Electronic Supplementary Material (ESI) for RSC Advances.

Supplenmtary Information

Effect of hydrothermal aging temperature on Cu-SSZ-13/H-SAPO-34 composite for the selective catalytic reduction of NOx by NH_3

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Table S1. Calculated areas of the area different divided peaks of the samples

Samples	243 °C [a]	333 °C [a]	396 °C [a]	Above 500 °C [a]
SSZ-SAPO	0.97	1.60		2.53
SSZ-SAPO-750	1.07	0.42	2.16	1.46
SSZ-SAPO-800	1.85		1.45	1.86
SSZ-SAPO-850	0.87		1.14	0.59

a. Obtained from Fig. 7. The peak was deconvoluted by Peakfit, and the values of area only represent the intensity.

The peaks at 243 and 333 °C represent the reduction of $Cu(OH)^+$ (also Cu^{2+}-new) and Cu^{2+}-2Al, respectively. The peak above 500 °C can be attributed to the reduction of Cu^+ to Cu^0. The details of the of the assignment of each reduction peak can be found in the main text (Section 3.3.3). The area values of the peak at 243 and 333 °C, representing the Cu^{2+} ions content in each sample, decreased after aging at 750 °C, and increased in the sample aged at 800 °C, and then decreased after aging at 850 °C, consistent with the trend of Cu^{2+} ions content changes in EPR results in Fig. 5.

The peak area of Cu^{2+} to Cu^+ in SSZ-SAPO-850 is higher than that of Cu^+ to Cu^0. This can be explained by that some highly stable Cu^+ ions are reduced at temperatures higher than 900 °C [1].

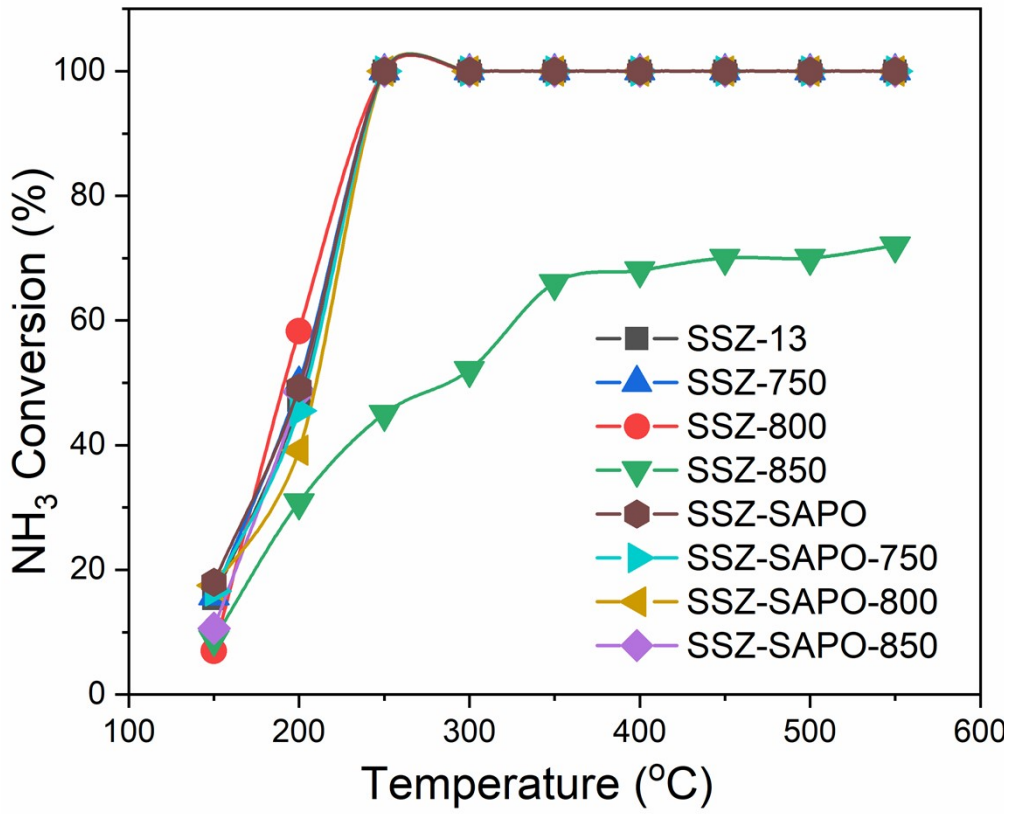

Fig. S1. NH_3 conversion as a function of temperature in NH_3-SCR reaction.

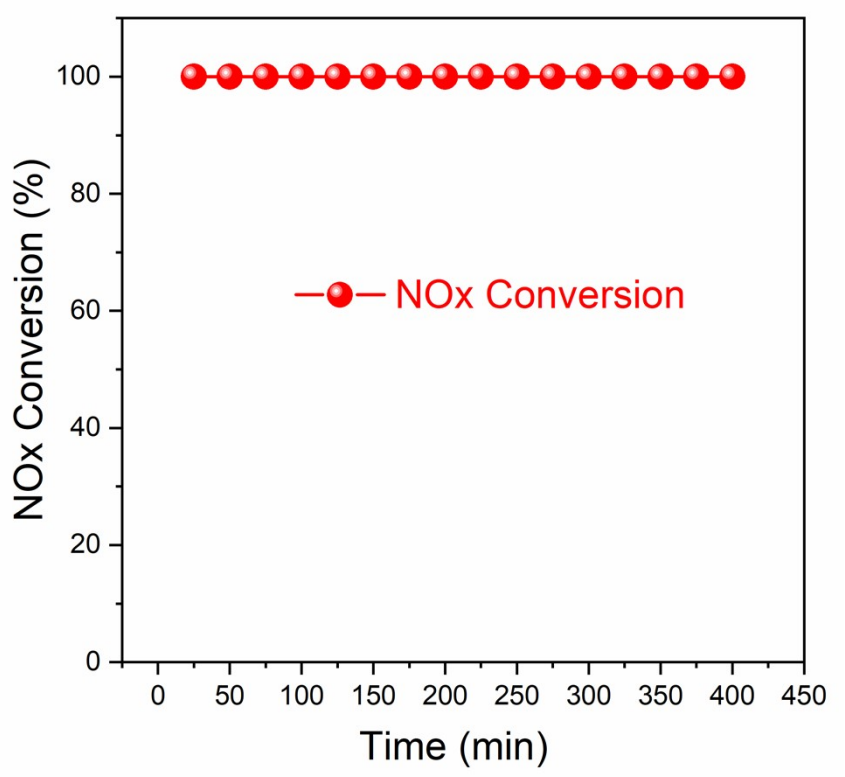

Fig. S2. Long-time NOx conversion of Cu-SSZ-13 at 250 °C.

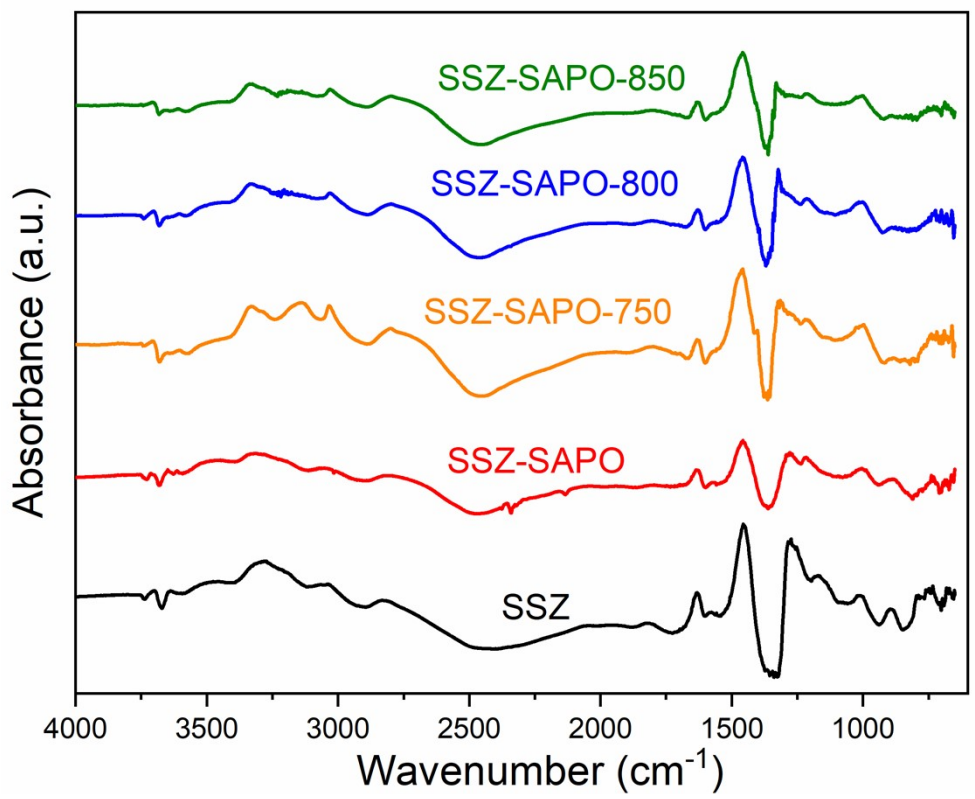

Fig. S3. DRIFTs spectra after NH_3 adsorption on the samples in 4000-500 cm^{-1} region. Conditions: 500 ppm NH_3/N_2 at 25 °C, then N_2 purge before spectra collection.

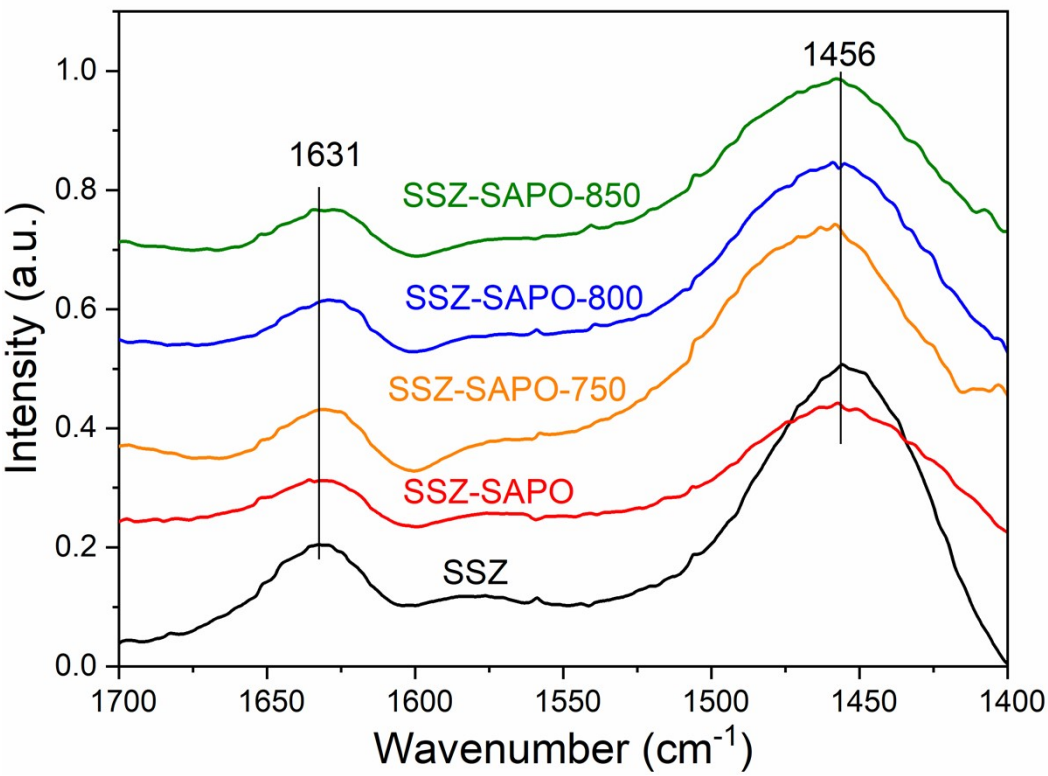

Fig. S4. DRIFTs spectra after NH_3 adsorption on the samples in 1700-1400 cm^{-1} region. Condition: 500 ppm NH_3/N_2 at 25 °C, then N_2 purge before spectra collection.

The bands at 1632 and 1475 cm^{-1} are attributed to molecular NH_3 absorbed onto exchanged Cu-related Lewis acid sites and NH_4^+ interacting with Brønsted acid sites (Si-OH-Al), respectively [2]. The aged samples exhibited intense NH_3 absorption peaks on Lewis acid and Brønsted acid sites, indicating the well-maintained Cu^{2+} ions and structure, consistent with the EPR (Fig. 5) and XRD (Fig. 2) results.

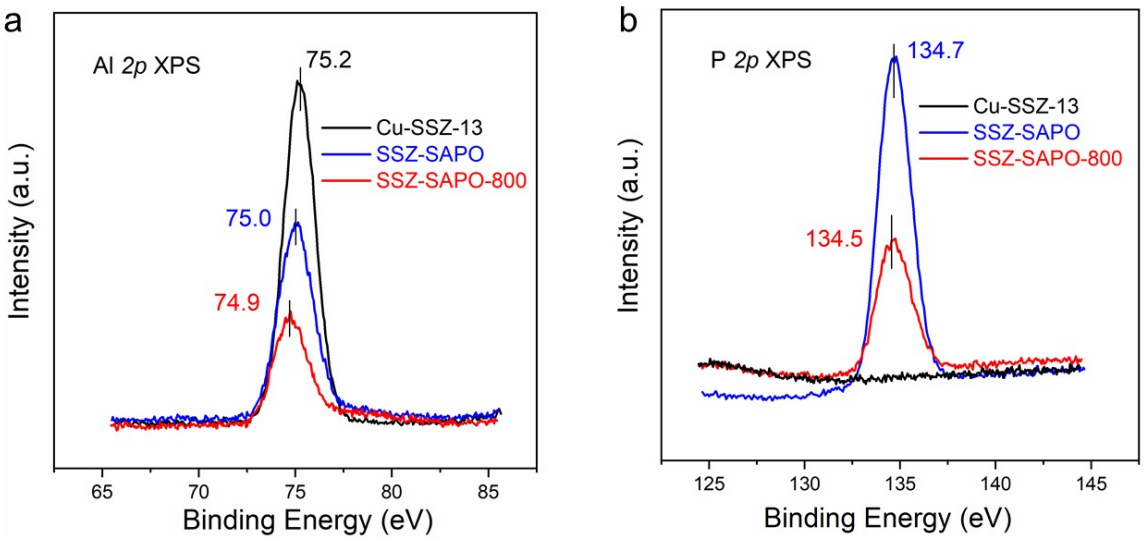

Fig. S5. Al 2p and P 2p XPS spectra of samples

The Cu-SSZ-13 and SSZ-SAPO samples showed the Al 2p XPS peaks at 75.2 and 75.0 eV, respectively. Clearly, the addition of H-SAPO-34 in the composite shifted the Al 2p XPS peak to lower angle, which is probably attributed to the presence of P-Al bond. The SSZ-SAPO-800 sample presented the Al 2p XPS peak at 74.9 eV, lower than that in SSZ-SAPO. This indicates that the Al in SSZ-13 might interact with P upon aging at 800 °C. The P 2p XPS results (Fig. S5b) support this hypothesis, which displayed that the XPS peak in SSZ-SAPO shifted from 134.7 to 13.5 eV after aging at 800 °C.

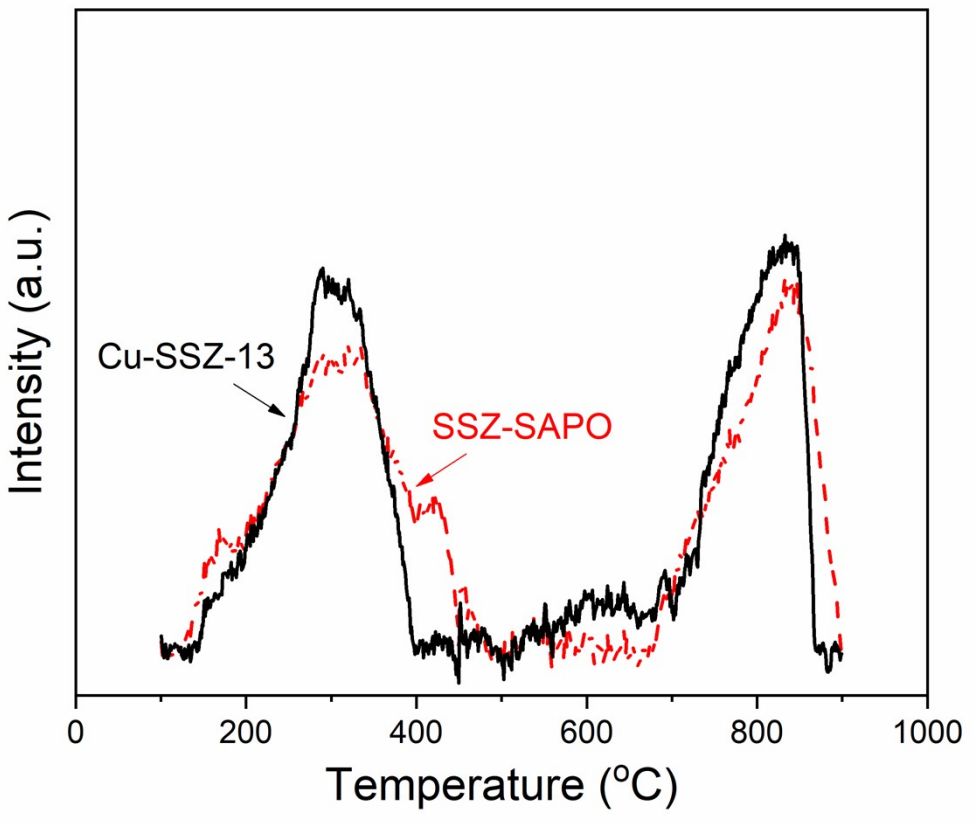

Fig. S6. H_2-TPR profiles of Cu-SSZ-13 and SSZ-SAPO samples. The curve of SSZ-SAPO was copied from Fig. 7.

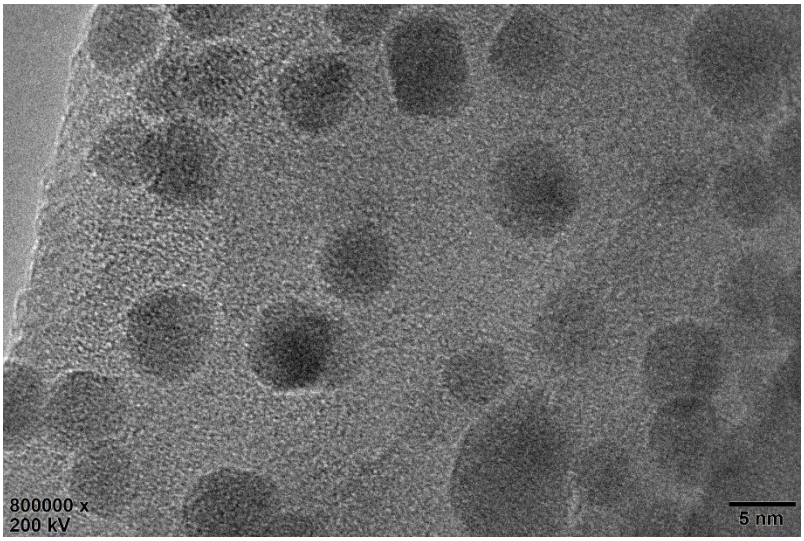

Fig. S7. The TEM image of SSZ-SAPO-750.

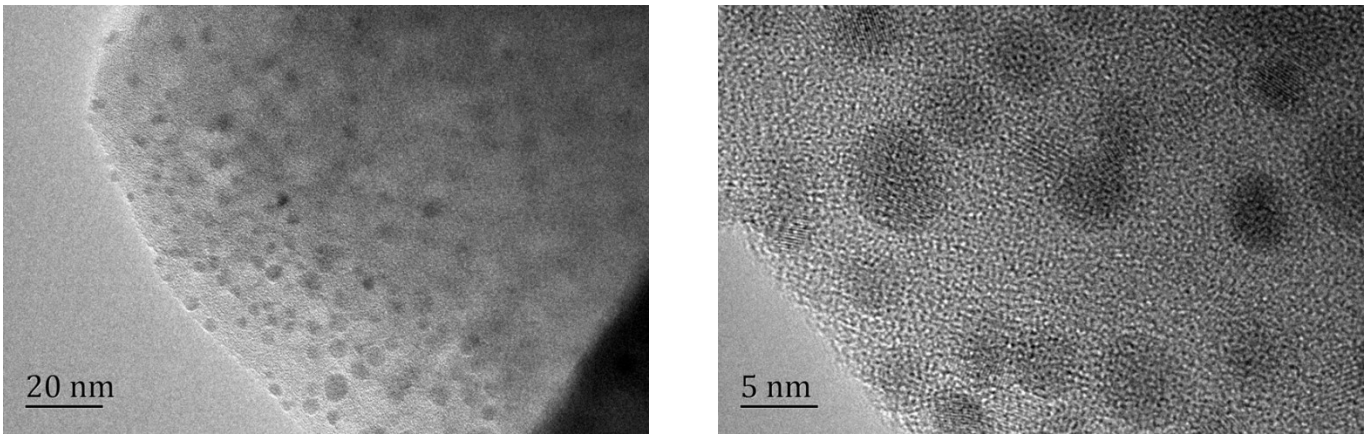

Fig. S8. The TEM image of SSZ-SAPO-850.

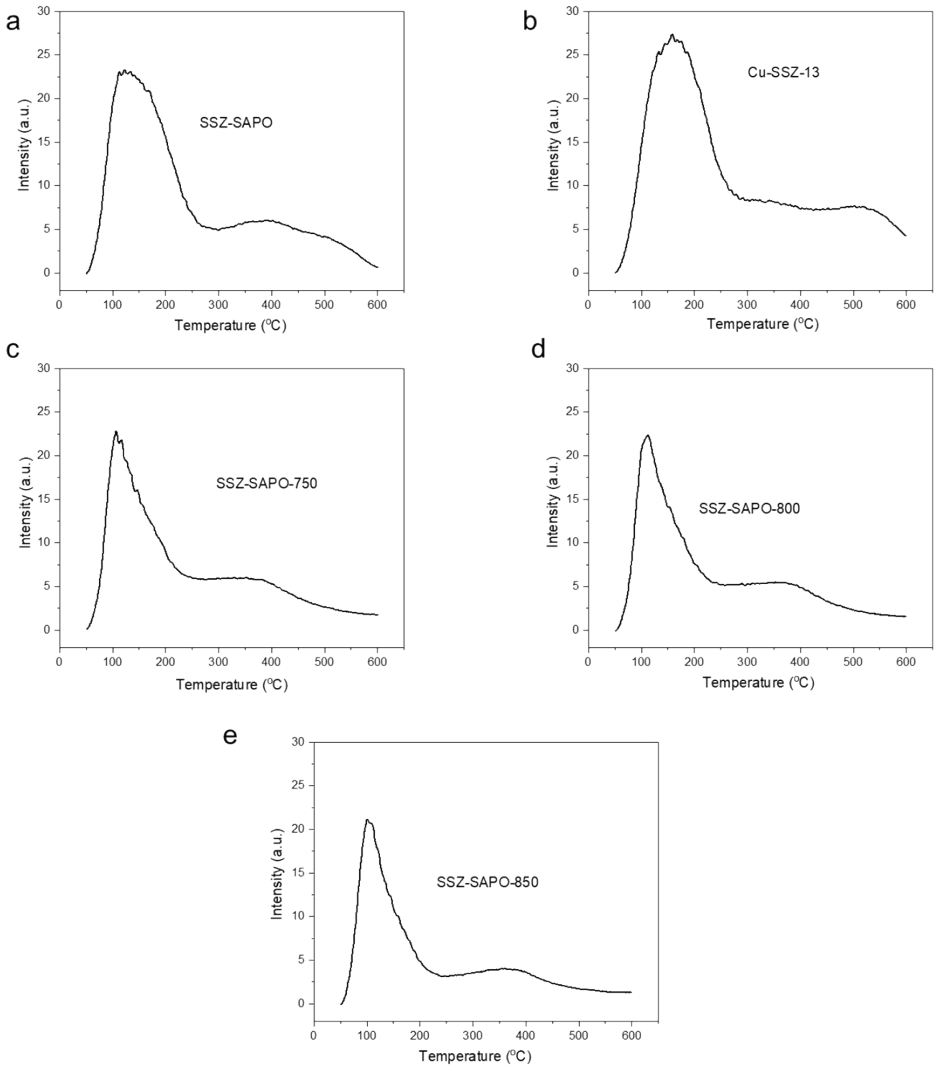

Fig. S9. NH_3-TPD results of the samples.

After HTA, the NH_3 desorption peak in 400-500 °C decreased in the composite samples, indicating the decrease of Brønsted sites.

References

[1] L. Xie, F. Liu, X. Shi, F.-S. Xiao, H. He, Appl. Catal. B: Environ. 179 (2015) 206-212.

[2] Y. Shan, J. Du, Y. Yu, W. Shan, X. Shi, H. He, Appl. Catal. B: Environ. 266 (2020) 118655.