Supporting Information

Template-free Amine-Bridged Silsesquioxane with dangling amino groups and its CO_2 adsorption performance

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No	Aminosilane precursors	Structure
1	Bis[3-(trimethoxysilyl)proyl]amine (BTMSPA)	$\begin{array}{ccc} CH_3 & CH_3 \\ O & O \\ H_3 CO Si & NH & SiO CH_3 \\ O CH_3 & O CH_3 \end{array}$
2	3-Aminopropyltriethoxysilane (APTES)	$H_{3}C^{-}O^{-}CH_{3}$ $H_{3}C^{-}O^{-}Si^{-}NH_{2}$ $H_{3}C^{-}O^{-}O^{-}NH_{2}$
3	[3-(2-Aminoethylamino)propyl]trimethoxysilane (DAMS)	$\begin{array}{c} & & & \\ & & & \\ H_3C^{-}O^{-}Si^{-} & & \\ H_3C^{-}O^{-}O^{-}Si^{-} & & \\ \end{array}$
4	N-(3-trimethoxysilylpropyl)diethylenetriamine (TAMS)	$\begin{array}{c} & & & \\ & & & \\ H_3C^{-}O^{-}Si^{-} & & \\ H_3C^{-}O^{-}O^{-}NH^{-$

Table S1. List of aminosilanes used for research and their structure



Figure S1. A schematic illustration of the CO₂ adsorption experiment



Figure S2. Experimental Setup of the packed bed reactor for investigating the adsorbent recyclability



Figure S3. SEM images of ABS (a), APTES-ABS (b), DAMS-ABS (c), and TAMS-ABS (d)



Figure S4. TGA profile of ABS and APTES-ABS



Figure S5. TEM images of samples with the APTES/MTMSPA molar ratio of 0.5 (A) and 0.75 (B)

$$R-Si-(OC_{2}H_{5})_{3} \xrightarrow{H_{2}O} R-Si-(OH)_{3} \xrightarrow{Self-condensation} \begin{cases} R-Si-(OH)_{2}-O-Si(OH)_{2}-R \\ T_{1} & T_{1} \\ R-Si-(OH)_{2}-O-Si(OH)R-OSi-...-OSi(OH)_{2}-R \\ T_{1} & T_{2} & T_{2} & T_{1} \\ & & & \\ T_{1} & T_{2} & T_{2} & T_{1} \\ & & & \\ R-Si-(OH)_{2}-O-Si(OH)R-OSiR-...-OSi(OH)_{2}-R \\ R-Si-(OH)_{2}-O-Si(OH)R-OSiR-...-OSi(OH)_{2}-R \\ T_{1} & T_{2} & T_{3} & T_{1} \\ \end{cases}$$

Figure S6. APTES self-condensation illustration. T_0 is the APTES hydrolyzed monomer with 3 unbonded OH groups, T_1 is singleside bonded APTES with 2 unbonded OH groups, T_2 is double-side bonded APTES with only one unbonded OH group and T_3 3-side bonded APTES with no unbonded OH group.



Figure S7. Nitrogen adsorption isotherms of APTES-ABS samples aged in water. Shift value for Y axis: 2h + 0, 8.5h +20, 24h + 30, 48h + 200, 72h + 380, 96h + 600, and 130h + 800.



Figure S8. Nitrogen adsorption isotherms of APTES-ABS samples aged in ethanol



Figure S9. TEM images of samples aged in water for 2h (A) and 130h (B)



Figure S10. Relation between surface area and CO₂ loading of APTES-ABS aged in water



Figure S11. FTIR spectra of APTES-ABS aged for 120h in ethanol before (a) and after (b) CO₂ adsorption. Sample was activated at 125 °C in nitrogen flow for 4h and then used for FTIR analysis and CO₂ adsorption.



Figure S12. CO₂ adsorption isotherms of samples aged 130h in water (after 14 months) and 144h in ethanol (after 12 months).



Figure S13. Pore size distribution of fresh APTES-ABS samples and after 3 month storage. Samples were aged in water for 8.5 h (A), 24h (B), 48 h (C) and 72 h (D).



Figure S14. Pore size distribution of fresh APTES-ABS samples and after 3 month storage. Samples were aged in water for 5 h (A), 24h (B), 48 h (C), 72 h (D), 120 h (E), and 144h (F).