

# Improved dehydrogenation activity and catalyst stability of Pd/carbon-coated Al<sub>2</sub>O<sub>3</sub>

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In recent years, liquid organic hydrogen carrier (LOHC) has been attracting much attention because it offers a viable hydrogen storage system [1]. LOHC selected in this study is perhydropyridinyltoluene that has the H<sub>2</sub> content of 6.2 wt% and 55 kg<sub>H<sub>2</sub></sub> m<sup>-3</sup> [2]. To apply for automobile, the three factors are essential, such as (1) a relatively low temperature for dehydrogenation, (2) repeated H<sub>2</sub> storage and discharge of LOHC, and (3) catalyst stability. In this respect, much effort would be made on catalytic dehydrogenation.

In the dehydrogenation reaction, the common support for metal loading is Al<sub>2</sub>O<sub>3</sub> because of thermal stability, physical strength and recoverability [3]. However, metal dispersion is not high on alumina, resulting in low dehydrogenation activities of Al<sub>2</sub>O<sub>3</sub>-supported catalysts. In the literature, carbon is a preferable support rather than alumina because of high surface area and, in turn, good metal dispersion. However, the catalytic activity decreases sharply since metal particles are aggregated at the carbon surface by weak metal-support interaction. Thus, we have tried combining the advantages of alumina and carbon; that is, carbon-coated alumina (CCA) is used as a support. In this study CCA was prepared by contacting alumina with a glycerol solution of 0, 1, 2 or 10 mol%, and then pyrolyzing in a tube furnace at 600 °C under N<sub>2</sub> [4]. The coke content in CCA supports was determined by EA and TGA.

Fig. 1 shows a faster dehydrogenation rate of Pd/CCA than Pd/C and Pd/Al<sub>2</sub>O<sub>3</sub> when the reaction temperature increased up to 250 °C. Additionally, the catalytic activities of Pd/C, Pd/Al<sub>2</sub>O<sub>3</sub> and Pd/CCA were measured in recycle tests (Fig. 2). In the first experiment, the H<sub>2</sub> efficiency of Pd/CCA (60.2%) was

slightly higher than that of Pd/C (58.4%) and much higher than that of Pd/Al<sub>2</sub>O<sub>3</sub> (49.5%). This indicates the positive effect of carbon coating. The third recycle experiment clearly demonstrated another beneficial effect of carbon coating because the activities of Pd/C and Pd/Al<sub>2</sub>O<sub>3</sub> were more declined compared to Pd/CCA. The activity change is in good agreement with Pd dispersion of the tested catalysts. Since this effect is believed to be associated with increase of the interaction between Pd metal and support surface, Pd/CCA catalyst has been characterized by TEM, H<sub>2</sub>-TPR and CO chemisorption.

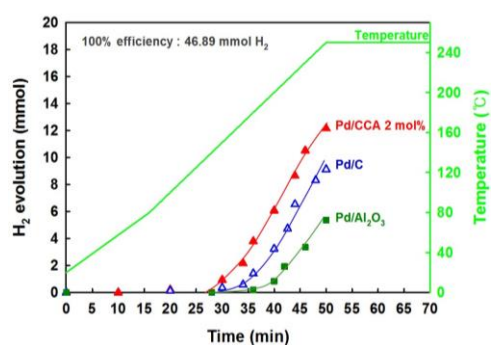


Fig. 1 Activities of Pd/CCA (2 mol% C), Pd/C and Pd/Al<sub>2</sub>O<sub>3</sub>. Reaction condition: Pd/reactant = 0.4 mol%, 50 min to 250 °C.

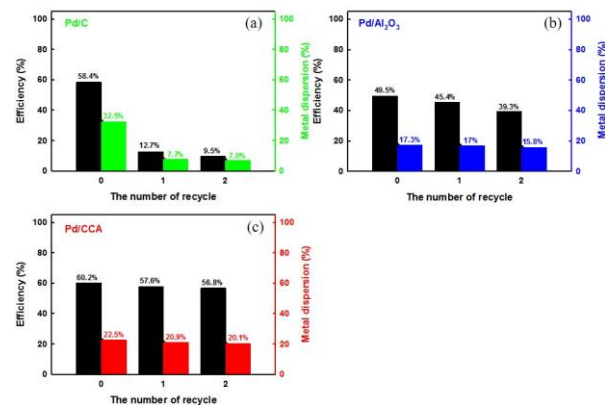


Fig. 2 H<sub>2</sub> efficiency and Pd dispersion of (a) Pd/C, (b) Pd/Al<sub>2</sub>O<sub>3</sub> and (c) Pd/CCA (2 mol% C) in repeated dehydrogenation tests. Reaction condition: Pd/reactant = 0.4 mol%, 4 h, 250 °C.

## REFERENCES

- [1] M. Momirlan, T.N. Veziroglu, *Renew. Sust. Energ. Rev.*, 6 (2002) 141.
- [2] M. Markiewicz, Y.Q. Zhang, A. Bosmann, N. Bruckner, J. Thoming, P. Wasserscheid, S. Stolte, *Energ. Environ. Sci.*, 8 (2015) 1035.
- [3] F. Rodríguez-Reinoso, *Carbon* 36 (1998) 159.
- [4] P.A. Nikulshin, V.A. Salnikov, A.V. Mozhaev, P.P. Minaev, V.M. Kogan, A.A. Pimerzin, *J. Catal.*, 309 (2014) 386-396.