Supporting Information

Designed Synthesis of MOR Zeolites using Gemini-type Bis(methylpyrrolidinium) Dications as Structure Directing Agents and Their DME Carbonylation Performance

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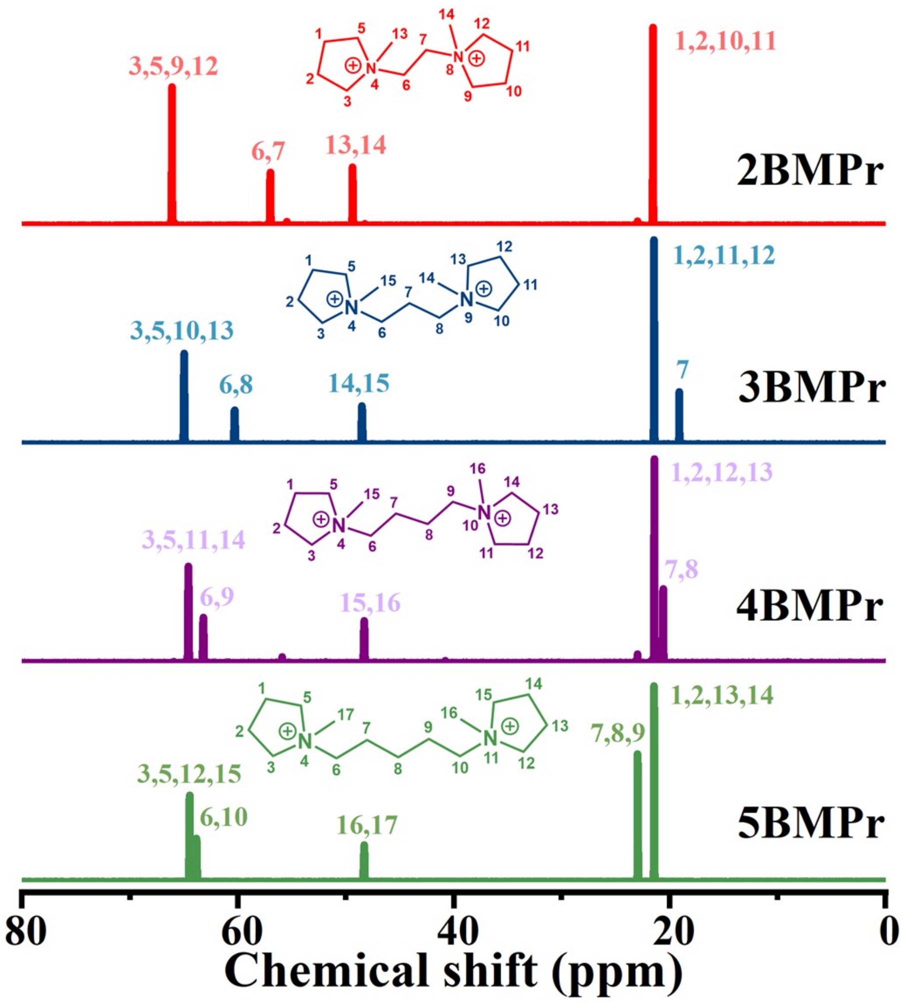

Fig. S1 The ^{13}C NMR spectra of the four organic dications solvated in D_2O.

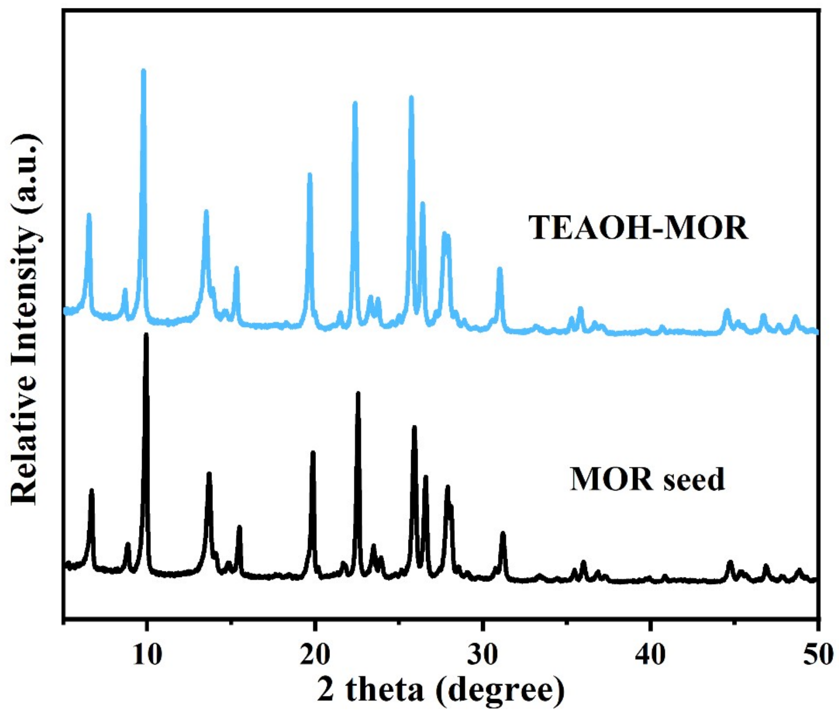

Fig. S2 XRD patterns of the seed and reference samples.

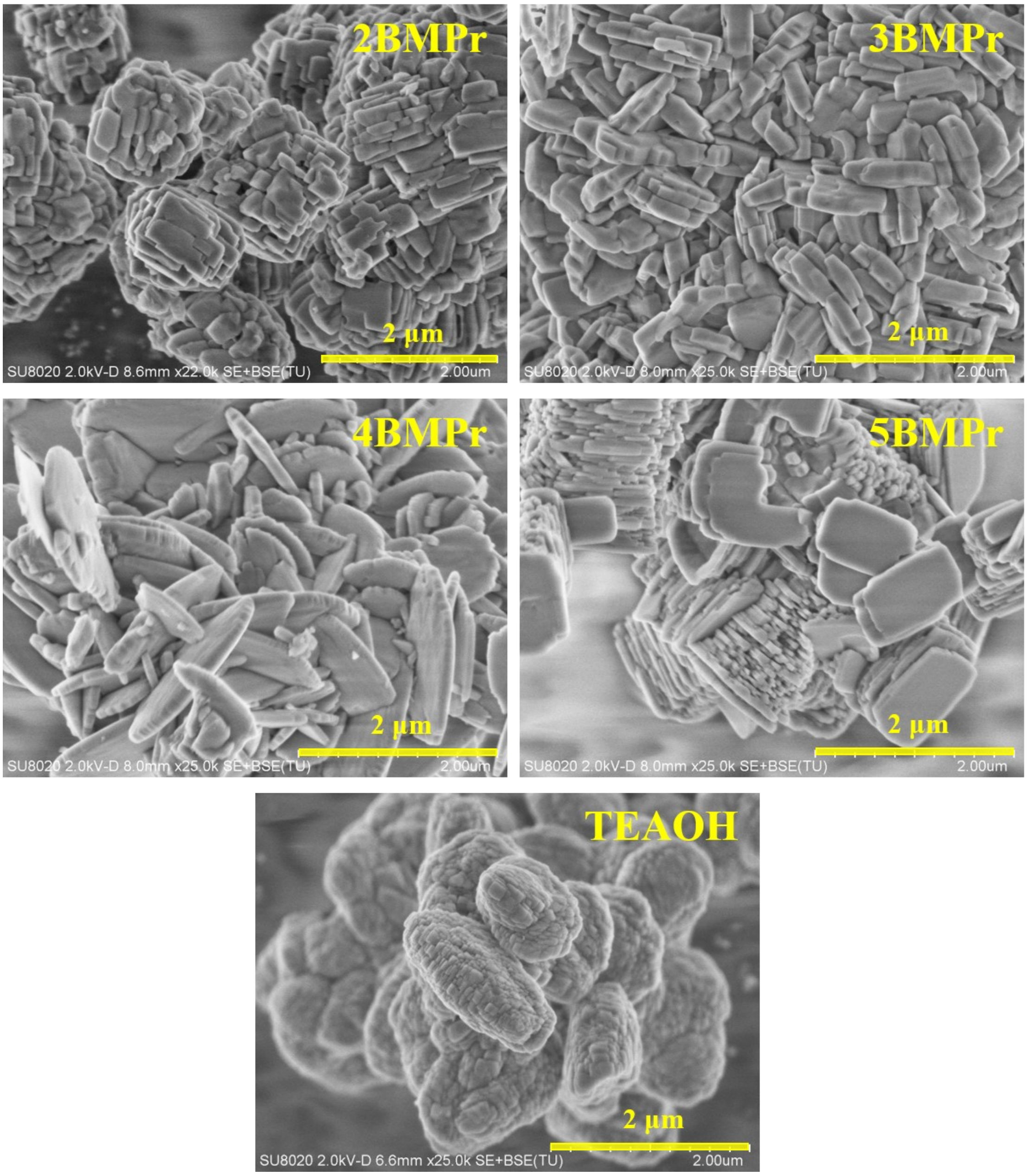

Fig. S3 SEM images of the as-synthesized MOR samples.

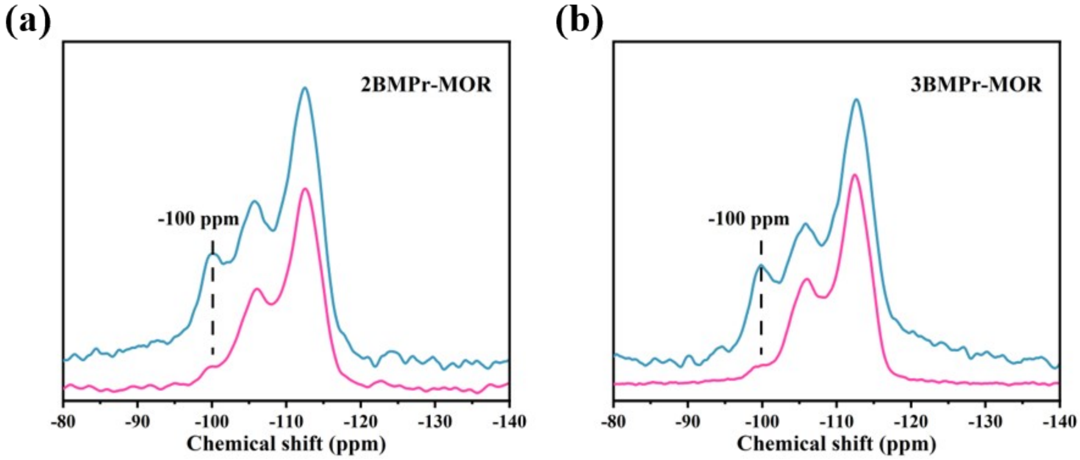

Fig. S4 The ^{29}Si MAS NMR (pink) and ^{1}H-^{29}Si CP MAS NMR spectra (blue) of the as-synthesized samples: (a) 2BMPr-MOR, (b) 3BMPr-MOR.

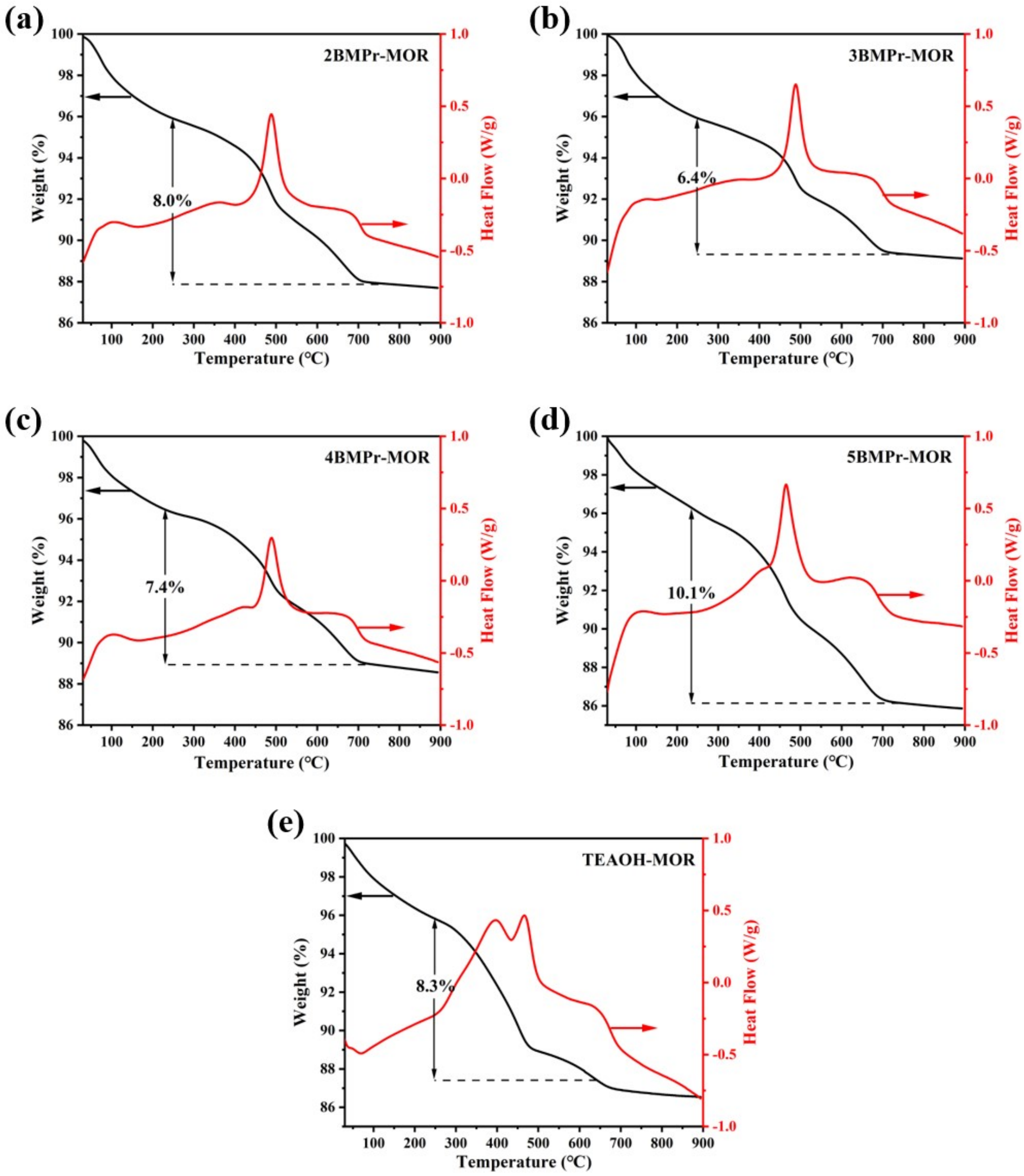

Fig. S5 Thermal analysis of the as-synthesized MOR samples: (a) 2BMPr-MOR, (b) 3BMPr-MOR, (c) 4BMPr-MOR, (d) 5BMPr-MOR, (e) TEAOH-MOR.

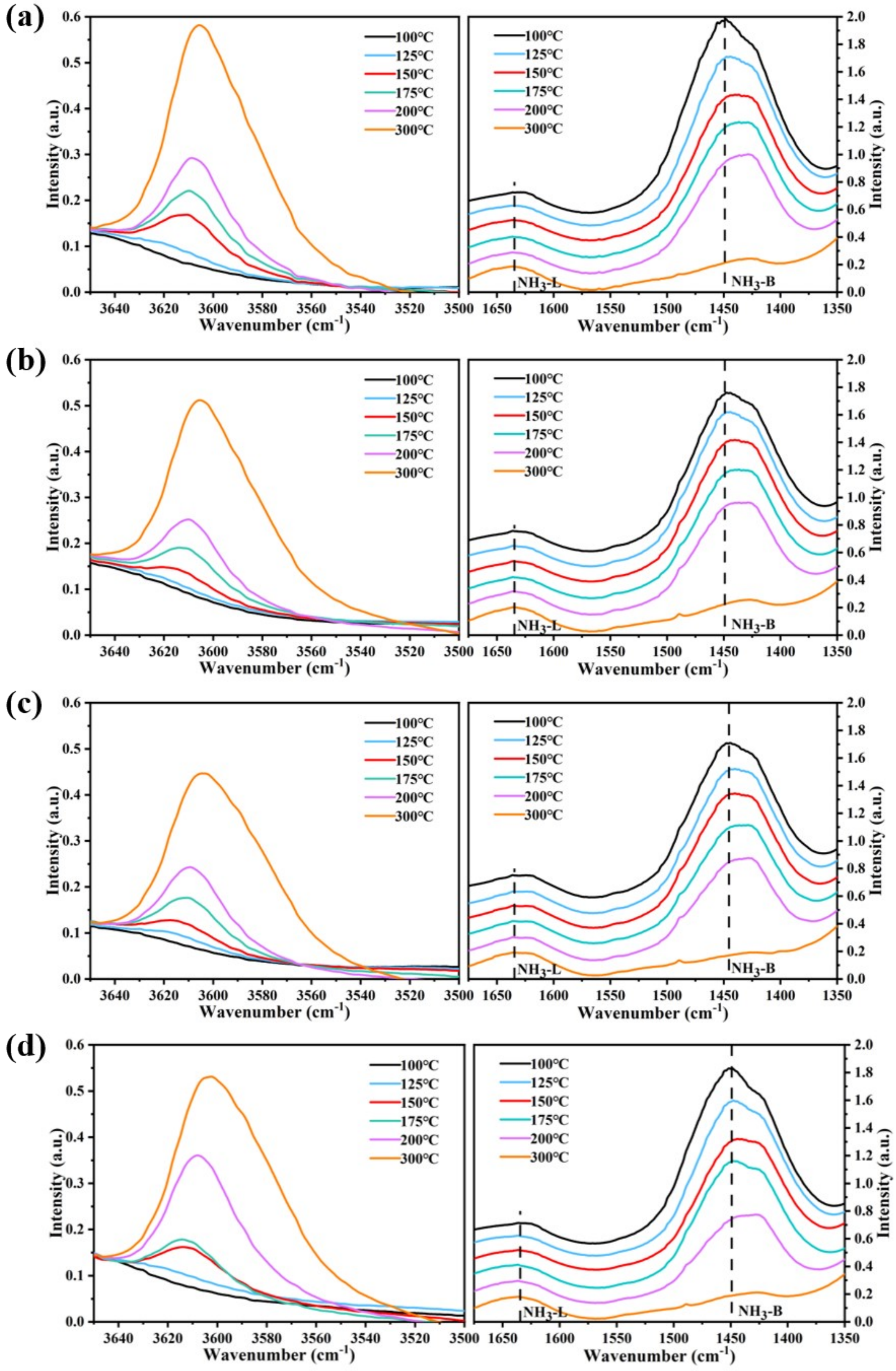

Fig. S6 NH_3-adsorbed FTIR spectra of H-MOR samples after NH_3 desorption at different temperatures: (a) 2BMPr-MOR, (b) 3BMPr-MOR, (c) 4BMPr-MOR, (d) TEAOH-MOR.

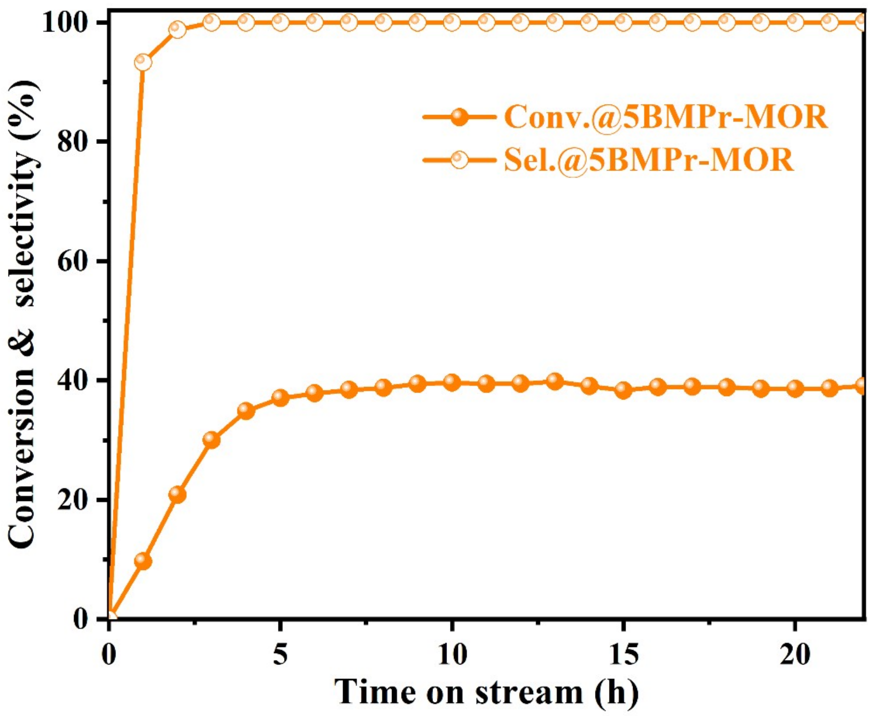

Fig. S7 The catalytic performance of DME carbonylation over 5BMPr-MOR. Reaction conditions: 200 °C, 2 MPa, DME/CO/N_2 = 5/35/60, GHSV = 7200 mL/g/h.

Table S1. Synthesis results for MOR zeolites using nBMPr [n=2, 3, 4, 5].

SDA	Gel molar composition [a]		Times(days)	product		Solid yield [c]
	SiO_2/Al_2O_3	Na_2O/SiO_2		Phase	SAR [b]	(%)
2BMPr	22	0.18	2	MOR	10.2	92
		0.2	2	MOR	9.5	84
	26	0.18	2	MOR	12.0(12.4)	86
		0.2	2	MOR	10.5	76
	30	0.2	2	MOR (amorphous)		
			2.5	MOR	12.9	85
			3	MOR	12.4	83
	34	0.2	2	Amorphous		
			2.5	MOR+Quartz		
3BMPr	22	0.2	2	MOR	9.4	82
	26	0.18	2	MOR	11.9(12.0)	84
		0.2	2	MOR	10.9	85
			3	MOR+ANA		
			4	MOR+Quartz		
	28	0.2	2	MOR	12.1	65
			2.5	MOR	11.9	78
	30	0.2	2	Amorphous		
			2.5	MOR	12.4	82
			3	MOR+ZSM-5		
	34	0.2	2	Amorphous		
			2.5	MOR+Nu-6		
			3	MOR+Quartz		
4BMPr	26	0.18	5	Amorphous		
		0.24	2	Amorphous		
	30	0.18	5	Amorphous		
		0.2	4	MOR+ZSM-5		
		0.22	3	Amorphous		
		0.24	4	MOR	9.7	65
	32	0.18	4.5	Amorphous		
			5	MOR	12.1(12.0)	73
			5.5	MOR+ZSM-5		
5BMPr	26	0.16	4	Amorphous		
			5	MOR+ZSM-5		
		0.18	3	Amorphous		
			4	MOR+ZSM-5+SSZ-24		
		0.2	3	MOR+ZSM-5		
		0.22	2	MOR	9.6(9.4)	64
		0.24	2	MOR	8.6	55
	30	0.18	5	MOR+ZSM-5+ZSM-57		
		0.24	2	MOR+ZSM-5+SUZ-4		

32	0.18	5	MOR+ZSM-5+SUZ-4
34	0.24	2	Amorphous

[a] Gel molar composition: H_2O/SiO_2=15, OSDA/SiO_2=0.12, seed addition: 6 wt% relative to SiO_2 resource; crystallization temperature: 180 °C. The SAR (Si/Al ratio) of MOR seed are 12.9 derived from XRF.
[b] The SAR was derived from XRF and the value in the bracket was calculated from ^{29}Si MAS NMR.
[c] The yield was calculated based on the mass of silica and alumina.

Table S2. OSDA stabilization energy for the four dications used in this work.

OSDA	OSDA stabilization energy (kJ/mol Si) [a]
2BMPr	-5.01
3BMPr	-4.85
4BMPr	-4.80
5BMPr	-4.60

[a] OSDA stabilization energy (nBMPr) = E(total) - E(zeolite) -E(nBMPr).

Table S3. Summary of the product STY in the MOR-catalyzed DME carbonylation reaction.

Literature	Reaction conditions				STY of MA	Pyridine
	DME : CO (%)	T (^{o}C)	P (MPa)	GHSV (mL/g/h)	(mmol/h/g)	modification
[1]	2:93	165	1	12024	1.9	
[2]	5:50	200	1	1250	0.8	Yes
[3]	5:50	200	1	1250	1.7	
[4]	3:95.5	210	1.5	5280	4.0	
[5]	5:35	200	2	1500	1.3	
[6]	1:47	200	1.5	4500	1.8	
[7]	5:35	200	3	1500	2.8	
[8]	2.4:50	210	2	2100	3.2	
[9]	1:49	200	1.5	6000	1.6	
[10]	5:50	200	1	1250	1.3	
[11]	5:35	200	2	1500	3.0	Yes
[12]	1:47	200	1.5	3000	1.1	Yes
[13]	5:76	200	1	2500	2.5	
[14]	1:49	200	1.5	6000	1.6	
[15]	2:98	190	2	2000	1.8	
[16]	3:95.5	190	1.5	2640	2.8	
[17]	5:35	200	2	3600	7.2	Yes
[18]	1:49	200	1.5	6000	6.5	
[19]	1:49	200	1.5	6000	7.2	
[20]	1:49	200	1.5	6000	6.4	
[21]	5:35	200	2	2250	4.5	
[22]	5:35	200	2	3600	6.8	Yes
[23]	5:35	200	2	3600	6.4	Yes
Our work	**5:35**	**200**	**2**	**7200**	**12.5**	**Yes**

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