## **Supporting Information**

## Assessment of Acid Catalytic Properties of Ferrosilicate MFI Zeolite by Methanol-to-Hydrocarbon Conversion

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**Figure S1.** Thermogravimetric analysis (TGA) data of the fully deactivated  $MFI_{TPAOH/(TEOS+Fe)}$  catalyst after the methanol-to-hydrocarbon reaction. The data was obtained by flowing the 40:60 mixture of N<sub>2</sub> and air gases by 100 cm<sup>3</sup> min<sup>-1</sup>, while heating the sample to 973 K at a ramping rate of 10 K min<sup>-1</sup>.



**Figure S2**. Regenerations of the MFI<sub>TPAOH/(TEOS+Fe)</sub> catalyst in the methanol-to-hydrocarbon reaction. Reaction conditions were as follows: 50 mg catalyst, 400 °C, and WHSV of methanol = 50 h<sup>-1</sup>. The reactivation conditions were as follows: (a) flowing 2 vol% O<sub>2</sub> in N<sub>2</sub> under 500 °C for 2 h, (b) flowing air under 550 °C for 1 h.



**Figure S3**. The MTH reaction catalytic performance of Fe-MFI synthesized by dropwisely adding H<sub>2</sub>O-TEOS-TPAOH mixture into iron nitrate solution.<sup>[36]</sup> Reaction conditions were as follows: 50 mg catalyst, 400 °C, and WHSV of methanol = 10 h<sup>-1</sup>.



Figure S4. FT-IR spectra of pyridine-adsorbed ZSM-5, and the calculated amount of Brønsted acid sites after the desorption at each temperature. Spectra were measured after the desorption of physisorbed pyridine at 150 °C (black line), 300 °C (blue line) and 500 °C (red line) for 1 h.



**Fig. S5.** The amount of Brønsted acid sites of Fe-MFI zeolites measured after the desorption of pyridine at 500  $^{\circ}$ C, and their C<sub>2+</sub> hydrocarbons yield at 4 h of reaction time-on-stream.

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	MFI <sub>TPAOH/(TEOS+Fe)</sub>	MFI <sub>Fe/(TEOS+TPAOH)</sub>	MFI <sub>Fe/(NaSil+TPABr)</sub>	MFI <sub>TPABr/(NaSil+Fe)</sub>	
Relative Crystallinity (%)	88.7	87.1	91.0	84.5	

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	Table S1. Relative cr	stallinity in com	parison	to the case of r	ourely	siliceous MF	I zeolite.



**Figure S6.** Solid state <sup>29</sup>Si MAS NMR spectra of the Fe-MFI zeolites, and purely siliceous MFI zeolite as reference. The measured spectra were drawn in solid red lines, and the calculated proportions of  $Q_3$  over  $Q_4$  were written over each NMR result. Grey curves indicate peak deconvolution, and the black dashed lines illustrate the integration of deconvoluted peaks.



Figure S7. SEM images of (a)  $MFI_{TPAOH/(TEOS+Fe)}$ , (b)  $MFI_{Fe/(TEOS+TPAOH)}$ , (c)  $MFI_{Fe/(NaSil+TPABr)}$  and (d)  $MFI_{TPABr/(NaSil+Fe)}$ , and the (e) particle size distributions of each sample.



Figure S8. X-ray absorption of Fe-MFI at Fe K-edge region.