**Supporting Information**

UV-Vis-IR irradiation driven CO$_2$ reduction with high light-to-fuel efficiency on a unique nanocomposite of Ni nanoparticles loaded on Ni doped Al$_2$O$_3$ nanosheets

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**Experimental**

The $\eta$ value was also calculated according to the standard molar Gibbs formation energy:

$$\eta = \frac{(r_{\text{H}_2} \times \Delta G^0_{\text{H}_2} + r_{\text{CO}} \times \Delta G^0_{\text{CO}} - r_{\text{CH}_4} \times \Delta G^0_{\text{CH}_4} - r_{\text{CO}_2} \times \Delta G^0_{\text{CO}_2})}{P}$$

The values of $\Delta G^0_{\text{H}_2}$, $\Delta G^0_{\text{CO}}$, $\Delta G^0_{\text{CH}_4}$, and $\Delta G^0_{\text{CO}_2}$ are 0, -137.2, -50.5, and -394.4 kJ mol$^{-1}$, respectively. The $\eta$ value of Ni/Ni-Al$_2$O$_3$ with the focused UV-Vis-IR irradiation is 13.5%.

The $\eta$ value according to $\Delta G^0$ being less than the corresponding values according to $\Delta H^0$ are ascribed to the following reason. As CRM is a reaction of entropy enhancement ($\Delta S_{298} = 256.6$ J K$^{-1}$ mol$^{-1}$), its $\Delta G_{298}$ (170.5 kJ mol$^{-1}$) is less than its $\Delta H_{298}$ (247 kJ mol$^{-1}$) according to the Gibbs free energy equation ($\Delta G = \Delta H - T \times \Delta S$).

For perform photocatalytic CRM on Ni/Ni-Al$_2$O$_3$ at near ambient temperature under the focused UV-Vis-IR irradiation, the reactor was put in an ice-water bath.

The thermodynamic maximum $\eta_{\text{max}}$ value of our reaction system is decided by Carnot efficiency as well as the solar absorption efficiency:

$$\eta_{\text{max}} = \left[1 - \sigma \times T_{\text{H}}^4 / (I_{\text{DNI}} \times C) \right] \times \left[1 - T_{\text{L}} / T_{\text{H}} \right]$$

Where $\sigma$ is Stefan–Boltzmann constant, $I_{\text{DNI}}$ is the direct normal solar irradiation (1 kWm$^{-2}$), C is the concentration ratio of solar flux, $T_{\text{H}}$ and $T_{\text{L}}$ are the high and low temperatures of the equal Carnot heat engine.

In the present case, the focused UV-Vis-IR irradiation intensity was 333.8 kWm$^{-2}$ (C is equal to 333.8). The focused UV-Vis-IR irradiation resulted in the surface temperature of Ni/Ni-Al$_2$O$_3$ being rapidly raised to an equilibrium temperature ($T_{\text{H}} = 764$ °C) from room temperature ($T_{\text{L}} = 25$ °C). The $\eta_{\text{max}}$ value is calculated to be 57.2%.
Figure S1. SEM image (A), TEM images (B, C), and HRTEM image (D) of Ni-Al$_2$O$_3$. 
Figure S2. HAADF image (A) and the corresponding element mappings of Ni (B), Al (C), and O (D) of Ni-Al₂O₃.
Figure S3. SEM image (A), TEM images (B, C), and HRTEM image (D) of Ni/Al₂O₃.
Figure S4. HAADF image (A) and the corresponding element mappings of Ni (B), Al (C), and O (D) of Ni/Al₂O₃.
Figure S5. XPS spectra of Ni2p, Al2p, and O1s in Ni-Al2O3, Ni/Al2O3, Ni/Al2O3, and the used Ni/Al2O3 catalyst after the 80 h photothermocatalytic durability test.
Figure S6. N\textsubscript{2} adsorption and desorption (A) and BJH adsorption pore size distribution (B) of Ni-Al\textsubscript{2}O\textsubscript{3}.
Figure S7. N$_2$ adsorption and desorption (A) and BJH adsorption pore size distribution (B) of Ni/Ni-Al$_2$O$_3$. 
Figure S8. N$_2$ adsorption and desorption (A) and BJH adsorption pore size distribution (B) of Ni/Al$_2$O$_3$. 
Figure S9. TG-MS profiles of the used catalysts of Ni/Al<sub>2</sub>O<sub>3</sub> (A) and Ni/Ni-Al<sub>2</sub>O<sub>3</sub> (B) after reacted for 14 and 80 h, respectively.
Figure S10. XRD patterns of the used catalysts of Ni/Al$_2$O$_3$ (A) and Ni/Ni-Al$_2$O$_3$ (B) after reacted for 14 and 80 h, respectively.
Figure S11. TEM image of the used catalysts of Ni/Al₂O₃ (A) and Ni/Ni-Al₂O₃ (B) after reacted for 14 and 80 h, respectively.
**Figure S12.** HRTEM image of the used Ni/Al$_2$O$_3$ catalyst after reacted for 14 h.

**Figure S13.** Time course of CO and H$_2$ production rates for photocatalytic CRM on Ni/Ni-Al$_2$O$_3$ at near room temperature under the UV-Vis-IR irradiation.
Figure S14. The equilibrium temperatures of Ni/Ni-Al$_2$O$_3$ under the focused Vis-IR irradiation.

Figure S15. The CH$_4$-TPD profiles of Ni/Ni-Al$_2$O$_3$ under the UV-Vis-IR irradiation and in the dark.

Figure S16. The CO$_2$-TPD profiles of Ni/Ni-Al$_2$O$_3$ under the UV-Vis-IR irradiation and in the dark.