

Supplementary Information For:

Selective Upcycling of Polypropylene Glycol to Propionaldehyde via Catalytic Cracking on Acid Sites of Mesoporous Y Zeolites

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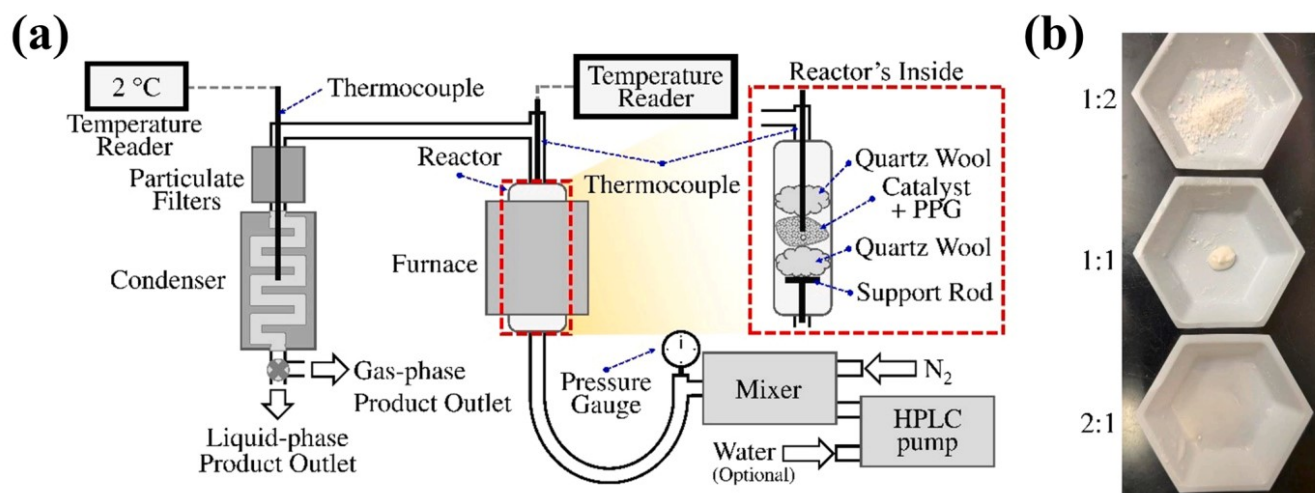


Figure S1: a) Original PPG reactor setup and b) images for different PPG polymer to zeolite mass ratios.
Reproduced from Ref [21].

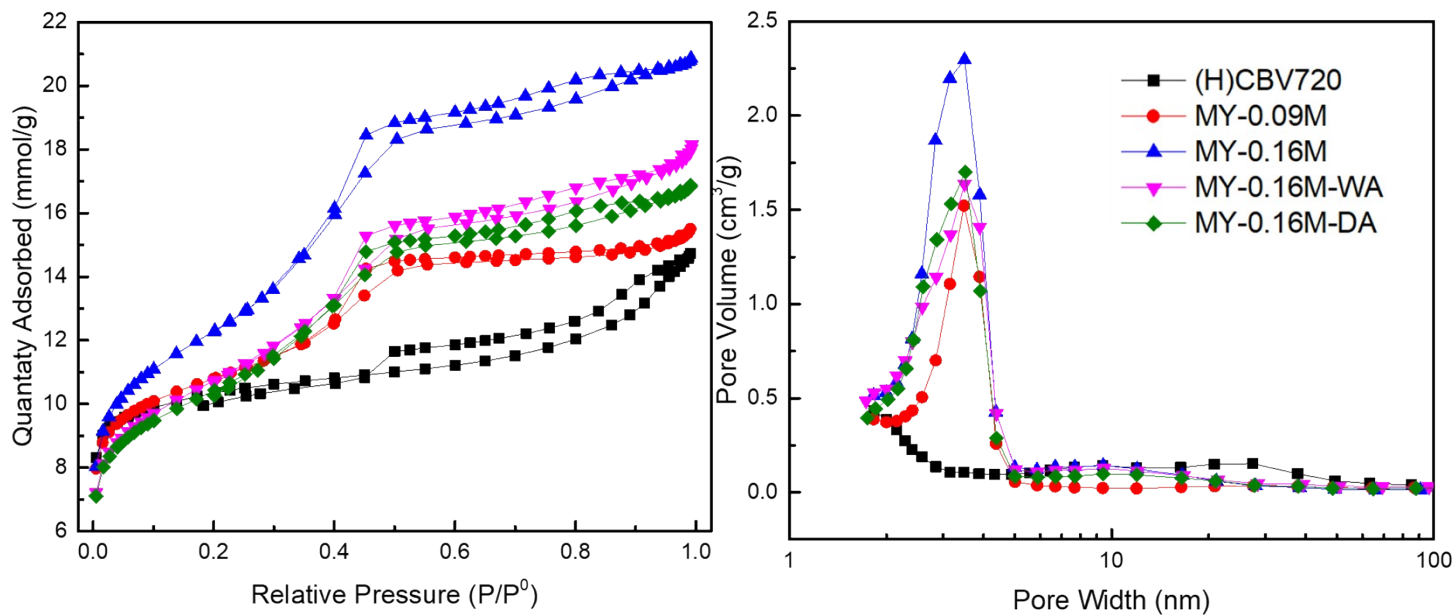


Figure S2: (Right) N_2 Physisorption isotherms at 77K and (Left) corresponding PSD plots.

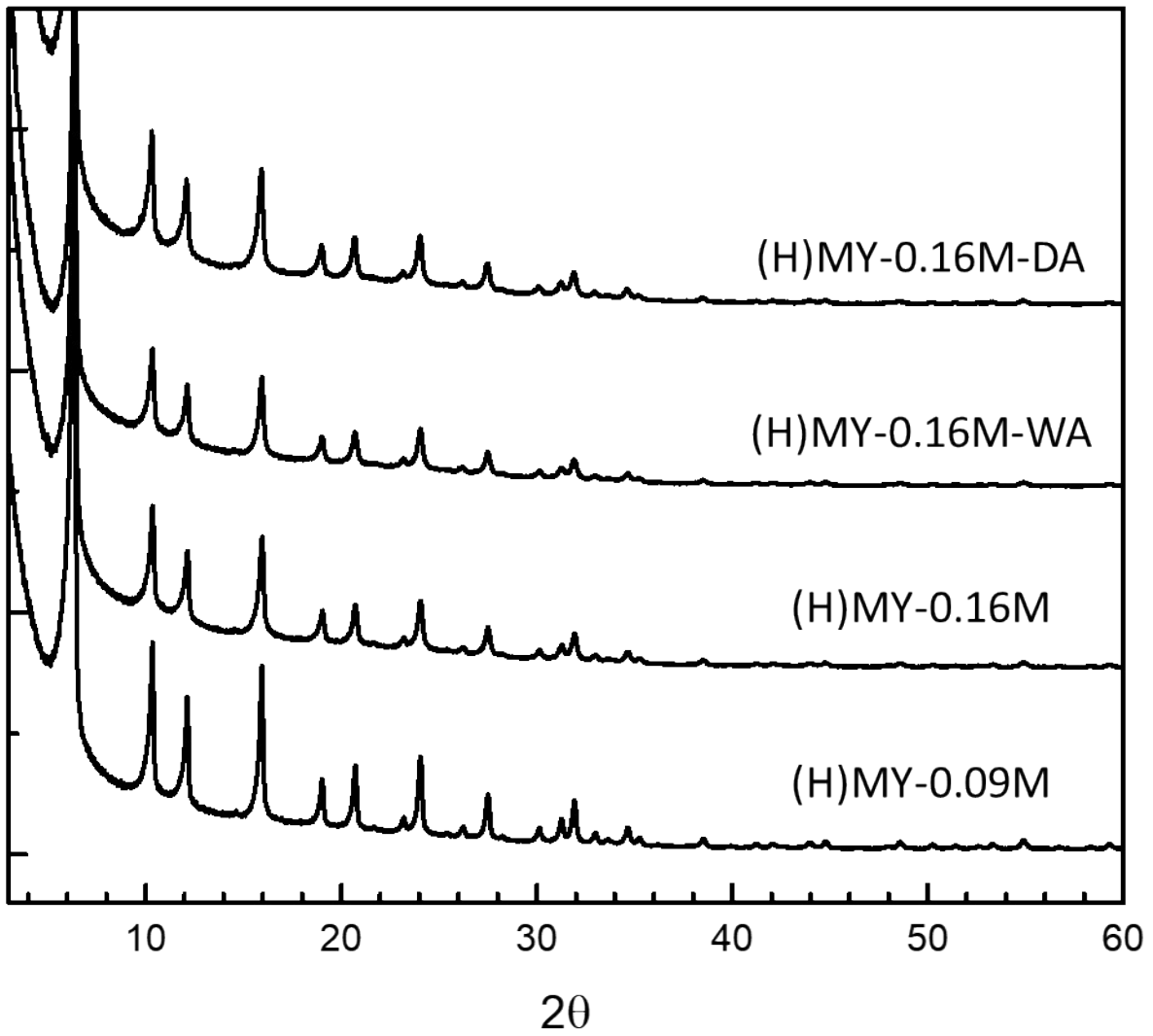


Figure S3: PXRD patterns of synthesized zeolite materials.

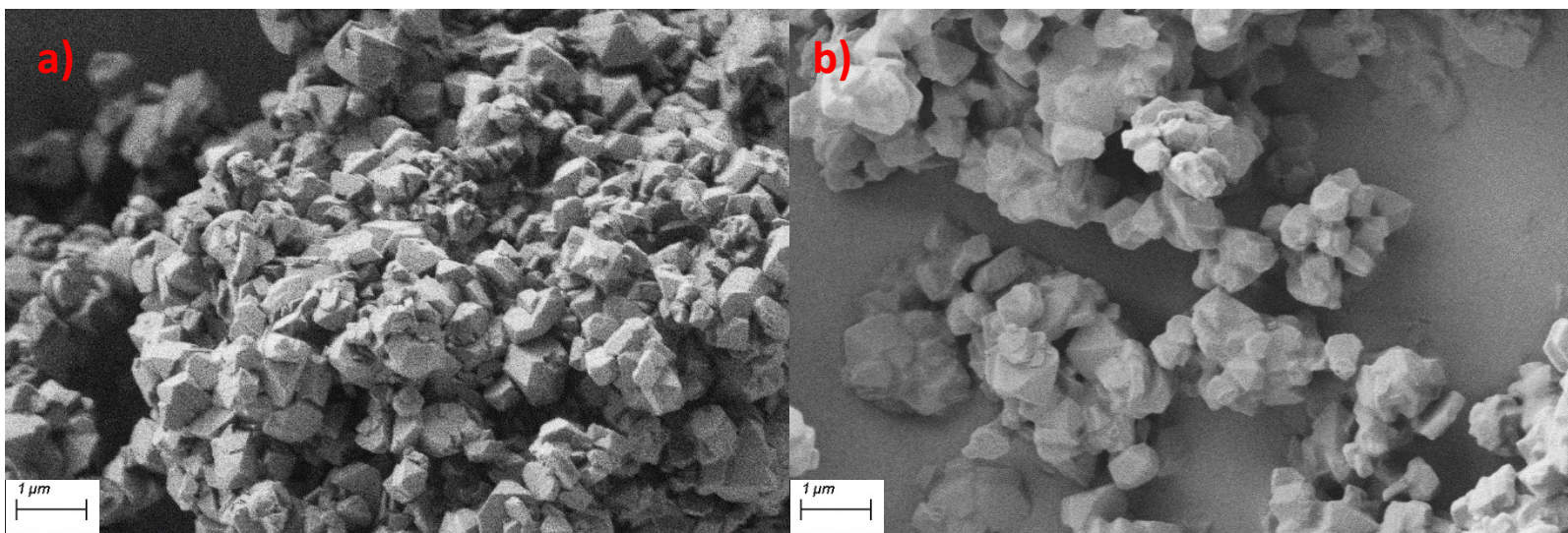


Figure S4: SEM images of zeolite materials. a) Commercial (H)CBV720; b) MY-0.16M-DirCal.

Table S1: Summary of aluminum distribution via area integrations of measured ^{27}Al MAS NMR Spectrum. Al_{EF} is summation of AlO_5^- and AlO_6^- .

Zeolite	$\text{AlO}_4^-/\text{Al}_{\text{tot}}$	$\text{AlO}_5^-/\text{Al}_{\text{tot}}$	$\text{AlO}_6^-/\text{Al}_{\text{tot}}$	$\text{Al}_{\text{F}}/\text{Al}_{\text{EF}}$	$\text{Al}_{\text{EF}}/\text{Al}_{\text{tot}}$
(H)CBV720	0.66	0.11	0.23	1.94	0.34
(H)MY-0.09M	0.53	0.18	0.28	1.13	0.47
(H)MY-0.16M	0.50	0.20	0.30	1.00	0.50
(H)MY-0.16M-WA	0.39	0.25	0.36	0.64	0.61
(H)MY-0.16M-DA	0.40	0.24	0.35	0.68	0.60

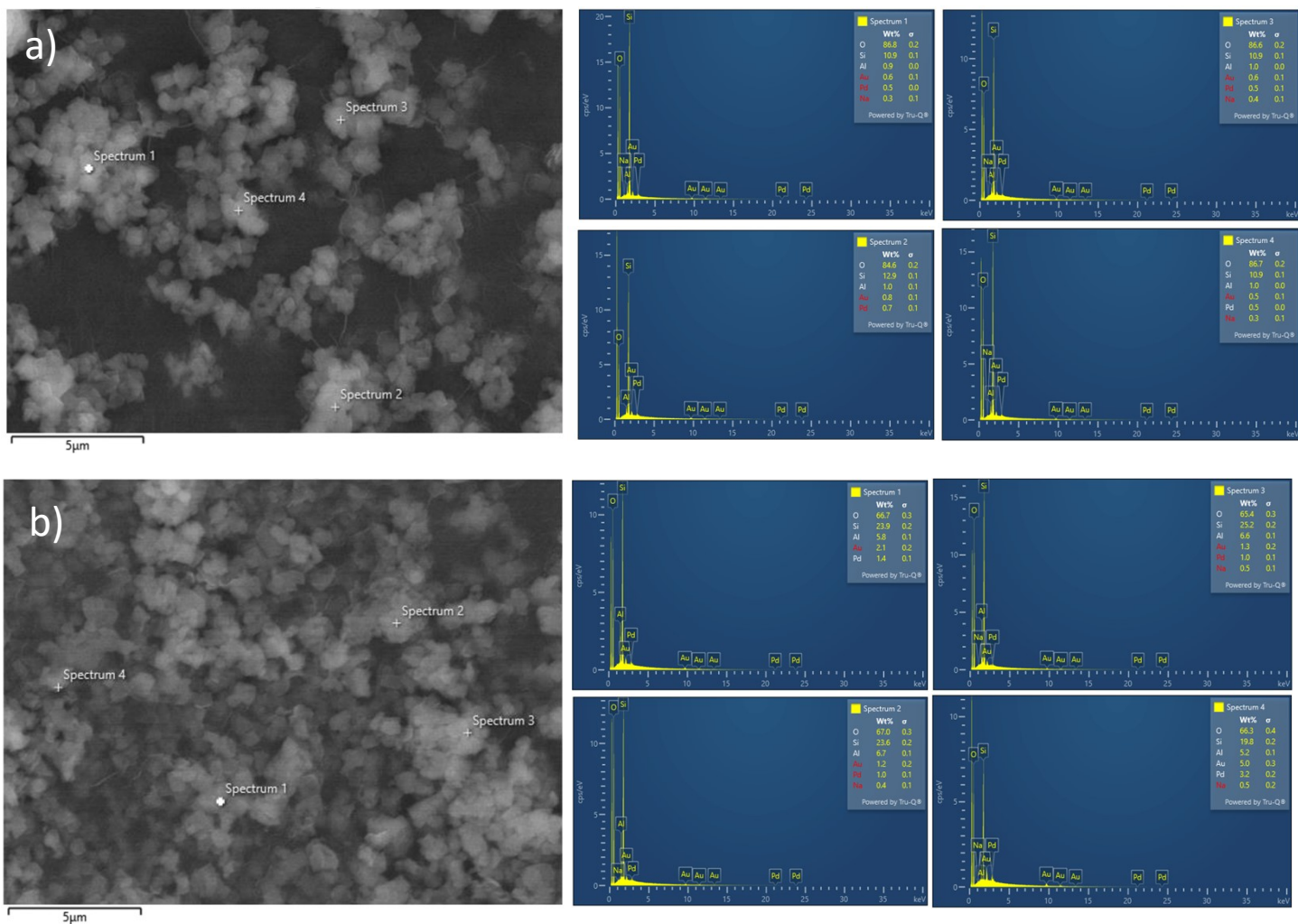


Figure S5: SEM/EDX Images for a) MY-0.16M and b) MY-0.16M-DA.

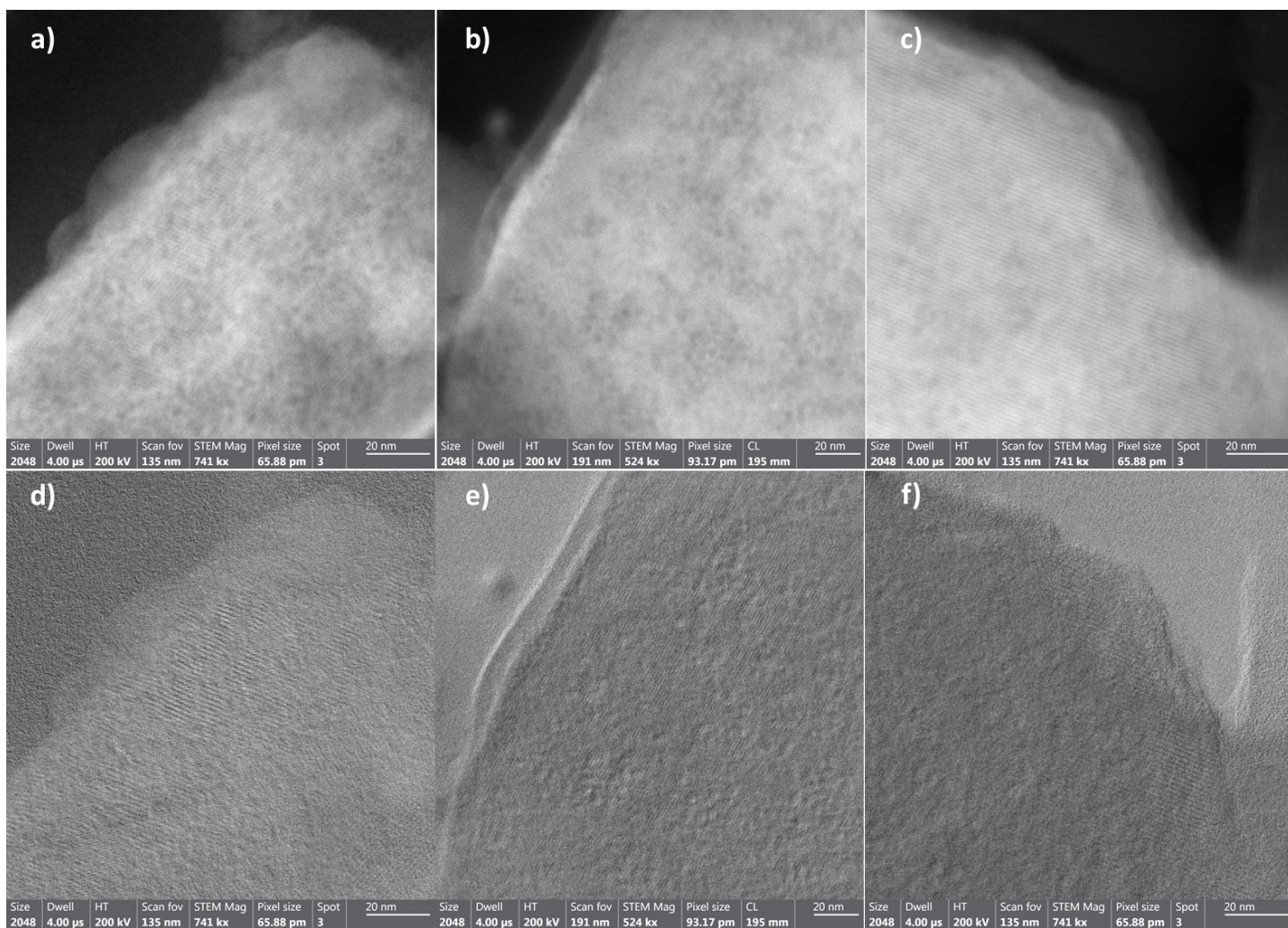


Figure S6: Microscopy characterization of MY-0.16M-DA. a-c) HAADF images and corresponding d-f) HRTEM images.

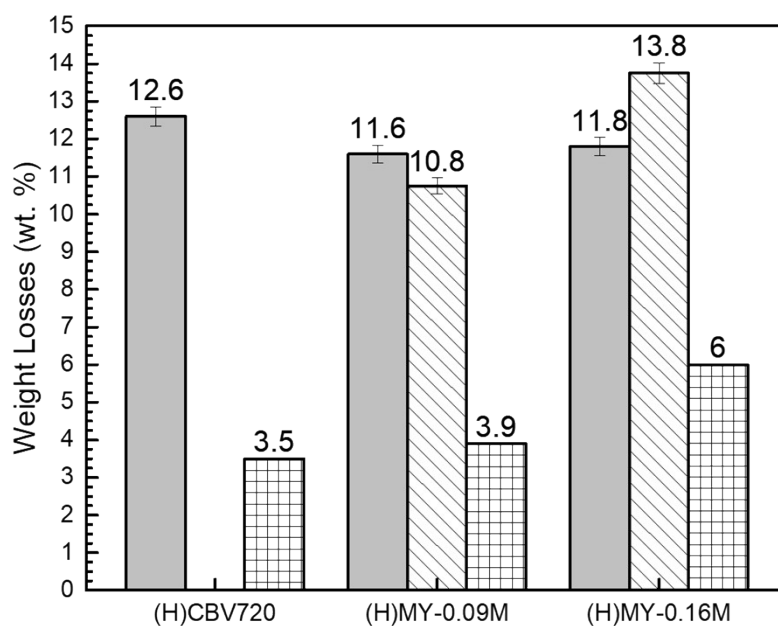


Figure S7: Solid residuals from PPG cracking for MY materials. (grey) Residue on spent catalyst that underwent pretreatment, (dashed pattern) residue on spent catalyst that underwent no pretreatment, (square pattern) water content on fresh catalysts. Original reaction setup conducted at 450 °C, 1:1:PPG:Catalyst ratio, 425 MW PPG polymer with 10-minute reaction time, under N₂ atmosphere. No TGA measurement was conducted for spent untreated (H)CBC720.

Product Selectivity (%)	Original setup Non-dried catalysts- PPG 425			Original setup Dried catalysts -PPG 425			Modified setup-PPG 425		Modified setup- 2000PPG				
	(H)CBV 720	(H)MY-0.09M	(H)MY-0.16M	(H)CBV 720	(H)MY-0.09M	(H)MY-0.16M	(H)CBV 720	(H)MY-0.16M	(H)CBV 720	(H)MY-0.16M	(H)MY-0.16M-WA	(H)MY-0.16M-DA	SBA 200
Propionaldehyde	75.05	68.4	63.1	71.53	80.73	83.46	73.5	94.86	73.5	93.38	88.5	86.7	25.52
1,3-Dioxane, 4,5-dimethyl-	7.87	16.21	15.17	15.46	10.48	8.61	8.63	1.95	8.63	1.42	3.62	4.19	20.29
1,3-Dioxolane, 2-ethyl-4-methyl-	1.75	3.38	3.4	3.4	1.76	1.32	0.58	0.59	0.58	1	1.4	1.5	0
Acetone	6.76	2.18	1.88	1.79	1.8	1.79	1.26	0.74	1.26	0.7	0.8	1	8.47
Ethyl glycidyl ether	4.26	1.33	1.86	1.27	2.36	0.68	0	0	0	0	0	0	0
1,2 propanediol	0	0	0	0	0	0	12.56	0.42	12.56	2.38	2.17	3.44	4.28
Ethandiol	1.91	2.8	8.53	0	0	0	0	0	0	0	0	0	0
Other liquid products	2.4	5.7	6.06	6.55	2.87	4.14	3.47	1.44	3.47	1.12	3.51	3.17	41.44
Solid Product Yield (%) (Solid Residues on Spent Catalysts)	N/A	10.75 (±0.24)	13.75 (±0.27)	12.59 (±0.25)	11.57 (±0.24)	11.78 (±0.24)	N/A	6.47 (±0.16)	7.29 (±0.15)	6.35 (±0.17)	8.27 (±0.20)	8.95 (±0.21)	5.24 (±0.23)
Total Liquid Product Yield (%)	N/A	86.7 (±1.22)	83.2 (±1.10)	85.1 (±0.97)	85.9 (±1.12)	86.1 (±0.92)	N/A	91.14 (±1.28)	90.66 (±1.16)	91.73 (±1.22)	88.89 (±0.78)	89.21 (±1.06)	91.33 (±1.51)
Total Recovered Yield of Solid and Liquid Products (%)	N/A	97.45	96.95	97.69	97.47	97.88	N/A	97.61	97.95	98.08	97.16	98.16	96.57

Table S2: Complete catalysis results. All reactions conducted at 450 °C, with 10-minute reaction time, under N₂ atmosphere.

