

Supplementary Materials

A New Mixed Metal Phosphate as Heterogeneous Catalyst for Knoevenagel Condensation

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Table S1. Indexing of triclinic phase of MALPO with space group *P*1.

h	k	l	d (Å)	2θ
0	0	1	8.768	10.08
1	0	1	8.010	11.04
2	0	1	6.927	12.77
2	1	0	6.700	13.21
3	1	1	4.677	18.96
3	0	-2	4.202	21.12
0	2	0	3.841	23.13
0	2	1	3.518	25.29
5	1	-2	3.333	26.72
7	1	1	3.098	28.79
0	0	3	2.922	30.56
9	1	-1	2.805	31.88
8	2	-1	2.528	35.48
8	1	2	2.394	37.53
3	1	-4	2.121	42.58

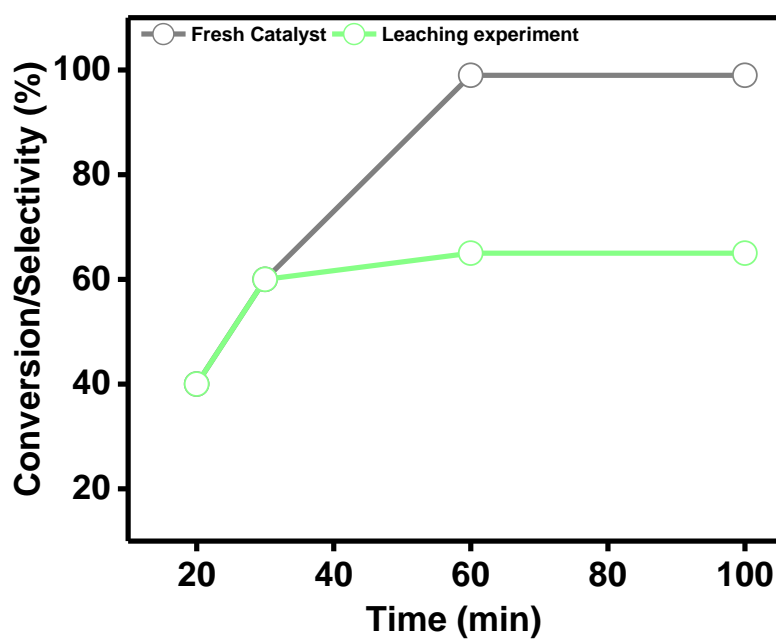


Figure S1. Leaching experiment indicating no leaching of active sites during catalysis.

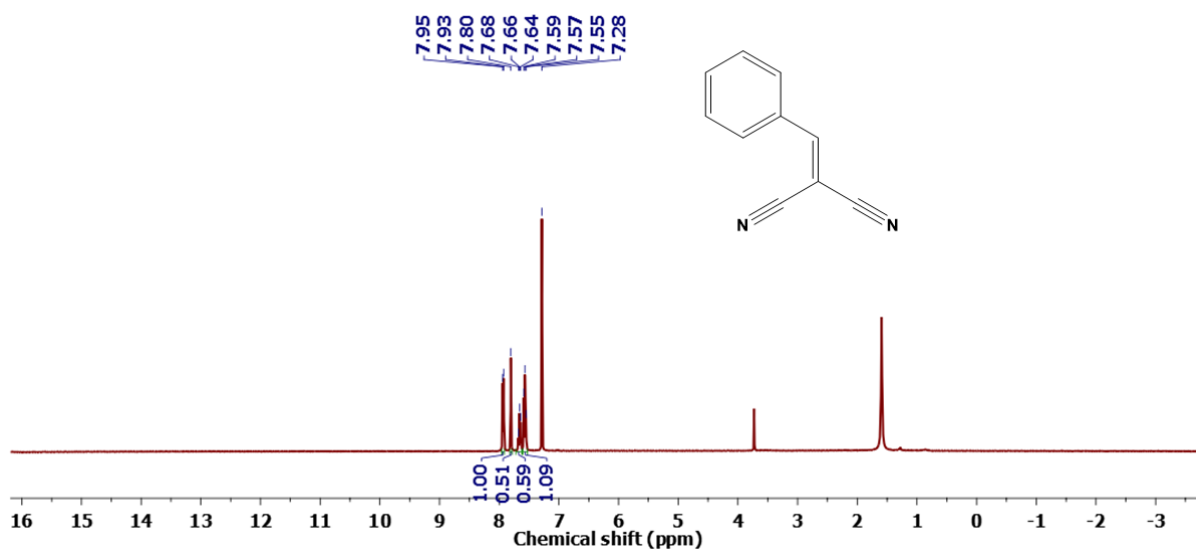


Figure S2. ¹H NMR of 2-benzylidenemalononitrile.

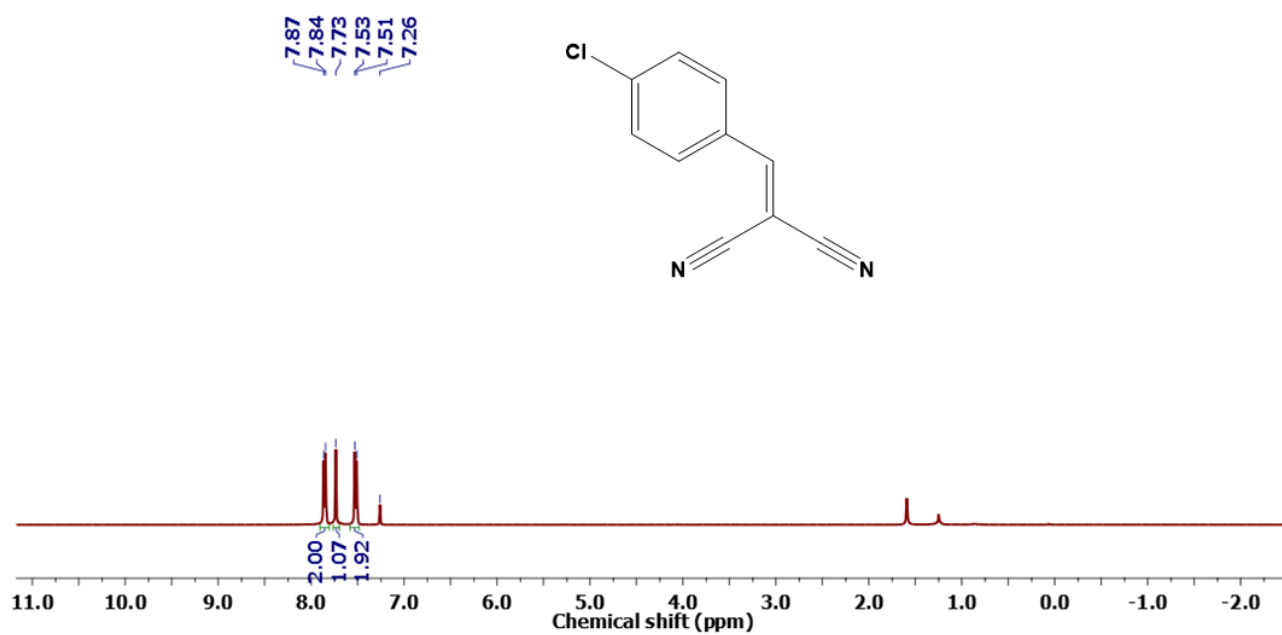


Figure S3. ¹H NMR of 2-(4-chlorobenzylidene)malononitrile.

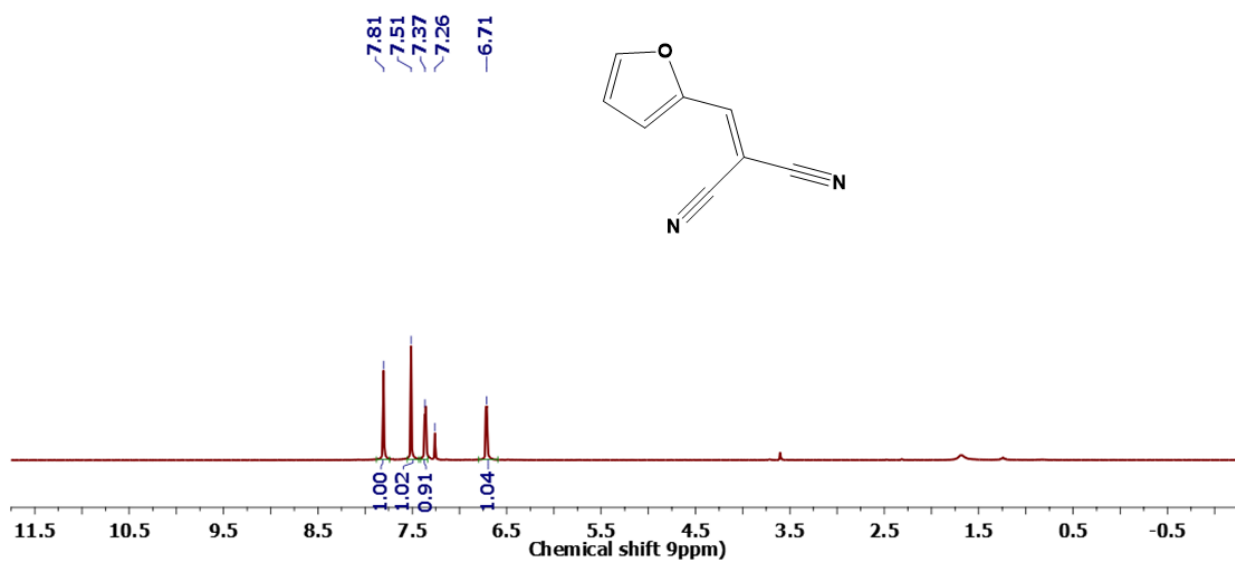


Figure S4. ¹H NMR of 2-(furan-2-ylmethylene)malononitrile.

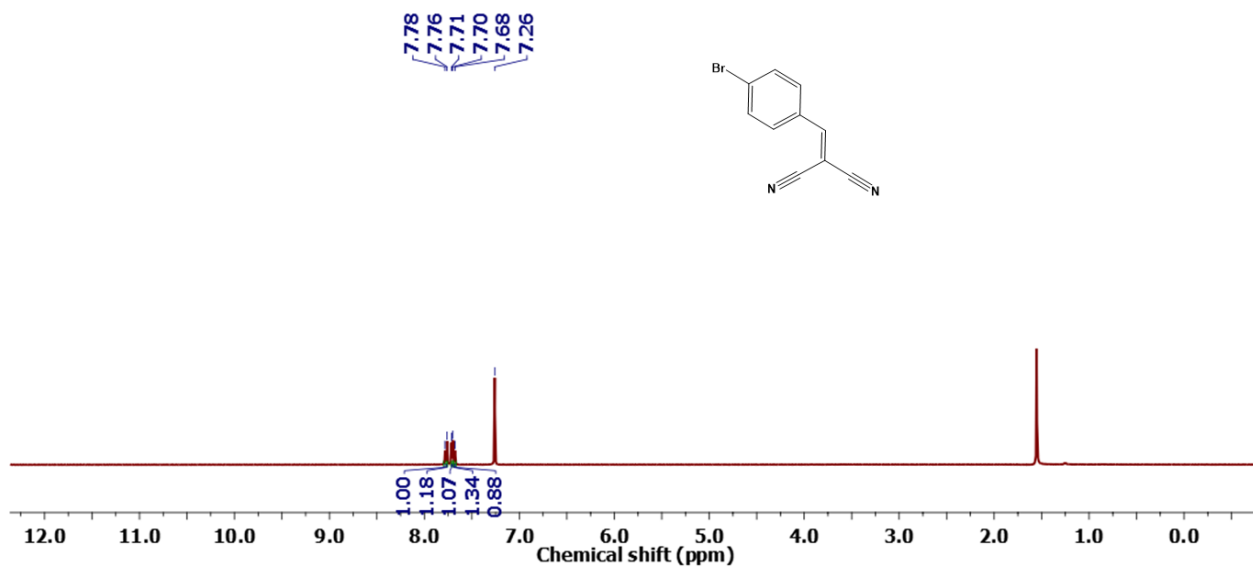


Figure S5. ¹H NMR of 2-(4-bromobenzylidene)malononitrile.

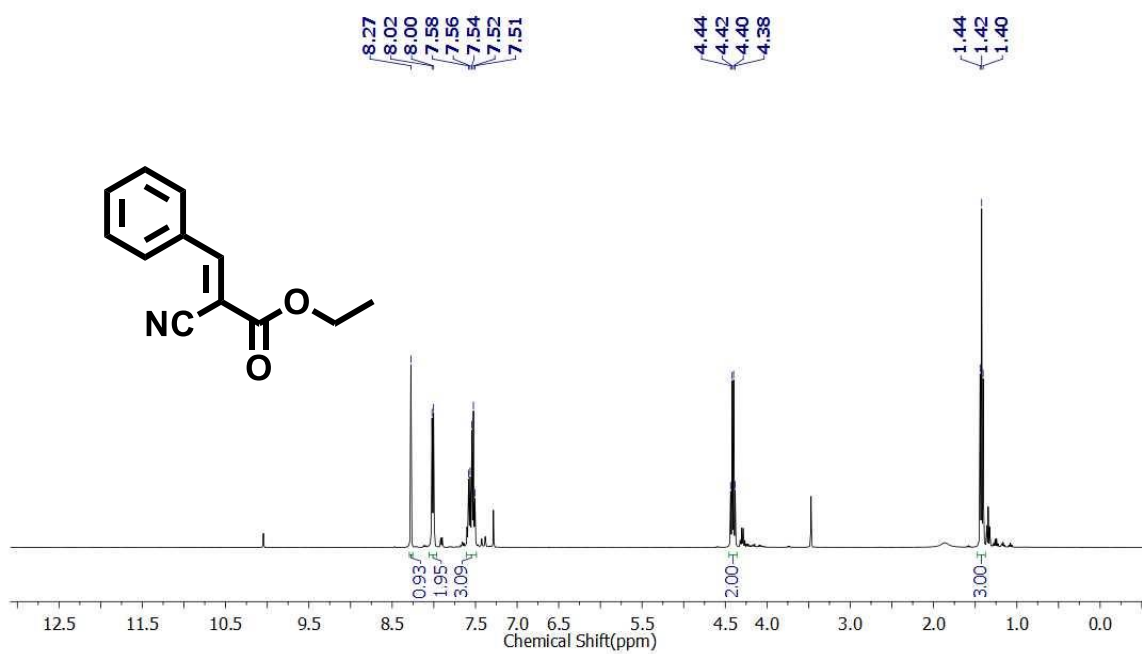


Figure S6. ¹H NMR of ethyl (E)-2-cyano-3-phenylacrylate.

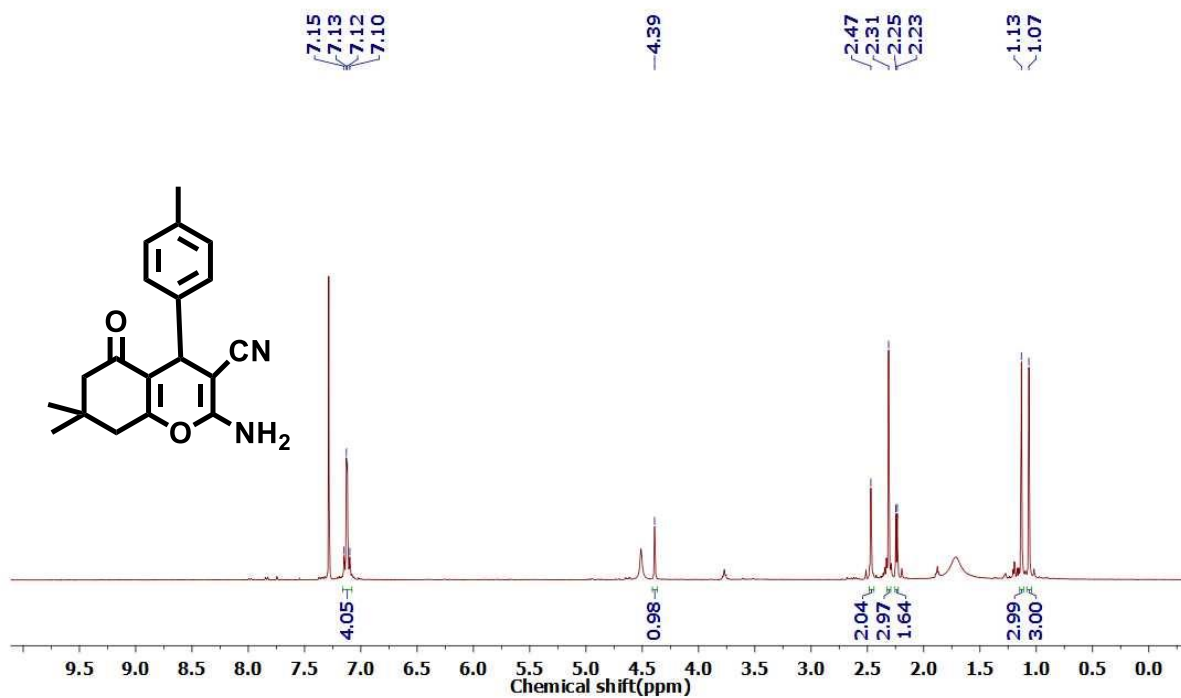


Figure S7. ^1H NMR of 2-amino-7,7-dimethyl-5-oxo-4-(p-tolyl)-5,6,7,8-tetrahydro-4H-chromene-3-carbonitrile.

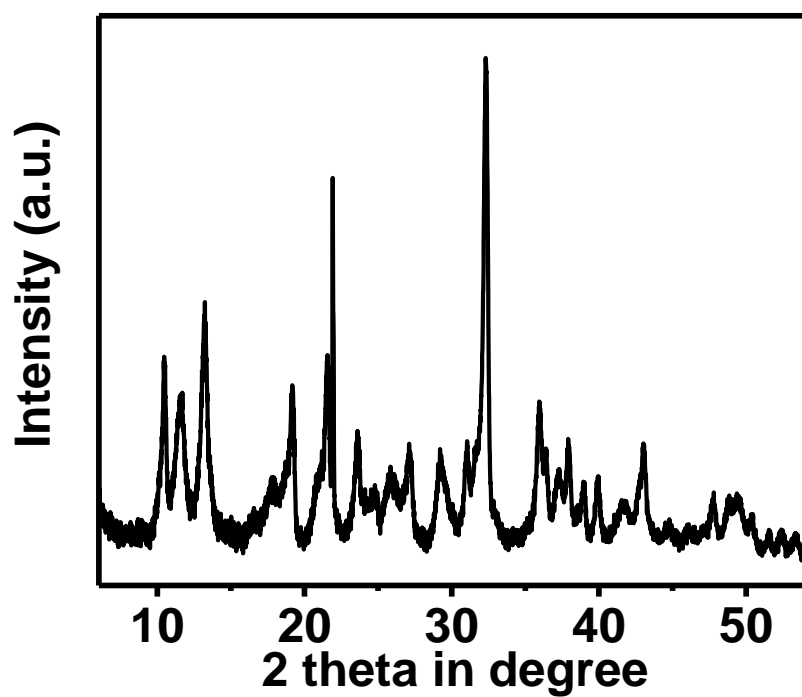


Figure S8. PXRD pattern of recycled catalyst.

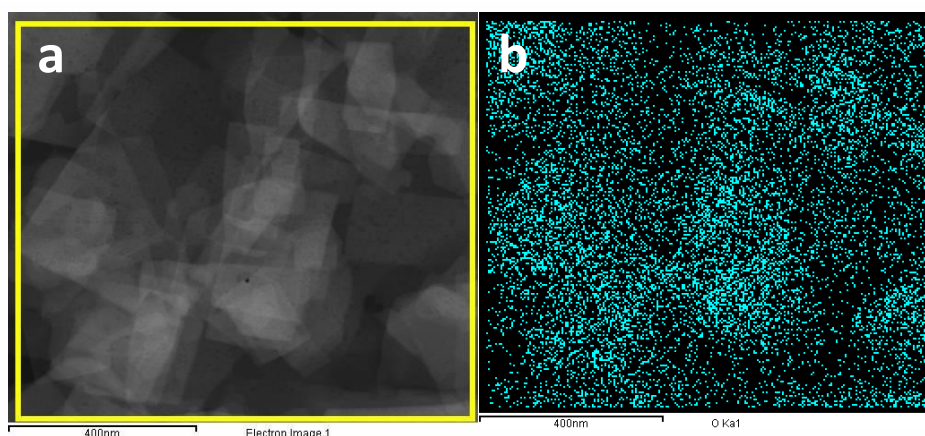


Figure S9. a) TEM image of MALPO; b) distribution of oxygen in MALPO.

Calculation of acidity

The acidity of MALPO had been calculated by indirect titration method using NaOH as base.

Initially, 60 mg of material MALPO was taken in 50 ml water and stirred for 24h at room temperature. Then 50 ml of standardized 0.0255 N NaOH solution was added and stirred for overnight at room temperature. Then filtered and the excess NaOH was back titrated with 0.08 N oxalic acid solution

Where the first equivalence point reached upon addition of 15.4 ml of 0.08 N oxalic acid

Now, 15.4 ml 0.08 N oxalic acid \equiv 15.4 ml 0.04 M oxalic acid \equiv 15.4 \times 0.04 mmol of oxalic acid.

50 ml 0.0255 N NaOH solution \equiv 50 ml 0.0255 M NaOH solution \equiv 50 \times 0.0255 mmol NaOH.

So, 15.4 \times 0.04 mmol of oxalic acid will neutralize 15.4 \times 0.04 \times 2 mmol of excess NaOH.

Then, [(50 \times 0.0255) – (15.4 \times 0.04 \times 2)] = 0.043 mmol of NaOH has been neutralized by 60 mg of MALPO.

Thus we can say, 60 mg of MALPO materials contains 0.043 mmol of acid.

Thus, the acid strength of MALPO would be, 0.7116 mmol g⁻¹.