Supporting Information

Ni-based catalysts supported on Mg-Al hydrotalcites with different morphologies for CO₂

methanation: Exploring the effect of metal-support interaction

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- 1. Fig. S1. TG and DTG curves of synthetic HT.
- 2. Fig. S2. Powder XRD patterns of different calcined Ni/HT catalysts.
- 3. Fig. S3. SEM images of the different synthesized Ni/HT catalysts.
- 4. Fig. S4. EDS images of designed Ni/HT catalysts.
- 5. Fig. S5. EF-TEM images of Ni/HT catalyst.
- 6. Fig. S6. STEM images of Ni/HT-10 catalyst.
- 7. Fig. S7. CO₂ conversion over different synthesized Ni/HT catalysts during 15 h.



Fig. S1 TG and DTG curves of synthetic HT.

The TG-DSC experiments of the HT supports were also carried under air atmosphere, and the results were illustrated in **Fig. S1**. TG-DSC curves of the four as-prepared supports confirmed the formation of HT structure. The release of interlayer water taken place in the temperature of 140-260 °C, and then the two endothermic peaks between 250 to 450 °C were the -OH elimination and CO₂ release. For HT-E, the ethanol and water molecules enter into the interlayer gallery resulted in the peak difference. The peak difference of HT-150 was resulted from the environments changes of water molecules when prepared at high temperature.



Fig. S2 Powder XRD patterns of different calcined Ni/HT catalysts. NiO crystallite size of Ni/HT-9.5, Ni/HT-10, Ni/HT-150, and Ni/HT-E are 7.5, 7.1, 8.6, and 6.8 nm, respectively.



Fig. S3 SEM images of the different synthesized Ni/HT catalysts.



Fig. S4 EDS images of designed Ni/HT catalysts.



Fig. S5 EF-TEM images of Ni/HT catalyst.



Fig. S6 STEM images of Ni/HT-10 catalyst.

Stability of the Ni/HT catalysts



Fig. S7 CO₂ conversion over different synthesized Ni/HT catalysts during 15 h.

After the optimum reaction temperature determined, the short-term stability of all the Ni/HT catalysts were tested for 15 h. The result of CO_2 conversion over different Ni/HT catalysts are shown in **Fig. S7**. At the beginning, the activity of four Ni/HT catalysts were increased sharply at the first 1 h and show great stability during 15 h reaction.