

Supporting Information

**Molecular Insight into the Electric Field Regulation of N₂ and CH₄ Adsorption in the
Trapdoor ZSM-25 Zeolites**

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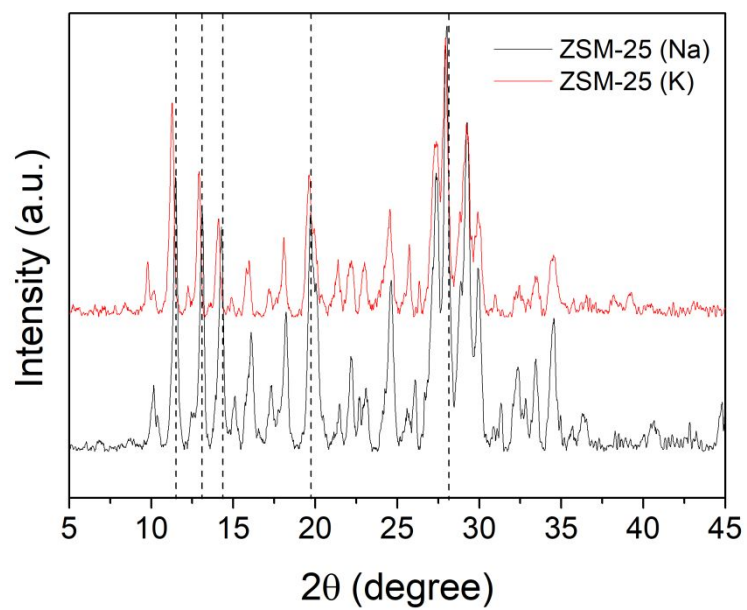


Figure S1 The PXRD patterns of the synthesized ZSM-25 (Na) and ZSM-25 (K).

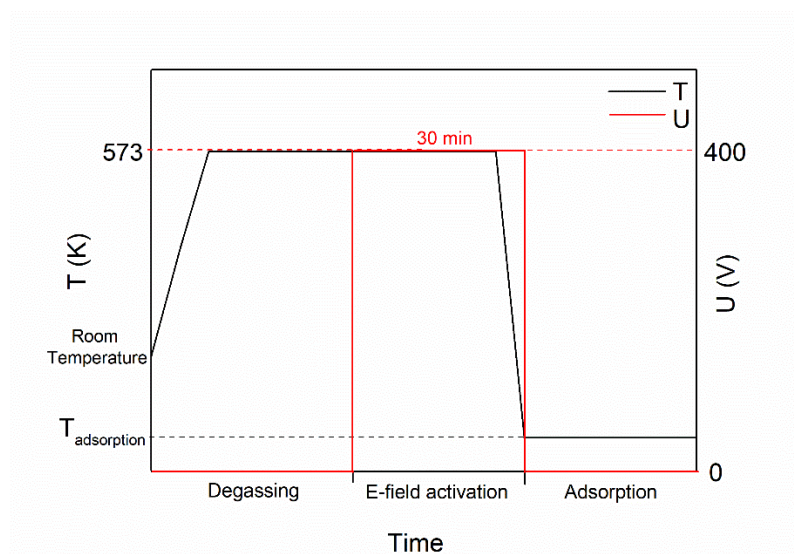


Figure S2 Schematic diagram of the E-field activation process of ZSM-25 before adsorption.

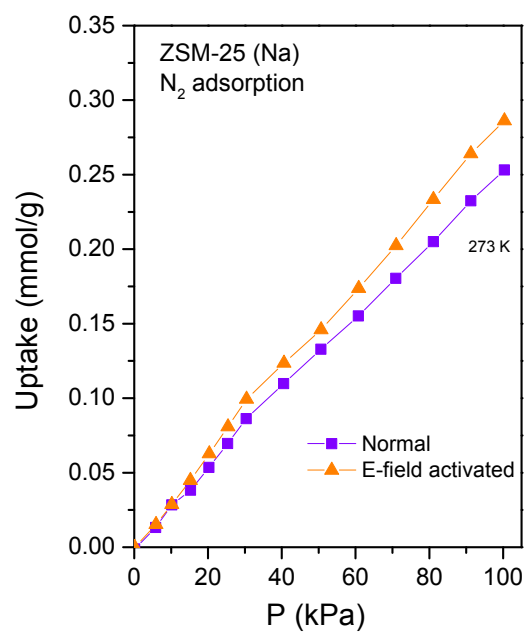


Figure S3 Adsorption isotherms of N_2 in ZSM-25 (Na) with or without E-field activation at 273 K.

The E-field intensity was 800 V/mm. Lines are guides for the eye.

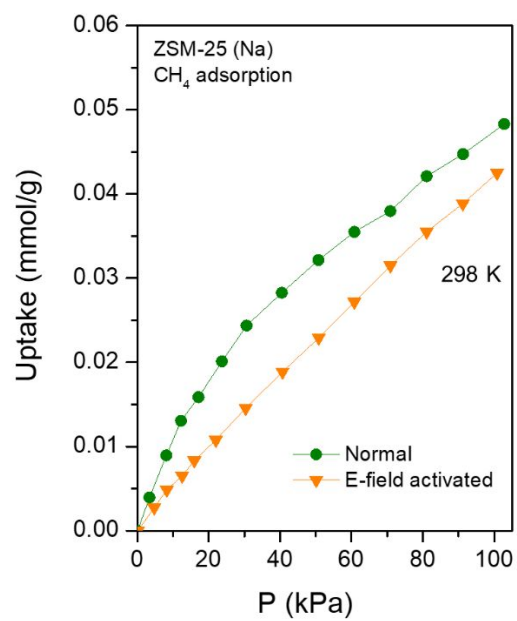


Figure S4 Adsorption isotherms of CH₄ in ZSM-25 (Na) with or without E-field activation at 298 K.

The E-field intensity was 800 V/mm. Lines are guides for the eye.

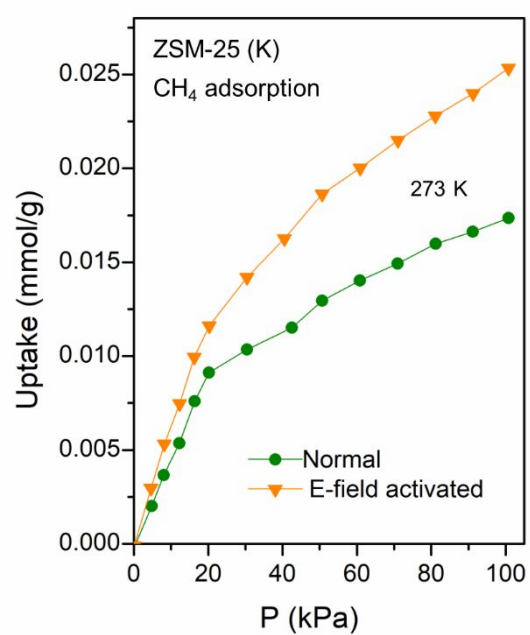


Figure S5 Adsorption isotherms of CH₄ in ZSM-25 (K) with or without E-field activation at 273 K.

The E-field intensity was 800 V/mm. Lines are guides for the eye.

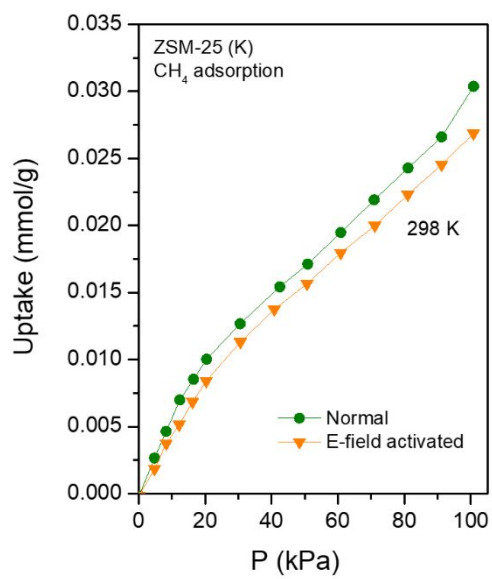


Figure S6 Adsorption isotherms of CH₄ in ZSM-25 (K) with or without E-field activation at 298 K.

The E-field intensity was 800 V/mm. Lines are guides for the eye.

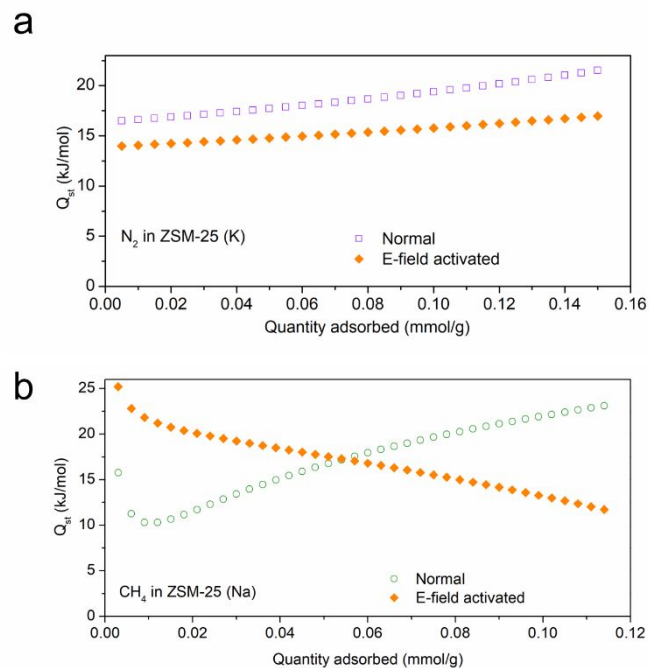


Figure S7 The isosteric heat of adsorption Q_{st} of (a) N_2 in ZSM-25 (K) and (b) CH_4 in ZSM-25 (Na) before and after the E-field activation. The Q_{st} of CH_4 in ZSM-25 (K) and N_2 in ZSM-25 (Na) can not be calculated by the Clausius–Clapeyron equation due to the trapdoor adsorption at the temperature range of 250–300 K.

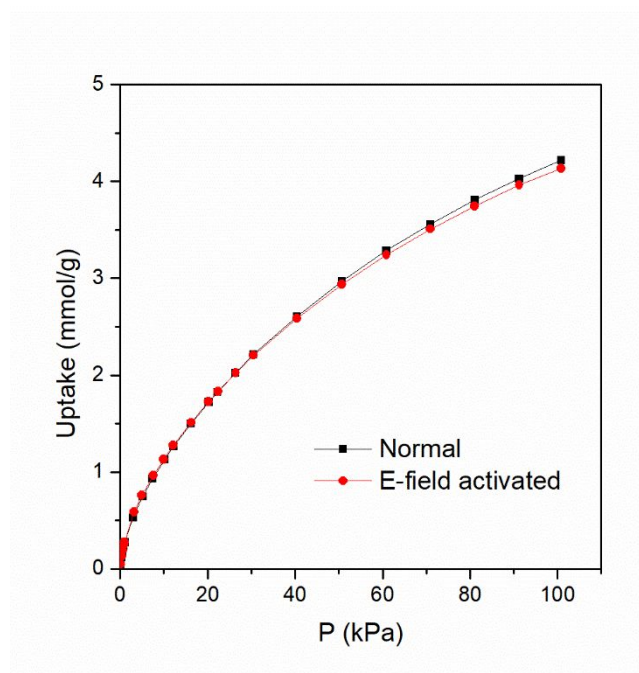


Figure S8 Adsorption isotherms of CO₂ in HY with or without E-field activation at 273 K. HY zeolite does not possess trapdoor cations. The E-field intensity was 800V/mm. Lines are guides for the eye.

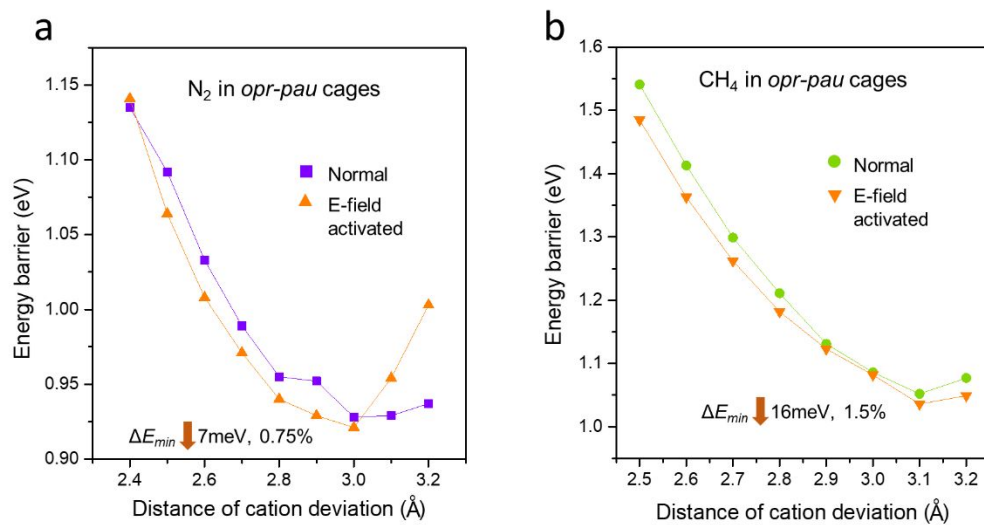


Figure S9 The energy profile for (a) N₂ and (b) CH₄ molecules passing through the 8MR of *opr-pau* cage pair in ZSM-25 (K).

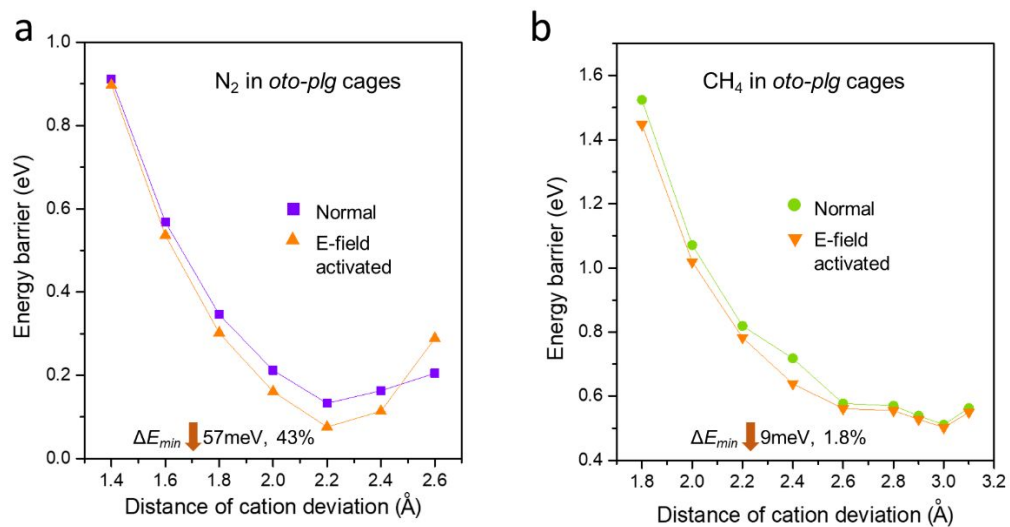


Figure S10 The energy profile for (a) N_2 and (b) CH_4 molecules passing through the 8MR of *oto-plg* cage pair in ZSM-25 (K).

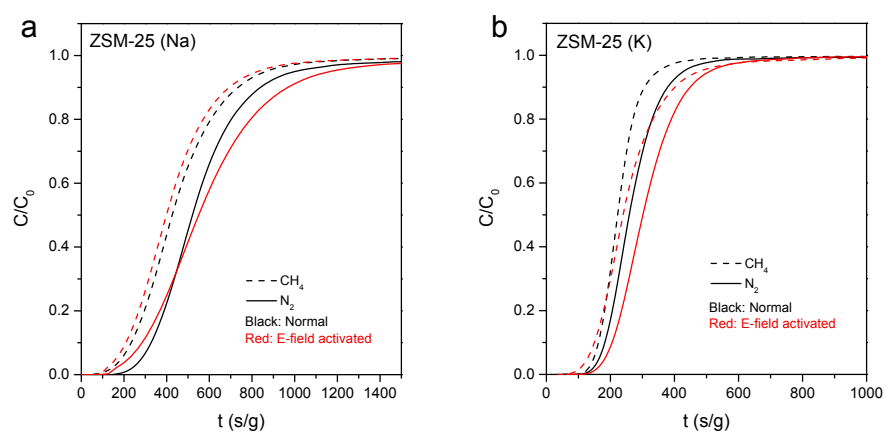


Figure S11 Breakthrough adsorption of N_2/CH_4 gas mixture (50:50, v/v) by the (a) ZSM-25 (Na) and (b) ZSM-25 (K) with or without E-field activation.

Supplementary Note

The isosteric heat of adsorption (Q_{st}) was calculated by the Clapeyron equation in Equation (S1).

$$\frac{Q_{st}}{RT^2} = \left[\frac{\partial \ln P}{\partial T} \right]_q \quad (S1)$$

where Q_{st} (kJ/mol) is the isosteric heat of adsorption at the loading q (mol/kg), P is the pressure (kPa), T is the system temperature (K), and R is the gas constant (kJ/kg·K). Equation (S1) can be integrated to get Equation (S2),

$$\ln P = \frac{Q_{st}}{RT} + C \quad (S2)$$

where C is an integration constant. At a certain coverage, if the isosteric heat is assumed to be independent of temperature, the Q_{st} can be calculated by the slopes of linear equations of $\ln(P)$ versus $1/T$ in Equation (S2). $\ln(P)$ was derived from the mathematical spline fitting of isotherms at different temperatures.

The single-component selectivity of N_2/CH_4 was calculated by Equation S3.

$$N_2/CH_4 \text{ selectivity} = \frac{x_{N_2}/y_{N_2}}{x_{CH_4}/y_{CH_4}} \quad (S3)$$

where x and y were the adsorbed-phase concentration and the corresponding gas-phase concentration, respectively.