## Direct Synthesis of Liquefied Petroleum Gas from Syngas over H-ZSM-5 Enwrapped Pd-based Zeolite Capsule Catalysts

<u>Peipei Zhang<sup>1</sup></u>, Guohui Yang<sup>1</sup>, Yoshiharu Yoneyama<sup>1</sup>, Ruiqin Yang<sup>2</sup>, Noritatsu Tsubaki<sup>1,2,\*</sup>

 <sup>1</sup>Department of Applied Chemistry, School of Engineering, University of Toyama, Toyama, Japan
 <sup>2</sup> Zhejiang Provincial Key Lab for Chem. & Bio. Processing Technology of Farm Product, School of Biological and Chemical Engineering, Zhejiang University of Science and Technology, China

## \*E-mail: tsubaki@eng.u-toyama.ac.jp

Currently, the global energy crisis and environmental contamination issues are greatly concerned all over the world. [1] Liquefied petroleum gas (LPG) is widely regarded as a promising candidate for fuel in household and industry fields since it is clean and renewable with advantages of high octane number of energy demand and easy storage. [2] LPG is mainly obtained during the exploitation of natural gas and crude oil from the earth or produced from refining petroleum, which both processes are considered as a by-product. Another progress for LPG synthesis is from syngas over a type of metal-zeolite hybrid catalyst, which highly attracted attention in recent years. [3, 4] In this report, different with the general hybrid catalyst, we want to design a core-shell-like zeolite capsule catalyst to realize LPG direct synthesis from syngas.

A facile synthesis route, named dual-layer crystal growth method, was utilized for preparing Pd-based zeolite capsule catalysts. The micrometer-sized zeolite capsule catalysts (Pd/SiO<sub>2</sub>-SZ), with a core-shell structure, were prepared by this dual-layer crystal growth method: coating Silicalite-1 and H-ZSM-5 zeolite orderly as double shell to encapsulate Pd/SiO<sub>2</sub> core. Fig. 1 exhibited the cross-section SEM image of the prepared Pd/SiO<sub>2</sub>-SZ zeolite capsule catalyst. We employed the prepared Pd/SiO<sub>2</sub>-SZ zeolite capsule catalyst for the direct synthesis of LPG from syngas.

This zeolite capsule catalyst showed excellent LPG selectivity than the single core catalysts Pd/SiO<sub>2</sub> or the physical mixture catalysts prepared by simply mixing core and shell catalysts. The Pd/SiO<sub>2</sub>-SZ realized the highest LPG selectivity of 34.4 % with CO conversion of 14.1 %. Moreover, the Pd/SiO<sub>2</sub>-SZ catalyst exhibited stable activity during 100 h time on stream reaction, as given by Fig. 2.



Fig.1 The cross-section SEM image of zeolite capsule catalyst Pd/SiO<sub>2</sub>-SZ.



Fig. 2 Time on stream result of Pd/SiO<sub>2</sub>-SZ on CO conversion and LPG selectivity.

## REFERENCES

[1] P. Corbo and F. Migliardini, Int. J. Hydrogen Energy., 32 (2007) 55.

[2] S. Takenaka, K. Kawashima, H. Matsune and M. Kishida, Appl. Catal. A, 321 (2007) 165.

[3] Q.W. Zhang, X.H. Li, K. Asami, S. Asaoka, K. Fujimoto, Fuel. Process. Technol., 85 (2004) 1139.
[4] X.G. Ma, Q.J. Ge, J.G. Ma, H.Y. Xu, Fuel. Process. Technol., 109 (2013) 1.