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Supplementary Information

Unraveling Separation Mechanisms of LTA Zeolite Depending on Regulated Particle Size and Pore Structure for Efficient Ethylene/Ethane Separation

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1 XRD patterns of LTA zeolites via different particle sizes and Ca exchange rate

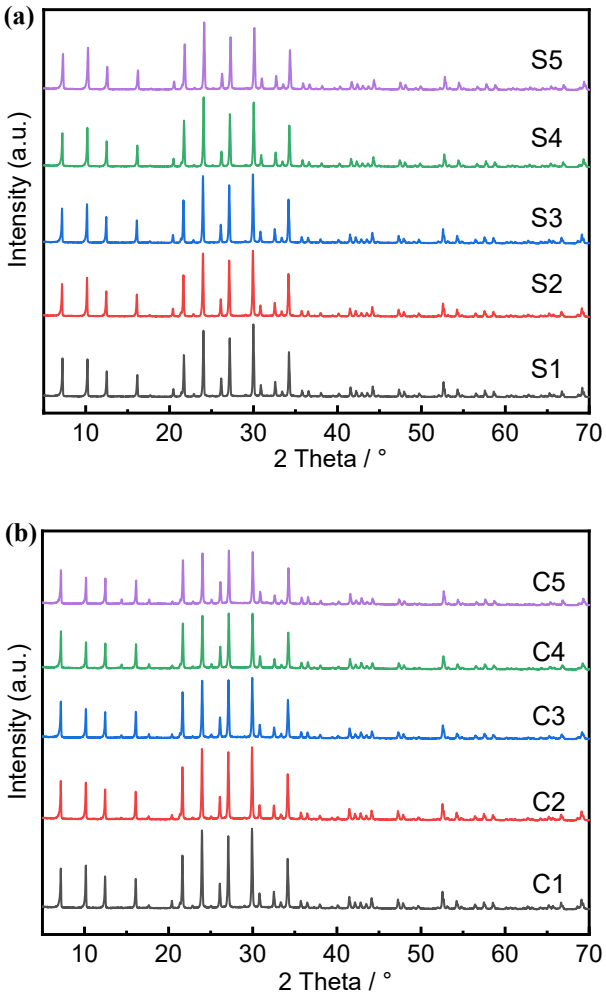

Fig. S1 XRD patterns of LTA zeolites via different particle sizes (a) and Ca exchange rate (b)

2 Dual-site Langmuir model for C_2H_4 and C_2H_6 adsorption isotherm fitting

The adsorption behaviors of C_2H_4 and C_2H_6 on the zeolites could be described from fitted adsorption isotherms by Dual-site Langmuir (DSL) equation:

$$q = \frac{q_{sat,A} b_A p}{1 + b_A p} + \frac{q_{sat,B} b_B p}{1 + b_B p}$$

wherein A and B represented two various adsorption sites, q_{sat} was saturation loading of adsorption sites, p was bulk gas phase pressure, and b was Langmuir coefficient.

Dual-site Langmuir parameter for adsorption of S1-S5 and C1-C5 as Table S1 and S2 showed.

Table S1 Dual-site Langmuir parameter for adsorption of S1-S5 and C1-C5 (C_2H_4,298K)

Sample	$q_{sat,A}$ (mmol/g)	$b_{sat,A}$ ($mbar^{-1}$)	$q_{sat,B}$ (mmol/g)	$b_{sat,B}$ ($mbar^{-1}$)	R^2
S1	1.1676	0.002901	2.6519	0.178897	0.999
S2	1.2036	0.003829	2.5663	0.292919	0.999
S3	1.2299	0.004050	2.5586	0.339424	0.999
S4	1.2469	0.004921	2.3794	0.349035	0.999
S5	1.1468	0.003398	2.5416	0.263521	0.999
C1	0.5207	0.001578	2.6340	0.059613	0.999
C2	0.9985	0.004630	2.2275	0.091779	0.999
C3	1.2294	0.005021	2.0529	0.164645	0.999
C4	1.0375	0.003087	2.3489	0.139171	0.999
C5	1.0180	0.002870	2.3988	0.149194	0.999

Table S2 Dual-site Langmuir parameter for adsorption of S1-S5 and C1-C5

(C_2H_6,298K)

Sample	$q_{sat,A}$ (mmol/g)	$b_{sat,A}$ ($mbar^{-1}$)	$q_{sat,B}$ (mmol/g)	$b_{sat,B}$ ($mbar^{-1}$)	R^2
S1	2.0503	0.004408	1.1827	0.033904	0.999
S2	1.8497	0.004687	1.4117	0.062496	0.999
S3	1.8049	0.004577	1.4682	0.063362	0.999
S4	1.8403	0.004878	1.3113	0.064303	0.999
S5	1.8311	0.004680	1.3155	0.064722	0.999
C1	1.1728	0.007251	1.3724	0.007252	0.999
C2	2.2656	0.007581	0.3718	0.043617	0.999
C3	0.7798	0.109373	1.9858	0.005990	0.999
C4	1.7975	0.005057	1.0569	0.058858	0.999
C5	1.3561	0.003384	1.5569	0.031608	0.999

3 Ideal adsorbed solution theory (IAST) calculation

The method of calculating selectivity was based on the premise that ethylene and ethane were at equal spreading pressure of the adsorbent material and used the pyIAST model [1] to calculate the molar percentages of ethylene and ethane on zeolite respectively. The IAST method was employed to evaluate the adsorption selectivity of modified zeolites for C_2H_4/C_2H_6 by equation [1]:

$$\alpha_{C_2H_4/C_2H_6} = \frac{x_{C_2H_4}/y_{C_2H_4}}{x_{C_2H_6}/y_{C_2H_6}}$$

wherein *x* and *y* represented the mole fractions of the gas and adsorbed phases, respectively.

The spreading pressure and $x_{C_2H_4}$ and $x_{C_2H_6}$ of S1-S5 and C1-C5 were shown, respectively, in Table S3.

Table S3 IAST calculation of S1-S5 and C1-C5 ($y(C_2H_4)/y(C_2H_6)$=50/50,1bar,298K)

Sample	spreading pressure	$x_{C_2H_4}$	$x_{C_2H_6}$	$\alpha_{C_2H_4/C_2H_6}$
S1	13.27	3.06	0.26	11.71
S2	13.86	2.97	0.29	10.41
S3	14.41	3.05	0.30	10.29
S4	14.80	3.10	0.28	11.13
S5	14.12	2.99	0.27	11.14
C1	9.58	2.53	0.31	8.17
C2	10.10	2.56	0.31	8.25
C3	11.03	2.53	0.38	6.70
C4	11.35	2.59	0.36	7.24
C5	11.62	2.66	0.32	8.36

Reference

1. Simon, C.M.;Smit, B.andHaranczyk, M. pyIAST: Ideal adsorbed solution theory (IAST) Python package. *Comput. Phys. Commun.* **2016,** *200*, 364-380.