

## Electronic Supplementary Information

# Efficient Conversion of CO<sub>2</sub> to Methane Using Thin-Layer SiO<sub>x</sub> Matrix Anchored Nickel Catalysts

Xieyi Huang,<sup>ab</sup> Peng Wang,<sup>bc</sup> Zhichao Zhang,<sup>d</sup> Shaoning Zhang,<sup>a</sup> Xianlong Du,<sup>\*e</sup> Qingyuan Bi<sup>\*a</sup>  
and Fuqiang Huang<sup>\*acf</sup>

<sup>a</sup> State Key Laboratory of High Performance Ceramics and Superfine Microstructure, Shanghai Institute of Ceramics, Chinese Academy of Sciences, Shanghai 200050, P. R. China  
*E-mail:* biqingyuan@mail.sic.ac.cn; huangfq@mail.sic.ac.cn

<sup>b</sup> University of Chinese Academy of Sciences, Beijing 100049, P. R. China

<sup>c</sup> School of Physical Science and Technology, ShanghaiTech University, Shanghai 200031, P. R. China

<sup>d</sup> Department of Materials Science and Engineering, University of Pennsylvania, Philadelphia, PA 19104-6272, USA

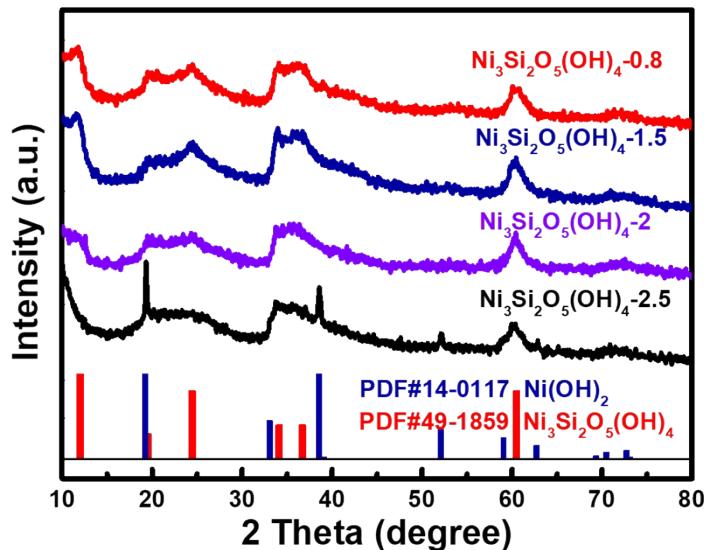
<sup>e</sup> Key Laboratory of Interfacial Physics and Technology, Shanghai Institute of Applied Physics, Chinese Academy of Sciences, Shanghai 201800, P. R. China

*E-mail:* duxianlong@sinap.ac.cn

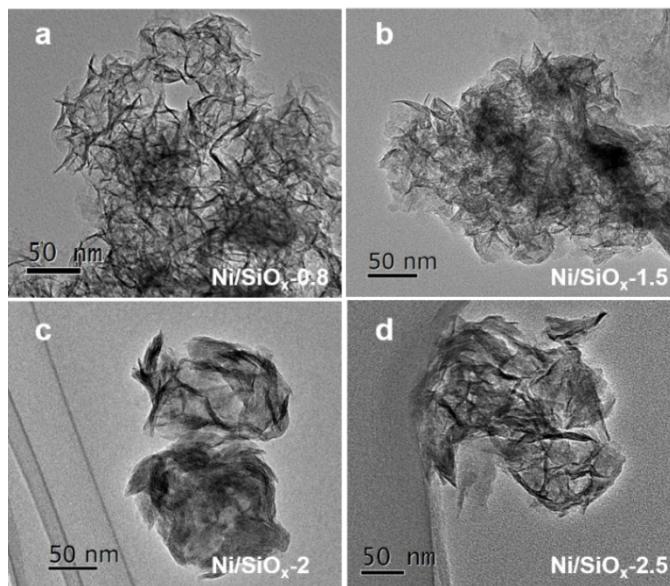
<sup>f</sup> Beijing National Laboratory for Molecular Sciences and State Key Laboratory of Rare Earth Materials Chemistry and Applications, College of Chemistry and Molecular Engineering, Peking University, Beijing 100871, P. R. China

**Table S1** Structure parameters of the optimized  $\text{Ni}_3\text{Si}_2\text{O}_5(\text{OH})_4$  basic cell.

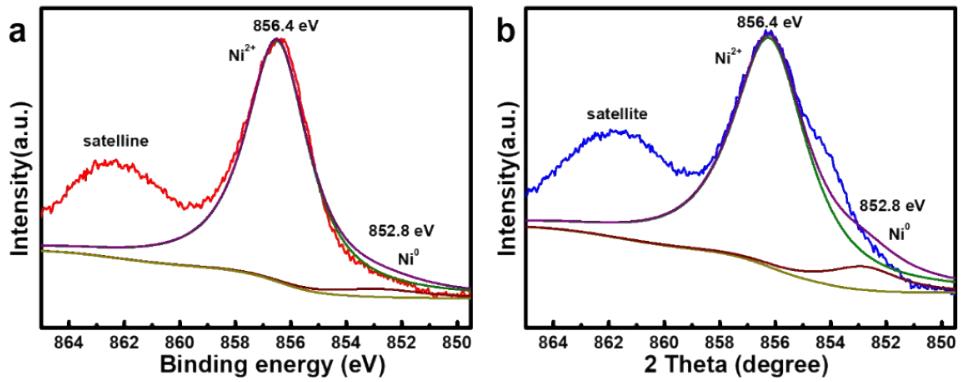
Symmetry	Space-group: $\text{P}_{31m}$ (157) - trigonal		
	$a = 5.3621 \text{ \AA}$	$\alpha = 90^\circ$	
Cell parameters	$b = 5.3621 \text{ \AA}$	$\beta = 90^\circ$	
	$c = 7.3784 \text{ \AA}$	$\gamma = 120^\circ$	



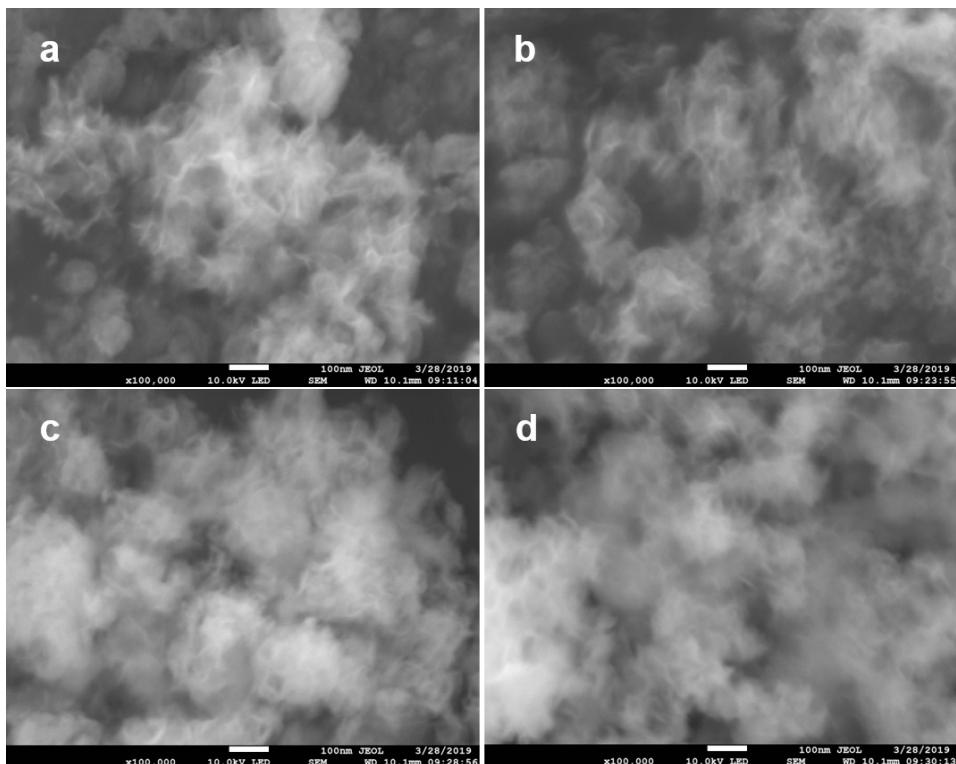
**Fig. S1** XRD patterns of  $\text{Ni}_3\text{Si}_2\text{O}_5(\text{OH})_4$  precursors with different Ni/Si ratios. Note that the coexistence of  $\text{Ni}(\text{OH})_2$  and  $\text{Ni}_3\text{Si}_2\text{O}_5(\text{OH})_4$  phases suggests that when the amount of nickel is too much, the suppression effect of  $\text{Ni}(\text{OH})_2$  formation by  $\text{NH}_4\text{Cl}$  becomes weak.<sup>S1</sup>



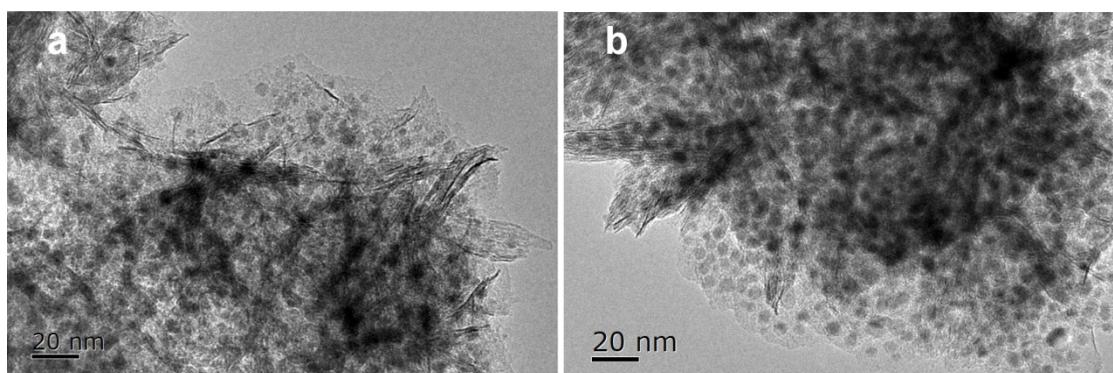
**Fig. S2** TEM images of layered structure  $\text{Ni}_3\text{Si}_2\text{O}_5(\text{OH})_4$  precursors with different Ni/Si ratios: (a)  $\text{Ni}/\text{SiO}_x$ -0.8, (b)  $\text{Ni}/\text{SiO}_x$ -1.5, (c)  $\text{Ni}/\text{SiO}_x$ -2, and (d)  $\text{Ni}/\text{SiO}_x$ -2.5.



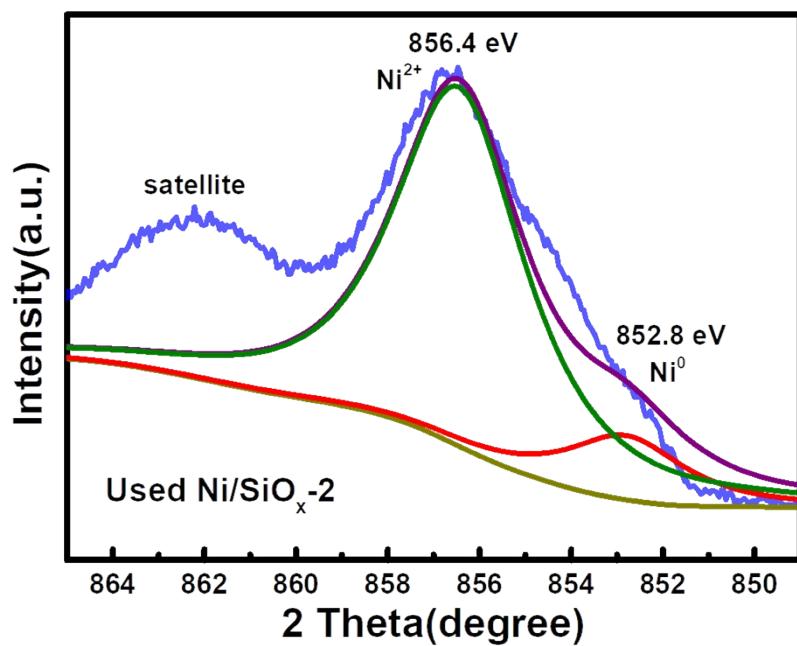
**Fig. S3** XPS Ni 2p<sub>3/2</sub> spectra of (a) Ni/SiO<sub>x</sub>-1.5 and (b) Ni/SiO<sub>x</sub>-2 catalysts.



**Fig. S4** SEM images of (a) Ni/SiO<sub>x</sub>-0.8, (b) Ni/SiO<sub>x</sub>-1.5, (c) Ni/SiO<sub>x</sub>-2, and (d) Ni/SiO<sub>x</sub>-2.5 catalysts.



**Fig. S5** HRTEM images of the used Ni/SiO<sub>x</sub>-2 catalyst.



**Fig. S6** XPS  $\text{Ni } 2\text{p}_{3/2}$  spectrum of used  $\text{Ni/SiO}_x$ -2 catalyst.

#### Reference

- (S1) C. J. Tang, J. Z. Sheng, C. Xu, S. M. B. Khajehbashi, X. P. Wang, P. Hu, X. J. Wei, Q. L. Wei, L. Zhou and L. Q. Mai, *J. Mater. Chem. A*, 2015, **3**, 19427–19432.