

Supporting Information for

Activation of hydrocarbons on acidic zeolites: superior selectivity of methylation of ethene with methanol to propene on weakly acidic catalysts

Qingjun Zhu ^a, Junko N. Kondo ^a, Tohru Setoyama ^b, Masashi Yamaguchi ^b, Kazunari Domen ^c and Takashi Tatsumi ^{a*}

^a Chemical Resources Laboratory, Tokyo Institute of Technology, 4259 Nagatsuta, Midori-ku, Yokohama 226-8503, Japan. Email address: ttatsumi@cat.res.titech.ac.jp.

^b Mitsubishi Chemical Group, Science and Technology Research Center, 1000 Kamoshida-cho, Aoba-ku, Yokohama 227-8502, Japan

^c Department of Chemical System Engineering, The University of Tokyo, 7-3-1 Hongo, Bunkyo-ku Tokyo 113-8656, Japan

Supporting Information Table of contents

Experimental details	Page S2
XRD, SEM and IR spectra of studied catalysts	Page S3-4
Reaction stability over H-[B]-MFI catalyst at 773 K	Page S4

Catalyst preparation

The MFI zeolites were prepared by hydrothermal synthesis in autoclaves. Cab-O-Sil M5, Al(NO₃)₃·9H₂O and H₃BO₃ were employed as silicon, aluminum and boron sources, respectively. Tetrapropylammonium bromide (TPABr) was used as the template. After the sol-gel was formed, the autoclave was kept at 443 K for 7 days under tumbling conditions. The obtained crystals were filtered, washed and dried at 373 K overnight. The template was removed by calcination at 823 K for 10 h. The sodium type of MFI zeolites was ion-exchanged with NH₄NO₃, followed by calcinations to produce proton type catalysts. The Si to Al ratio in the resultant H-ZSM-5 was approximately the same as that in the starting gel. However, the Si to B ratio was 41 in the H-[B]-MFI product whereas the Si to B ratio was 5 in the sol-gel mixture. H-ZSM-5 and H-[B]-MFI are heteroatom (Al and B, respectively) substituted zeolite materials with the same crystal structure of MFI topology, and thus they showed similar XRD patterns.

Catalyst characterization

Powder X-ray diffraction was measured using a Rigaku diffractometer. Typically, an XRD diffractogram was recorded in the range of 2° < 2θ < 50° using Cu Kα radiation at a scanning speed of 0.04° min⁻¹.

The microscopic features of synthesized zeolites were observed by scanning electron microscopy (SEM). Images were recorded by a Hitachi S-4700 microscope for samples without any metal coating.

The catalyst sample of approximately 10 mg was pressed into a self-supporting wafer. All the IR spectra were recorded on an FT/IR 7300 spectrometer with an MCT detector.

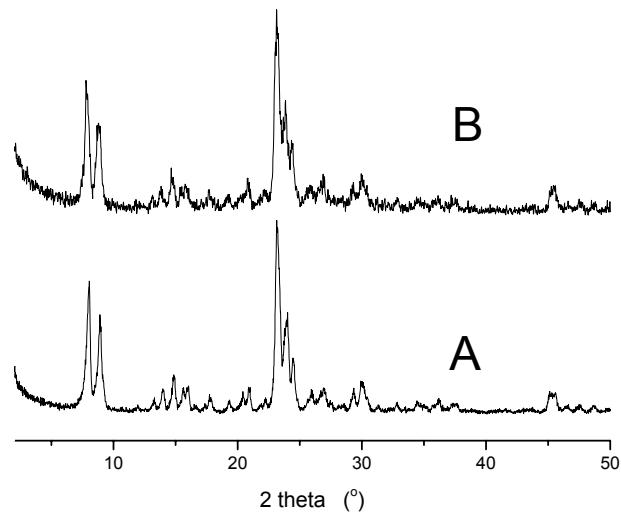
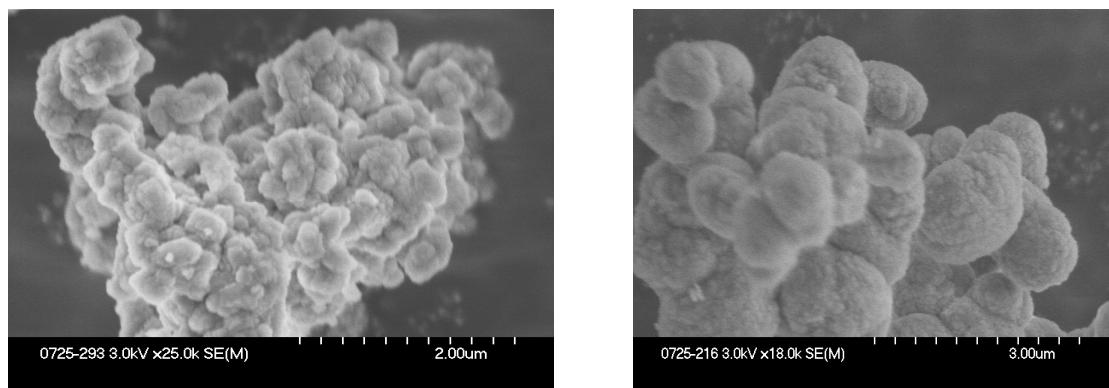


Fig.S1 Powder x-ray diffraction patterns for (A) H-ZSM-5, (B) H-[B]-MFI.



(A)

(B)

Fig.S2 SEM images of (A) H-ZSM-5, (B) H-[B]-MFI.

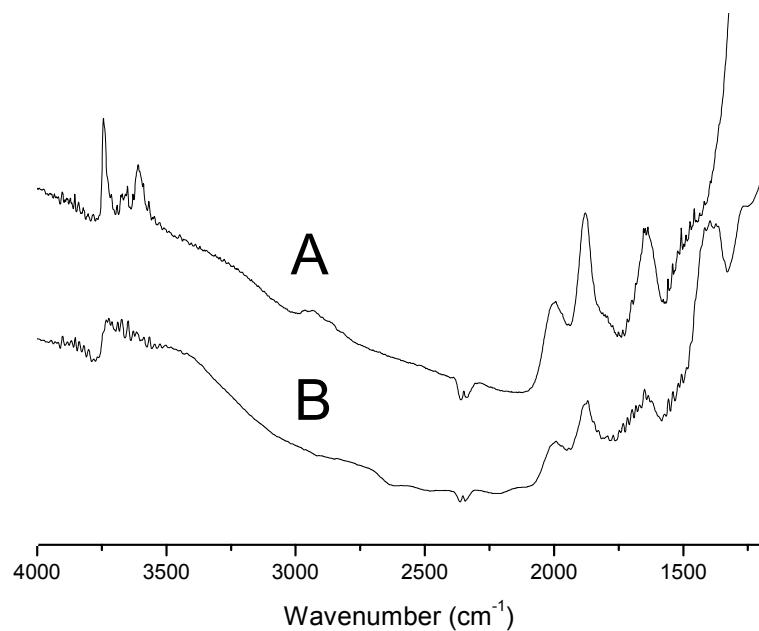


Fig.S3 IR spectra of (A) H-ZSM-5, (B) H-[B]-MFI.

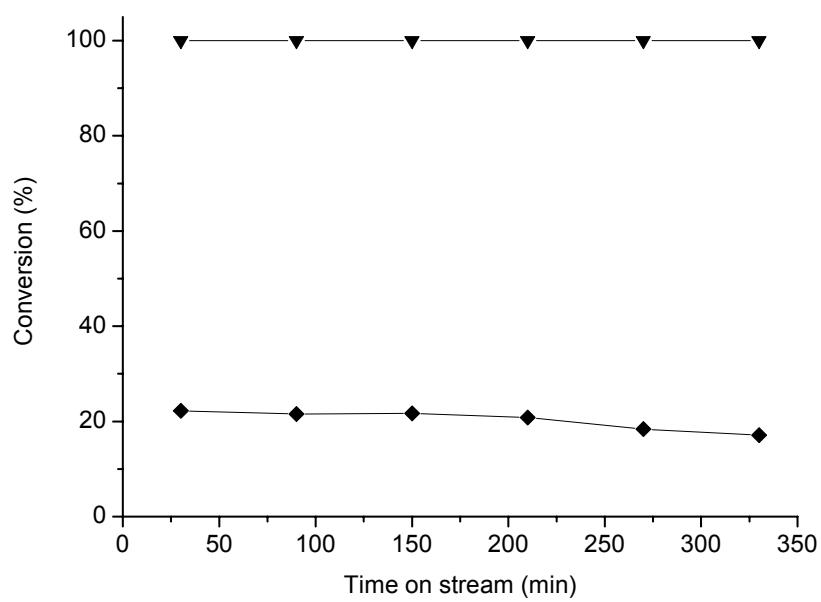


Fig.S4 Reaction stability of co-reaction of methanol and ethene over H-[B]-MFI catalyst at 773 K, (▼) conversion of methanol, (♦)conversion of ethylene.