Supporting information

A Novel Mechanism of Tuning Pore Size Distribution in Amorphous Carbon Molecular Sieves at Sub-angstrom level for Efficiently Separating Similar Gases

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List of contents

Section 1. The heating procedures for Preparation of samples RCMS-x

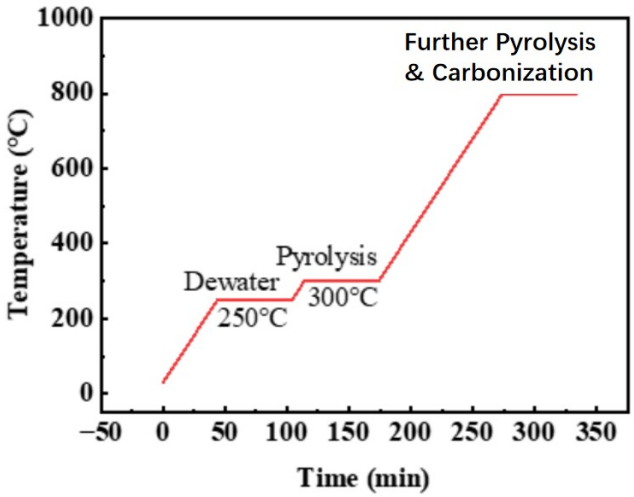

Figure S1. The heating procedures for Preparation of samples RCMS-x

Figure S1 presents a flow chart illustrating the preparation process of granular carbon molecular sieves (RCMS) from rice grains. First, 1 g of rice grains was placed in a tube furnace, which was then programmed to heat to the desired temperatures, following the prescribed heating procedures. **Figure S1** shows the heating procedures, which were as follows: After the rice grains were placed in a tube furnace, they were heated to 200-250℃ at a rate of 5℃/min for slow removal of water under a nitrogen atmosphere, lasting 1 hour. Next, the furnace was heated to 300 ℃ at the same heating rate of 5℃/min for pyrolysis, lasting 1 hour under nitrogen. After that, the temperature was increased to the desired values (700, 800, 900, or 1000 ℃) at a rate of 5℃/min for further pyrolysis and carbonization, lasting 1 hour under nitrogen atmosphere. Finally, these samples were cooled to room temperature in the N_2 atmosphere. The resulting RCMS samples were marked as RCMS-*x*, where X represents the given carbonization temperature at which the sample was prepared.

Section 2 The breakthrough experiment setup.

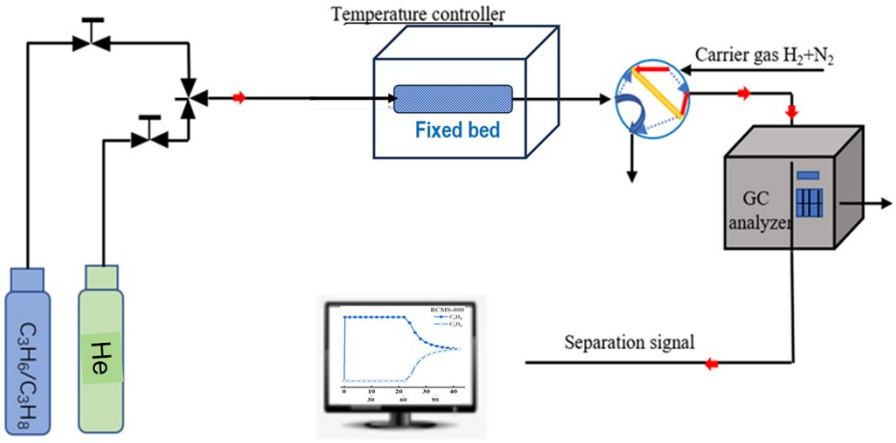

Figure S2. The breakthrough experiment setup

Section 3 Surface morphology and textural parameters of RCMS-*x*

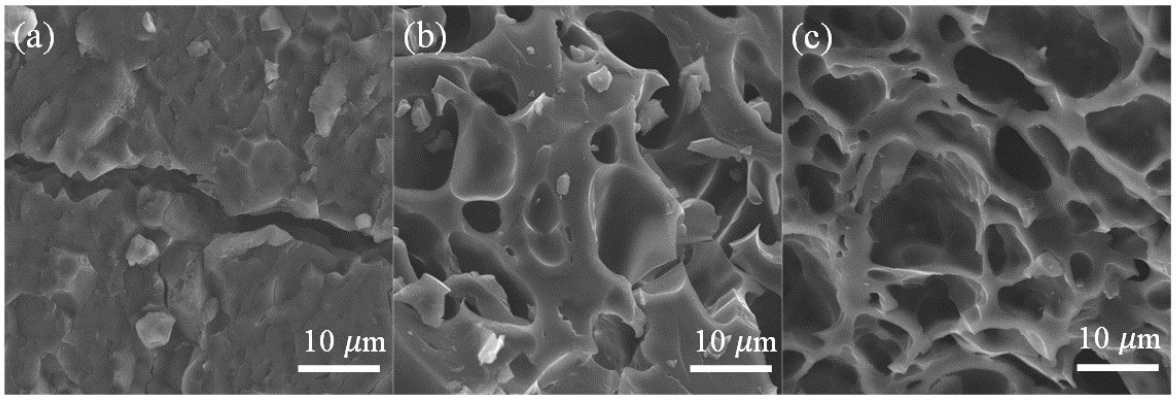

Figure S3. SEM of the samples: (a) rice, (b) RC250, (c) RCMS-800.

Figure S3 present the SEM image of rice, RC250 and RCMS-800. The sample exhibits rough surface and irregular macropores after elevated temperature radiation. This is attributed to some functional group decomposition, and subsequent escape of resulting volatile gases as well as water evaporation.

Table S1 The pore parameters of RCMS-*x* calculated by CO_2 isotherm at 195 K

Sample	S_{BET}(m^2/g)	V_{micro}(cm^3/g)
RCMS-700	246.65	0.140
RCMS-800	264.45	0.149
RCMS-900	236.14	0.134
RCMS-1000	210.01	0.126

Section 4. Kinetic diffusion coefficient of C_3H_6 and C_3H_8 on RCMS-*x*.

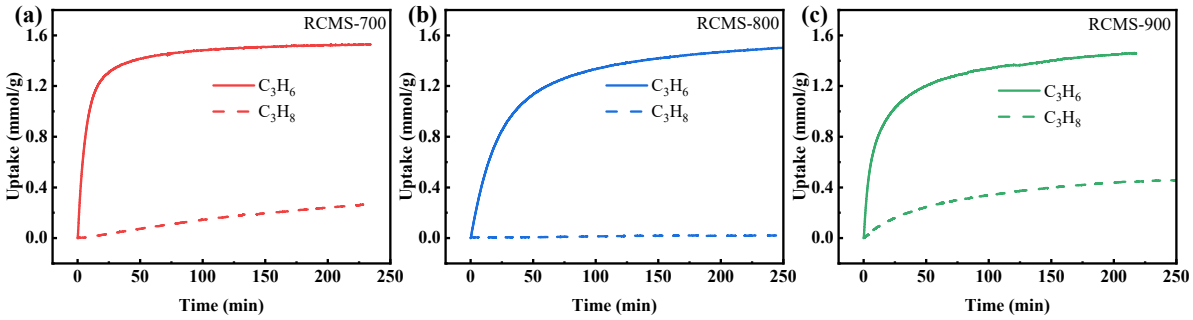

Figure S4. Kinetics curves of C_3H_6 and C_3H_8 adsorption on the samples RCMS-*x* at 308 K and 0.5 bar

Figure S4 presents adsorption kinetic curves of C_3H_6 and C_3H_8 on RCMS-*x*. According to the kinetic curves, diffusional time constants (D/r^2) for C_3H_6 and C_3H_8 can be calculated on the basis of following micropore diffusion model [1-4]:

$$\frac{q_t}{q_e} \approx \frac{6}{\sqrt{\pi}}\sqrt{\frac{Dt}{r^2}} \left(\frac{q_t}{q_e} < 0.3\right)$$

Where t is the adsorption time, min; q_t is transient uptakes at time t, mmol/g; q_e is the equilibrium adsorption amount of the adsorbent, mmol/g; D is the diffusivity, and r is the radius of the equivalent spherical sectional.

The relationship between q_t/q_e and $\sqrt{t}$ was plotted based on the kinetic curves, revealing a straight line with a slope of $\frac{6}{\sqrt{\pi}}\sqrt{\frac{D}{r^2}}$, as depicted in Figure S4. Then, the diffusion time constant D/r^2 can be determined from the slope of the straight lines. Table S2 lists the diffusion time constant D/r^2 of C_3H_6 and C_3H_8 on and the kinetic selectivity of three samples for C_3H_6 and C_3H_8.

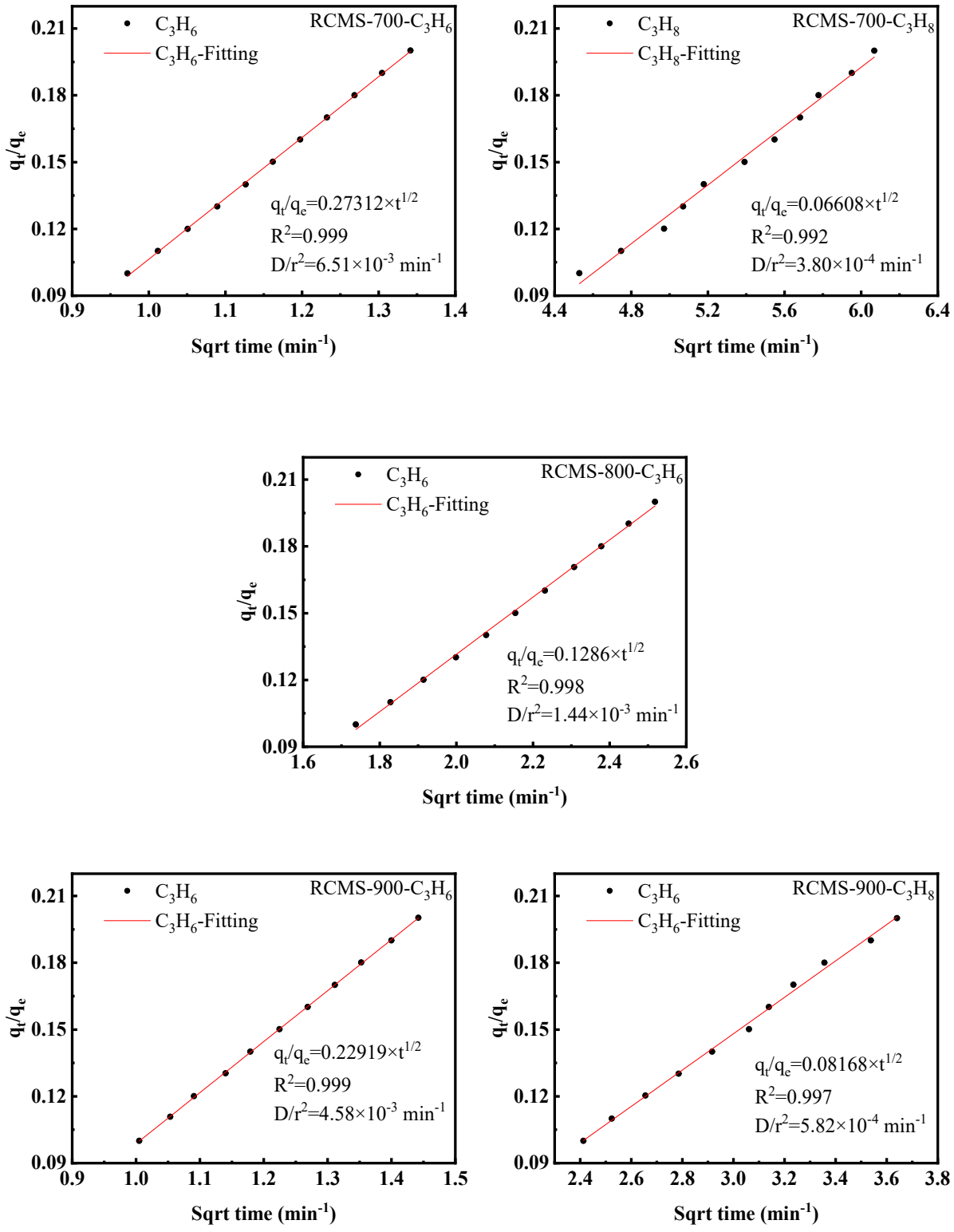

Figure S5. Plot of the fractional uptake (q_t/q_e) against the square root of adsorption time at 0.5 bar for C_3H_6 and C_3H_8 adsorption on the samples RCMS-*x*

Table S2 lists the diffusion time constant D/r^2 of C_3H_6 and C_3H_8 on and the kinetic selectivity of three samples for C_3H_6 and C_3H_8.

Table S2. The diffusional time constant and kinetic selectivity of RCMS-*x*.

Sample	$D/r^2(C_3H_6)$	$D/r^2(C_3H_8)$	Selectivity
RCMS-700	6.51×10^{-3}	3.80×10^{-4}	17.13
RCMS-800	1.44×10^{-3}	——	∞
RCMS-900	4.58×10^{-3}	5.82×10^{-4}	7.87

Section 5. Isotherms of C_3H_6 on RCMS-800 at different temperatures

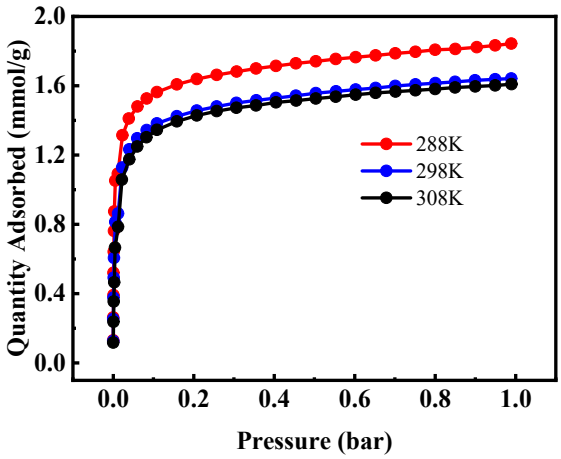

Figure S6. C_3H_6 isotherms of RCMS-800 at different temperatures

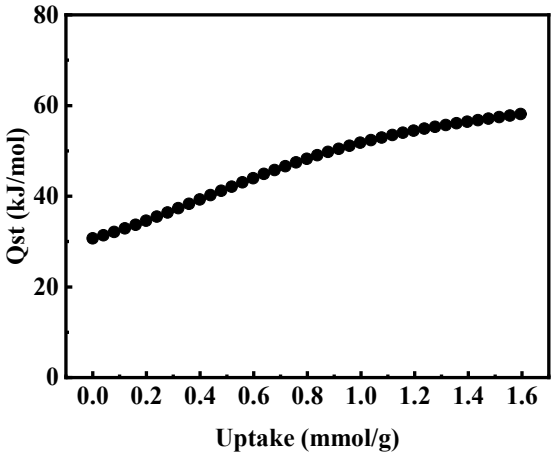

Figure S7. Isosteric heat of C_3H_6 adsorption on RCMS-800

Section 6 Breakthrough curves of C_3H_6 and C_3H_8 mixture in the fixed bed of RCMS-*x*

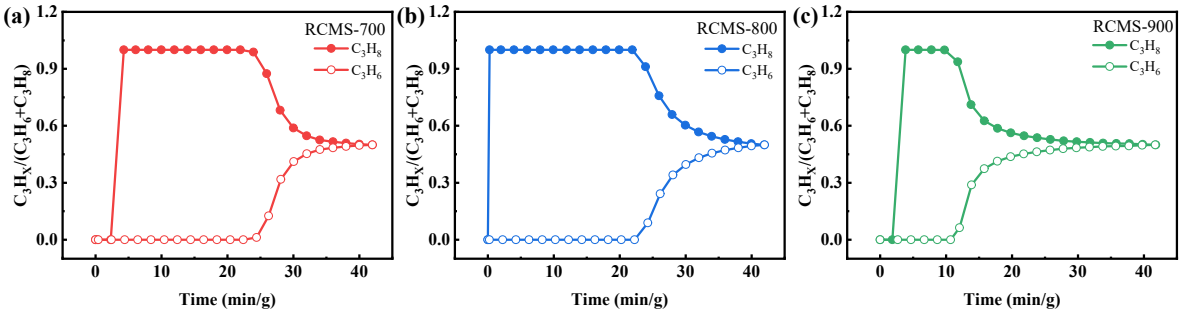

Figure S8. Breakthrough curves of C_3H_6 and C_3H_8 mixture (v/v=50/50) through the fixed bed packed with RCMS-*x* at 298 K and normal pressure.

Figure S8 shows Breakthrough curves of C_3H_6 and C_3H_8 mixture (v/v=50/50) through the fixed bed packed with RCMS-*x* at 298 K and normal pressure

References

1. C. Y. Lee, Y.-S. Bae, N. C. Jeong, O. K. Farha, A. A. Sarjeant, C. L. Stern, P. Nickias, R. Q. Snurr, J. T. Hupp and S. T. Nguyen, *Journal of the American Chemical Society*, 2011, **133**, 5228-5231.

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