Table S1. Elemental Composition

Parameter/ Sample	Unit	2MgAl	20K_2O/2MgAl
SiO_2	wt.%	<0.1	0.3
Al_2O_3	wt.%	38.3	28.1
MgO	wt.%	59.7	43.9
K_2O	wt.%	0.1	24.7
Other impurities[a]	wt.%	1.9	3.0
Mg/Al	mol/mol	2.0	2.0

[a] other impurities including Cl, SO_3, P_2O_5, CaO, Na_2O, MnO, CuO, ZnO, Fe_2O_3, Ga_2O_3, NiO

Table S2. Physicochemical properties of Hydrotalcite (HTC) catalysts

Catalyst	Base and Acid site loading (mmol g^{-1})				S_{BET} [c] (m^2 g^{-1})	S_{MIC} [d] (m^2 g^{-1})	S_{EXT} [e] (m^2 g^{-1})	V_p [f] (cm^3 g^{-1})	V_{mic} [d] (cm^3 g^{-1})	V_{mes} [g] (cm^3 g^{-1})
	T_{max} (°C)	Basic site loading [a] (mmol g^{-1})	T_{max} (°C)	Acid site loading [b] (mmol g^{-1})						
20K_2O/2MgAl	100, 404, 642	0.498	300	1.200	34	0	4	0.45	0	0.45
2MgAl	140-230, 648	0.566	400, 536	3.100	54	0	35	0.57	0	0.57

[a] from CO_2-TPD; [b] from propylamine-TGA-MS; [c] S_{BET} total surface area determined using BET equation with $P/P_0 < 0.05$; [d] S_{MIC} micropore surface area and V_{MIC} micropore volume determined from t-plot analysis [e] S_{EXT} external surface area was calculated from the difference between S_{BET}-S_{MIC}, [f] V_p total pore volume obtained from isotherm at P/P_0=0.99 (Error = ±10%) and [g] V_{MES} mesopore volume obtained from the difference between V_p-V_{MIC}.

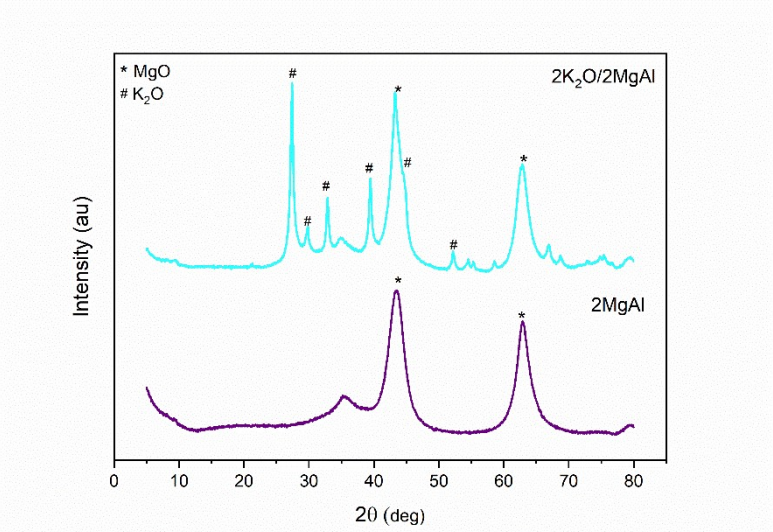


Fig S1. Wide-angle XRD of parent and modified 2MgAl Hydrotalcite

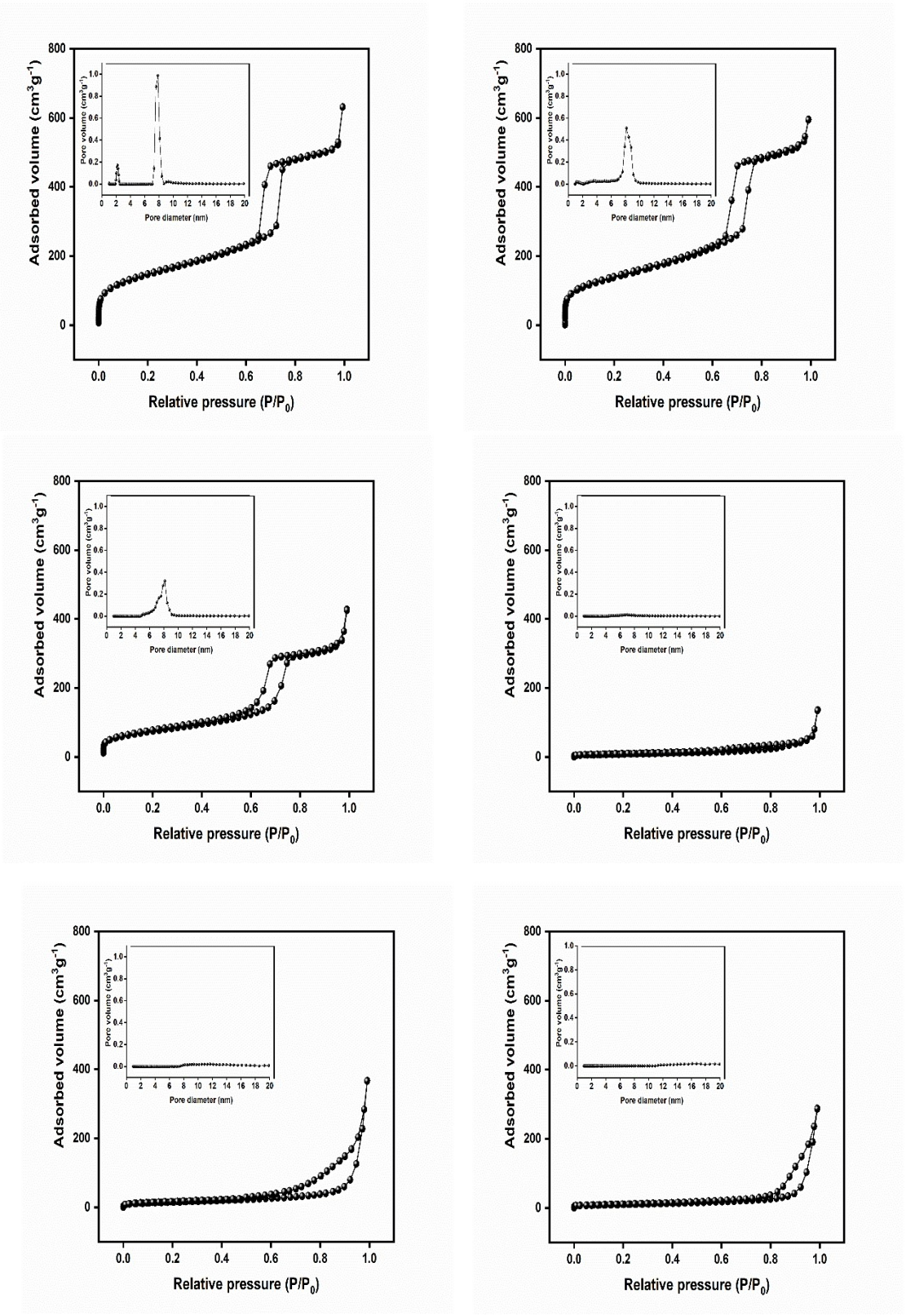


Fig S2. Adsorption–desorption isotherms of N_2 molecules at 77 K and pore size distribution (inset) of a) SBA-15, b) 12MgAl-SBA-15, c) $8K_2O$/12MgAl-SBA-15, d) $20K_2O$/12MgAl-SBA-15, (e) 2MgAl and (f) $2K_2O$/2MgAl

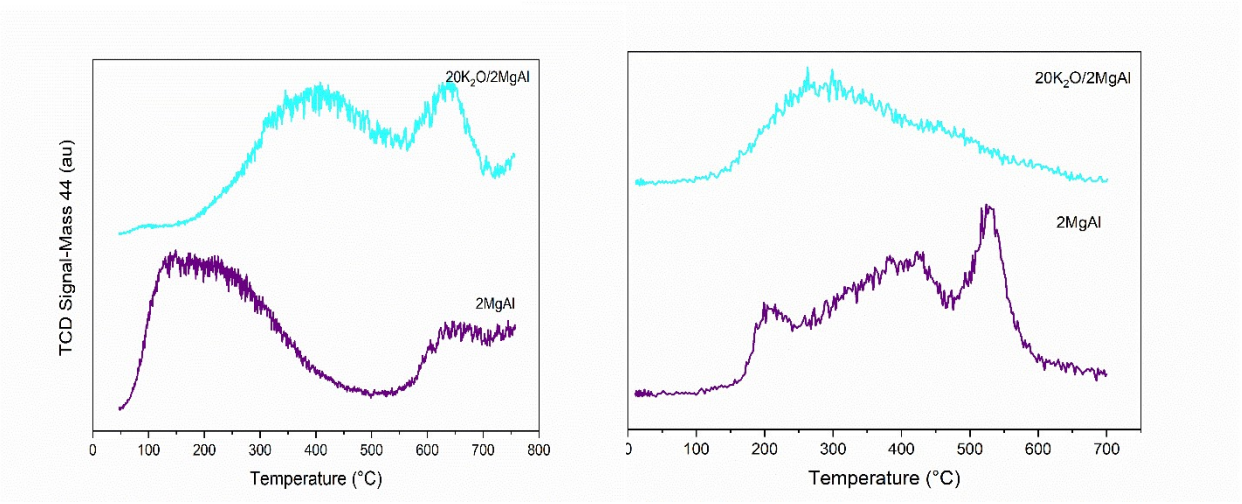
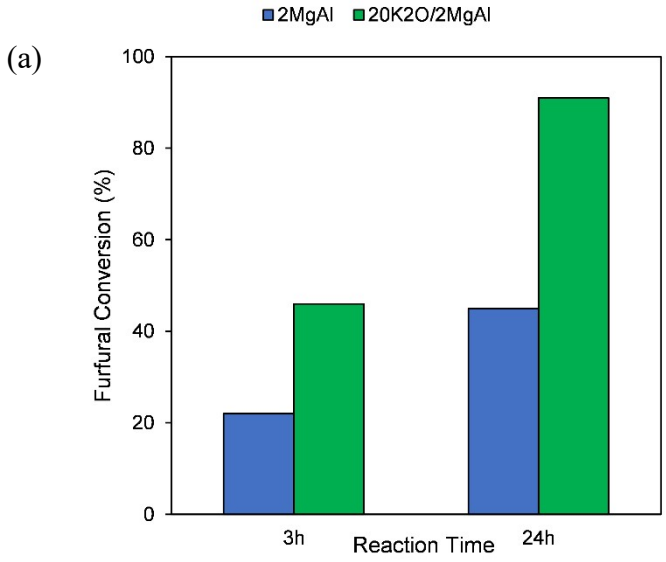


Fig S3. (a) CO_2-TPD and (b) TGA-MS-propylamine of parent and modified 2MgAl



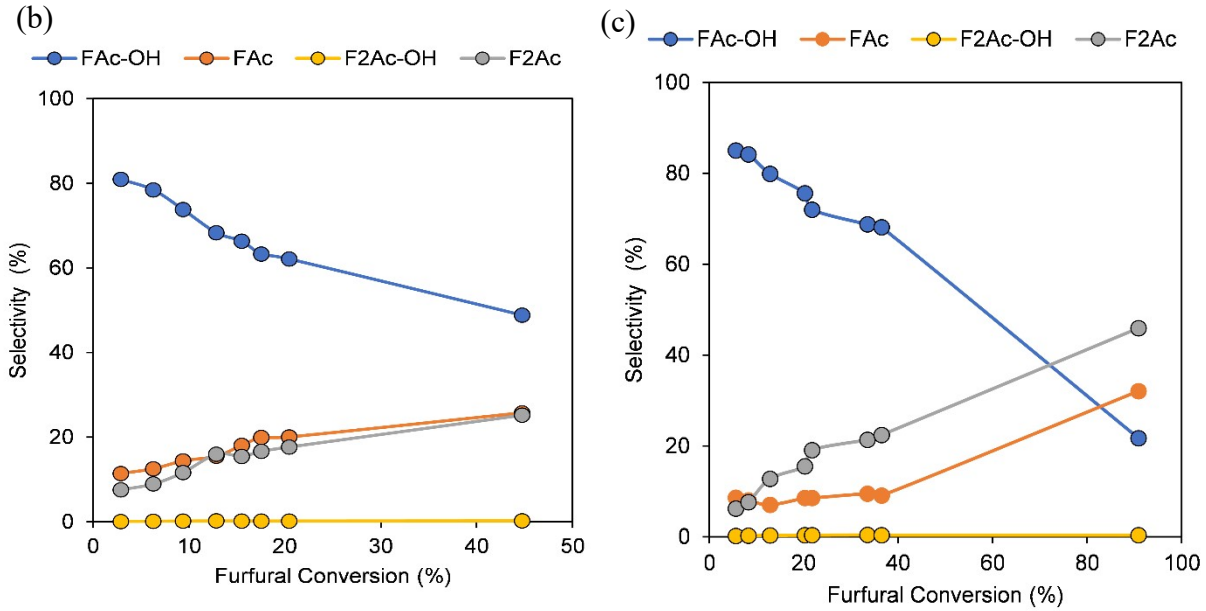


Fig. S4 (a) Comparison of furfural conversion for 2MgAl and 20K_2O/2MgAl and (b and c) product selectivity as a function of furfural conversion achieved over 24 h for 2MgAl and 20K_2O/2MgAl catalysts

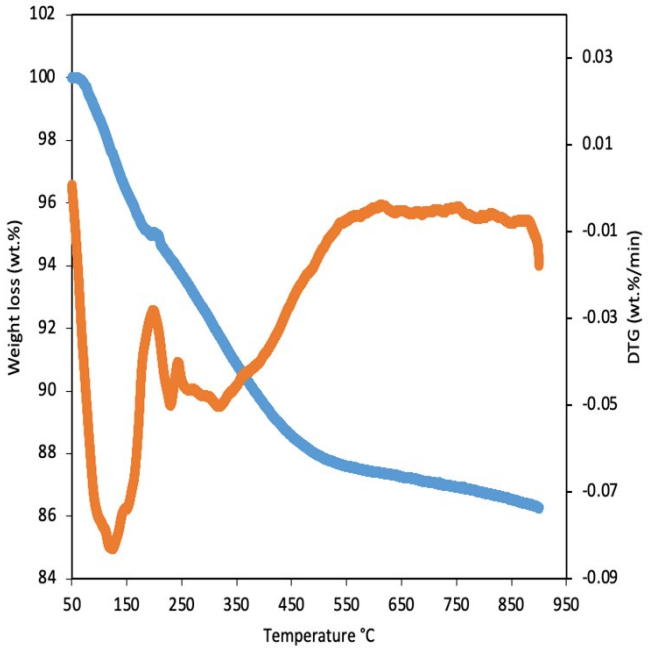


Fig. S5 TGA-DTG measurement of regenerated 20K_2O/12MgAl-SBA-15 by thermal treatment under an N_2