SUPPORTING INFORMATION

In situ growth of ZIF-8 nanocrystals on layered double hydroxide

nanosheets for enhanced CO₂ capture

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Experimental Section

1. Synthesis of composite nanosheet

Preparation of Zn–Al layered double hydroxide

Zn-Al LDH was prepared by a modified homogeneous precipitation method, using urea as the ammonia releasing reagent. In a typical synthesis, an aqueous solution (100 mL) of Zn and Al nitrates (Zn : Al molar ratio equals to 2 : 1) with a total metal ion concentration of 0.15 mol/L in the final solution was added into a round bottom flask. Subsequently, the aqueous solution of urea (three times the [Al³⁺]) was added carefully. The white mixture was heated up to 100°C with vigorous stirring under reflux conditions in inert atmosphere for 48 hours. *Preparation of Zn-Al LDH@ZIF-8 composite nanosheet*

Zn-Al LDH@ZIF-8 composite nanosheet was prepared by *in situ* growth of ZIF-8 on Zn-Al LDH. In a typical procedure, 0.245g of 2-methylimidazole and 0.135g of HCOONa were dissolved in 40 mL methanol in a beaker. Then, 0.12g of Zn-Al LDH was added to the above solution. The mixture was mechanically stirred at

room temperature for several minutes before transferring to the Teflon-lined stainless steel autoclave followed by heating at 100°C for 2 hours. The product was collected by centrifugation and washed by methanol three times. The sample was then dried in a vacuum oven at 150°C for 4 h prior to further analysis.

2. Characterization.

Scanning electron microscopy (SEM) images were taken on Hitachi TM-4000. X-ray diffraction (XRD) patterns were recorded on a Bruker D8 diffractometer using Cu K α radiation. The nitrogen adsorption measurements of pore size and surface area were made at 77 K using an ASAP 2020 M apparatus. Before each measurement, the sample was outgassed at 473 K for 2 h under vacuum. CO₂ uptake was also measured on the ASAP 2020 M apparatus at atmospheric pressure (1 bar). Prior to uptake measurements, the sample was outgassed at 473 K for 2 h under vacuum.



Figure S1. FT-IR spectra of ZIF-8, Zn-Al LDH and Zn-Al LDH@ZIF-8.



Figure S2. EDX spectrum of the Zn-Al LDH@ZIF-8.



Figure S3. The EDX mapping of Zn-Al LDH@ZIF-8.



Figure S4. Nitrogen adsorption-desorption isotherms of Zn-Al LDH.



Figure S5. Nitrogen adsorption-desorption isotherms of ZIF-8.

Sample	Zn-Al LDH@ZIF-8	Zn-Al LDH	ZIF-8
BET surface (m ² ·g ⁻¹)	1136.87	556.32	1460
CO2 sorption (mmol·g ⁻¹)	1.0	0.1	0.65

 Table S1. The comparison of ZIF-8, Zn-Al LDH and Zn-Al LDH@ZIF-8.