## Electronic supplementary information (ESI) for: Precisely designed layered silicate as an effective and highly selective CO<sub>2</sub> adsorbent

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## Experimental details

NH<sub>2</sub>-C<sub>18</sub>-mag synthesized reaction C<sub>12</sub><sup>+</sup>-mag was by the of with octadecyltrichlorosilane (ODS) and 3-(aminopropyl)trimethoxysilane (APS). 1.0 g of  $C_{12}^{+}$ -mag, prepared according to the reported method,<sup>23</sup> was mixed with a solution of ODS (0.305 mL) in dehydrated toluene (80 mL) at 333 K for 3 days. After the evaporation of the solvent, the solid was mixed with a solution of APS (10 g) in toluene (50 mL) and the mixture was refluxed for 2 days. The product was separated by centrifugation, washed with toluene, a toluene/ethanol mixture, and ethanol, sequentially, and finally dried at 333 K. NH<sub>2</sub>-mag was synthesized by the method similar to that conducted for the synthesis of  $NH_2$ - $C_{18}$ -mag; 1.0 g of C<sub>12</sub><sup>+</sup>-mag was refluxed in a solution of APS (10 g) in toluene (50 mL). NH<sub>2</sub>-SBA and Au@CeO2 was prepared by the method reported in our previous literature.<sup>7</sup> Magadiite modified only with ODS (named C<sub>18</sub>-mag) was also synthesized by the similar way.

Gas adsorption was performed by a BELSORP-mini II (BEL, Japan). Before the adsorption, the sample was heated at 120  $^{\circ}$ C in vacuo for 3 h.

Photocatalytic oxidations were done as follows. Catalyst (30 mg) and an O<sub>2</sub>-saturated aqueous solution (25 mL) of formic acid (244 ppm) were added into a stainless-made closed container (50 mL) equipped with Pyrex glass window. The CO<sub>2</sub> adsorbent (50 mg) was placed in the gas phase of the reactor. The mixture was irradiated by a solar simulator (San-Ei Electric Co., Ltd., Japan) at 315 K, under magnetic stirring. After the removal of the catalyst by filtration, the amount of formic acid remained in the supernatant was determined by HPLC. The photocatalysis under a CO<sub>2</sub> atmosphere was conducted by adding dry ice into the starting mixture.



Fig. S1  $^{13}$ C CP/MAS NMR spectra of (a)  $C_{12}^{+}$ -mag, (b) NH<sub>2</sub>-mag, and (c) NH<sub>2</sub>-C<sub>18</sub>-mag.



Fig. S2 XRD patterns of (a)  $NH_2$ - $C_{18}$ -mag and (b)  $NH_2$ -mag after mixing with liquid  $H_2O$ .



**Fig. S3** CO<sub>2</sub> adsorption isotherms measured at 298 K of ( $\Box$ ) C<sub>18</sub>-mag (the amount of the grafted octadecyl group is 1.8 groups per Si<sub>14</sub>O<sub>29</sub>), ( $\Box$ ) NH<sub>2</sub>-mag, ( $\Box$ ) NH<sub>2</sub>-C<sub>18</sub>-mag and ( $\blacktriangle$ ) NH<sub>2</sub>-SBA.