Straightforward synthesis of MTW-type magnesium silicalite for CO_2 fixation with epoxides under mild conditions†

Haimeng Wen,[a] Jingyan Xie,[a,b] Yang Zhou,[a] Yu Zhou*[a] and Jun Wang*[a]

[a]State Key Laboratory of Materials-Oriented Chemical Engineering, College of Chemical Engineering, Nanjing Tech University, Nanjing 210009, Jiangsu, P. R. China.

[b]Maoming Branch R&D Institute, SINOPEC, Maoming 525011, P. R. China.

† These two authors contributed equally: Haimeng Wen and Jingyan Xie.

* Corresponding authors

E-mails: njutzhouyu@njtech.edu.cn (Y. Zhou), junwang@njtech.edu.cn (J. Wang)

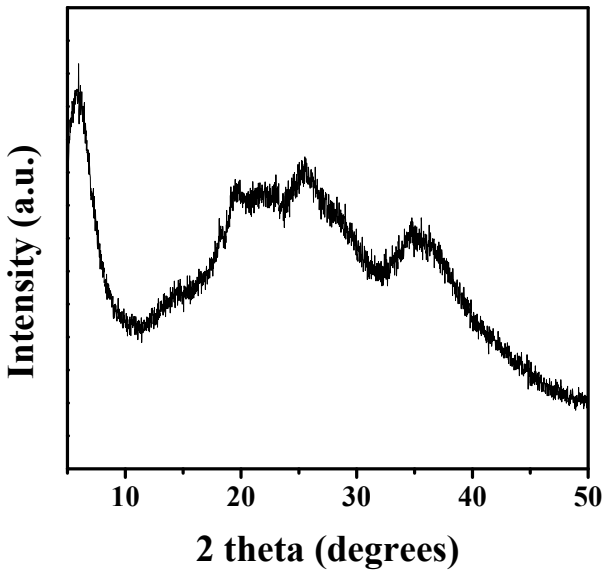


Figure S1 XRD pattern of Mg-Si-ZSM-12(20) synthesized by using NaOH as the alkali source. The initial gel composition: 40 SiO_2: 2 MgO: 8 TEAOH: 320 H_2O; Crystallization: 140 °C, 14 d.

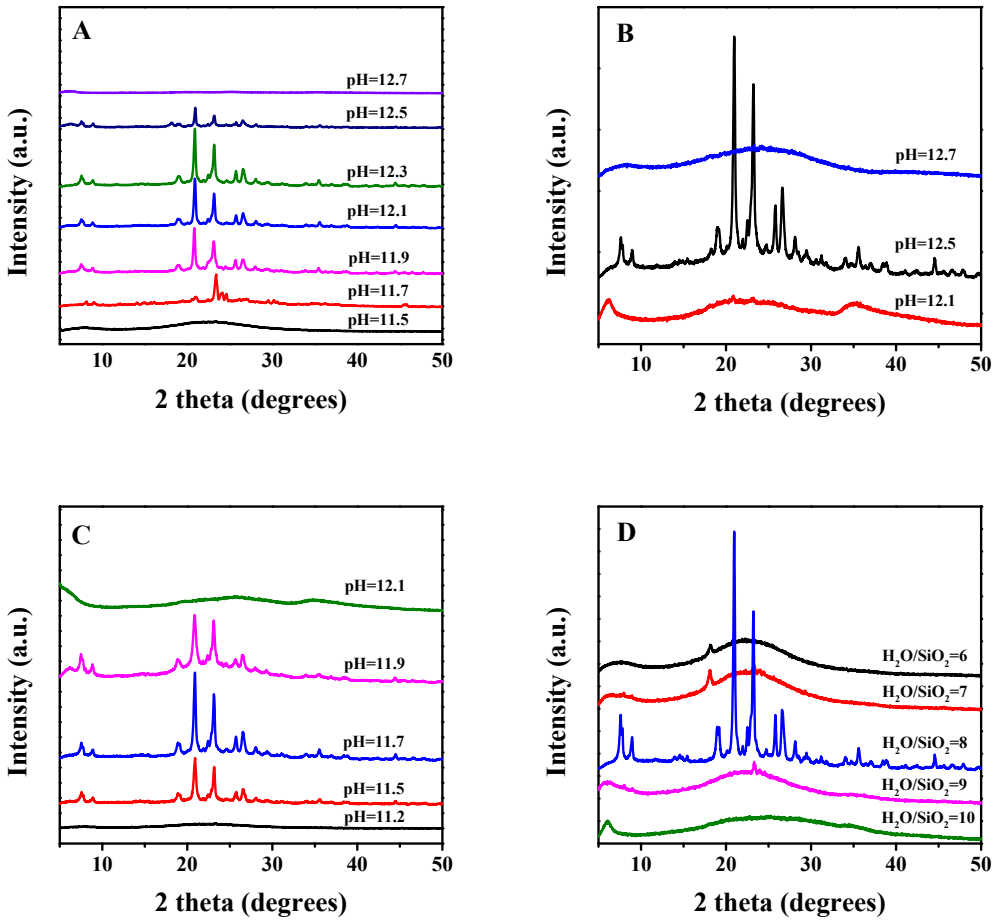


Figure S2 XRD patterns of the samples synthesized by using (A) NaOH, (B) KOH and (C) NaOH and KOH as the alkali sources (NaOH/KOH mass ratio: 1:1). The initial gel composition: 40 SiO_2: 0.8 MgO: 8 TEAOH: 320 H_2O; Crystallization: 140 °C, 14 d. (D) XRD patterns of the samples synthesized with different H_2O/SiO_2 ratio. The initial gel composition: 40 SiO_2: 0.8 MgO: 8 TEAOH: *y* H_2O; Crystallization: 140 °C, 14 d, pH=12.3.

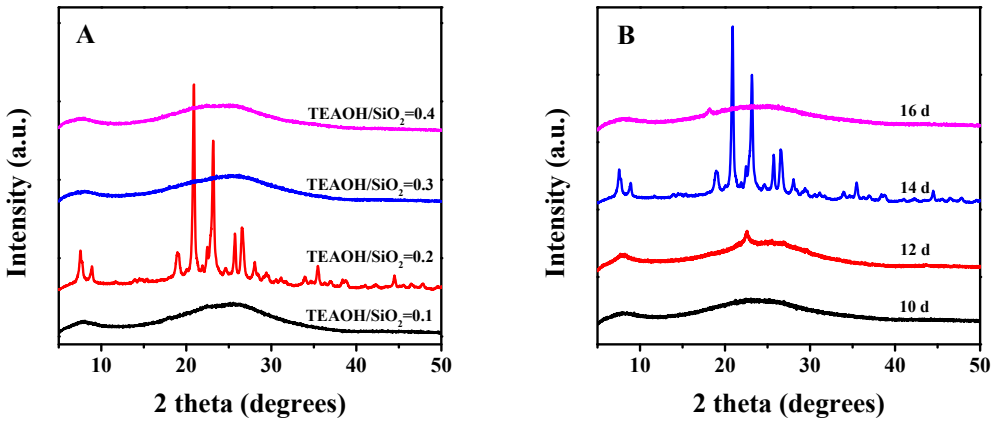


Figure S3 (A) XRD patterns of the samples synthesized with different TEAOH/SiO_2. The initial gel composition: 40 SiO_2: 0.8 MgO: *x* TEAOH: 320 H_2O; Crystallization: 140 °C, 14 d, pH=12.3. (B) XRD patterns of the samples synthesized under different crystallization time, synthetic parameters: 40 SiO_2:0.8 MgO: 8 TEAOH: 320 H_2O; Crystallization: 140 °C, pH=12.3.

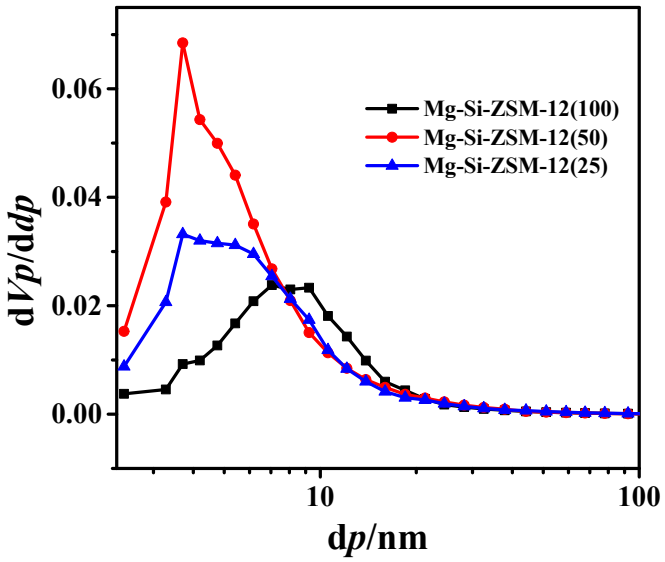


Figure S4 Pore size distribution curves of Mg-Si-ZSM-12(*n*) series.

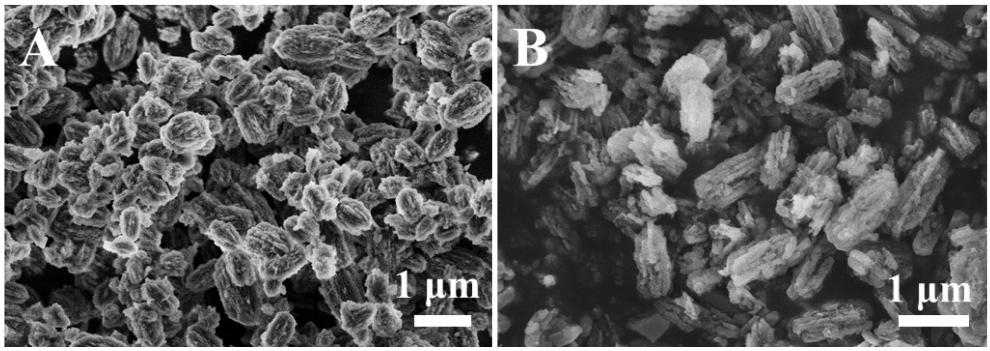


Figure S5 SEM images of (A) Mg-Si-ZSM-12(100) and (B) Mg-Si-ZSM-12(25).

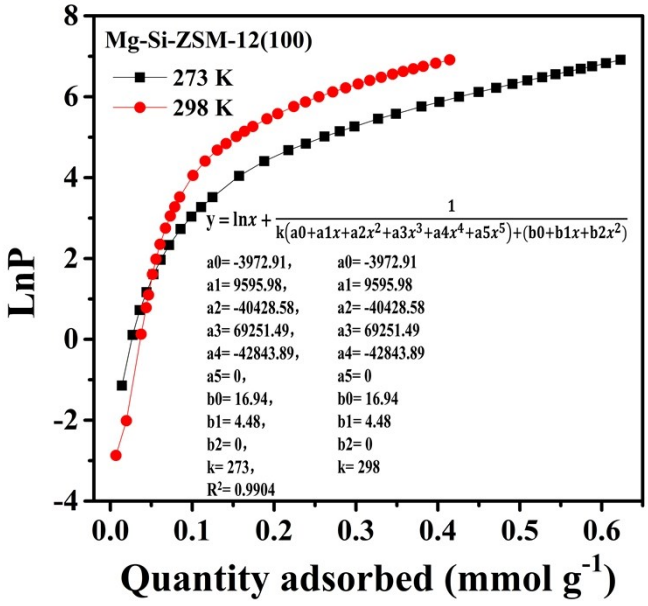


Figure S6 Fitting parameters of virial equation to CO_2 uptakes at 273/298 K for Mg-Si-ZSM-12(100).

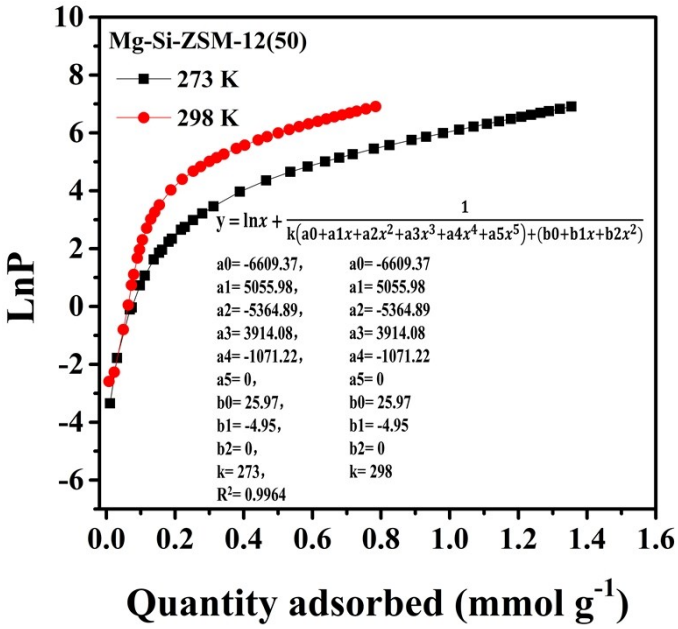


Figure S7 Fitting parameters of virial equation to CO_2 uptakes at 273/298 K for Mg-Si-ZSM-12(50).

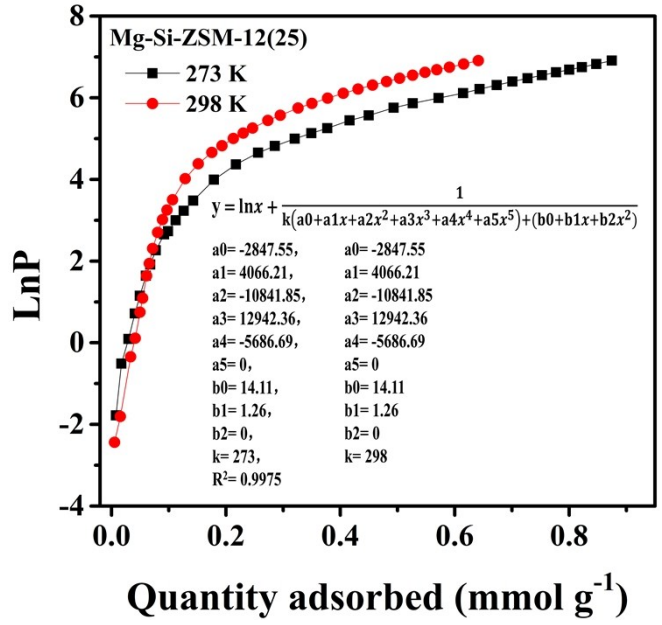


Figure S8 Fitting parameters of virial equation to CO_2 uptakes at 273/298 K for Mg-Si-ZSM-12(25).

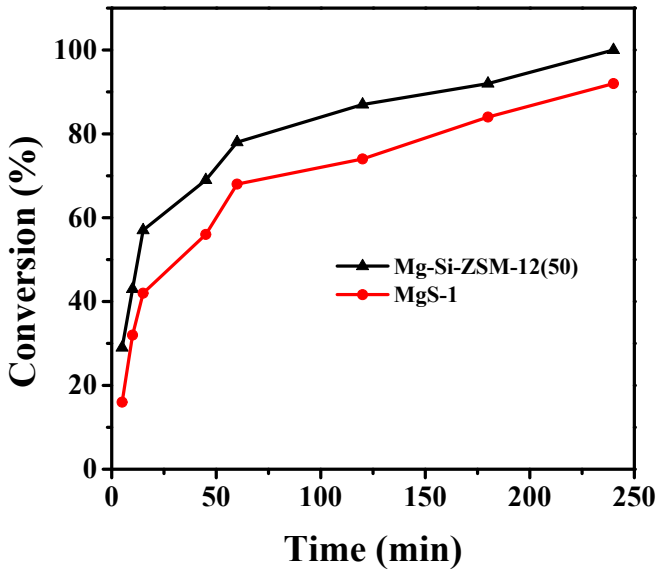


Figure S9 Kinetic curves in Mg-Si-ZSM-12(50) and MgS-1 catalyzed Knoevenagel condensation of benzaldehyde and ethyl cyanoacetate. Reaction condition: benzaldehyde (8 mmol), ethyl cyanoacetate (8 mmol), catalyst (0.1 g), ethanol (5 mL), 70 °C, 4 h.

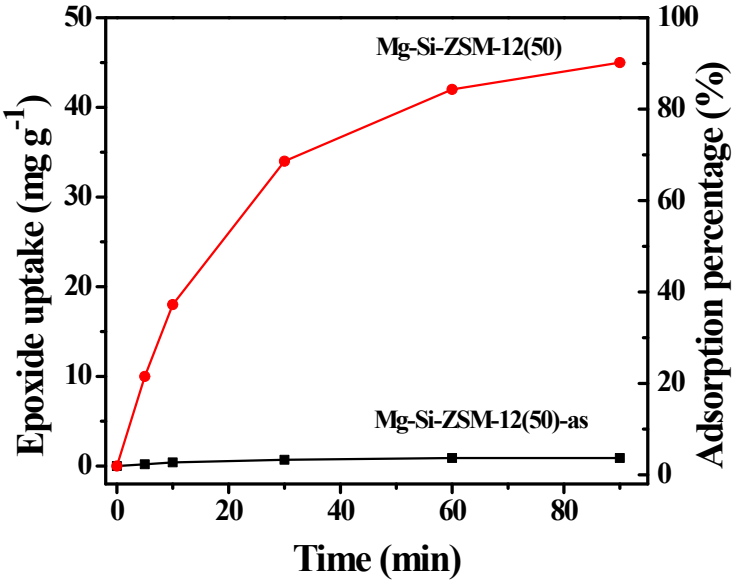


Figure S10 Adsorption kinetic curves of 1,2-epoxytetradecane. The adsorption test was performed by placing 50 mg catalyst in 1 mL ethyl acetate containing 5 mg 1,2-epoxytetradecane. The left Y-axis represents the adsorption amount (mg g^{-1}), while the right one shows the corresponding adsorption percentage (%).

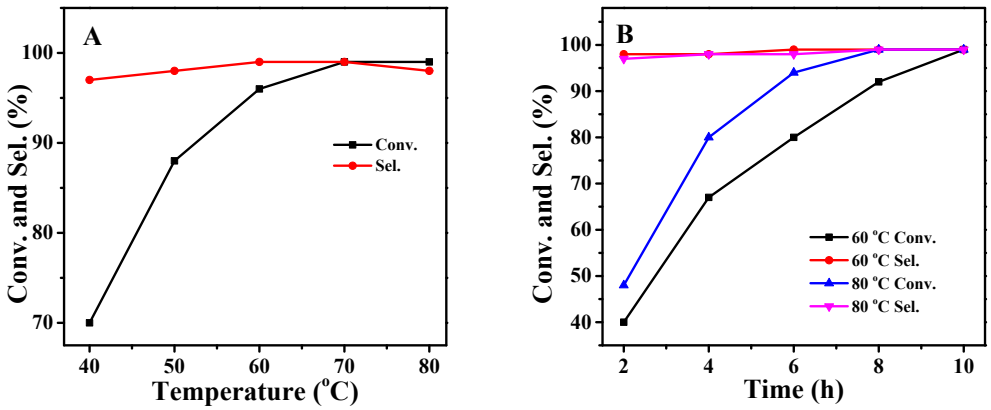


Figure S11 (A) Cycloaddition of CO_2 with epichlorohydrin at different temperature for 10h; (B) Kinetic curves in cycloaddition of CO_2 with epichlorohydrin at different temperature. Reaction condition: epichlorohydrin (5 mmol), catalyst (0.12 g), CO_2 (1.0 bar), TBAB (3 mol%, 0.048 g).

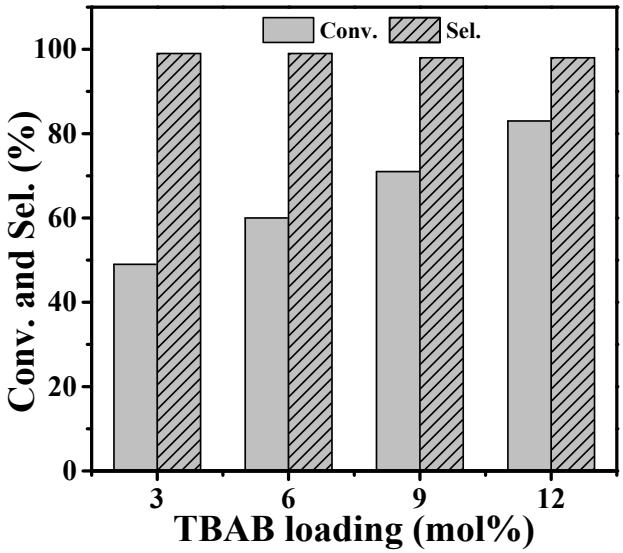


Figure S12 Cycloaddition of CO_2 with 1,2-epoxyhexane with TBAB alone in the reaction. Reaction condition: 1,2-epoxyhexane (5 mmol), catalyst (0.12 g), CO_2 (1.0 bar), 90 °C, 9 h.

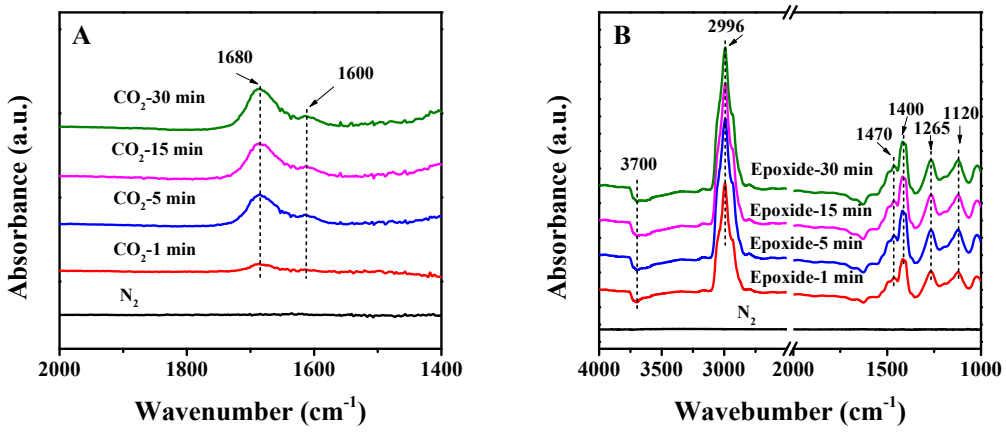
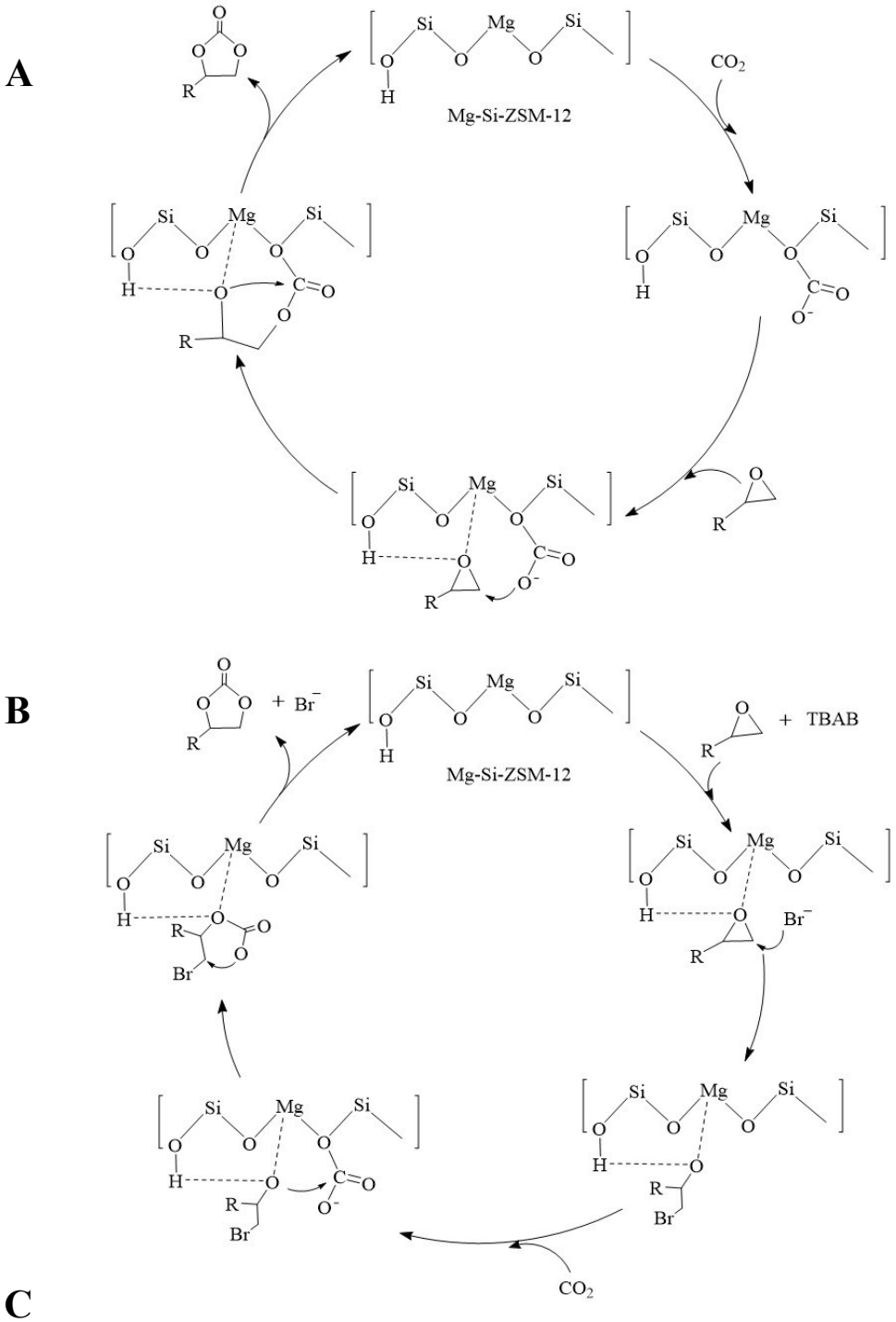


Figure S13 In situ FT-IR spectra of adsorbed (A) CO_2 and (B) epoxide (epoxypropane) on Mg-Si-ZSM-12(50).



Scheme S1 Probable catalytic routes for Mg-Si-ZSM-12 catalyzed CO_2 cycloaddition with epoxide in the (A) absence and (B) presence of TBAB. (C) The formation of side-product 1,2-diol in the presence of trace water.