Experimental Section

Before the experiment, 1.5 g H-ZSM-5 (Si/Al = 22) (20-40 mesh) was pretreated at 623 K for 60 min in N₂ flow, and then cooled down to room temperature. The whole experiment included two stages. At the adsorption stage (plasma off), a flowing mixed gas containing 1000 ppm NO, 5% O₂, 2% H₂O (when used) and balanced N₂ was introduced into the catalyst-packed DBD reactor at a total flow rate of 120 mL/min. This stage lasted for 0.5-2.0 h at room temperature and NO was adsorbed completely on H-ZSM-5. When 2% H₂O was introduced into the system, the adsorption stage lasted for 0.5 h and NO was still adsorbed completely on H-ZSM-5 (Fig 6S). 0.16 mmol NO was adsorbed on 1.5 g H-ZSM-5. At the discharge stage (plasma on), the gas stream containing oxygen-deficient air (5% O₂+95% N₂) with or without NH₃ was switched into the DBD reactor. This discharge process lasted for 15 minutes at room temperature. The definitions of the conversion of NO_x to N₂ as follows:

$$Conv._{NOx} \rightarrow _{N2}(\%) = \frac{n_{NOx}^{absorbed} - n_{NOx}^{emitted} - 2 \times n_{N_2O}^{produced} - n_{NOx}^{residual}}{n_{NOx}^{absorbed}} \times 100\%$$

Here, $n_{NOx}^{absorbed}$ is the amount of NOx adsorbed on H-ZSM-5 at the adsorption stage; $n_{NOx}^{emitted}$ is the amount of NOx (NO, NO₂) emitted to the exhaust during the plasma stage; $n_{N_2O}^{produced}$ is the

amounts of N₂O produced during the plasma stage; $n_{NOx}^{residual}$ is the amount of NOx species remained on the catalyst after the plasma stage, which was determined by a temperature-programmed desorption (TPD) process. The concentrations of NO_x (NO and NO₂) were analyzed online by a chemiluminescent NO/NO₂/NO_x analyzer (Eco Physics). N₂O was analyzed using a Fuli-9750 gas chromatograph (GC) with TCD and a Porapak Q column.



Fig. 1S Effect of discharge power on the selectivity to N_2 and N_2O in NO_x storage and decomposition process at different discharge power. Conditions : at adsorption stage, 1.5 g H-ZSM-5 adsorb 1000 ppm NO for 0.5 h. At discharge stage, 30 mL/min flow rate of oxygen-deficient air (5% O_2 +95% N_2).



Fig. 2S Effect of NH_3/O_2 molar ratio on (a) the conversion of adsorbed NO_x to N_2 , and (b) the residual NO_x on H-ZSM-5 in NO_x storage and reduction with NH_3 process. Conditions : at adsorption stage, 1.5 g H-ZSM-5 adsorb 1000 ppm NO for 0.5 h. At discharge stage, 30 mL/min flow rate of oxygen-deficient air (5% O_2 +95% N_2) with various NH_3/O_2 molar ratios, discharge power is fixed at 7.8 W.



Fig. 3S Cyclic operation of the present adsorption–discharge process for NO_x removal: percentage of NO_x abatement in the adsorption stage (black) and conversion of the adsorbed NO_x to N₂ in the discharge stage (slash). Conditions : at adsorption stage, 1.5 g H-ZSM-5 adsorb 1000 ppm NO, 5% O₂ and 2% H₂O for 0.5 h. At discharge stage, 30 mL/min flow rate of oxygen-deficient air (5% O₂+95%N₂) and 2.5% NH₃. Between the intermission of two cycles, H-ZSM-5 should be treated in N₂ at 350 °C for 30 min.



Fig. 4S XRD patterns of H-ZSM-5 before and after cyclic operation



Fig. 5S. SEM patterns of H-ZSM-5 (A,B)before and (C,D)after cyclic operation



Fig. 6S NO_x adsorption curve on H-ZSM-5. Conditions: 1g H-ZSM-5 adsorb 1000ppm NO, 5% O₂, 2% H₂O (when used), 13% CO₂ (when used) and balanced N₂ at 30 °C.

Fig. 6S. displays the adsorption curve of NO in the presence of 13% CO₂ and 2% H₂O on H-ZSM-5 at 30 °C. Firstly, 2% H₂O was introduced into the system and the NO_x breakthrough time of H-ZSM-5 decreased from 112 min to 21 min. Furthermore, both 13% CO₂ and 2% H₂O was introduced into the system and the NO_x breakthrough time of H-ZSM-5 merely decreased from 21 min to 18 min (breakthrough adsorption capacity from 0.11 mmol·g⁻¹ to 0.10 mmol·g⁻¹). The result indicated that the effect of H₂O was primary factor for the adsorption stage.

When 2% H_2O was introduced into the system at the adsorption stage, it lasted for 0.5 h and 0.16 mmol NO was adsorbed completely on 1.5 g H-ZSM-5.