

Available online at [www.sciencedirect.com](http://www.sciencedirect.com)**ScienceDirect**

Energy Procedia 79 (2015) 137 – 142

Energy

**Procedia**

2015 International Conference on Alternative Energy in Developing Countries and  
Emerging Economies

## Dry Methane Reforming Performance of Ni-based Catalyst Coated onto Stainless Steel Substrate

Suntorn Sangsong<sup>a,b</sup>, Monrudee Phongaksorn<sup>a,b\*</sup>, Sabaithip Tungkamani<sup>a,b</sup>,  
Thana Sornchamni<sup>c</sup>, Rungroj Chuvaree<sup>c</sup>

<sup>a</sup>Department of Industrial Chemistry, Faculty of Applied Science,

King Mongkut's University of Technology North Bangkok, Bangkok, 10800, Thailand

<sup>b</sup>Research and Development Center for Chemical Engineering Unit Operation and Catalyst Design (RCC),

King Mongkut's University of Technology North Bangkok, Bangkok, 10800, Thailand

<sup>c</sup>PTT Public Company Limited, PTT Research and Technology Institute, Ayutthaya, 13170, Thailand

### Abstract

In this work, the Ni-based catalysts (10wt.%Ni/Al<sub>2</sub>O<sub>3</sub>-MgO and 10wt.%Ni/MgO) were synthesized by sol-gel method. Each catalyst was prepared as catalyst pellets and catalyst-coated plate, stainless steel plate coated with catalyst. The catalyst-coated plates were fabricated using spray coating. The dry methane reforming (DMR) performances of catalyst-coated plate were investigated and compared with DMR performances of catalyst pellets. As the mass transfer was improved, the methane consumption rate per gram of catalyst coated onto metal plate was ten times greater than those of catalyst pellets approximately. The carbon deposition was also significantly prevented when the catalyst was constructed as a thin-film layer on the plate under our developing condition. Therefore, this catalyst-coated plate can be further developed for multichannel reactor.

© 2015 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY-NC-ND license

(<http://creativecommons.org/licenses/by-nc-nd/4.0/>).

Peer-review under responsibility of the Organizing Committee of 2015 AEDCEE

**Keywords:** Nickel Based Catalysts, Spray Coating, Dry Methane Reforming

### 1. Introduction

Syngas or synthesis gas mainly consists of hydrogen and carbon monoxide which are widely used chemical industries and electricity generation. Syngas also plays a vital role in alternative energy production as it is a reactant for liquid fuel synthesis process (Fischer-Tropsch process). Nonetheless,

\* Corresponding author. Tel.: +66 255 52000 (4822); fax: +66 258 78251

E-mail address: [monrudee.p@sci.kmutnb.ac.th](mailto:monrudee.p@sci.kmutnb.ac.th)

hydrogen is considered as a direct high efficiency alternative fuel in near future. Syngas can be produced from biomass, biological gas, natural gas or coal through the catalytic reforming processes such as steam reforming, dry reforming, partial oxidation and autothermal reforming with different metal supported catalyst and reactants. Normally, catalysts for reforming are pelletized Ni based catalysts, placed in a fixed-bed reactor [1]. However, the pellet catalysts have been concerned with a stability issue due to heat and mass transfer limitations, causes of sintering and carbon formation problems [2,3]. Therefore, researchers have overcome these limitations by employing a new type of reactor, called multichannel reactor, which improves temperature control and mass diffusion. In the coated-wall of multichannel reactor, the physical size of channel in the reactor is decreased and the surface-to-volume ratio is increased. As a result, the relative surface area per fluid volume is high which improves heat and mass transfer performance [4-7]. The requirement for this type of reactor is the proper coating condition and technique to develop thin-film layer of heterogeneous catalyst onto the reactor walls. Recently, the three major techniques; wash coating, spin coating and spray coating, have been applied [8]. Advantages of these techniques are high efficiency, low operating temperature, simple installation, simple handling, and simple maintenance.

This research aims to investigate the dry methane reforming (DMR) performance of our Ni-based supported catalysts coated onto stainless steel plate by spray coating and to compare with DMR performance of the similar catalyst in pellet form under the same reaction condition.

## 2. Experimental

### 2.1 Catalyst Preparation

10wt.%Ni/Al<sub>2</sub>O<sub>3</sub>-MgO (10NAM) and 10wt.% Ni/MgO (10NM) were prepared by sol-gel method followed by dried at 60°C for overnight and calcined at 650°C for 4 h. Then, 10NAM and 10NM powder was pelletized or fabricated as a coated stainless steel (SS 316) plate (6.9 mm x 60.0 mm x 4.0 mm). To produce a catalyst-coated plate, the catalyst powder was prepared as a slurry by ball mill technique. After coating process, the coated plate was dried at 60°C for overnight and calcined at 650°C for 4 h.

### 2.2 Catalyst Tests

DMR performance were carried out in a tubular reactor. Catalyst pellets (~200 mg) were packed as a fixed-bed, whereas catalyst-coated plates (~15 mg) were located inside the reactor vertically with stainless steel housing. Catalysts were reduced in-situ in H<sub>2</sub> flow (30 mL min<sup>-1</sup>) at 620°C for 2 h. The reforming was performed at 620°C for 6 h with the mixed flows of CH<sub>4</sub> flow (15 mL min<sup>-1</sup>), CO<sub>2</sub> flow (25 mL min<sup>-1</sup>), and N<sub>2</sub> flow (20 mL min<sup>-1</sup>). The outlet gas was analysed using an on-line gas chromatograph (Agilent GC7890A) equipped with a thermal conductivity detector (TCD).

### 2.3 Catalyst Characterization

The catalyst pellets and catalyst-coated plate morphology were depicted by stereo microscopy (SM, NIKON® SMZ800) and scanning electron microscopy, (SEM, FEI® QUANTA 250).

## 3. Results

### 3.1 Surface Morphology of Coating

The micrographs of 10NAM and 10NM powder are illustrated in Fig. 1 (a) and (b) respectively. As seen in the figure, the particle of the catalyst powder is different in particle size. Although catalyst particle on the coated plate is more uniform (Fig. 1 (c,d)), the agglomeration of the powder still exists due to the characteristic of the spray coating.

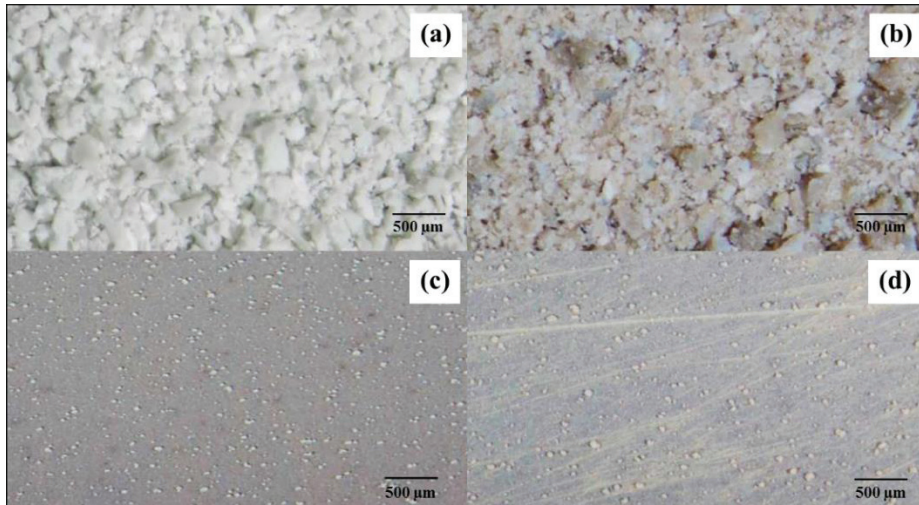


Fig. 1. SM micrographs of fresh catalysts: (a) 10NAM powder, (b) 10NM powder, (c) 10NAM-coated plate (10NAMC) and (d) 10NM-coated plate (10NMC)

### 3.2 DMR Performance

DMR results are presented in Fig. 2. It shows that the methane consumption rate per gram of 10NAM increases from 0.05-0.06  $[\text{molCH}_4]\text{g}^{-1}\text{min}^{-1}$  for pellet (10NAMP) to 0.55-0.98  $[\text{molCH}_4]\text{g}^{-1}\text{min}^{-1}$  for coated plate (10NAMC) and this rate per gram of 10NM increases from 0.04-0.06  $[\text{molCH}_4]\text{g}^{-1}\text{min}^{-1}$  for pellet (10NMP) to 0.49-0.89  $[\text{molCH}_4]\text{g}^{-1}\text{min}^{-1}$  for coated plate (10NMC). The results reveal that the improvement of mass transfer by surface expansion of metal active site enhances activity for ten times, approximately. In the results, 10NAMC provides greater methane consumption rate than 10NMC slightly. It can be attributed to the low surface area and low porosity of MgO, compared to those of  $\text{Al}_2\text{O}_3\text{-MgO}$  [1,9,10].

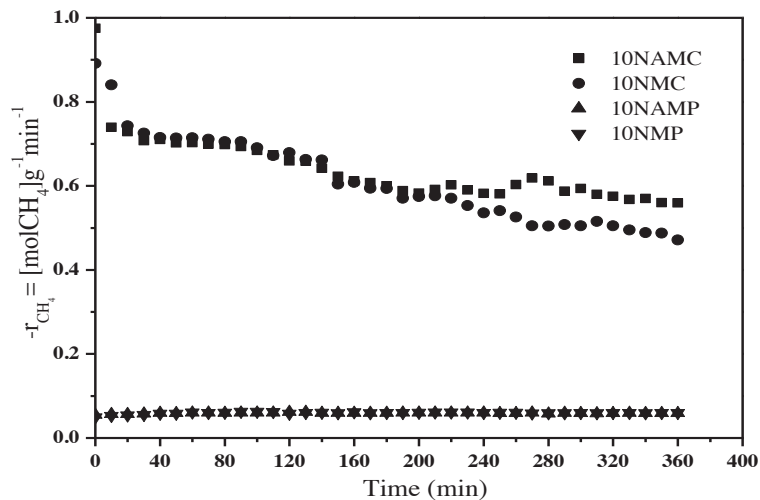


Fig. 2. The methane consumption rate per gram of catalyst over 10NAM pellet (10NAMP),

10NM pellet (10NMP), 10NAM-coated plate (10NAMC) and 10NM-coated plate (10NMC)

### 3.3 Carbon Formation

SM and SEM micrographs (Fig. 3) of the spent catalyst pellets (10NAMP, Fig. 3 (a)) and the spent catalyst-coated plate (10NAMC, Fig. 3 (b)) show that the filament carbon were formed on the surface of the spent catalyst pellet (Fig. 3 (c)). This type of carbon has not grown on the surface of catalyst-coated plate (Fig. 3 (d)). The reason of carbon deposition on catalyst pellet is the limitation of heat and mass transfer which constructs a hot spot and a local rich carbon species over some active area inside the pore of catalyst. In the case of spent catalyst-coated plate, the average thickness of catalyst layer on 10NAMC is less than  $10\ \mu\text{m}$  (Fig. 4 (a)) which is in the range of thin-film. Thus, the active area exposes to the surface of a thin-film (Fig. 4 (b)). Therefore, the local heat and mass accumulation due to the pore structure is negligible.

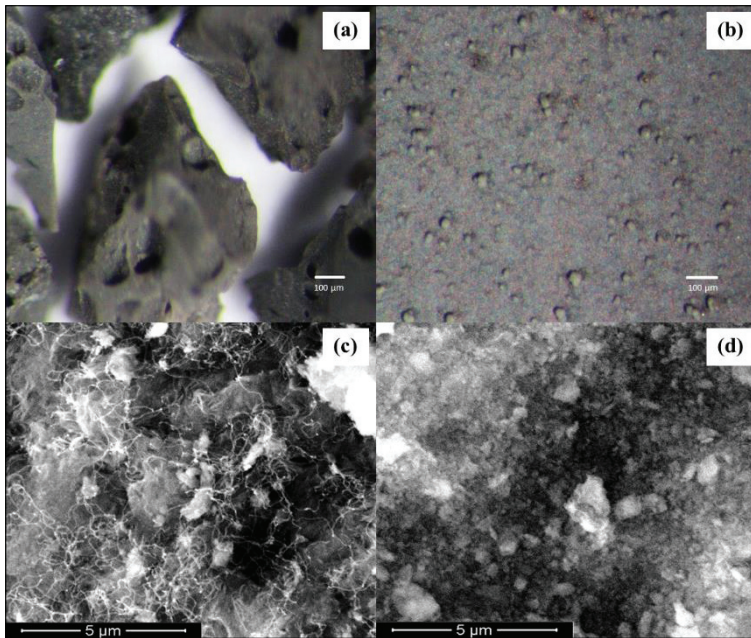


Fig. 3. (a) SM micrograph of spent 10NAMP, (b) SM micrograph of spent 10NAMC, (c) SEM micrograph of spent 10NAMP and (d) SEM micrograph of spent 10NAMC

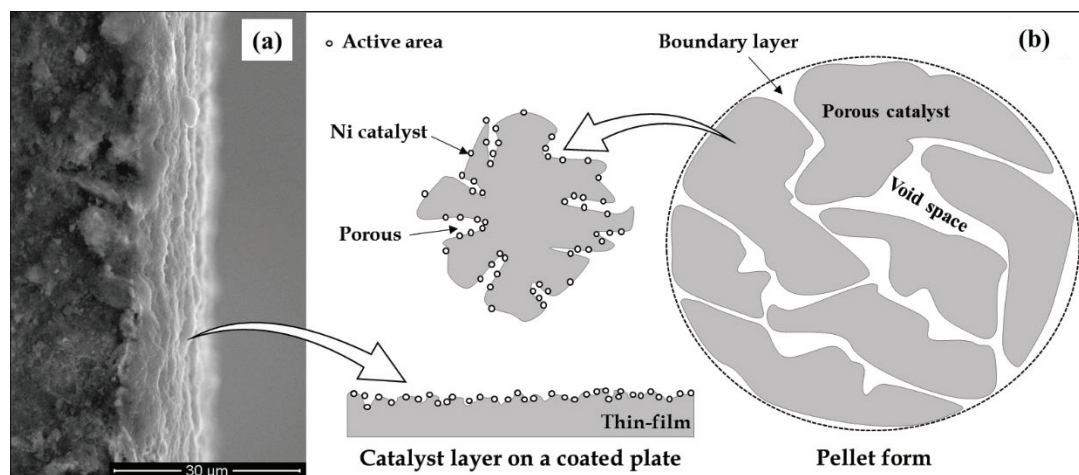


Fig. 4. (a) SEM micrographs for the thickness of 10NAMC, (b) schematic of porous pellet catalyst and thin-film catalyst

#### 4. Conclusion

The layer of 10NAM catalyst was fabricated on the stainless steel plate using spray coating using our condition. It is sufficiently thin to convey the metal active area onto the surface of thin-film. With this form, the catalyst activity was ten times higher than the pellet form and the carbon formation was also prevented effectively due to the improvement of heat and mass transfer performance. Consequently, this catalyst-coated plate can be developed to a coated-wall multichannel reactor.

#### Acknowledgements

The authors gratefully acknowledge PTT Public Company Limited, Thailand for the financial support, Department of Industrial Chemistry and Research and Development Center for Chemical Engineering Unit Operation and Catalyst Design (RCC) for supporting this research work.

#### References

- [1] S. Tungkamani, M. Phongaksorn, P. Narataruksa, T. Sornchamni, N. Kanja-nabat, N. Siri-Nguan, "Developing carbon tolerance catalyst for dry methane reforming", *Chemical Engineering Transition*. **32**, 2013, pp. 745-750.
- [2] X. Lv, J. F. Chen, Y. Tan, Y. Zhang, "A highly dispersed nickel supported catalyst for dry reforming of methane", *Catalysis Communications*, **20**, 2012, pp. 6-11.
- [3] A. R. McFarlane, I. P. Silverwood, R. Warringham, E. L. Norris, R. M. Ormerod, C. D. Frost, D. Lennon, "The application of inelastic neutron scattering to investigate the 'dry' reforming of methane over an alumina-supported nickel catalyst operating under conditions where filamentous carbon formation is prevalent", *RSC Advances*. **3**, 2013, pp. 16577-16589.
- [4] J. J. Lerou, A. L. Tonkovich, L. Silva, S. Perry, J. McDaniel, "Microchannel reactor architecture enables greener processes", *Chemical Engineering Science*. **65**, 2010, pp. 380-385.
- [5] C. W. Liu, H. S. Ko, C. Gau, "Microchannel Heat Transfer: Heat Transfer - Theoretical Analysis, Experimental Investigations and Industrial Systems, Prof. Aziz Belmiloudi (Ed.)", *In Tech*, 2011.
- [6] A.K. Avci, D.L. Trimm, M. Karakaya, "Microreactor catalytic combustion for chemicals processing", *Catalysis Today*. **155**, 2010, pp. 66-74.
- [7] J. J. Lerou, A. L. Tonkovich, L. Silva, S. Perry, J. McDaniel, "Microchannel reactor architecture enables greener processes", *Chemical Engineering Science*. **65**, 2010, pp. 380-385.

- [8] V. Hessel, A. Renken, J. Schouten, J. Yoshida “Micro Process Engineering: A Comprehensive Handbook 3 Volume Set”, *Wiley-VCH: Weinheim, Germany*, 2009.
- [9] S. W. Kim, I. Kim, D. J. Moon, “Synthesis and Characterization of Magnesium Oxide Supported Catalysts with a Meso-Macropore Structure”, *Journal of nanoscience and nanotechnology*. **13**, 2013, pp. 5750-5753.
- [10] Y. H. Wang, H. M. Liu, B. Q. Xu, “Durable Ni/MgO catalysts for CO<sub>2</sub> reforming of methane: activity and metal–support Interaction”, *Journal of Molecular Catalysis A: Chemical*. **299**, 2009, pp. 44-52.