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(Supplementary information)

## A lamellar structure zeolite LTA for CO2 capture

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## **Contents:**



Fig. S1. The NH<sub>3</sub>-TPD of 2D and 3D zeolite.

There were many solid acid centers on the surface of the zeolites after calcination, such as Brønsted and Lewis acid.<sup>46</sup> These solid acid centers were actually the active sites of the zeolites and the acid strength was tested by NH<sub>3</sub>-TPD. The active sites of zeolite were usually distributed on the trivalent Al<sup>+</sup> inside the skeleton. The advantage of the 2D zeolite LTA with a large specific surface area compared to 3D zeolite was that the skeleton aluminum ions were more easily exposed to the surface, resulting in more active sites.



Fig. S2. CO<sub>2</sub> adsorption-desorption isotherms of the zeolites with (a)3D, (b)2D.



Fig. S3. Heat of adsorption of zeolites. Closed symbol: 2D-LTA; open symbol: 3D-LTA. (Square: 273-298 K; triangle: 298-373 K; circular:273-373 K)

The heat of adsorption was calculated by Clausius-Clapeyron equation:

$$Q = \frac{RT_1T_2\ln(P_2/P_1)}{T_2 - T_1}$$

Where Q is heat of adsorption, KJ/mol; R is gas constant, 8.314J/mol; T is adsorption temperature, K; P is adsorption pressure, bar.

| Sample            | Temperature | CO <sub>2</sub> Capacity | CO <sub>2</sub> /CH <sub>4</sub> | Reference |
|-------------------|-------------|--------------------------|----------------------------------|-----------|
|                   | /K          | /mmol·g <sup>-1</sup>    | Selectivity                      |           |
| 2D-LTA            | 273         | 2.55ª                    | -                                | This work |
|                   | 298         | 2.23ª                    | 76.76                            | This work |
| 3D-LTA            | 273         | 2.20 <sup>a</sup>        | -                                | This work |
|                   | 298         | 1.92ª                    | 5.93                             | This work |
| ZIF-68            | 298         | 1.68                     | 5.0                              | 9         |
| ZIF-69            | 273         | 1.81                     | 5.1                              | 9         |
| ZIF-70            | 298         | 2.45                     | 5.2                              | 9         |
| ZIF-78            | 298         | 2.30                     | 10.6                             | 9         |
| ZIF-79            | 298         | 1.49                     | 5.4                              | 9         |
| ZIF-81            | 298         | 1.70                     | 5.7                              | 9         |
| ZIF-82            | 298         | 2.35                     | 9.6                              | 9         |
| NaA-RS            | 273         | 2.26                     | -                                | 10        |
|                   | 303         | 1.15                     | -                                | 10        |
|                   | 333         | 0.53                     | -                                | 10        |
| TEPA-ZSM-5        | 298         | 0.15                     | -                                | 11        |
| TEPA-ZY           | 298         | 1.12                     | -                                | 11        |
| TEPA-SAOP-34      | 298         | 0.45                     | -                                | 11        |
| NaA               | 273         | 3.5                      | -                                | 12        |
|                   | 298         | 3.1                      | -                                | 12        |
| 5A                | 293         | 1.86                     | 24.9                             | 13        |
| SAPO-34           | 303         | 2.4 <sup>b</sup>         | 3.8                              | 14        |
| ZSM-5             | 303         | 1.5 <sup>b</sup>         | 2.4                              | 14        |
| K-ETS-10          | 298         | 2.53                     | 52.5                             | 15        |
| As-synthesized-10 | 298         | 2.01                     | 49.6                             | 15        |
| PEHA/CDMC         | 373         | 3.72                     | -                                | 16        |
|                   | 298         | 2.50                     | -                                | 16        |

Table S1. Comparison of  $CO_2$  and  $CO_2/CH_4$  selectivity with different materials.

<sup>a</sup>  $(x \text{ mL} \cdot \text{g}^{-1})/(22.4 \text{L} \cdot \text{mol}^{-1}) = y \text{ mmol} \cdot \text{g}^{-1}$ <sup>b</sup> P=2bar