Supporting information

Controlling the Distribution of Aluminum in a Cu-Zeolite Catalyst by Seed-Assisted Synthesis to Improve its NH$_3$-SCR Activity

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Table S1. The composition of NH_4-KFI obtained by SEM-EDX.

mol/mol %	KFI parent	KFI-SA1	KFI-SA2	KFI-SA3
Si	27.53	27.10	29.29	25.79
Al	5.64	5.66	6.06	5.20
K	0.19	0.35	0.34	0.29
Si/Al	4.88	4.79	4.83	4.96

Table S2. The composition of Cu-KFI obtained by ICP-AES.

	Cu-KFI parent	Cu-KFI -SA1	Cu-KFI-SA2	Cu-KFI-SA3
K (wt%)	0.24	0.33	0.27	0.30
Cu/Al (mol/mol)	0.20	0.20	0.20	0.20

Table S3. The Si/Al$_f$ ratio of NH$_4$-KFI, Cu-KFI and Cu-KFI HTA.

	Si/Al$_f$ ratio				
	Parent	SA1	SA2	SA3	Average
NH$_4$-KFI	4.38	4.57	4.42	4.28	4.41 (±0.10)
Cu-KFI	6.01	5.41	5.64	5.77	5.71 (±0.22)
Cu-KFI HTA	7.35	7.53	7.37	7.37	7.41 (±0.07)
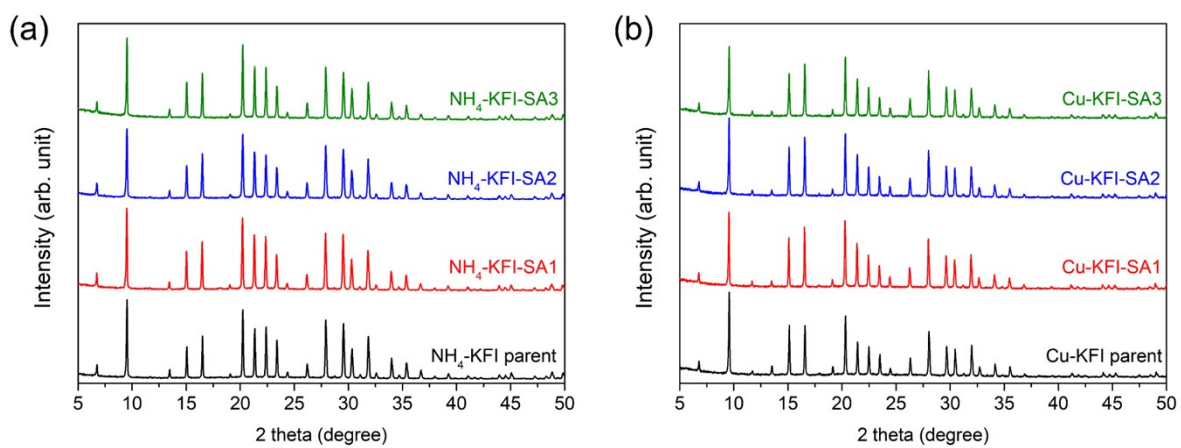
Fig. S1. The XRD profiles of (a) NH_4-KFI and (b) Cu-KFI.
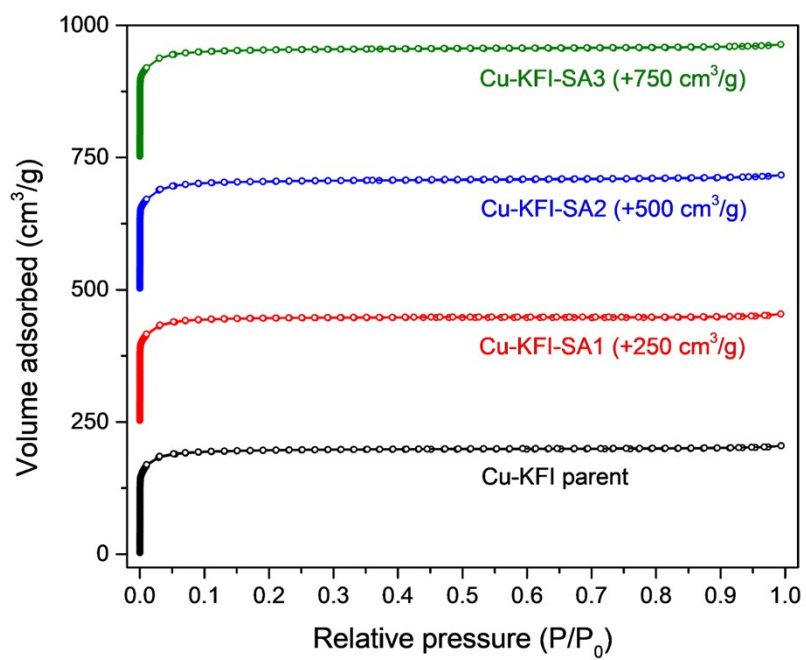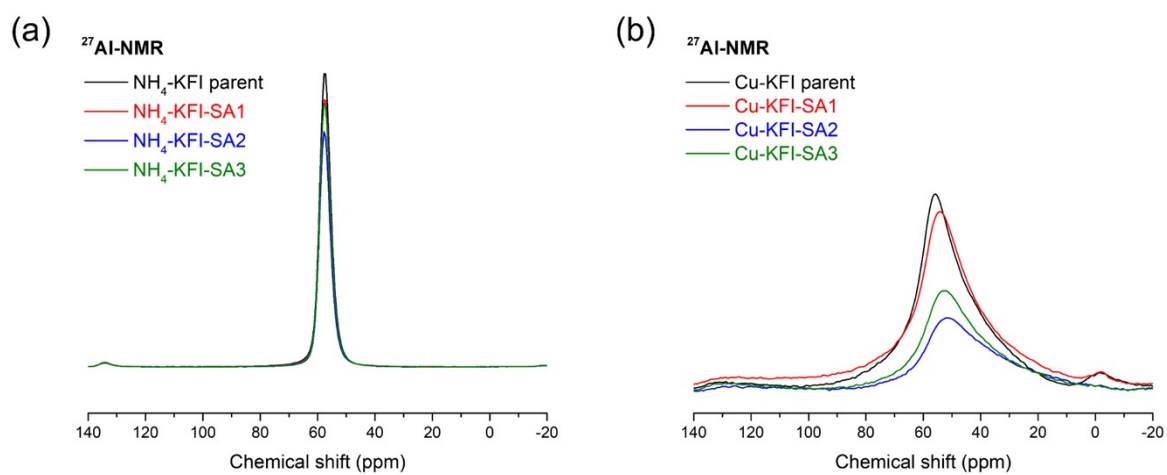
Fig. S2. The Ar-BET isotherms of the Cu-KFI.

Fig. S3. ^{27}Al-NMR spectra of the (a) NH$_4$-KFI and (b) Cu-KFI.
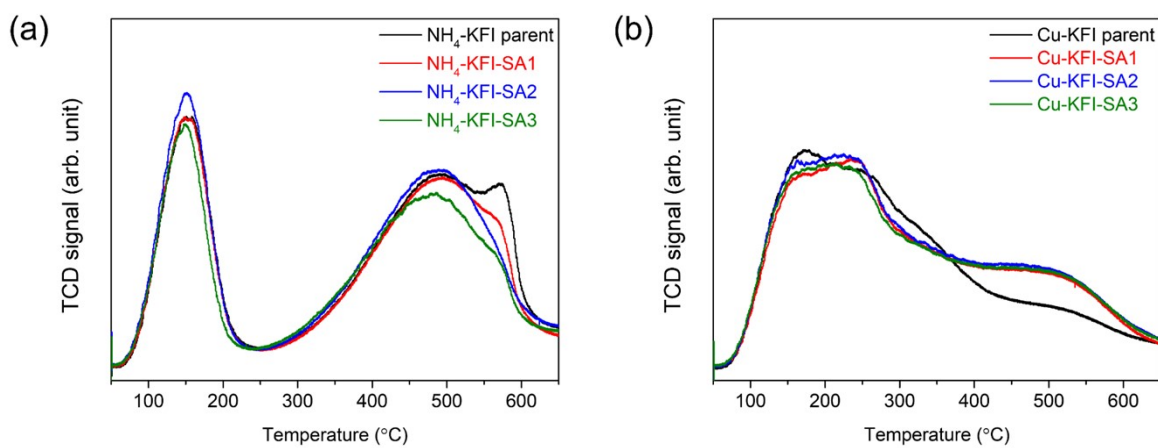
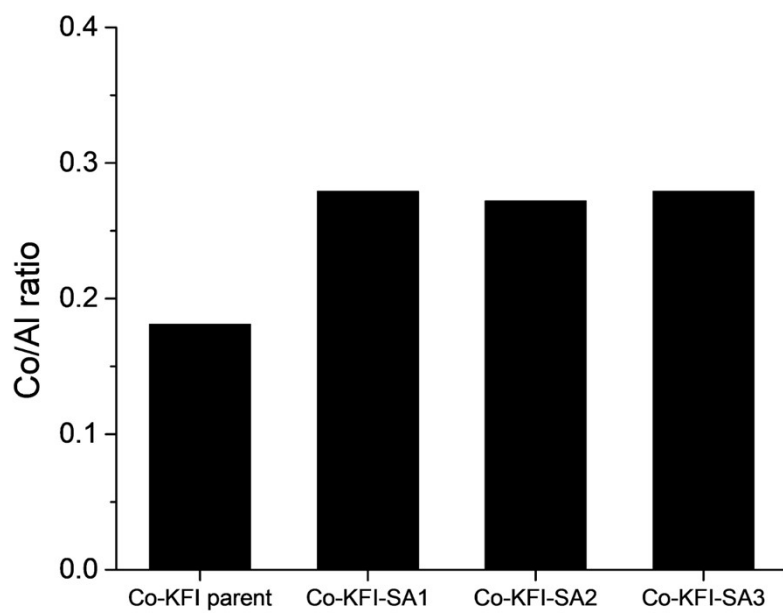**Fig. S4.** The NH$_3$-TPD profiles of the (a) NH$_4$-KFI and (b) Cu-KFI.

Fig. S5. The result of cobalt ions titration for measuring the amount of paired Al sites in the parent and seed added Cu-KFI.
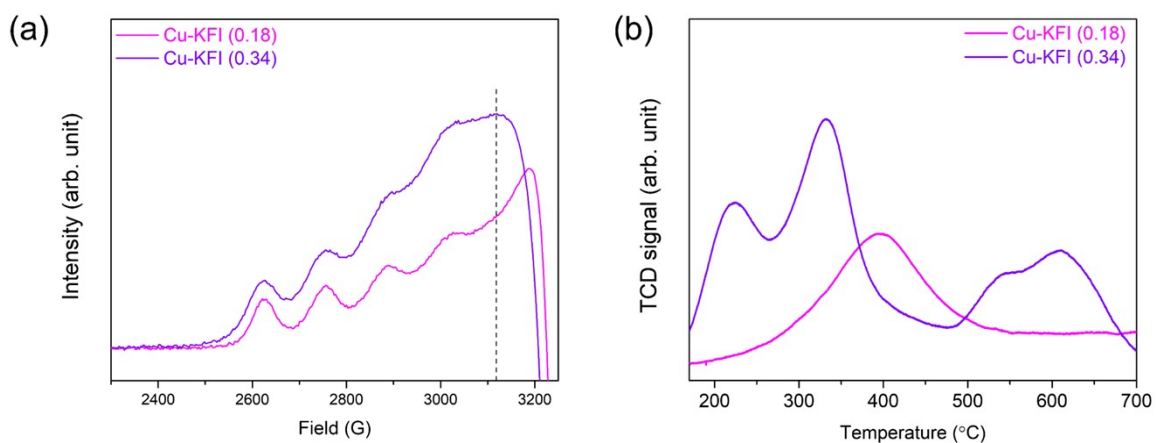
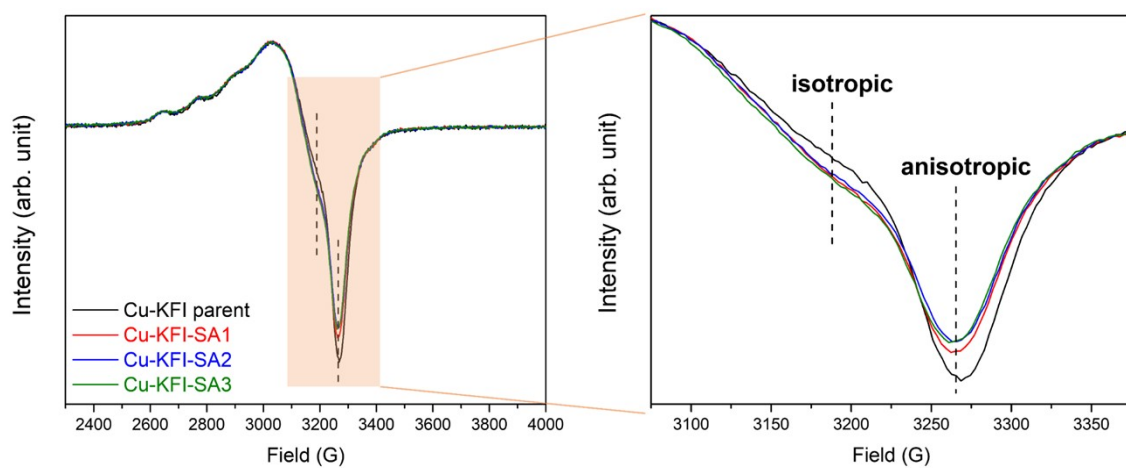**Fig. S6.** (a) The EPR spectra and (b) H_2-TPR profiles of Cu-KFI with different loadings.

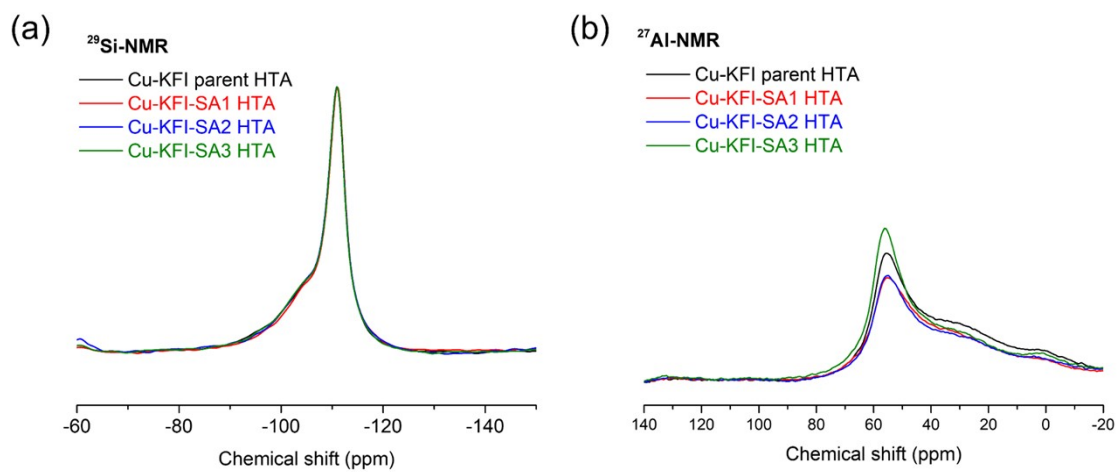**Fig. S7.** The EPR spectra of Cu-KFI at room temperature.

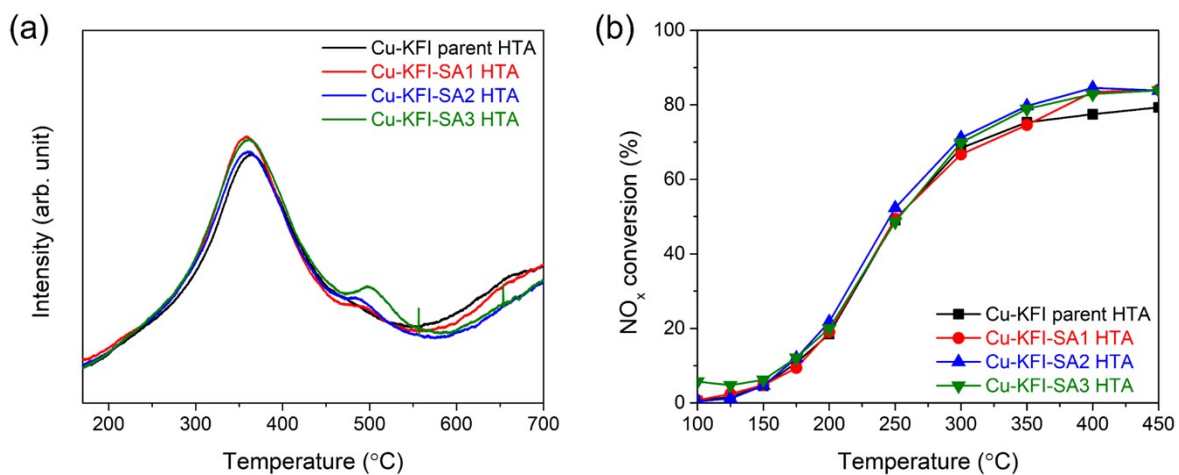**Fig. S8.** The ^{29}Si and ^{27}Al-NMR spectra of hydrothermally aged Cu-KFI catalysts.

Fig. S9. (a) The H$_2$-TPR and (b) SCR activity of hydrothermally aged Cu-KFI catalysts.
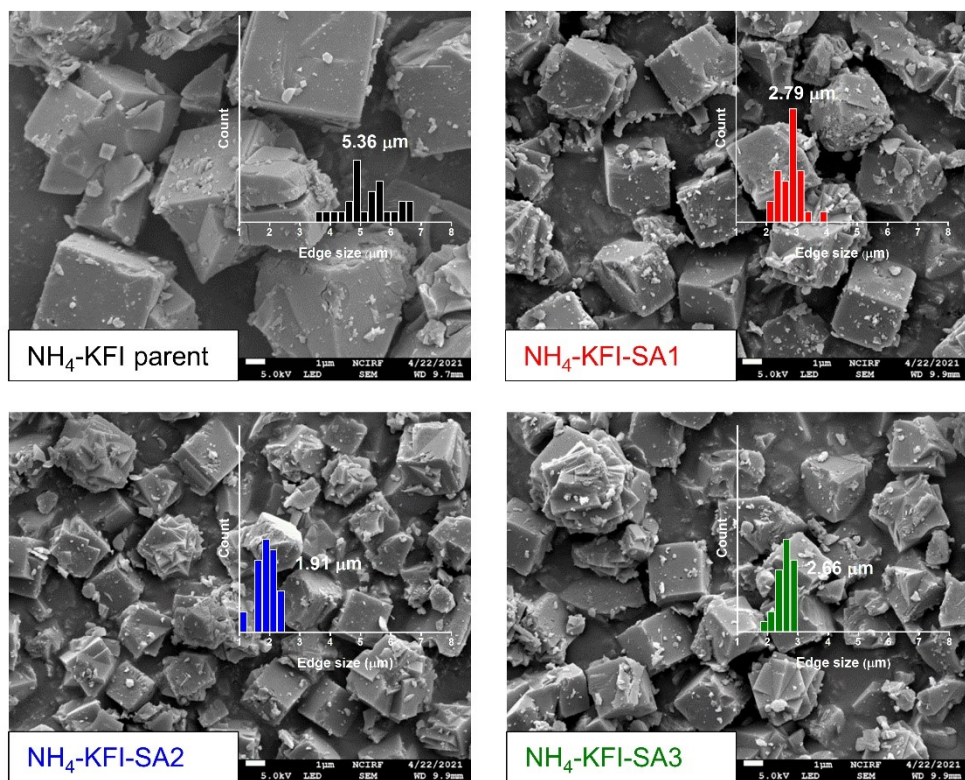
Fig. S10. The SEM image of the hydrothermally aged (a) Cu-KFI parent, (b) Cu-KFI-SA1 (c) SA2, and (d) SA3.
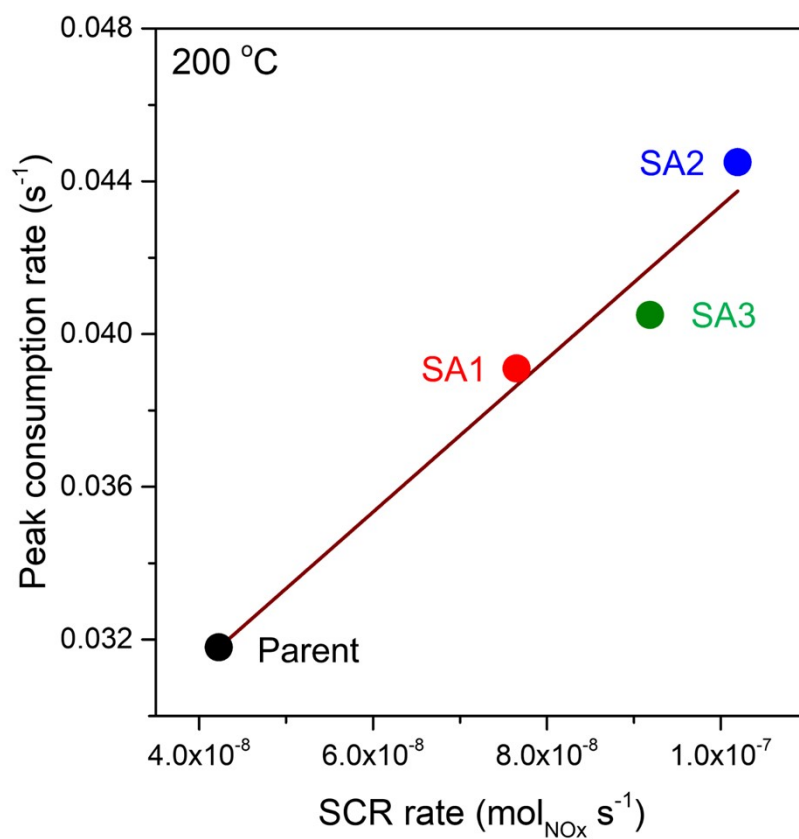
Fig. S11. The correlation between the SCR rates in the reaction and peak consumption rates of Lewis NH_3 during DRIFT analysis.
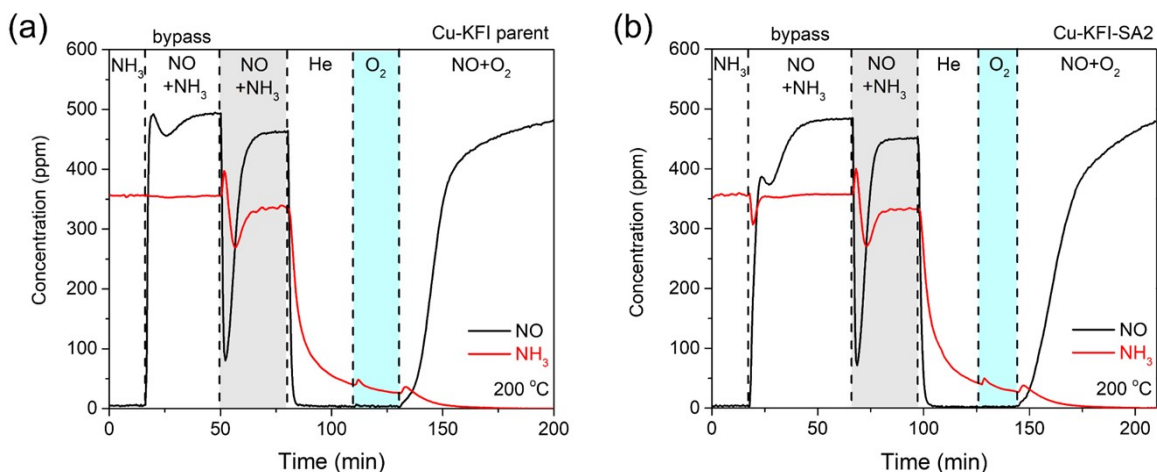
Fig. S12. The NO and NH$_3$ concentration profiles during the simulated SCR protocol at 200 °C. The (a) Cu-KFI parent and (b) Cu-KFI seed #2 catalysts were pretreated under 20% O$_2$/He for 30 min, and fully saturated by 350 ppm NH$_3$. After confirming the steady-state concentration at bypass, the 500 ppm NO and 350 ppm NH$_3$ were introduced into the catalysts to reduce the Cu ions (gray box). After purging with He, the reduced catalysts were oxidized with O$_2$ (sky blue box). Then, the NO was introduced to consume the all adsorbed NH$_3$.
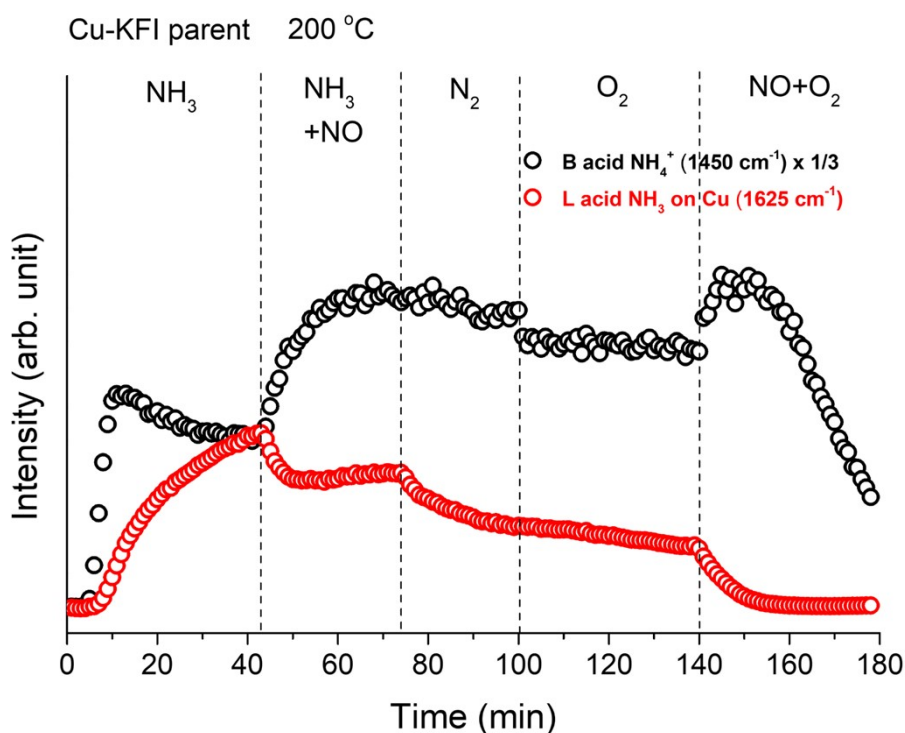
16

Fig. S13. The DRIFTS peak profiles of Brønsted acid NH_4^+ and Lewis acid NH_3 in the Cu-KFI parent during the simulated SCR reaction protocol.

In oxidation process (100-140 min), the Brønsted acid NH_4^+ slightly decreased at initial. The amount of consumed NH_4^+ was quantified combining the DRIFT and NH_3-TPD results in Figure S4. The amount of Brønsted acid sites of Cu-KFI parent is 1.34 mmol/g, which is obtained by deconvoluting the desorption profile in Figure S4. This would correspond to the intensity of B acid NH_4^+ at 40 min in the DRIFT profile. Therefore, we could estimate the decreased amount of B acid sites at 100 min by following equation.

$$\text{Consumed } NH_4^+ = \frac{\text{Decreased intensity at 100 min}}{\text{Intensity at 40 min}} \times amount\ of\ NH_4^+ \text{ on Cu-KFI parent in } NH_3\text{-TPD}$$
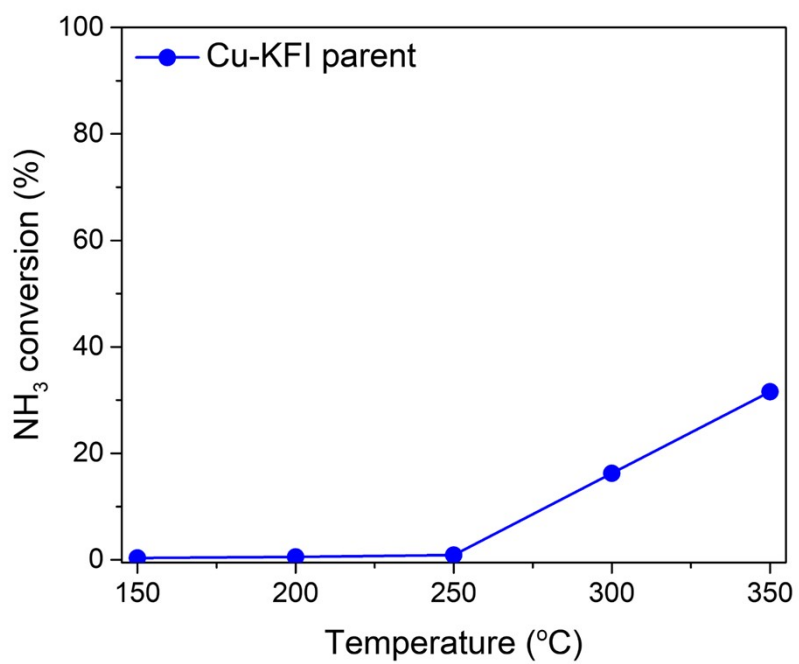
Fig. S14. NH_3 oxidation of Cu-KFI parent under 500 ppm NH_3, 10% O_2 and N_2 balance (GHSV = 240,000 h^{-1}).