

Figure S1. UV-vis spectra of H-ZSM-5 (a) and 0.52% Fe-ZSM-5 (b). The raw data and the deconvoluted data are shown as a black line and a dashed line, respectively.

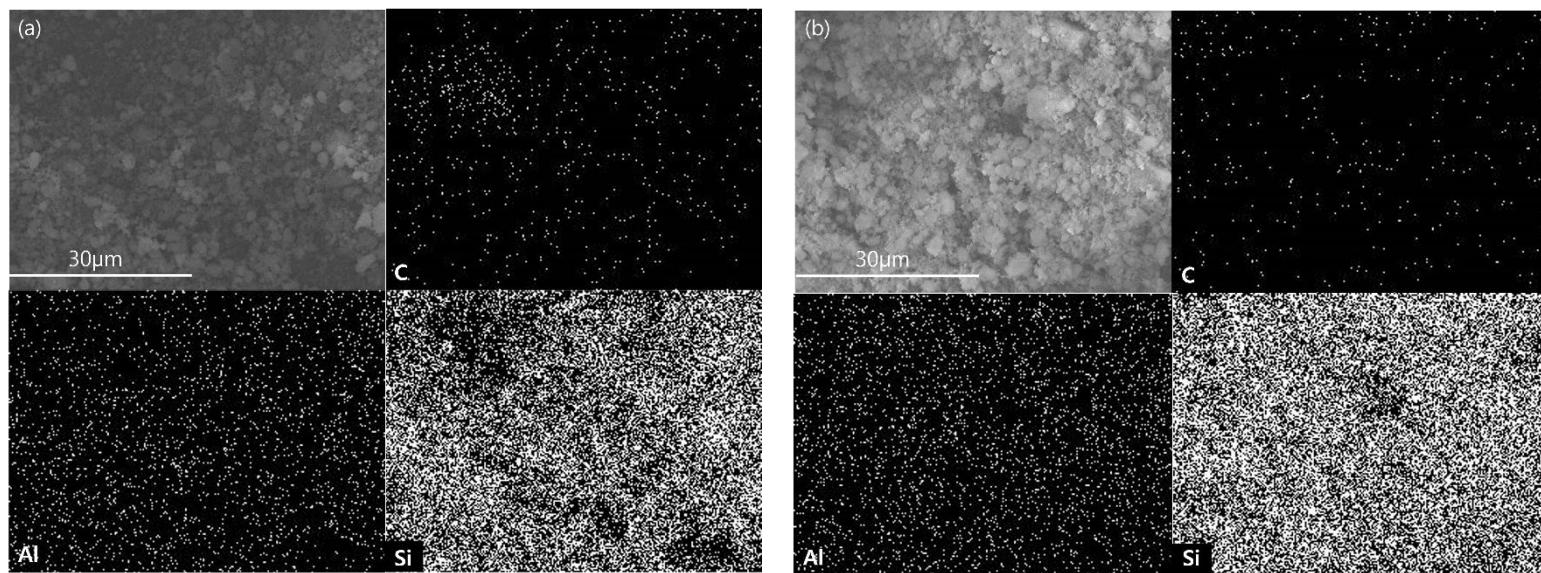


Figure S2. SEM-EDX results of (a) C0.45-H-ZSM-5, (b) C1.08-Fe-ZSM-5

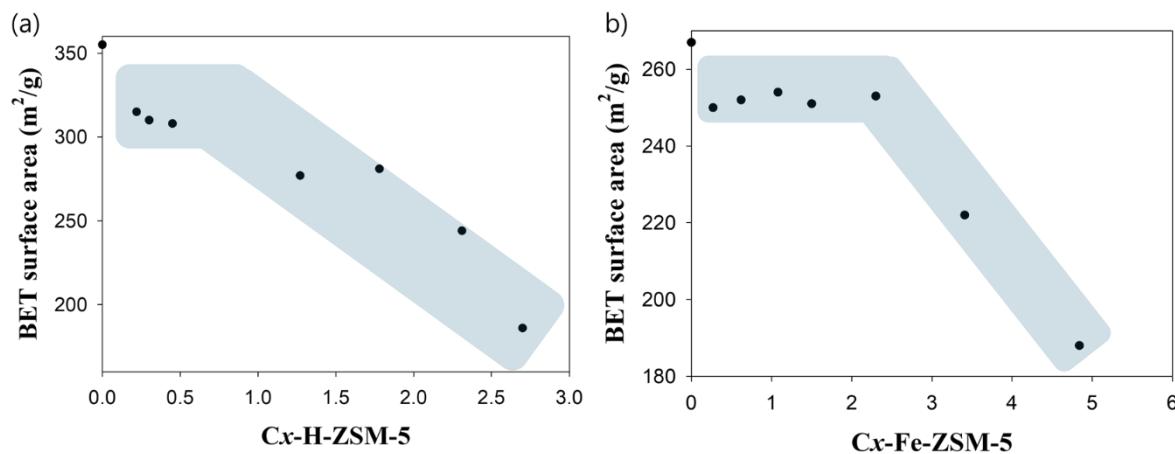


Figure S3. The changes in surface area according to the carbon content in (a) Cx-H-ZSM-5 and (b) Cx-Fe-ZSM-5 .

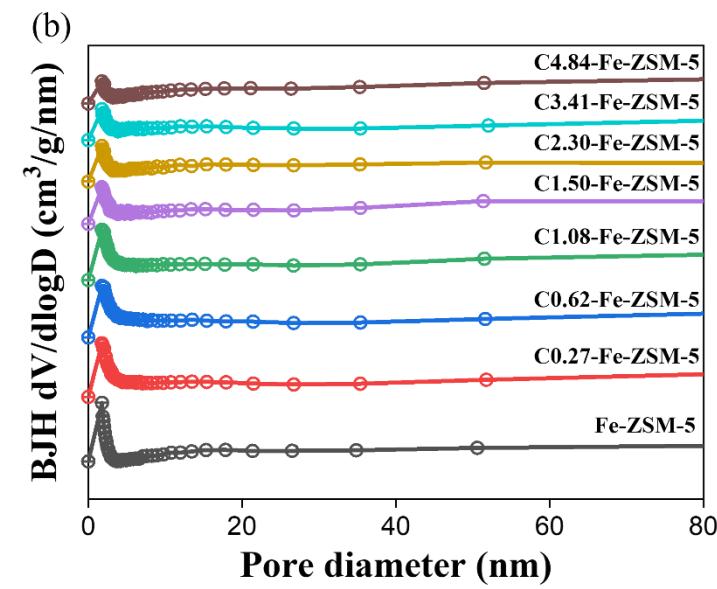
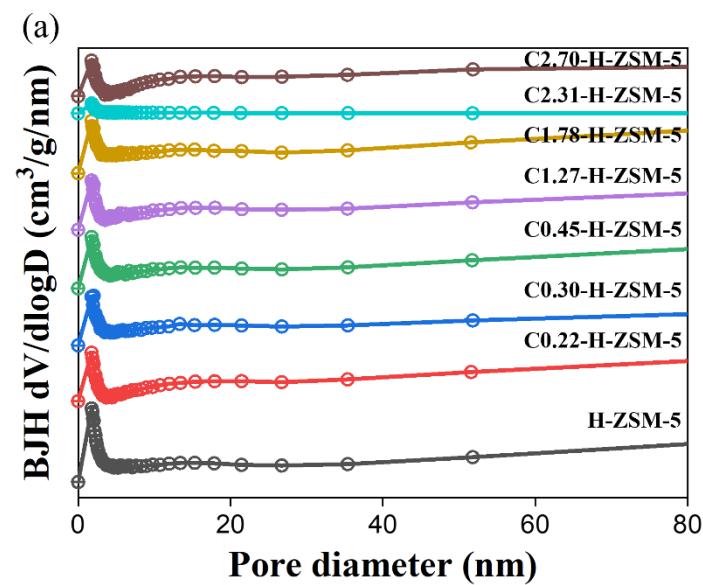


Figure S4. The pore size distribution of (a) Cx-H-ZSM-5 and (b) Cx-Fe-ZSM-5.

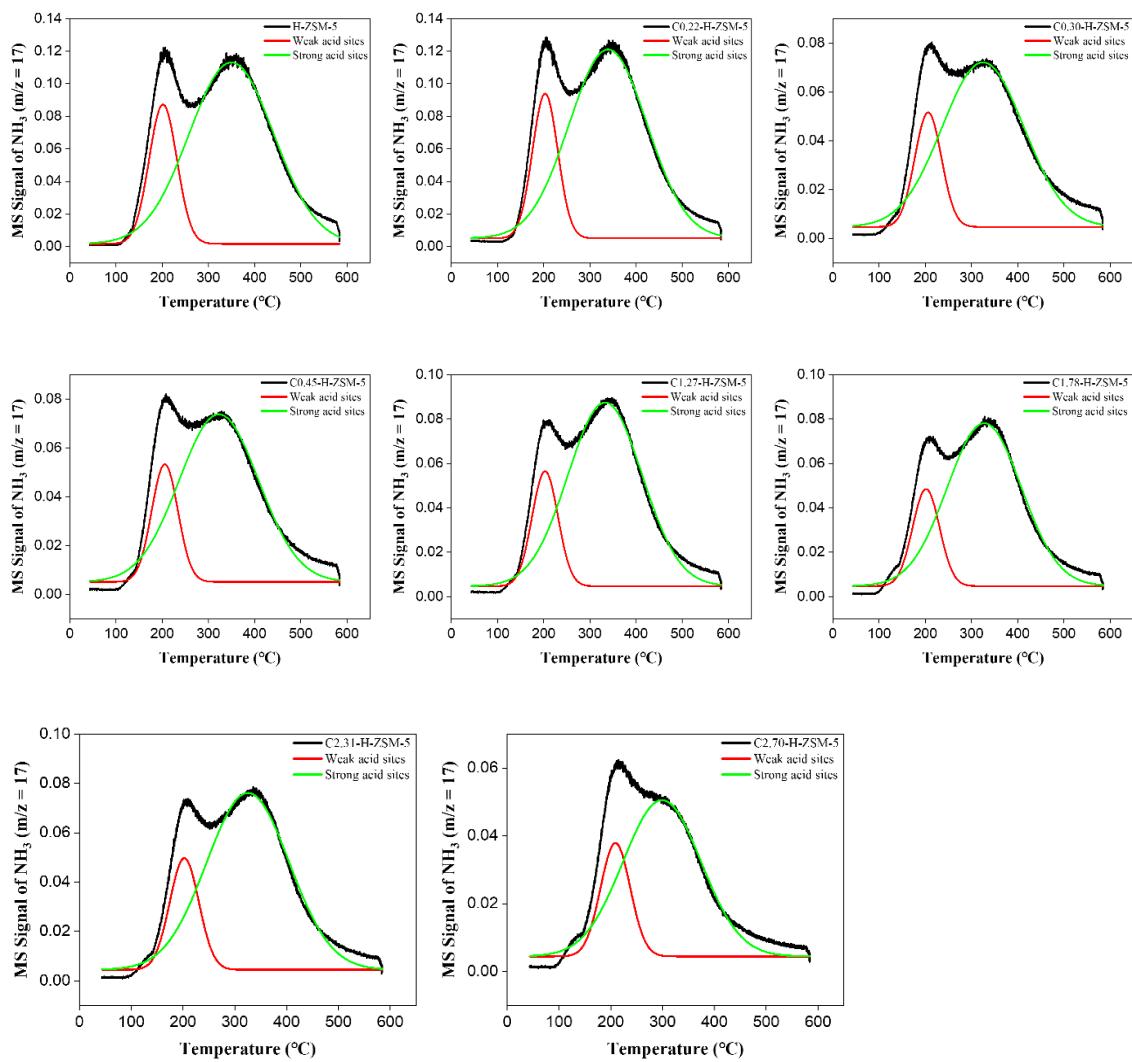


Figure S5. Deconvolution of NH_3 -TPD profiles for H-ZSM-5 and carbon-coated H-ZSM-5 with different carbon contents.

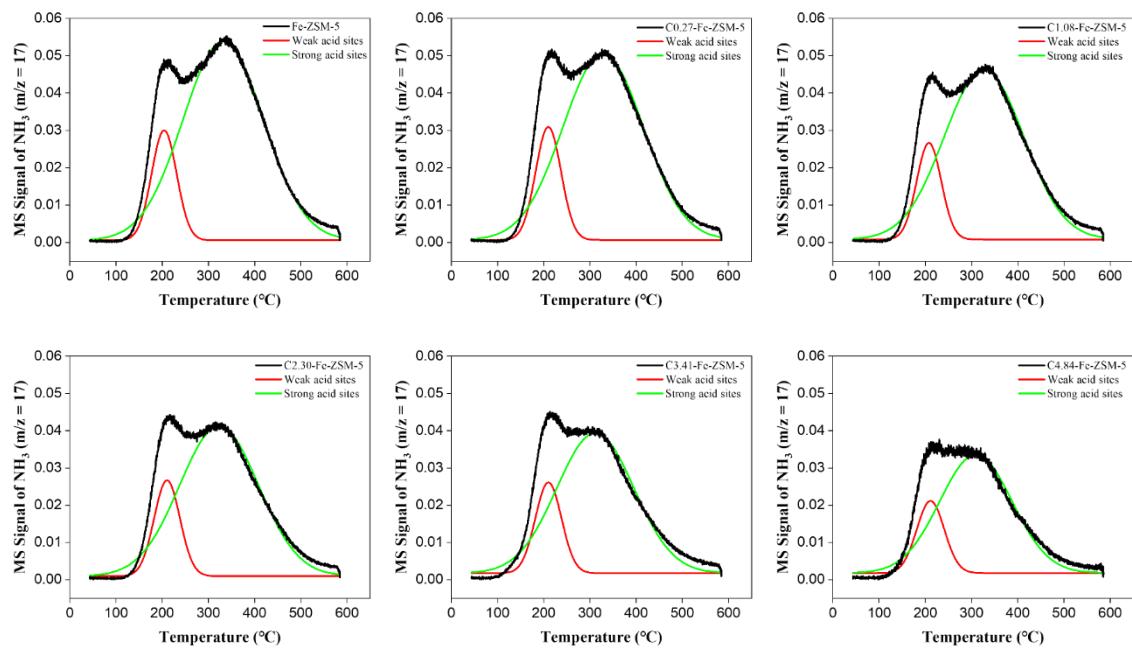


Figure S6. Deconvolution of NH₃-TPD profiles for Fe-ZSM-5 and carbon-coated Fe-ZSM-5 with different carbon contents.

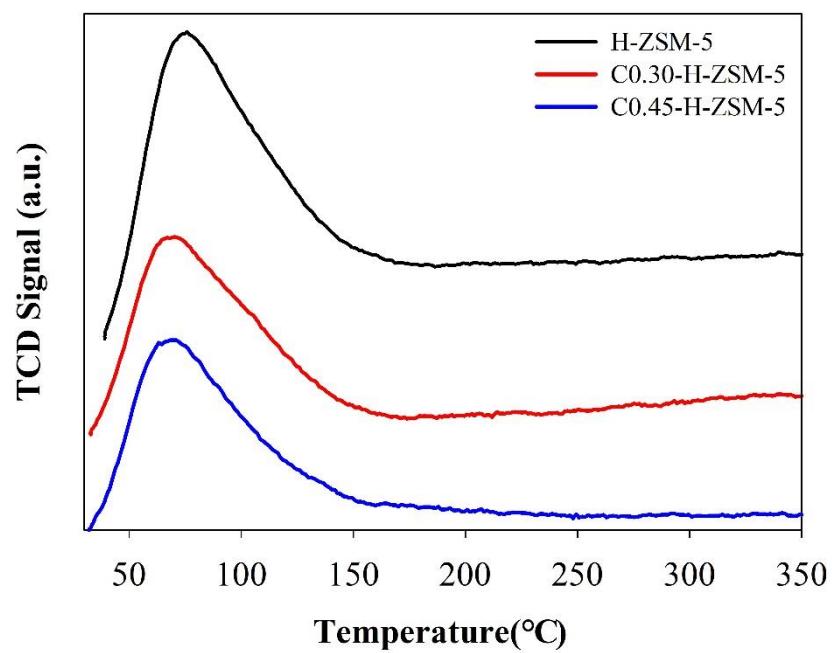


Figure S7. H₂O-TPD profiles for H-ZSM-5 and carbon-coated H-ZSM-5 with different carbon contents.

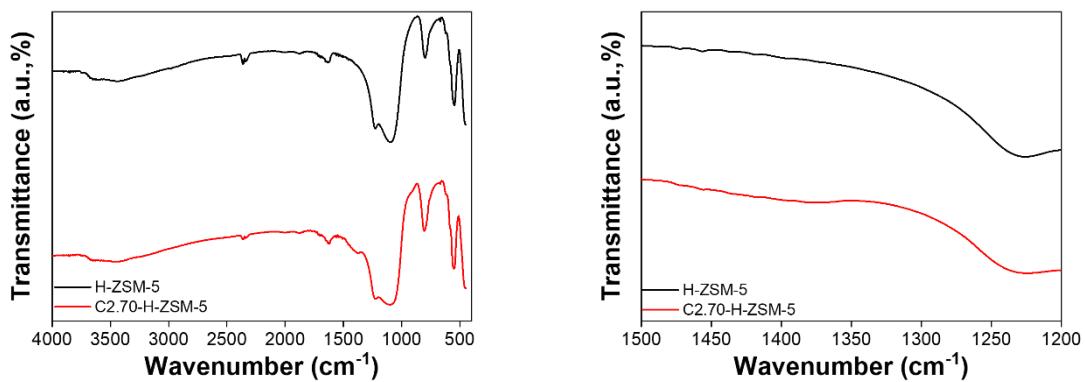


Figure S8. FT-IR spectra for H-ZSM-5 and carbon-coated H-ZSM-5 (C2.70-H-ZSM-5).

The FT-IR spectra of H-ZSM-5 and carbon-coated H-ZSM-5 show the bands at 1085–1100 cm⁻¹ and 798–810 cm⁻¹ due to the zeolite framework, i.e., the asymmetric and symmetric vibration modes of T-O-T (T = Si and Al) groups [1]. The broad but distinct vibration band at 542–552 cm⁻¹ is attributed to the asymmetric stretching mode of the double-five ring in ZSM-5 [1]. Also, the broad band at 3413 cm⁻¹ and the sharp peak at 1635 cm⁻¹ are attributed to the structural hydroxyl groups and bending mode of physically adsorbed water, respectively [1]. The weak band at 1377 cm⁻¹ observed over carbon-coated H-ZSM-5 (C2.70-H-ZSM-5) can be ascribed to the bending mode of vibration of –C-H (CH₃) [2]. This species may be due to a carbon species formed through the polymerization and subsequent thermal decomposition of furfuryl alcohol.

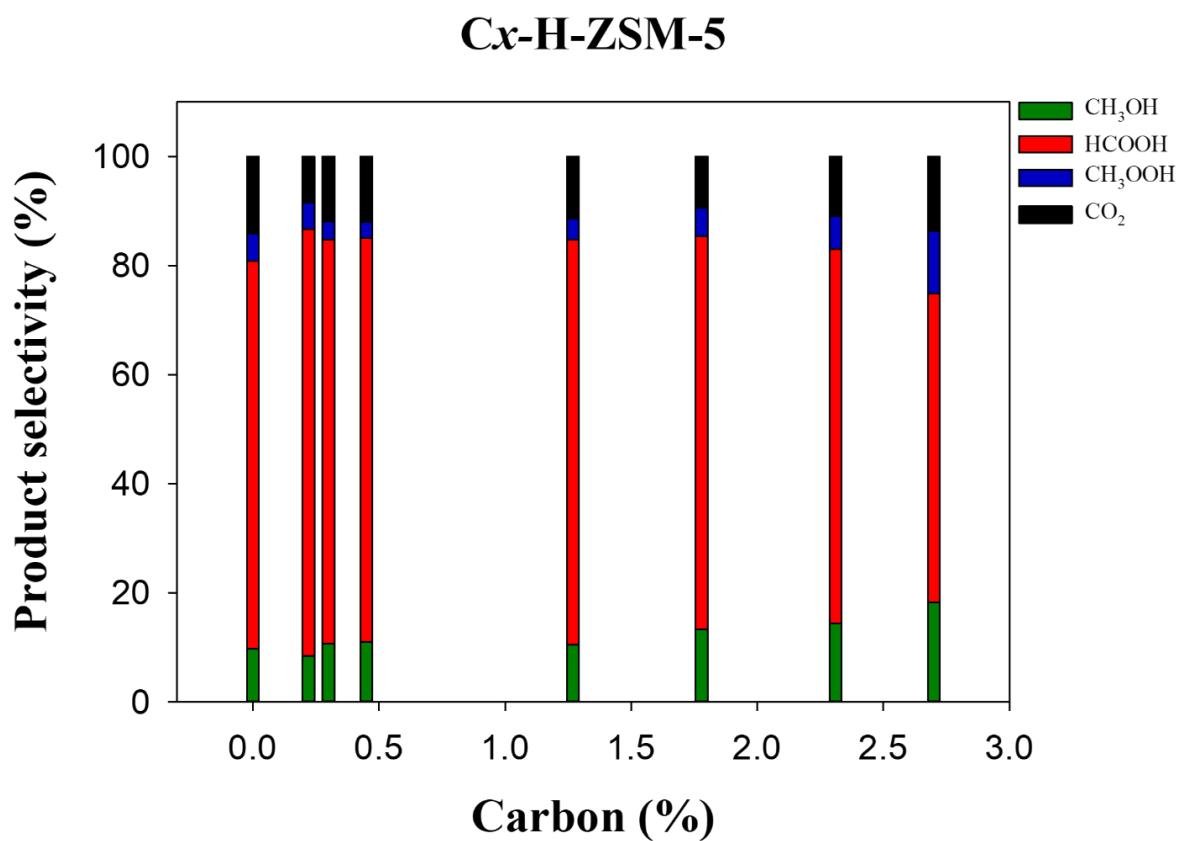


Figure S9. Product selectivity for partial oxidation of methane with H₂O₂ over H-ZSM-5 and Cx-H-ZSM-5.

Cx-Fe-ZSM-5

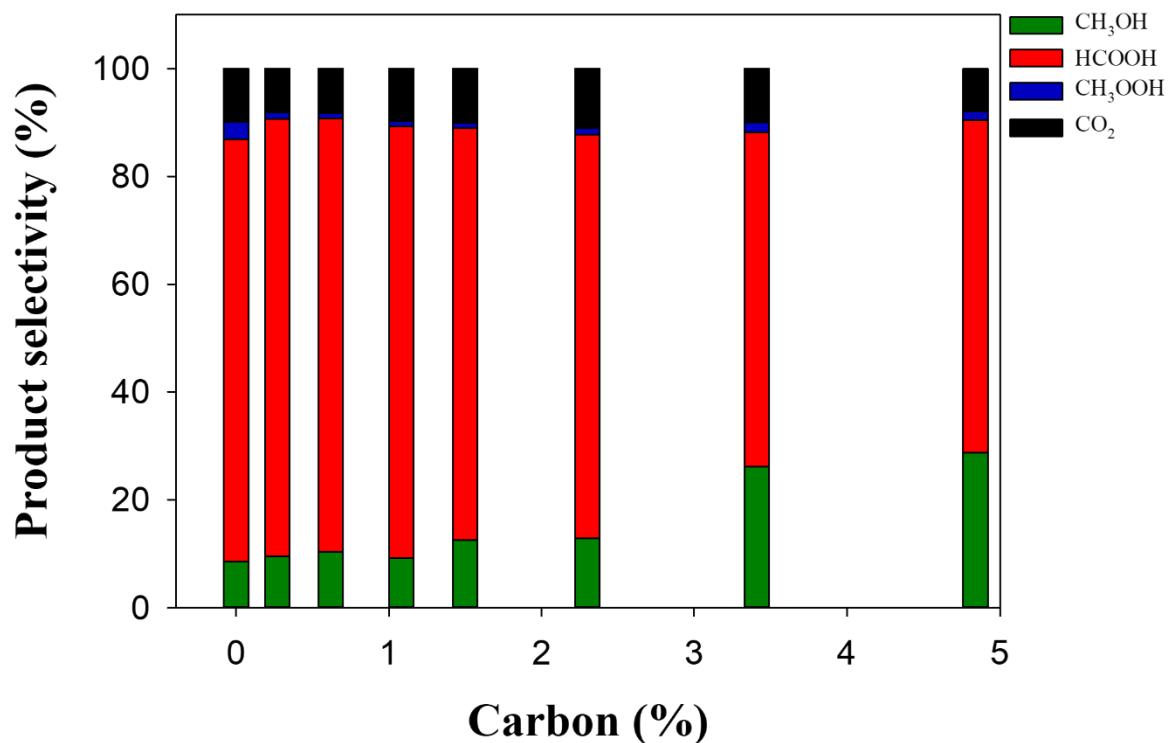


Figure S10. Product selectivity for partial oxidation of methane with H₂O₂ over Fe-ZSM-5 and Cx-Fe-ZSM-5.

Table S1. The fraction of UV-vis band for H-ZSM-5 and 0.52% Fe-ZSM-5.

Catalyst	Fraction of UV-vis band (%)		
	250~350 nm	350~450 nm	>450 nm
H-ZSM-5	89	11	-
0.52% Fe-ZSM-5	59	31	10

Table S2. The surface acid properties of H-ZSM-5 and carbon-coated H-ZSM-5 with different carbon contents.

Catalyst	Weak acid sites			Strong acid sites			Desorbed NH ₃ mmol/g _{cat}
	Temp. (°C)	Area	Area (%)	Temp. (°C)	Area	Area (%)	
H-ZSM-5	201	6.46	20.2	349	25.6	79.8	1.49
C0.22-H-ZSM-5	202	6.10	20.0	339	24.5	80.0	1.16
C0.30-H-ZSM-5	205	3.38	18.4	324	15.0	81.6	0.862
C0.45-H-ZSM-5	205	3.42	18.8	322	14.8	81.2	0.880
C1.27-H-ZSM-5	202	3.63	17.9	332	16.6	82.1	0.900
C1.78-H-ZSM-5	201	3.17	17.7	327	14.7	82.3	0.897
C2.31-H-ZSM-5	202	3.19	18.2	325	14.4	81.8	0.849
C2.70-H-ZSM-5	210	2.06	16.1	296	10.7	83.8	0.578

Table S3. The surface acid properties of Fe-ZSM-5 and carbon-coated Fe-ZSM-5 with different carbon contents.

Catalyst	Weak acid sites			Strong acid sites			Desorbed NH ₃ mmol/g _{cat}
	Temp. (°C)	Area	Area (%)	Temp. (°C)	Area	Area (%)	
Fe-ZSM-5	204	2.00	14.66	332	11.66	85.34	1.03
C0.27-Fe-ZSM-5	209	2.08	16.10	328	10.87	89.90	0.981
C1.08-Fe-ZSM-5	208	1.72	14.96	327	9.79	85.04	0.934
C2.30-Fe-ZSM-5	210	1.79	17.27	323	8.61	82.73	0.879
C3.41-Fe-ZSM-5	210	1.72	17.99	312	7.89	82.01	0.857
C4.84-Fe-ZSM-5	211	1.44	18.83	309	6.24	81.17	0.672

Table S4. The contact angle of water-droplet for H-ZSM-5, carbon-coated H-ZSM-5, Fe-ZSM-5, and carbon-coated Fe-ZSM-5.

Catalyst	Contact angle (°)
H-ZSM-5	11.6±0.2
C0.30-H-ZSM-5	11.8±0.8
C1.27-H-ZSM-5	12.7±0.5
C2.70-H-ZSM-5	19.1±1.2
Fe-ZSM-5	17.4±0.6
C0.62-Fe-ZSM-5	18.2±0.5
C3.41-Fe-ZSM-5	20.4±0.6
C4.84-Fe-ZSM-5	29.2±0.3

Table S5. The specific amount of furfuryl alcohol used for the synthesis of carbon-coated H-ZSM-5 and carbon-coated Fe-ZSM-5.

Catalyst	Volume of furfuryl alcohol (μL)
C0.22-H-ZSM-5	5.1
C0.30-H-ZSM-5	13
C0.45-H-ZSM-5	25
C1.27-H-ZSM-5	51
C1.78-H-ZSM-5	76
C2.31-H-ZSM-5	101
C0.27-Fe-ZSM-5	5.0
C0.62-Fe-ZSM-5	9.3
C1.08-Fe-ZSM-5	19
C1.50-Fe-ZSM-5	37
C2.30-Fe-ZSM-5	56
C3.41-Fe-ZSM-5	93
C4.84-Fe-ZSM-5	130

Reference

- [1] Asghari, A.; Khorrami, M.K.; Kazemi, S.H. Hierarchical H-ZSM5 zeolites based on natural kaolinite as a high-performance catalyst for methanol to aromatic hydrocarbons conversion. *Sci Rep* **2019**, *9*, 17526, doi:10.1038/s41598-019-54089-y.
- [2] Setyaningrum, D.L.; Riyanto, S.; Rohman, A. Analysis of corn and soybean oils in red fruit oil using FTIR spectroscopy in combination with partial least square. *Int. Food Res. J.* **2013**, *20*(4), 1977-1981.