

## *Supporting Information*

# Cu-Erionite Zeolite Achieves High Yield in Direct Oxidation of Methane to Methanol by Isothermal Chemical Looping

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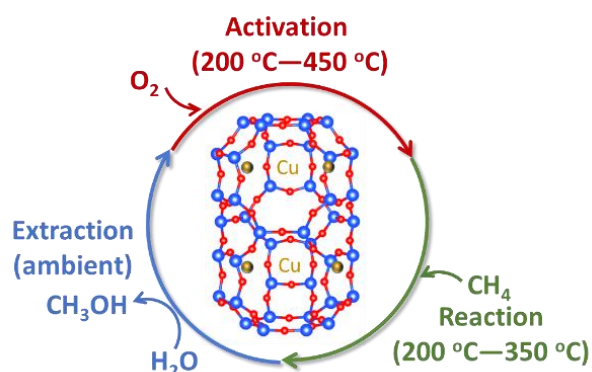
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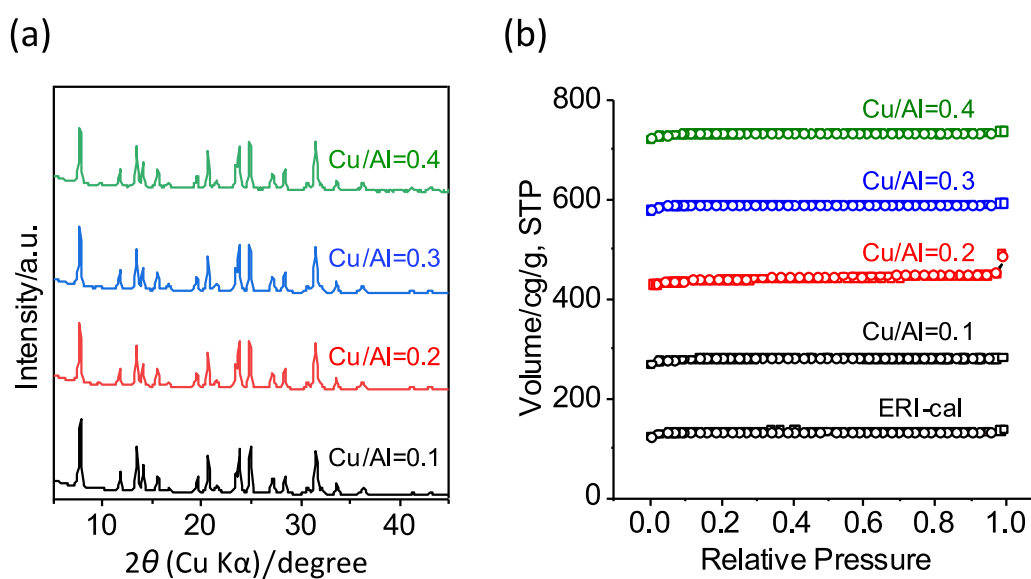
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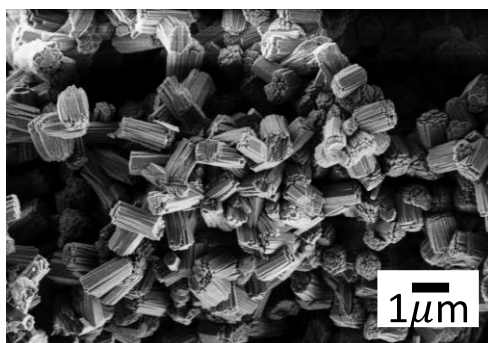
## Supplementary Figures



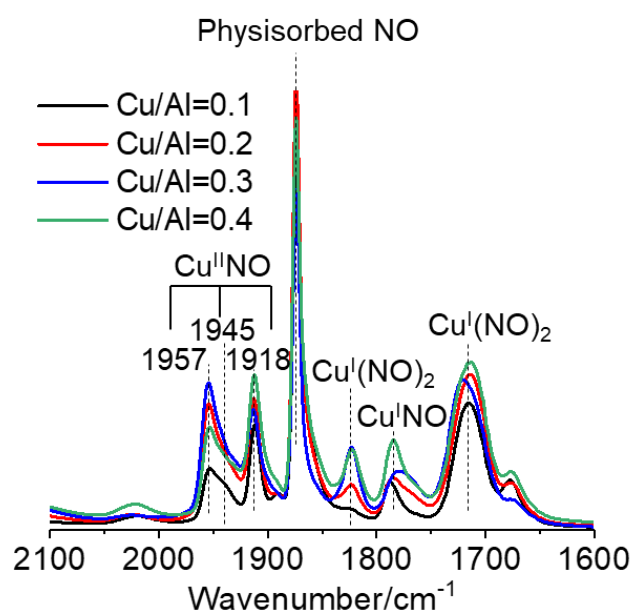
**Figure S1.** Illustration of stepwise conversion of methane to methanol including activation with oxygen, methane oxidation and extraction of methanol.



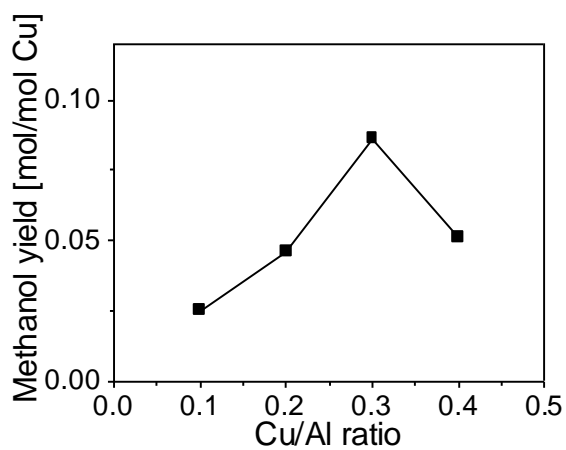
**Figure S2.** XRD patterns and nitrogen adsorption-desorption isotherms of the Cu-ERI materials. (a) Powder X-ray diffraction patterns of Cu-ERI zeolites with different copper loadings. (b) Nitrogen adsorption-desorption isotherms of Cu-ERI zeolites with different copper loadings.



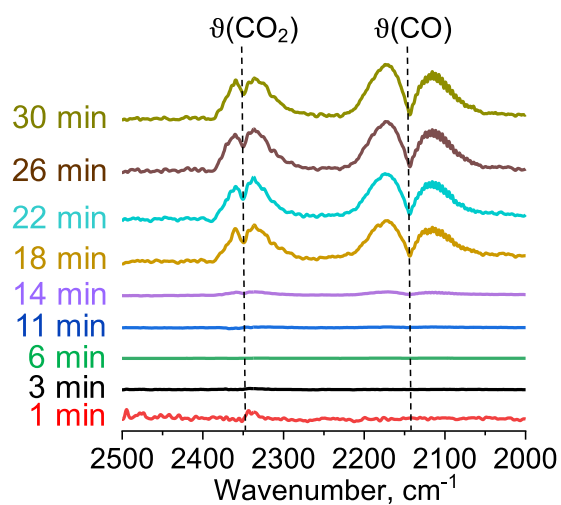
**Figure S3.** SEM image of the parent ERI zeolite.



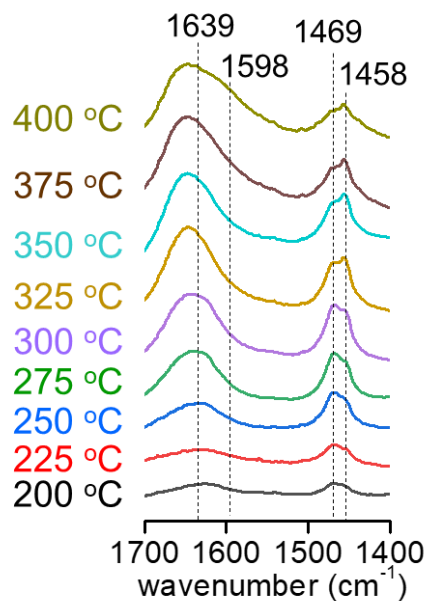
**Figure S4.** In-situ FTIR spectra of nitrogen monoxide over Cu-ERI zeolites after activation in 300 torr of oxygen at 673 K for 1 h.



**Figure S5.** Methanol yield over the Cu-ERI zeolites normalized by copper after reaction at 200 °C and methane partial pressure of 30 bar.

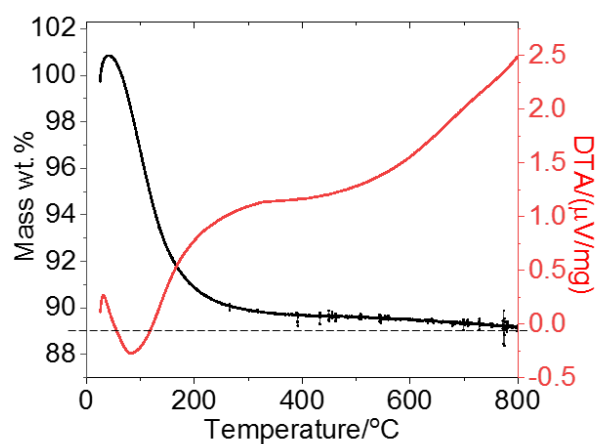


**Figure S6.** In-situ FTIR spectra of surface species formed after the reaction with methane at different temperatures for 30 min.

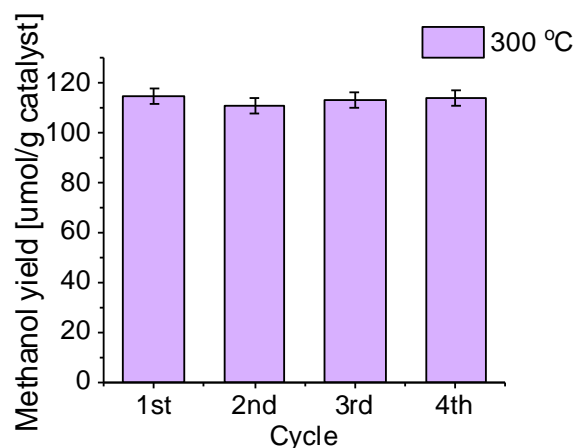


**Figure S7.** In-situ FTIR spectra of species formed in the gas phase through the reaction with methane in 30 min at 300 °C. The amount of carbon monoxide and carbon dioxide formed as gas phase are calculated as 9.4  $\mu\text{mol/g-zeolite}$  and 2.5  $\mu\text{mol/g-zeolite}$ , respectively. A methanol yield of 80  $\mu\text{mol/g-zeolite}$  was achieved, thus the selectivity for Cu-ERI(0.3) was calculated as

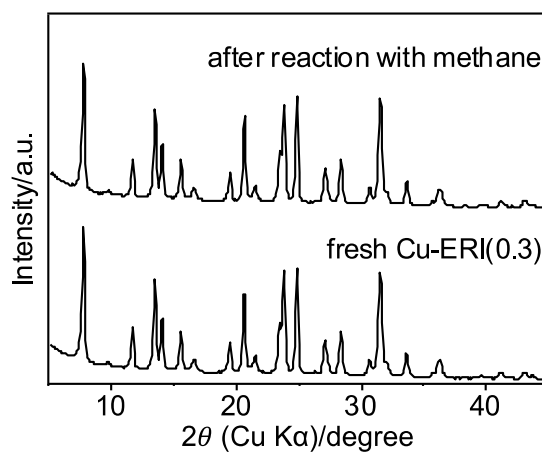
$$\frac{80 \mu\text{mol/g-zeolite}}{(80+9.4+2.5) \mu\text{mol/g-zeolite}} = 87\%.$$



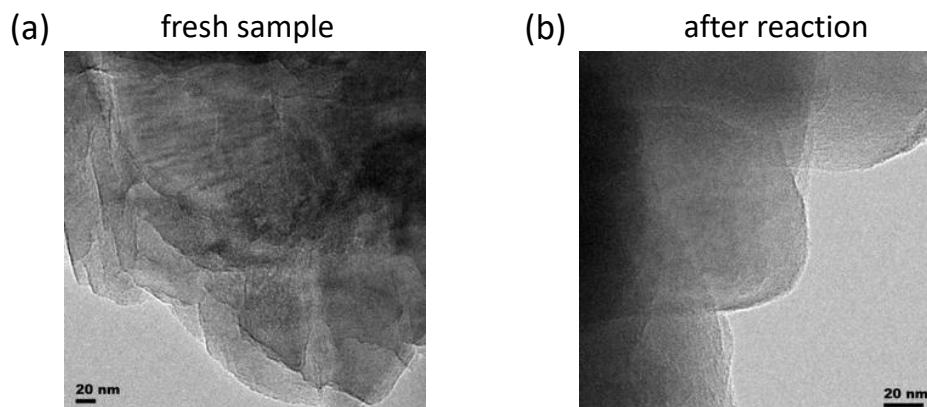
**Figure S8.** TG-DTA curves of the Cu-ERI(0.3).



**Figure S9.** Methanol yields of the Cu-ERI(0.3) at 300 °C in four cycles. (Note that only one methanol extraction was performed to calculate the methanol yield.)



**Figure S10.** XRD patterns of Cu-ERI(0.3) before activation and after reaction according to the conventional procedure with methane partial pressure of 30 bar.



**Figure S11.** TEM images of the Cu-ERI(0.3) zeolite before and after reaction. (a) Before activation in oxygen. (b) After the reaction according to the conventional procedure at a methane partial pressure of 30 bar.