# CO and $CO_2$ methanation over Ni/SiC and Ni/$SiO_2$ catalysts

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**Abstract:** Various Ni catalysts supported on SiC and $SiO_2$ were prepared by a wet impregnation (WI) and deposition-precipitation (DP) method and applied them to CO and $CO_2$ methanation. In the case of CO methanation, a noticeable increase in the specific catalytic activity was observed with increasing Ni content from 10 to 20 wt% irrespective of the preparation method and support. For supported Ni catalysts prepared with WI method, Ni/$SiO_2$ catalyst is superior to Ni/SiC catalyst for both CO and $CO_2$ methanation. However, Ni/SiC catalyst prepared with DP method showed the highest catalytic activity for CO methanation among tested catalysts. These results clearly imply that both the preparation method and support are important factors to determine the catalytic activity for CO and $CO_2$ methanation.

**Keywords:** CO methanation, $CO_2$ methanation, Ni catalyst, SiC, $SiO_2$.

## 1. Introduction

CO and $CO_2$ methanation, which are important reactions in energy conversion technology, have been investigated to develop highly efficient catalysts [1-6]. Ni-based catalysts are more preferred for the methanation process owing to their low cost, good activity, and high availability compared with noble metal catalysts [2-4]. The catalytic activity over supported Ni catalysts has been reported to be strongly dependent on the nature of the support [3, 4] and preparation methods [5]. $SiO_2$ is a well-known conventional support for Ni-based catalysts with high activity [2]. However, its low thermal conductivity might cause the sintering of Ni metal during the highly exothermic reactions including CO and $CO_2$ methanation. On the other hand, SiC owns the excellent mechanical strength, superior thermal stability, high heating conductivity, and chemical inertness. Therefore, SiC can be considered as a promising catalyst support [6]. Besides, the deposition-precipitation (DP) method is regarded to be more effective for achieving high metal dispersion than a simple wet impregnation (WI) method [5]. Therefore, Ni/$SiO_2$ and Ni/SiC catalysts with different Ni contents were prepared by the WI and DP method and compared for CO and $CO_2$ methanation.

## 2. Experimental

Ni/$SiO_2$ and Ni/SiC catalysts were prepared by a WI method and DP with urea (DPU) method from an aqueous $Ni(NO_3)_2$ solution and $SiO_2$ (Zeochem, ZEOprep 60, $S_{BET}$ = 542 $m^2/g$) or SiC (US Nano, $S_{BET}$ = 43 $m^2/g$). The catalysts were calcined in air at 500 ℃ and reduced in hydrogen at 500 ℃ before the activity test. The catalytic activity was measured at atmospheric pressure in the reaction temperature range 140 - 450 $^oC$. The feed gas composed of 1 mol% CO (or $CO_2$), 50 mol% $H_2$, and 49 mol% He was contacted with 0.10 g of the catalyst at a flow rate of 100 mL/min. Various techniques were employed to characterize the catalysts including $N_2$ physisorption, $CO_2$, $H_2$ chemisorption, $H_2$-TPR, chemisorption, $CO_2$-TPD, XRD, ICP-OES, STEM-EDX, and TEM.

## 3. Results and discussion

The catalytic activity for CO and $CO_2$ methanation over Ni catalysts supported on different supports with WI and DP methods are shown in **Figure 1**. The 20 wt% Ni/SiC prepared by DP method showed the highest CO methanation activity: 100% conversion at 300 $^oC$ with the smallest $C_2H_6$ yield (0.4%) (**Table 1**).

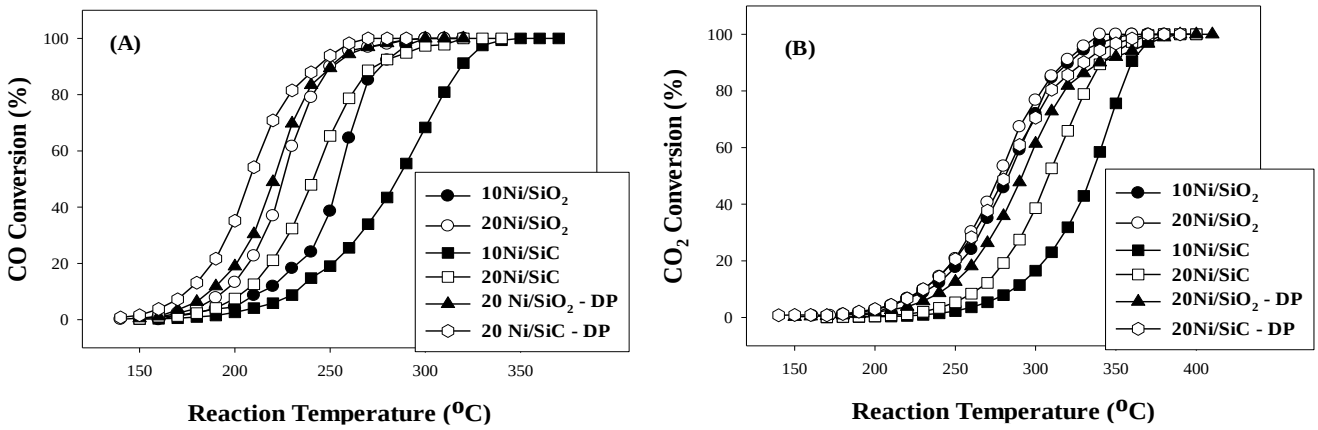


**Figure 1.** Catalytic performance of Ni supported catalysts via different kinds of supports and preparation methods on CO methanation (A) and $CO_2$ methanation (B). Reaction conditions: 1 mol% $CO_x$, 50 mol% $H_2$, 49 mol% He, F/W = 1,000 mL/min/$g_{cat}$.

Regarding $CO_2$ methanation, both Ni/SiC and Ni/$SiO_2$ exhibited 100% $CH_4$ selectivity. Ni/$SiO_2$ catalyst appears to be much superior to Ni/SiC catalyst as long as the catalysts are prepared with WI method. However, no noticeable difference in the catalytic activity can be found among Ni/$SiO_2$ prepared with WI method, Ni/$SiO_2$ prepared with DP method, and Ni/SiC prepared with DP method.

**Table 1.** The maximum products yield detected in CO methanation over Ni supported catalysts.

| Catalysts | Products Yield (%) | | |
|---|---|---|---|
| | $CH_4$ | $C_2H_6$ | $C_3H_8$ |
| 20 Ni/$SiO_2$ - WI | 98.6 | 1.1 | 0.3 |
| 20 Ni/$SiO_2$ - DP | 99.3 | 0.4 | 0.3 |
| 20 Ni/SiC - WI | 99.5 | 0.5 | 0 |
| 20 Ni/SiC - DP | 99.6 | 0.4 | 0 |

## 4. Conclusions

Ni/$SiO_2$ and Ni/SiC catalysts with different Ni contents were prepared by the WI and DP method and compared for CO and $CO_2$ methanation. In the case of CO methanation, a noticeable increase in the specific catalytic activity was observed with increasing Ni content from 10 to 20 wt% irrespective of the preparation method and support. For supported Ni catalysts prepared with WI method, Ni/$SiO_2$ catalyst is superior to Ni/SiC catalyst for both CO and $CO_2$ methanation. However, Ni/SiC catalyst prepared with DP method showed the highest catalytic activity for CO methanation among tested catalysts.

## References

1. J. Gao, Q. Liu, F. Gu, B. Liu, Z. Zhong, F. Su, RSC Adv. 5 (2015) 22759.
2. P. Lakshmanan, M.S. Kim, E.D. Park, Appl. Catal. A 513 (2016) 98.
3. T.A. Le, M.S. Kim, S.H. Lee, T.W. Kim, E.D. Park, Cat. Today 293-294 (2017) 89.
4. T.A. Le, T.W. Kim, S.H. Lee, E.D. Park, Cat. Today (2017) https://doi.org/10.1016/j.cattod.2017.09.031.
5. P. Burattin, M. Che, C. Louis, J. Phys. Chem. B 102 (1998) 2722.
6. G. Zhang, T. Sun, J. Peng, S. Wang, S. Wang, Appl. Catal. A 462-463 (2013) 75.