Supplementary Information

Modeling and analysis of ammonia oxidation and nitrous oxide formation on dual-layer ammonia slip catalyst for diesel after-treatment

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The developed low-dimensional kinetic model was compared with the full 1+1D kinetic model from reference to confirm the validity of the approximation of the internal Sherwood number.[1] The optimized kinetic parameters from the developed low-dimensional kinetic model were used in the full 1+1D kinetic model without further optimization. As shown in Fig. S1-S3, the simulated results indicated that the developed low-dimensional kinetic model has good prediction in conversion and product selectivity as well as the full 1+1D kinetic model. The biggest prediction errors of the developed low-dimensional kinetic model for Cu/SSZ-13 catalyst under Standard SCR conditions, Pt/Al$_2$O$_3$ and dual-layer ASC catalysts under ammonia oxidation conditions were 6.2%, 6.4% and 4.7%, respectively. While the biggest prediction errors of the full 1+1D kinetic model were 6.0%, 8.3% and 5.0%, respectively. The quantitative comparison results of model prediction error confirmed the validity of approximation.
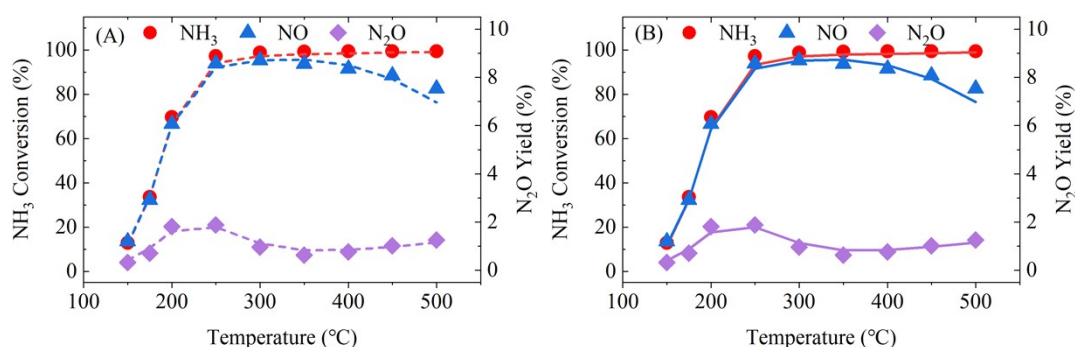

Fig. S1. (A) The developed low-dimensional kinetic model and (B) full 1+1D kinetic model simulated NH$_3$, NO conversions and N$_2$O yields under Standard SCR conditions over Cu/SSZ-13 catalyst at GHSV 60,000/hour. Feed: 300 ppm NH$_3$, 300 ppm NO, 10% O$_2$, 5% H$_2$O, balance N$_2$. (symbols: experiments; dash lines: developed low-dimensional kinetic model simulation; lines: full 1+1D kinetic model simulation)
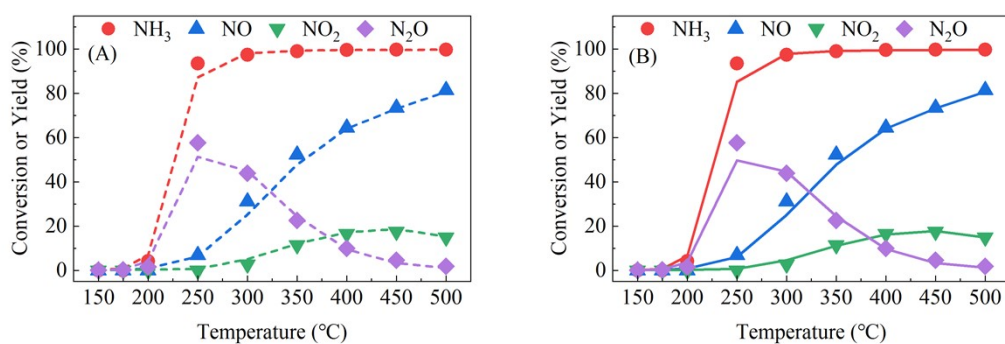

Fig. S2. (A) The developed low-dimensional kinetic model and (B) full 1+1D kinetic model simulated NH$_3$ conversions and product yields under NH$_3$ oxidation conditions over Pt/Al$_2$O$_3$ catalyst at GHSV 90,000/hour. Feed: 300 ppm NH$_3$, 10%O$_2$, 5%H$_2$O, balance N$_2$. (symbols: experiments; dash lines: developed low-dimensional kinetic model simulation; lines: full 1+1D kinetic model simulation)
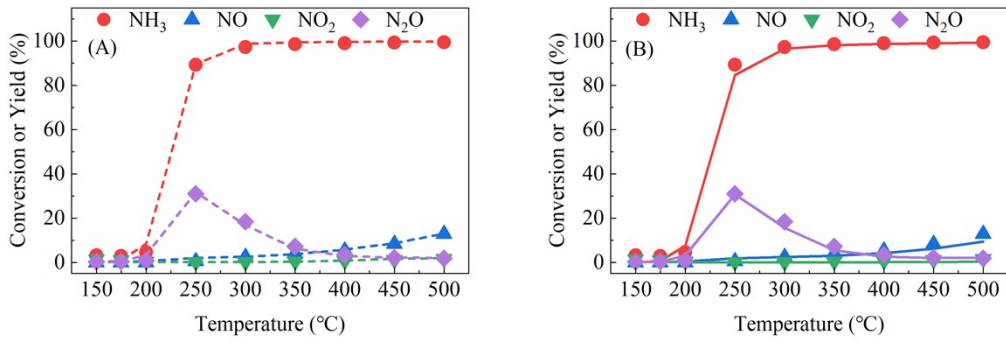
Fig. S3. (A) The developed low-dimensional kinetic model and (B) full 1+1D kinetic model simulated NH_3 conversions and product yields under NH_3 oxidation conditions over dual-layer ASC catalyst at GHSV 90,000/hour. Feed: 300 ppm NH_3, 10%O_2, 5%H_2O, balance N_2. (symbols: experiments; dash lines: developed low-dimensional kinetic model simulation; lines: full 1+1D kinetic model simulation)
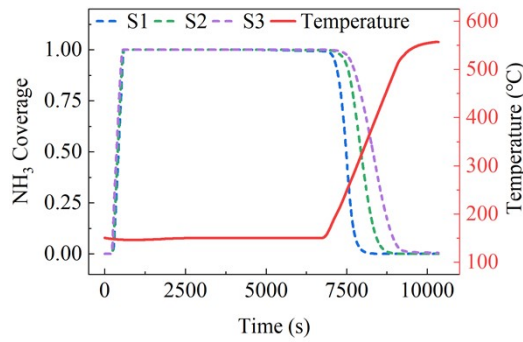

Fig. S4. Simulated NH_3 coverages during NH_3-TPD experiment over Cu/SSZ-13 catalyst at GHSV 60,000/hour. Feed: 300 ppm NH_3, 5% H_2O, balance N_2 during adsorption; 5% H_2O, balance N_2 during purge process and temperature programmed desorption process.
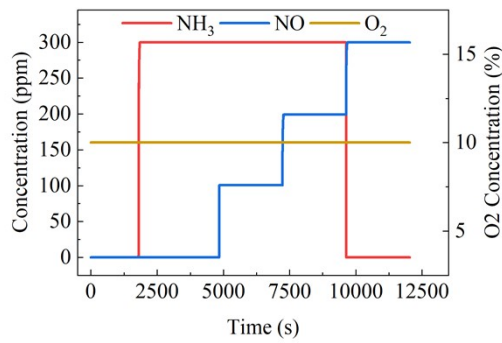

Fig. S5 Isothermal transient experiment inlet gas mole fraction concentrations at 250°C with N_2 as balance gas.
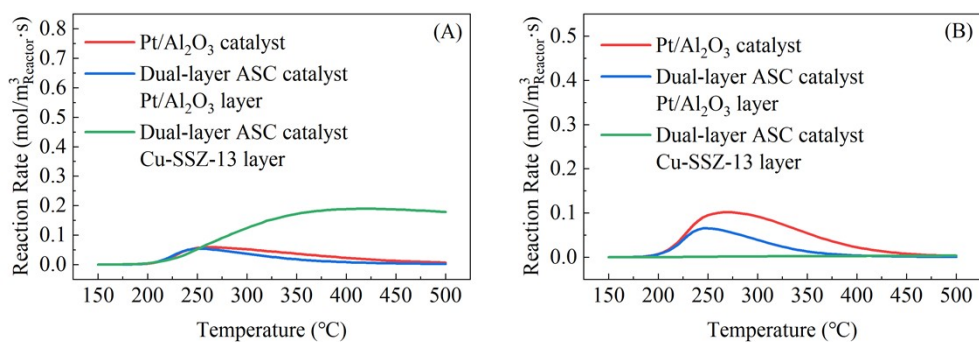
Fig. S6 Model predicted (A) N$_2$ and (B) N$_2$O formation rates under NH$_3$ oxidation conditions at GHSV 90,000/hour. Feed: 300 ppm NH$_3$, 10% O$_2$, 5% H$_2$O, balance N$_2$.
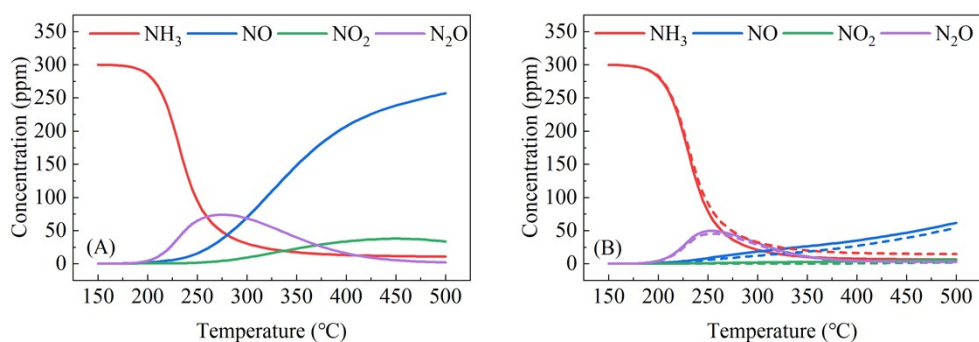

Fig. S7 Model predicted species concentrations within washcoat over (A) Pt/Al$_2$O$_3$ catalyst and (B) dual-layer ASC catalyst under NH$_3$ oxidation conditions at GHSV 90,000/hour. Feed: 300 ppm NH$_3$, 10% O$_2$, 5% H$_2$O, balance N$_2$. (lines: Pt/Al$_2$O$_3$ layer; dash lines: Cu/SSZ-13 layer)

Reference

1   P. S. Dhillon, M. P. Harold, D. Wang, A. Kumar and S. Y. Joshi, *React. Chem. Eng.*, 2019, **4**, 1103–1115.