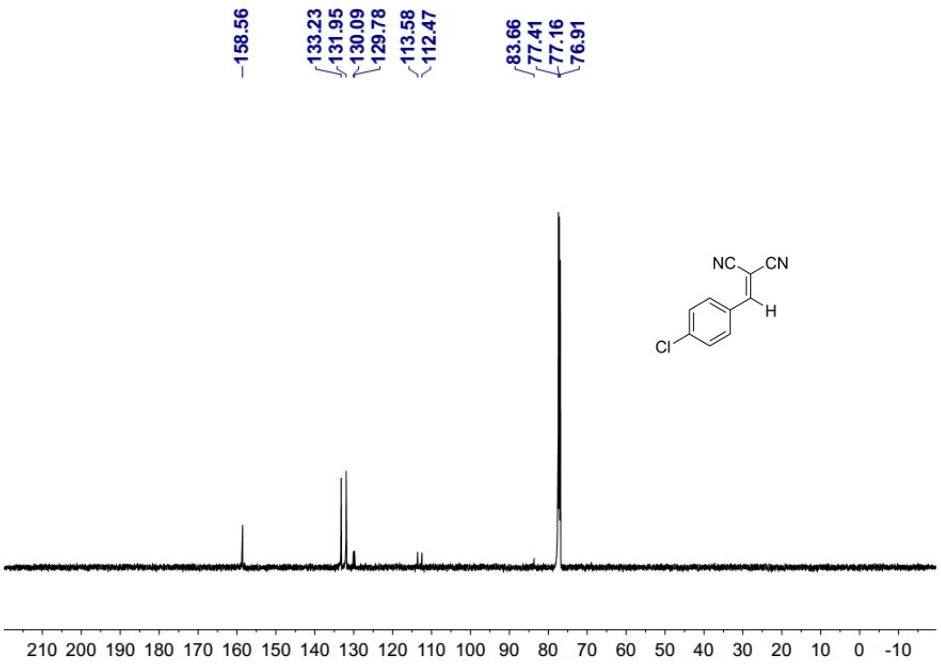

Figure S13. ^{13}C NMR spectrum of 2-(4-chlorobenzylidene)malononitrile (entry 3) in $CDCl_3$.

Entry 4. 2-(4-bromobenzylidene)malononitrile.

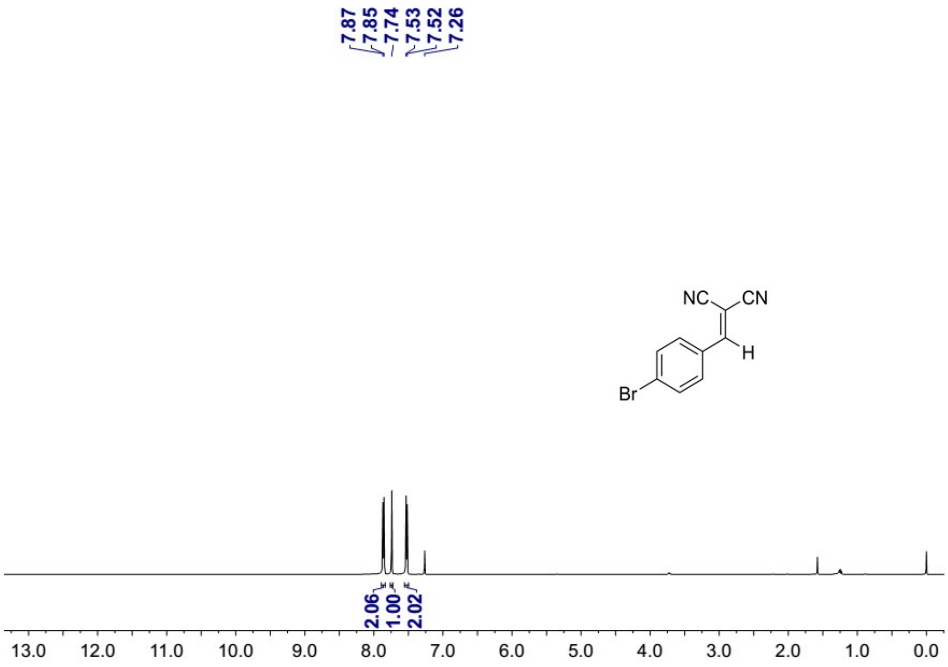

Figure S14. ^{1}H NMR spectrum of 2-(4-bromobenzylidene)malononitrile (entry 4) in $CDCl_3$.

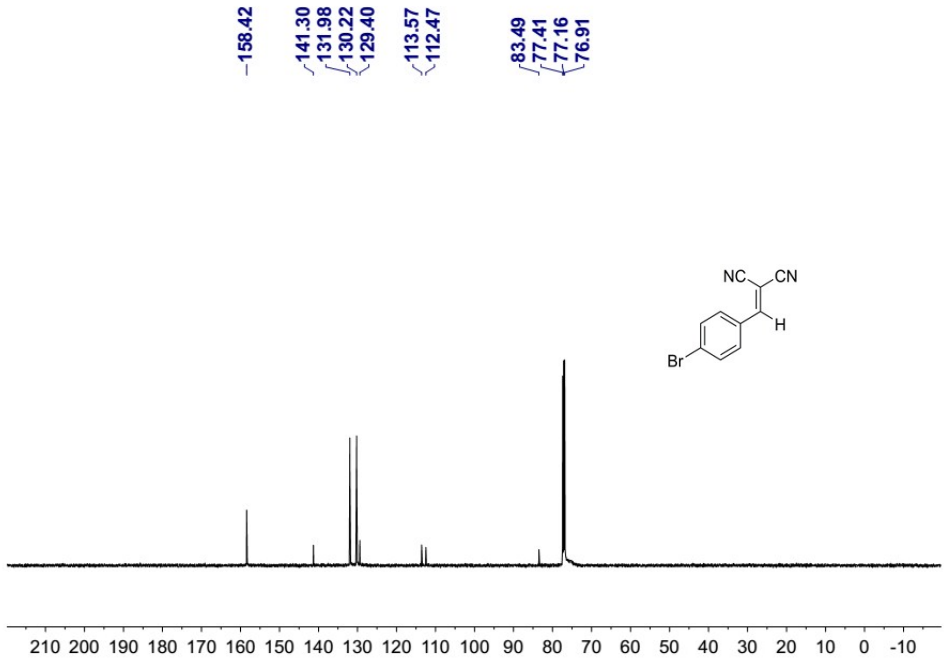

Figure S15. ^{13}C NMR spectrum of 2-(4-bromobenzylidene)malononitrile (entry 4) in $CDCl_3$.

Entry 5. 2-(2-bromobenzylidene)malononitrile.

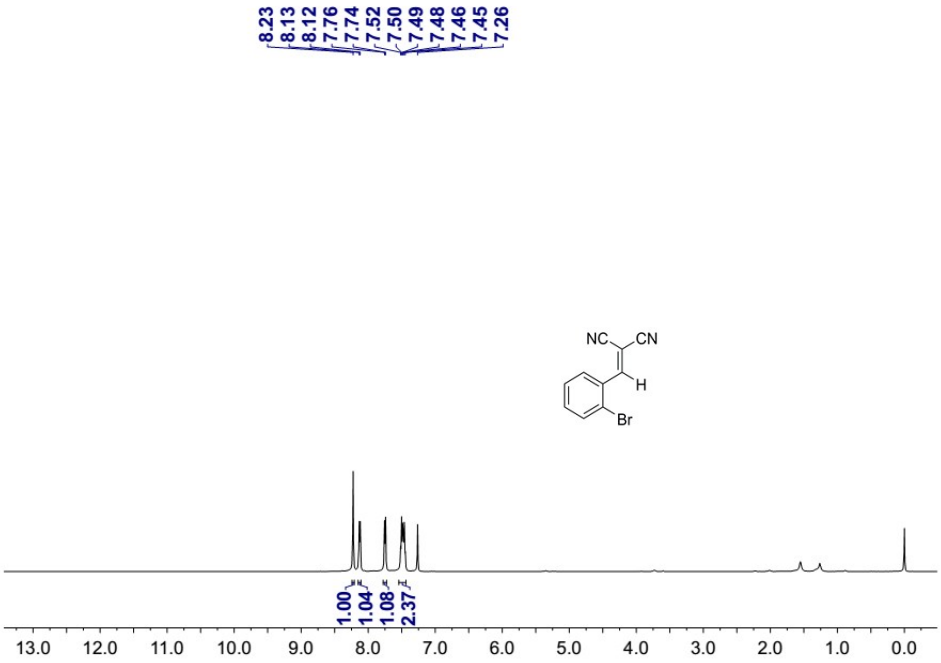

Figure S16. ^{1}H NMR spectrum of 2-(2-bromobenzylidene)malononitrile (entry 5) in $CDCl_3$.

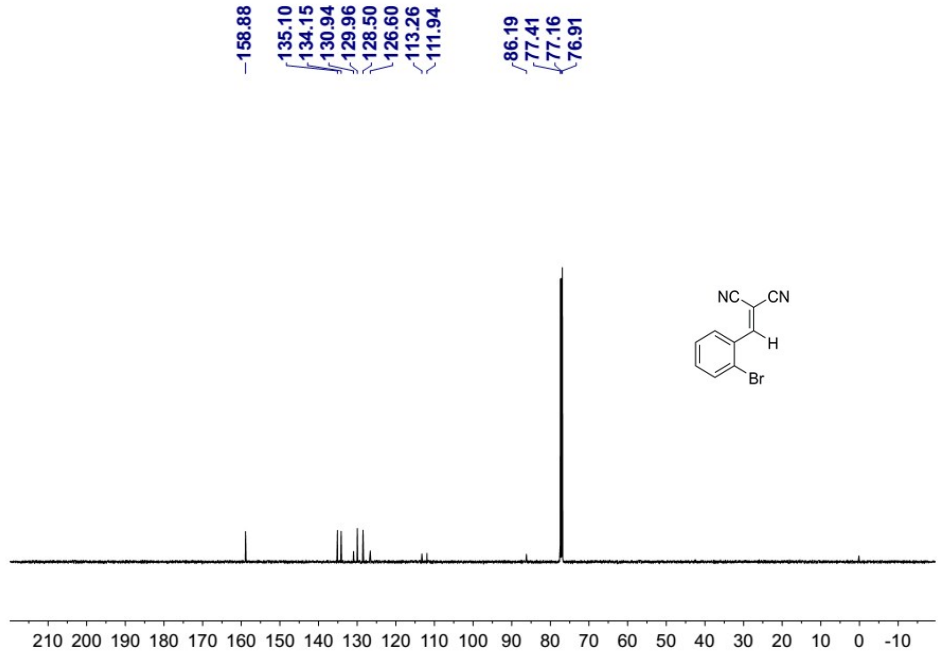

Figure S17. ^{13}C NMR spectrum of 2-(2-bromobenzylidene)malononitrile (entry 5) in $CDCl_3$.

Entry 6. 2-(4-methylbenzylidene)malononitrile.

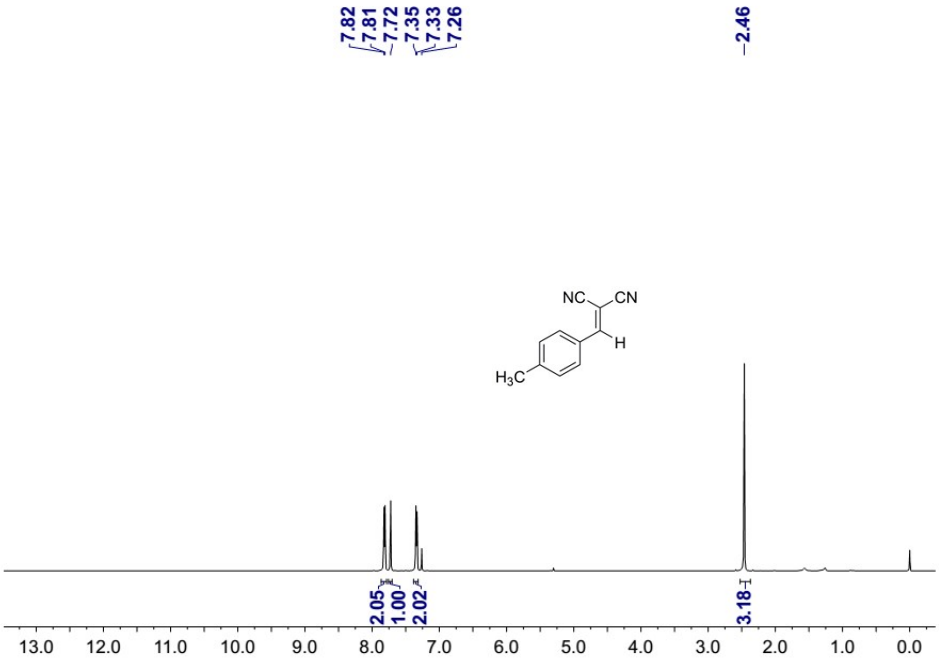

Figure S18. ^{1}H NMR spectrum of 2-(4-methylbenzylidene)malononitrile (entry 6) in $CDCl_3$.

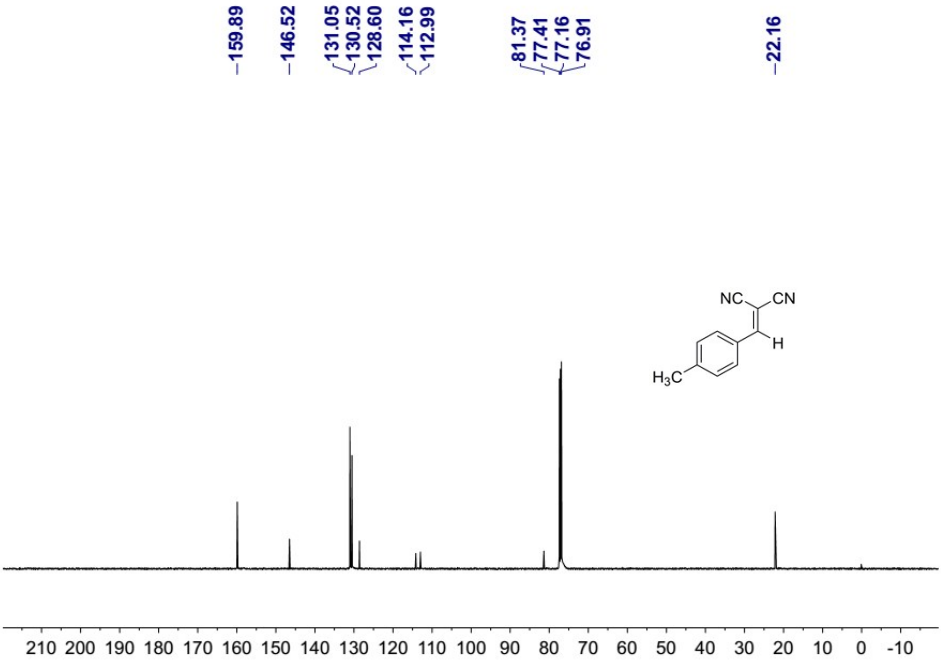

Figure S19. ^{13}C NMR spectrum of Entry 2-(4-methylbenzylidene)malononitrile (entry 6) in $CDCl_3$.

Entry 7. 2-(4-methoxybenzylidene)malononitrile.

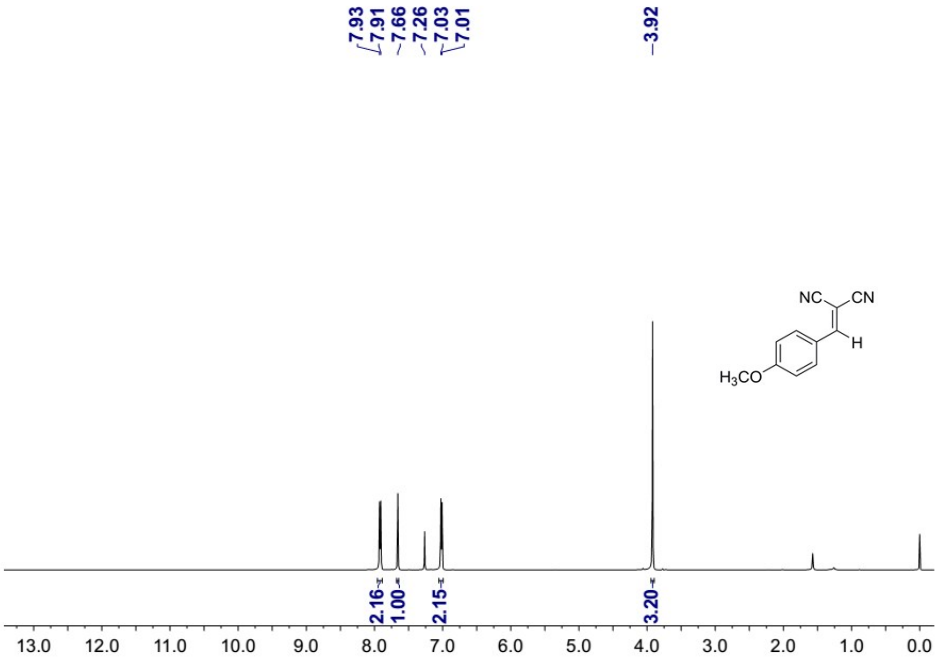

Figure S20. ^{1}H NMR spectrum of 2-(4-methoxybenzylidene)malononitrile (entry 7) in $CDCl_3$.

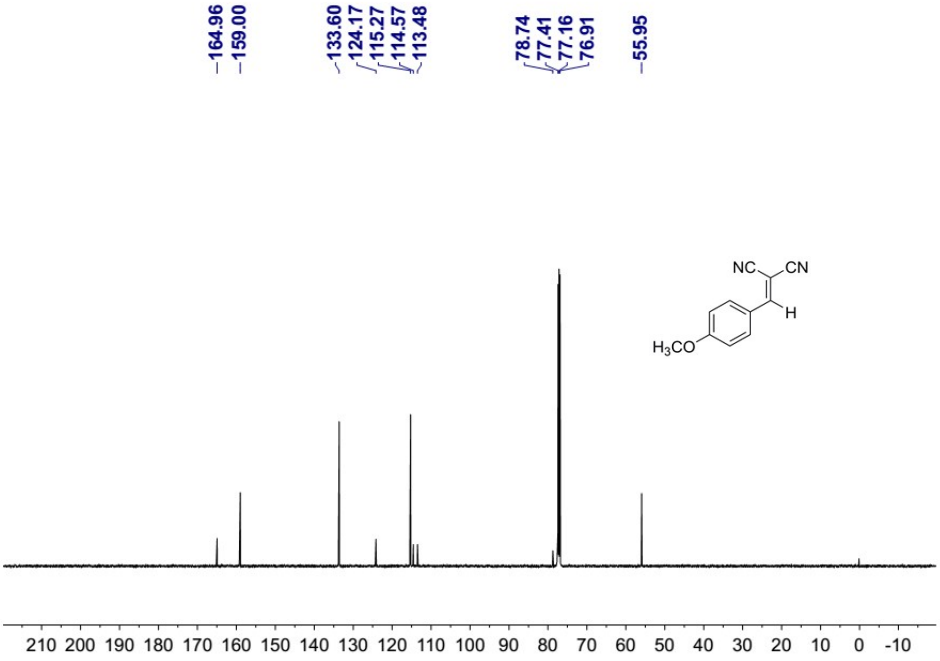

Figure S21. ^{13}C NMR spectrum of 2-(4-methoxybenzylidene)malononitrile (entry 7) in $CDCl_3$.