Designed Pd nanoparticles for direct synthesis of hydrogen peroxide

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Hydrogen peroxide (H₂O₂) has been used in various chemical industries such as bleaching agent of pulp, paper and raw material for producing chemicals etc. H₂O₂ can be synthesized by direct synthesis process from H₂ and O₂, and the palladium (Pd) is widely used active metal. In reaction mechanism of direct H₂O₂ synthesis, decomposition of O₂ and H₂O₂ should be inhibited to increase H₂O₂ selectivity.

Density functional theory (DFT) study has allowed deeper understanding of palladium’s excellent activity in correlation with the physical structure and electronic state and serve as a guide for optimized design of the catalyst. Therefore, optimal catalysts of direct H₂O₂ synthesis could be synthesized as a combined result of DFT predictions and experiments.

According to the DFT results, {100} facet-enclosed cubic Pd has lower O₂ decomposition energy barrier than {111} facet-enclosed octahedral Pd [1]. Cube- and octahedron-shaped Pd catalysts were synthesized and applied in direct synthesis of H₂O₂, and Pd octahedral catalyst yielded higher H₂O₂ productivity than Pd cubic catalyst [2]. In addition, size-controlled Pd octahedral catalysts were synthesized to reduce the energetic O₂ dissociative corner & edge sites and to increase less energetic terrace sites. The largest 18-nm-sized catalysts among the tested Pd octahedral nanoparticles of varying sizes showed the highest productivity owing to the highest proportion of the terrace sites [3]. In addition, bimetallic catalysts are being investigated to further increase the activity. Galvanic-replaced Pt-doped Pd octahedral catalysts were examined on H₂O₂ direct synthesis to determine synergistic effect. Also, cubic Pd core-Au shell catalysts were synthesized to study strain effects.

However, the shape- and size-controlled Pd nanoparticles (NPs) were physically detached (55-77%) from support after reaction [2]. Core- and yolk-shell structures were examined as an alternative to prevent the loss of Pd particles. Both structures significantly reduced the loss of NPs (below 5%) during the reaction. Especially, the catalytic activity of yolk-shell structured Pd catalyst was superior because of higher exposed Pd surface area [4]. The micropore channels of the shell, however, limited diffusion of reactant gas molecules and compromised productivity. Therefore, grafted Pd nanoparticles on mesoporous SiO₂ shell (SiO₂@Pd@m-SiO₂) were synthesized to enhance mass diffusion through the pores. Consequently, SiO₂@Pd@m-SiO₂ showed the highest productivity (1090 mmol/gPd h) among applied catalysts for direct H₂O₂ synthesis [5].

REFERENCES