Supporting Information

Size and Function Influence Study on Enhanced Catalytic Performance of a Cooperative MOF for Mild, Green and Fast C-C Bond Formation

Najmeh Varnaseri [†a], Farzaneh Rouhani [†b], Ali Morsali [b*], Ali Ramazani [a, c*]

[a] Department of Chemistry, Faculty of Sciences, University of Zanjan, P.O. Box 45195-313, Zanjan, Iran

[b] Department of Chemistry, Faculty of Sciences, Tarbiat Modares University, P.O. Box 14155-4838, Tehran, Iran

[c] Research Institute of Modern Biological Techniques (RIMBT), University of Zanjan, P.O. Box 10

45195-313, Zanjan, Iran

[†] Najmeh Varnaseri and Farzaneh Rouhani contributed equally to this work.

Materials and methods

All raw materials were purchased from Merck and Aldrich and were used without further purification. X-Ray powder diffraction (XRD) patterns were recorded using a Philips X'pert diffractometer with monochromated Cu-kα radiation. The IR spectra (in KBr) have been recorded by Nicolet Fourier Transform IR, Nicolet 100 spectrometer in the range of 4000-400 cm^{-1}. The thermogravimetric behavior of the structures (TGA) was evaluated using a thermal gravimetric analysis instrument (PL-STA 1500) into alumina pans with the gradient of 10 ^{o}C/ min under neutral N_2 atmosphere. The morphologies of the nanostructures samples were examined with scanning electron microscopy (SEM) (Philips XL 30). ^{1}H NMR spectra were recorded on a BRUKER DRX-250 AVANCE spectrometer operating at 250 MHz for the proton nucleus at room temperature.

1. Synthesis of N,N'-(oxybis(4,1-phenylene))bis(1-(pyridin-4-yl)methanimine) (OPP)

* *E-mail address*: morsali_a@modares.ac.ir (A. Morsali);

aliramazani@znu.ac.ir (A. Ramazani).

4,4′-Oxydianiline (1mmol, 0.2 g) and 4-Pyridinecarboxaldehyde (2.1 mmol, 1mL) was dissolved in 15 ml ethanol and the mixture was stirred for 4 h in room temperature. The yellow residue was filtered and washed with ethanol and dried at 80 °C in oven overnight.

2. Synthesis of N_1, N_2-Bis (pyridin-4-ylmethylene) ethane-1,2-diamine(L)

1,2-Ethylenediamine (1mmol, 0.66mL) dissolved in 20 mL of ethanol then 4-pyridinecarboxaldehyde (1mmol, 1mL) added to the mixture and refluxed for 3h. The yellow solid washed with hexane and dried.

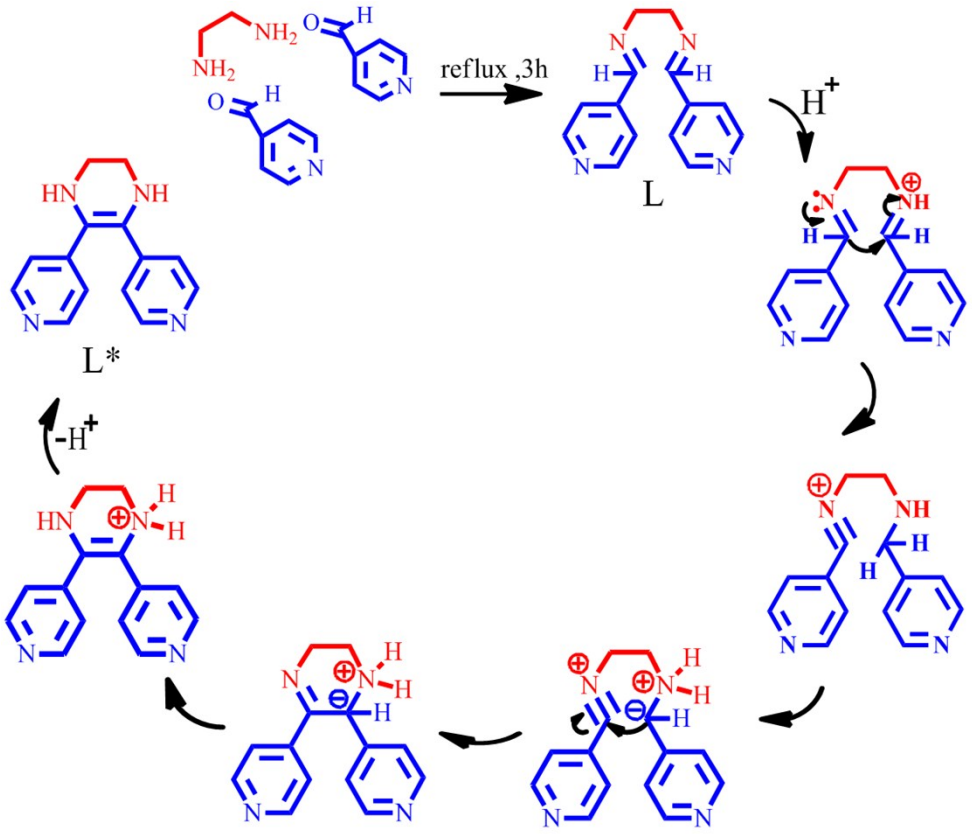

Figure S1. Synthesis rout of L and C=C coupling reaction of L to L**

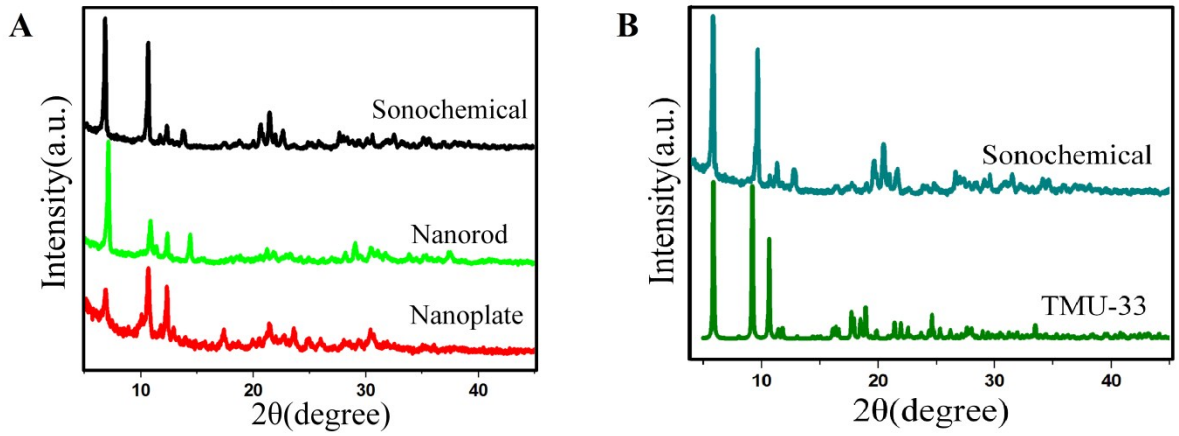

Figure S2. PXRD patterns for A) nanorod and nanoplate sample B) the as-synthesized sonochemical

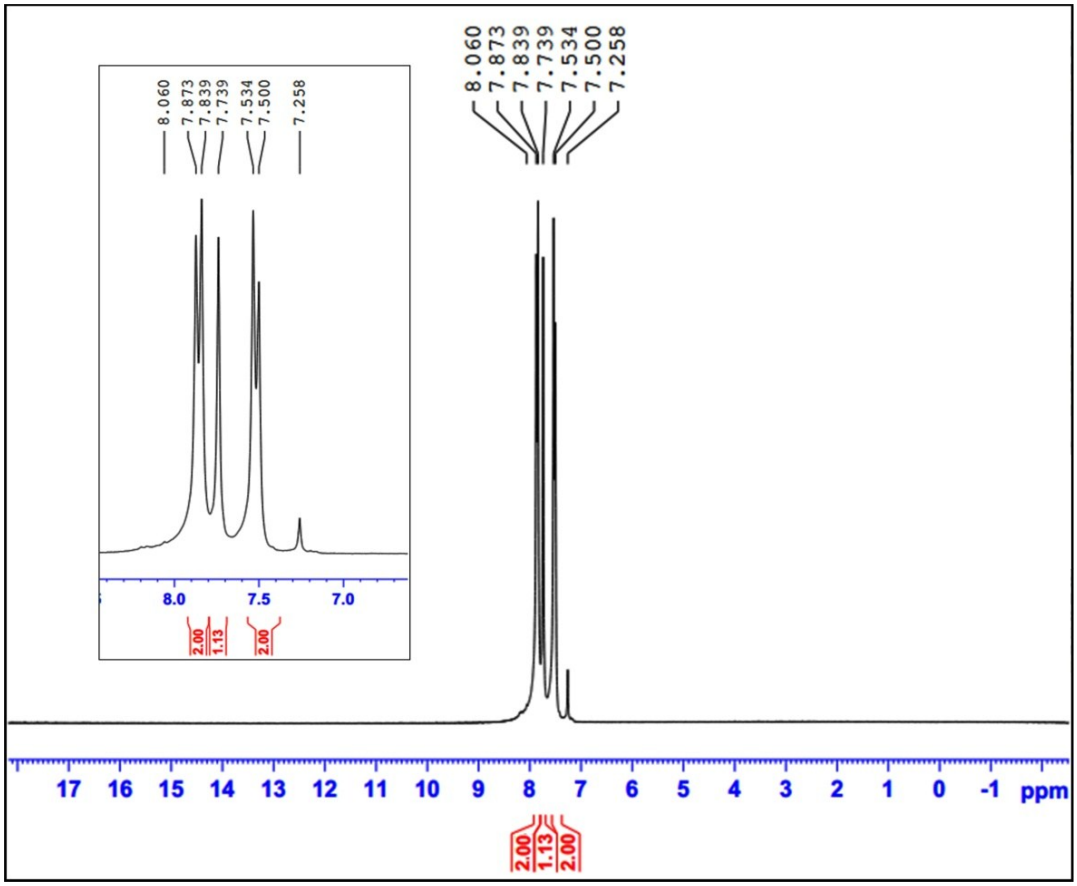

Figure S3. ^{1}H NMR spectrum of as synthesized 2-(4-Chlorobenzylidene) malononitrile in $CDCl_3$.

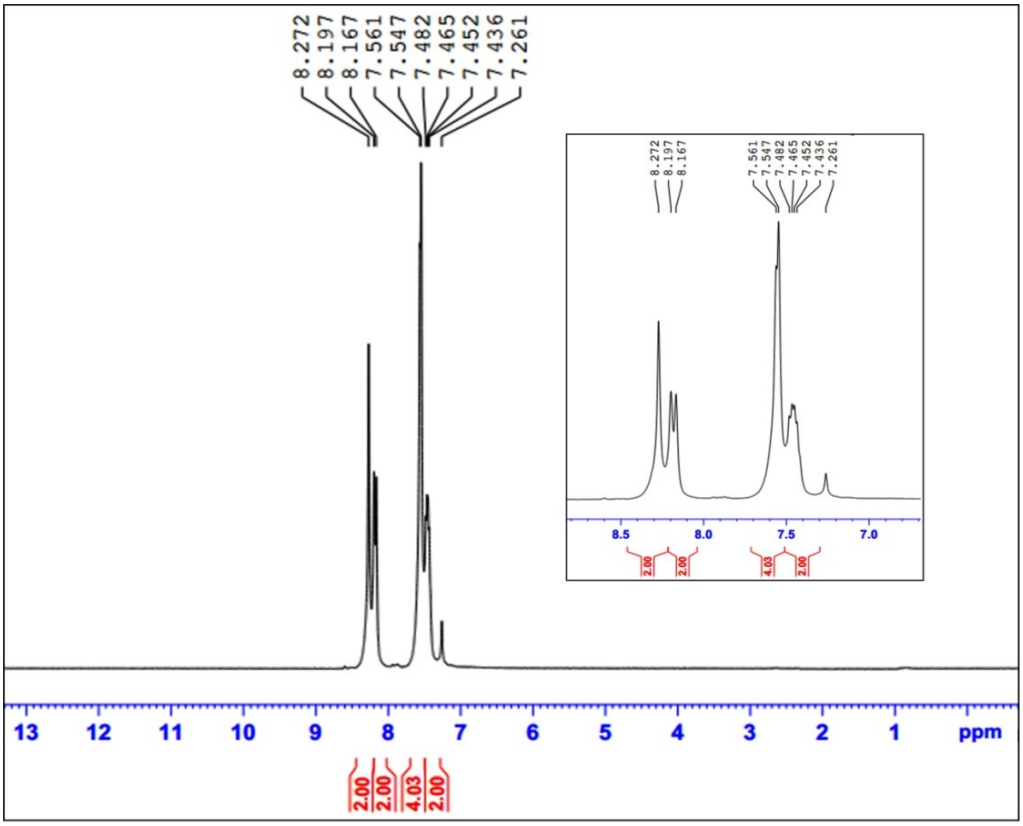

Figure S4. ^{1}H NMR spectrum of as synthesized 2-(2-Chlorobenzylidene) malononitrile in $CDCl_3$.

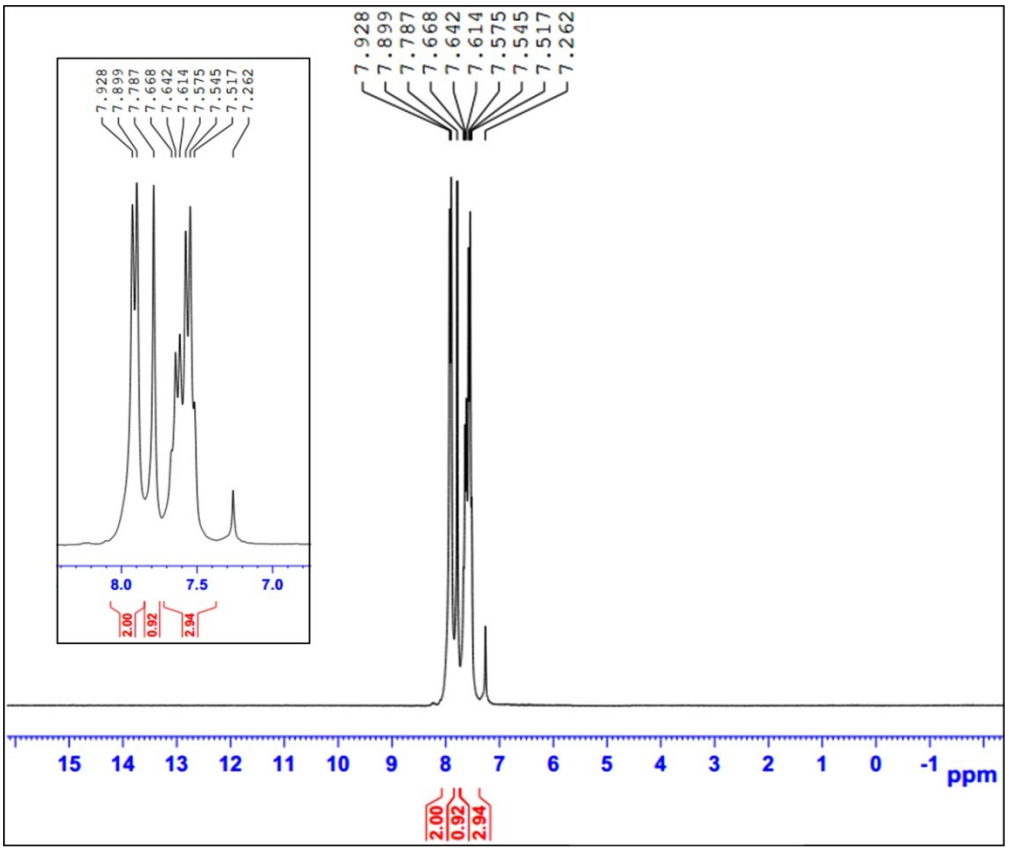

Figure S5. ^{1}H NMR spectrum of as synthesized 2-benzylidenemalononitrile in $CDCl_3$.

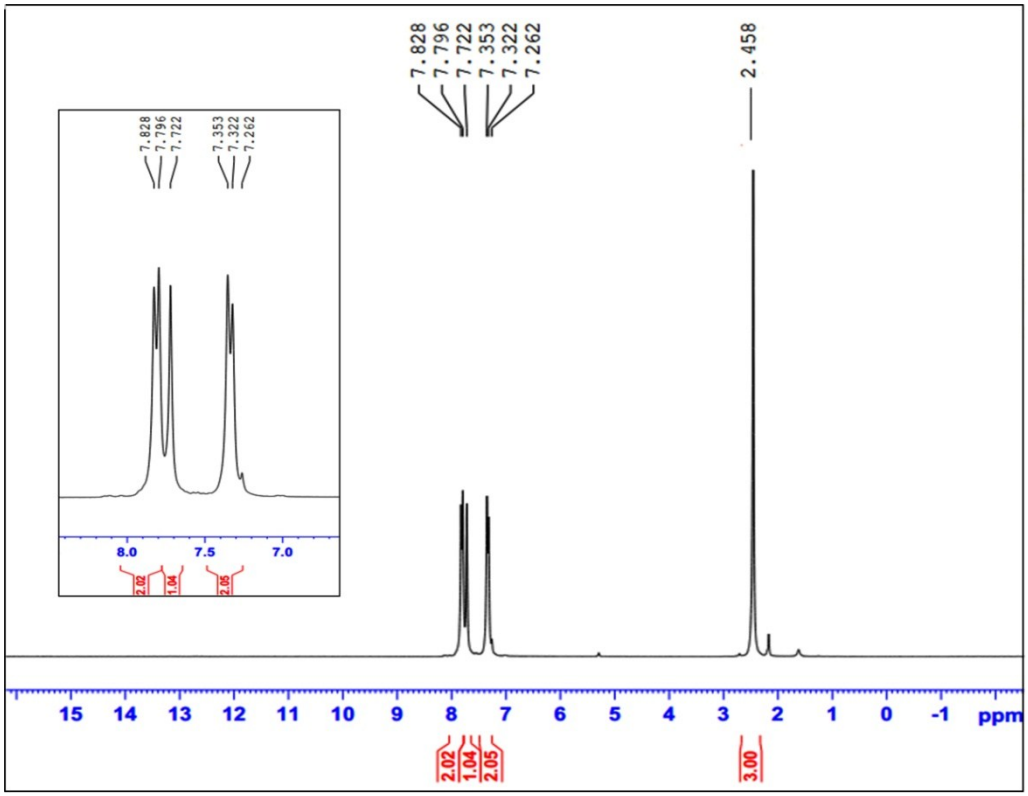

Figure S6. [1]H NMR spectrum of as synthesized 2-(4-methylbenzylidene) malononitrile in $CDCl_3$.

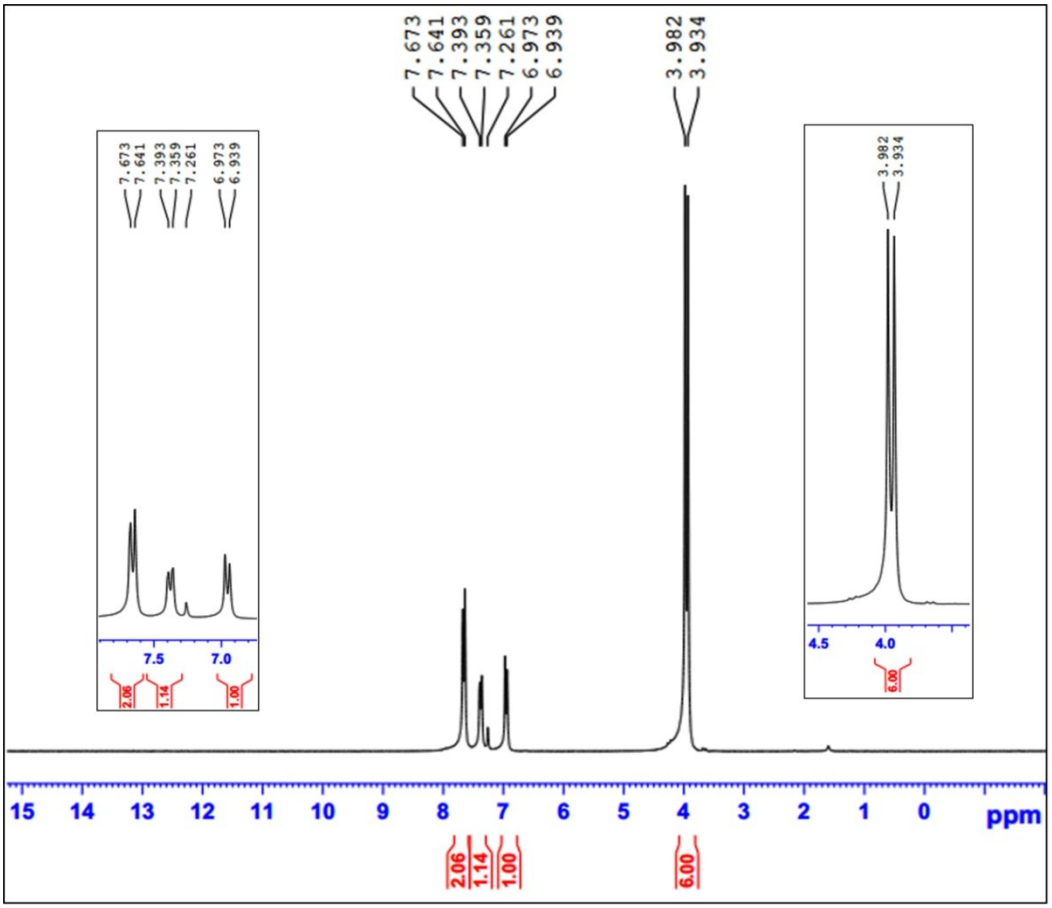

Figure S7. [1]H NMR spectrum of as synthesized 2-(3,4-methoxybenzylidene)malononitrile in $CDCl_3$.

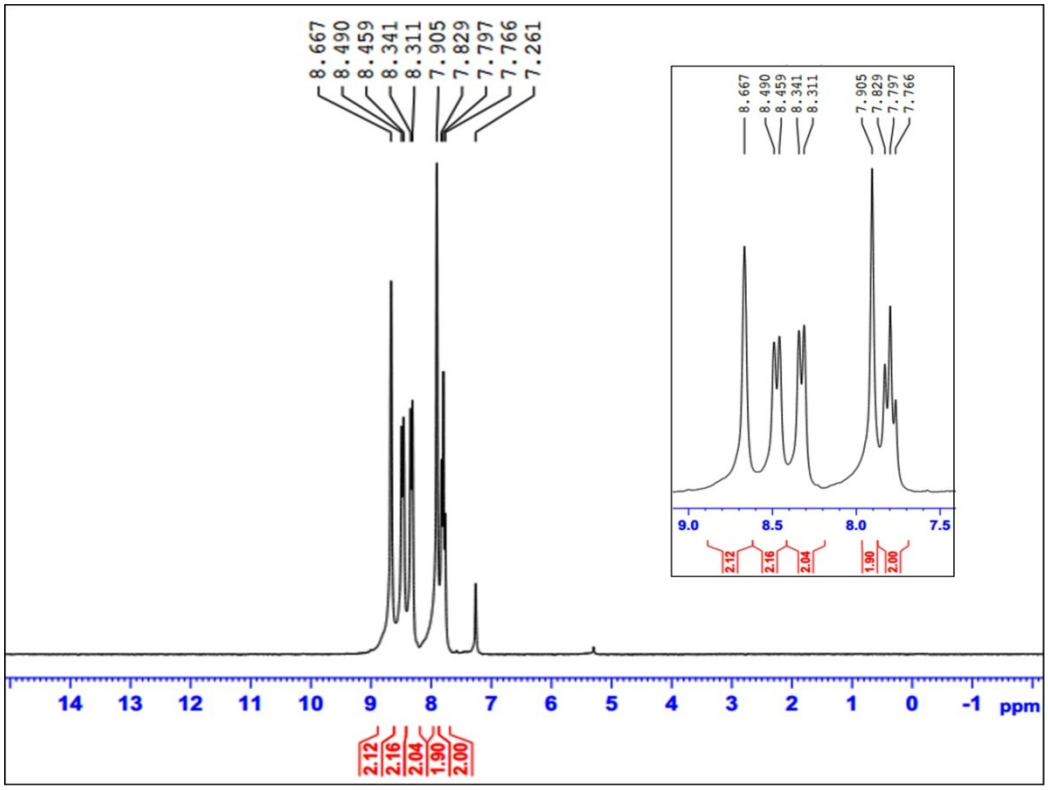

Figure S8. ^{1}H NMR spectrum of as synthesized 2-(3-nitrobenzylidene) malononitrile in $CDCl_3$.

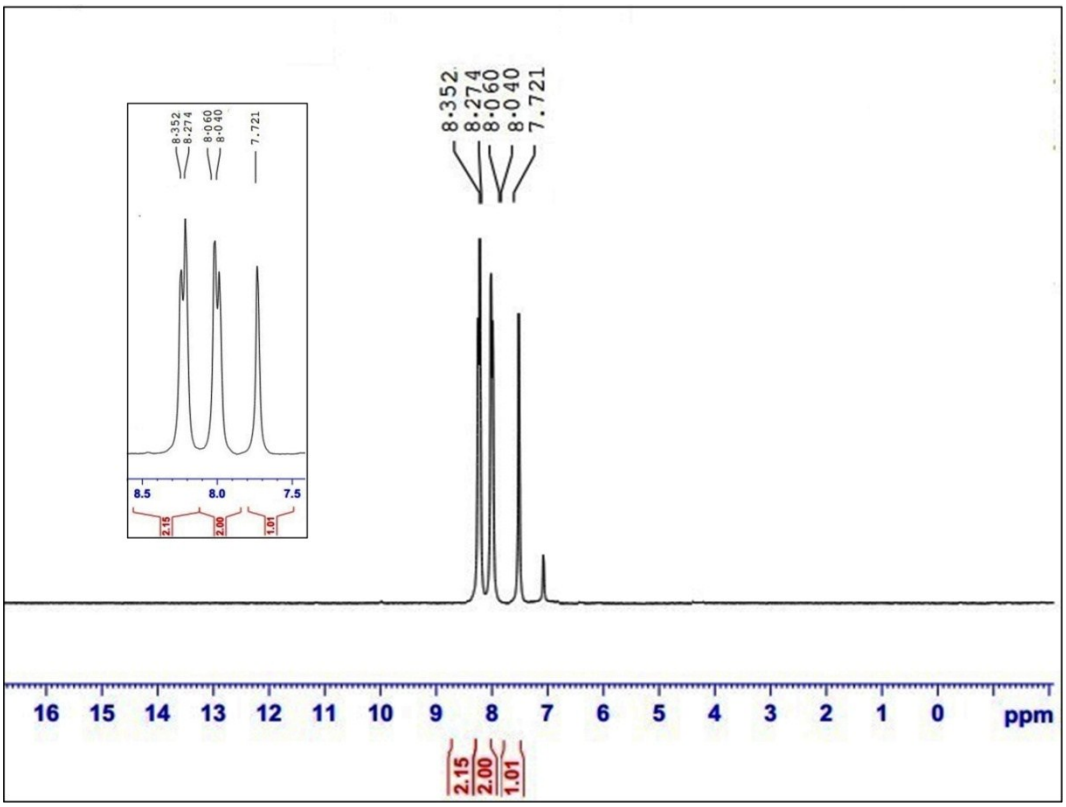

Figure S9. ^{1}H NMR spectrum of as synthesized 2-(4-nitrobenzylidene) malononitrile in $CDCl_3$.

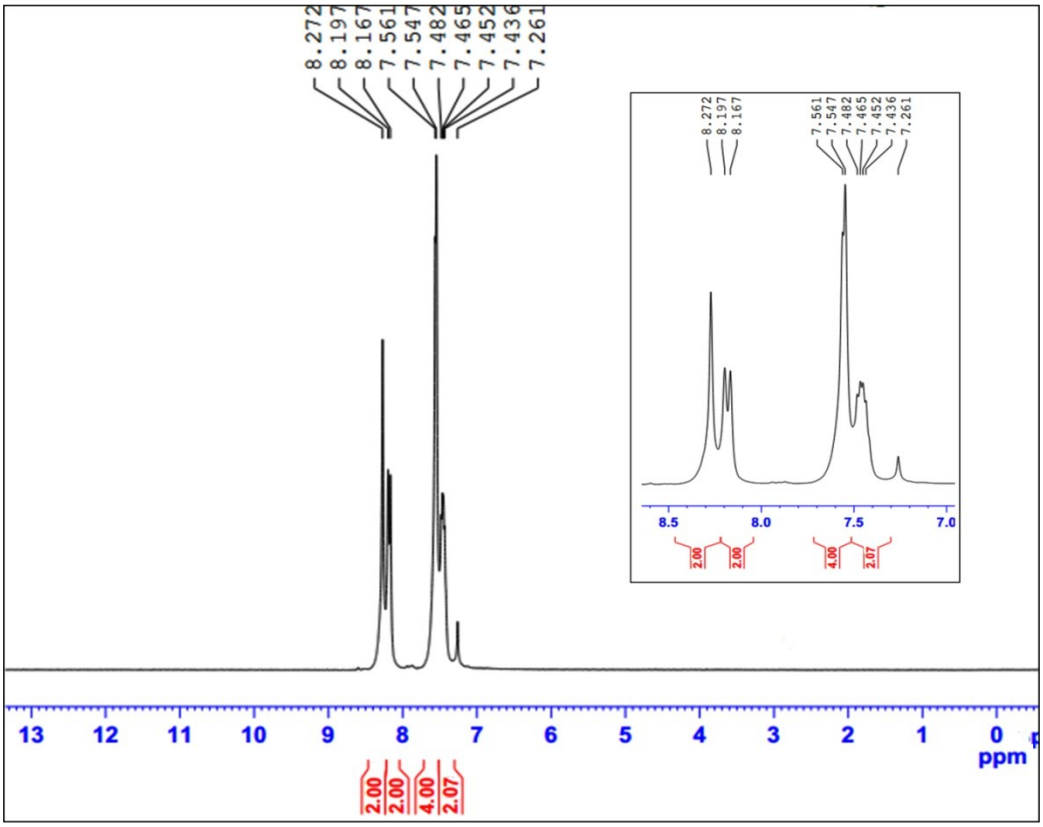

Figure S10. [1]HNMR spectrum of as synthesized 2-(2,4-dichlorobenzylidene)malononitrile in $CDCl_3$.

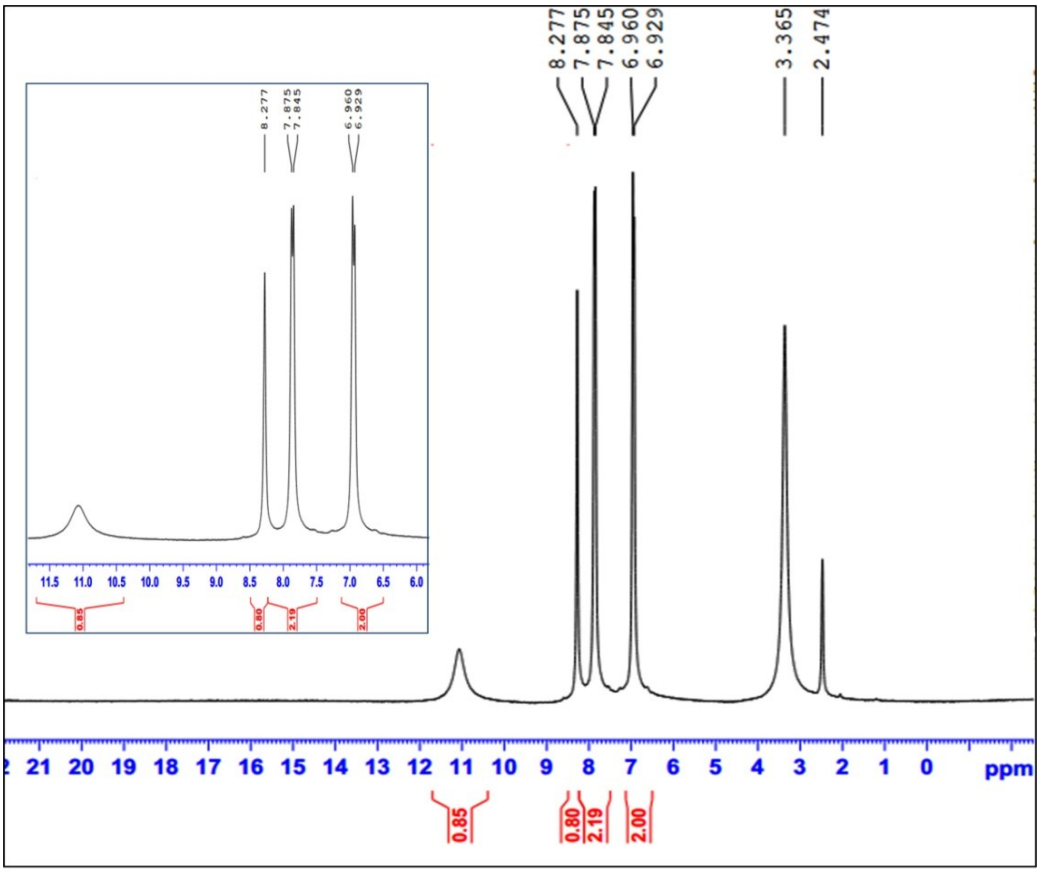

Figure S11. [1]H NMR spectrum of as synthesized 2-(4-hydroxybenzylidene) malononitrile in DMSO.

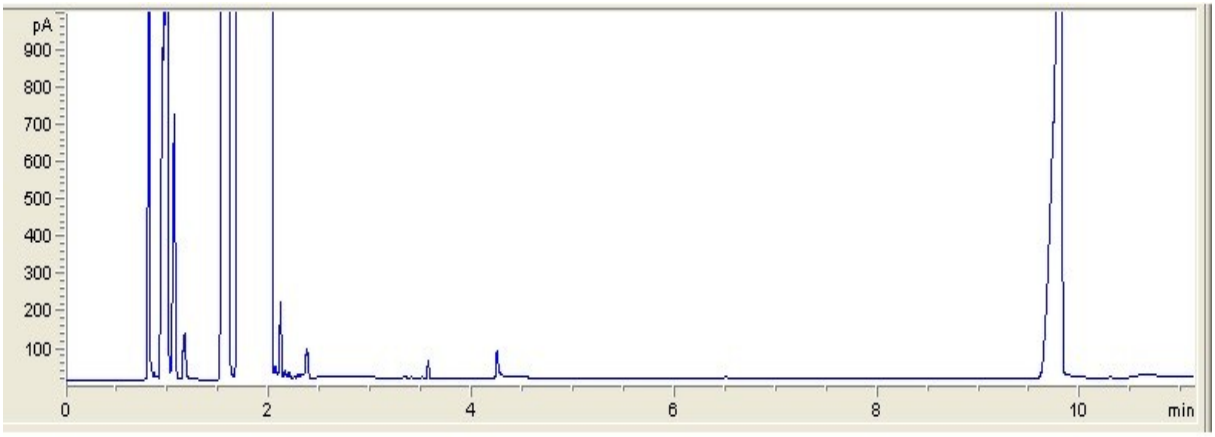

Figure S12. Photograph of reaction progress of 4-chlorobenzaldehyde and malononitrile.

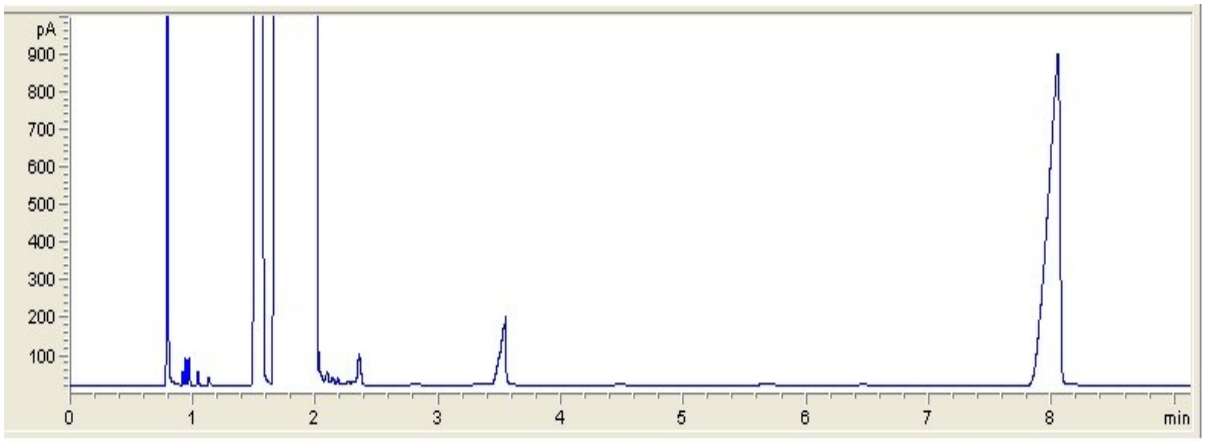

Figure S13. Photograph of reaction progress of 2-chlorobenzaldehyde and malononitrile.

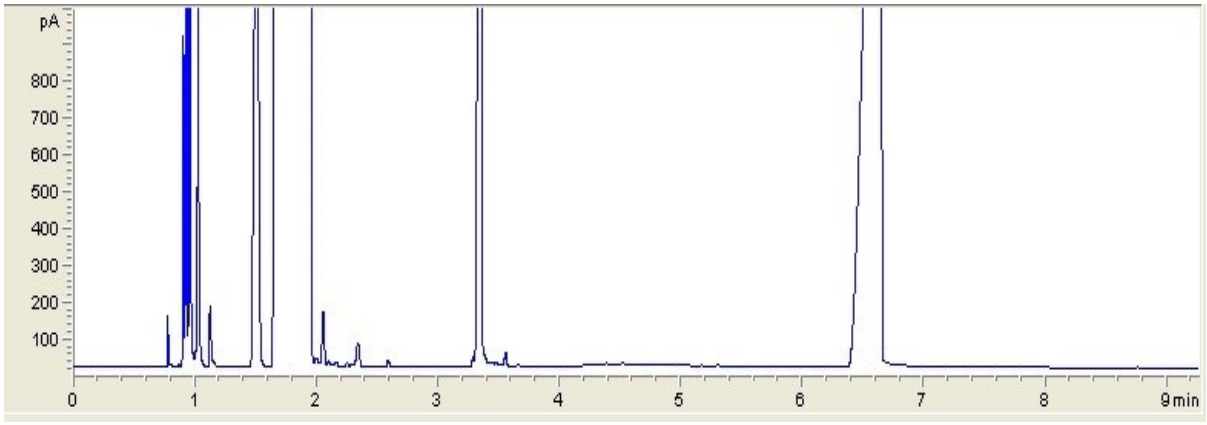

Figure S14. Photograph of reaction progress of benzaldehyde and malononitrile.

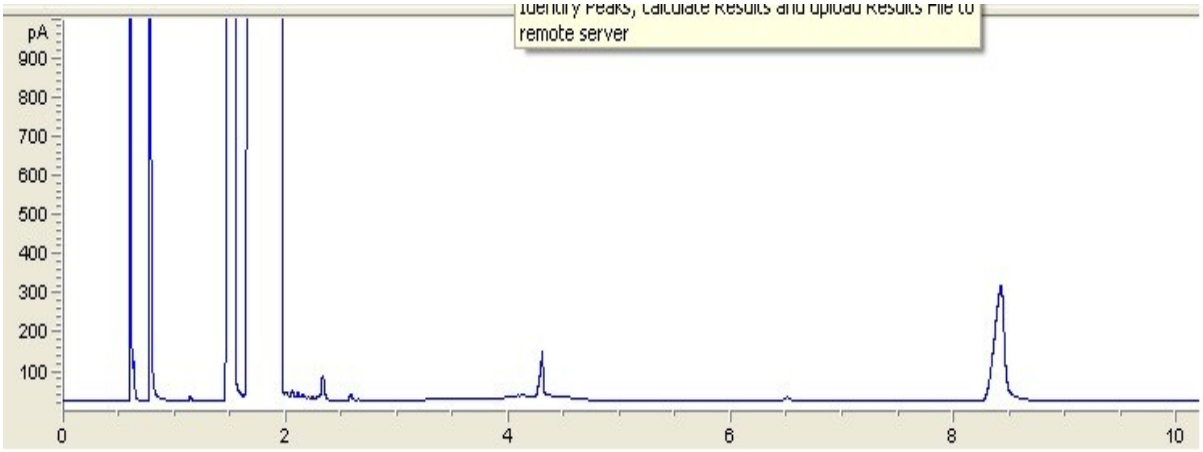

Figure S15. Photograph of reaction progress of 4-methylbenzaldehyde and malononitrile.

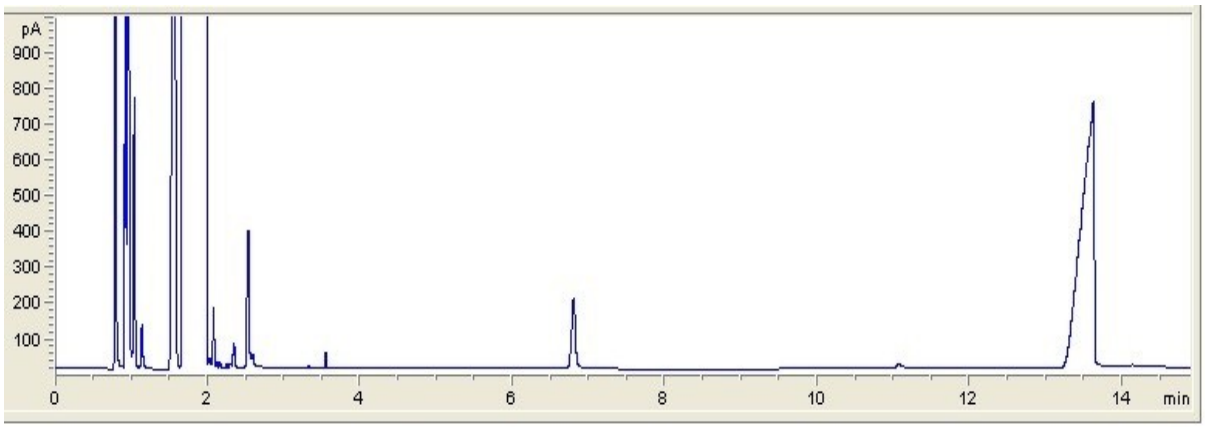

Figure S16. Photograph of reaction progress of 3,4-methoxylbenzaldehyde and malononitrile.

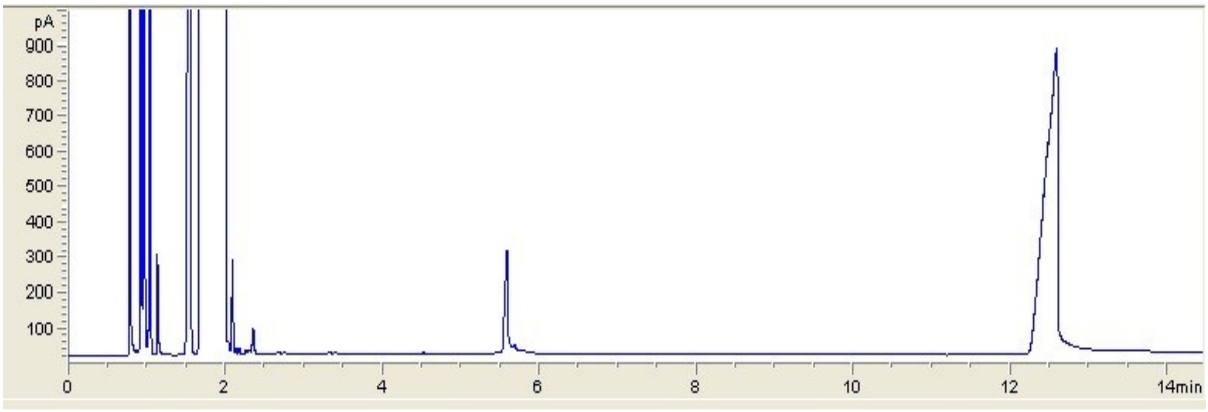

Figure S17. Photograph of reaction progress 3-nitrolbenzaldehyde and malononitrile.

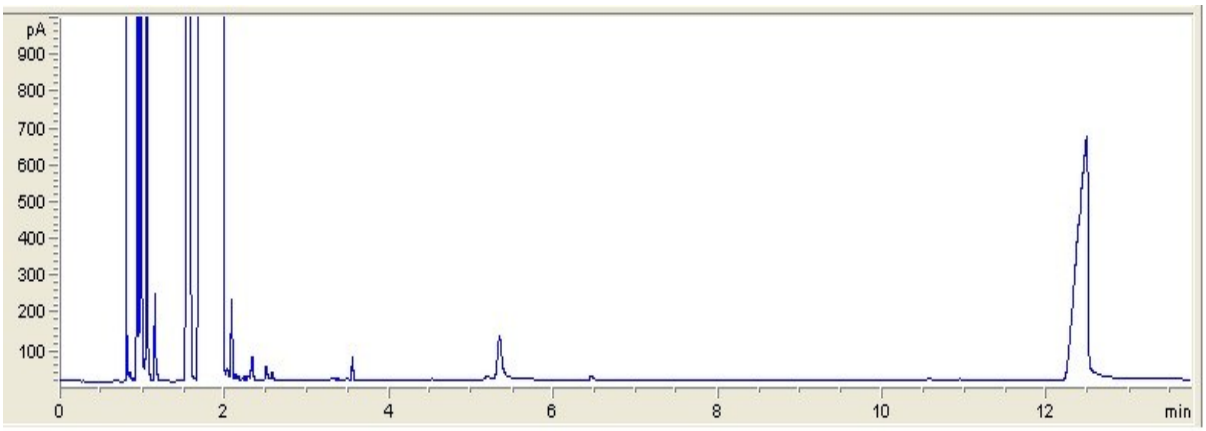

Figure S18. Photograph of reaction progress 3-nitrolbenzaldehyde and malononitrile.

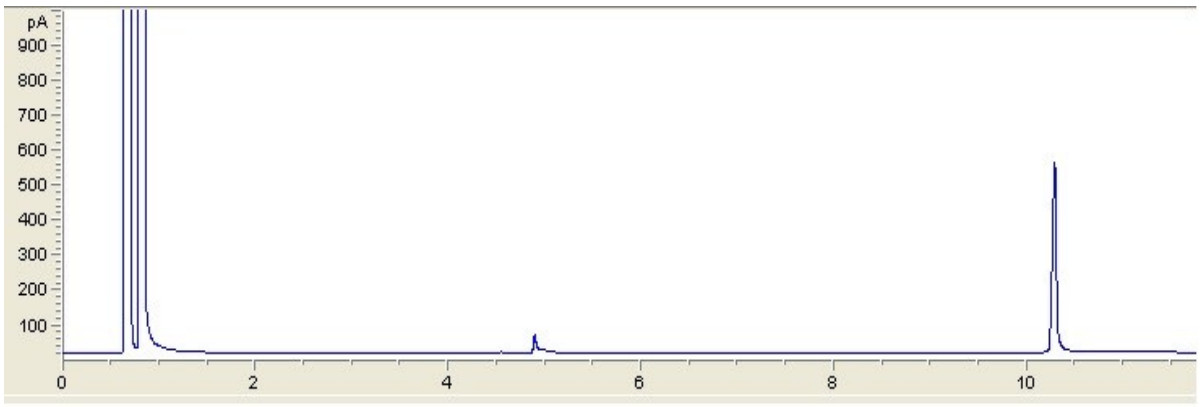

Figure S19. Photograph of reaction progress 2,4-dichlorobenzaldehyde and malononitrile.

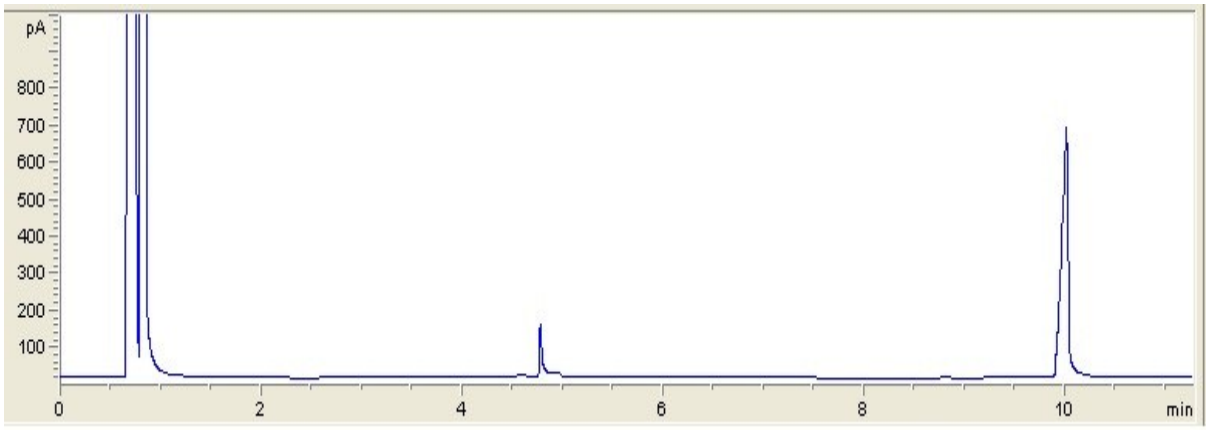

Figure S20. Photograph of reaction progress 4-hydroxybenzaldehyde and malononitrile.

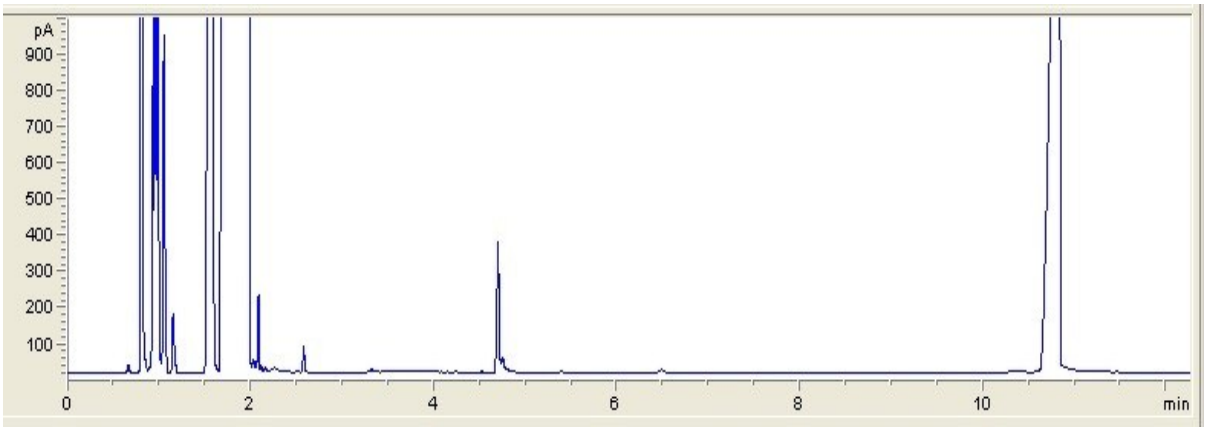

Figure S21. Photograph of reaction progress 4-bromobenzaldehyde and malononitrile

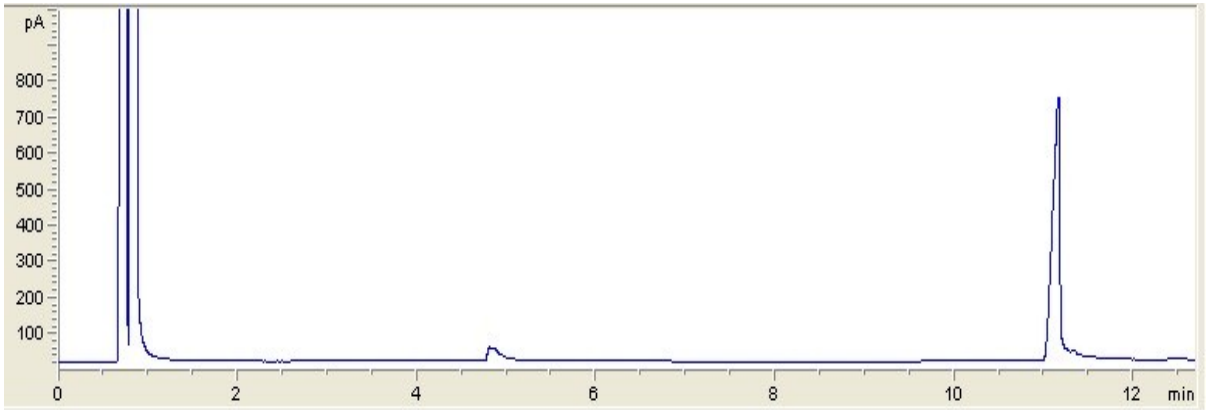

Figure S22. Photograph of reaction progress 4-cyanobenzaldehyde and malononitrile.

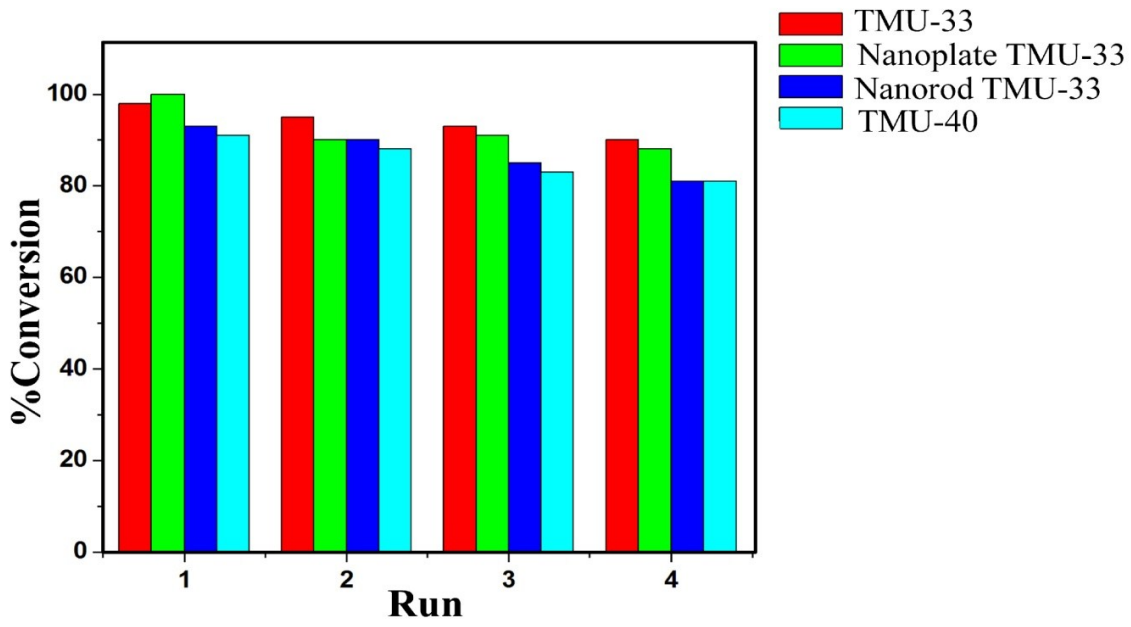

Figure S23. Bar chart of GC conversion (%) for various runs in the recycle and reuse of structures