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学位論文内容の要旨

博士の専攻分野の名称 博士（工学） 氏名 邢 飛龍

学位論文題名

Developing a Highly Efficient Multimetallic Catalyst for CO₂-assisted oxidative dehydrogenation and dry reforming of propane

(CO₂ によるプロパンの酸化脱水素および改質反応に有効な多元素合金触媒の開発)

Attempts have long been made to improve the performance of metal catalysts by alloying, but it is difficult to achieve performance improvements by simply combining two metals. Therefore, fundamental innovation in catalyst design guidelines is necessary to develop high-performance catalysts that can meet the growing needs of society. In this regard, two approaches have been used: (i) pseudo-binary alloy, which is ternary alloys represented as A_m(B_{1-x}C_x)_n, where one metal site of the intermetallic A_mB_n is partially replaced by third metal C, and (ii) High-entropy intermetallics (HEIs), which are multi-metallic alloys with five or more elements whose specific crystal structure is derived from the parent binary intermetallics.

Chapter 2 showed that Pt–Co–In ternary alloy on CeO₂ exhibited a highly efficient catalyst for CO₂-ODP. Both the C₃H₈ dehydrogenation and CO₂ reduction smoothly proceed on the Pt–Co active site to form 2H and O, as well as the main products: C₃H₆ and CO. Alloying with In enhances the C₃H₆ selectivity due to the ensemble effect and decreases the amount of coke that accumulates on the catalyst surface. The doped Co improves the adsorption strengths of the C and H species, which promotes the activation of C₃H₈ and CO₂. The O atoms oxidize the eliminated H to form H₂O or combust coke on the alloy surface or neighboring CeO₂ sites. Moreover, CeO₂ can facilitate coke combustion via a Mars-van Krevelen-type mechanism, which significantly enhances the coke resistance and catalyst stability. Pt–Co–In/CeO₂ exhibits outstanding catalytic activity, C₃H₆ selectivity, long-term stability, and CO₂ utilization efficiency, all of which are greater than typical values ever reported. In **Chapter 3**, I further designed and prepared a PtSn-based HEIs catalyst for higher coke resistant and thermal stability than those of Pt–Co–In/CeO₂. I focused on intermetallic PtSn and its Pt and Sn sites were partially substituted with Ni/Co and In/Ga, resulting in nanoparticles with a HEI structure of (PtCoNi)(SnInGa). The multi-metallization sufficiently isolates Pt atoms, preventing propylene decomposition and coke formation. The incorporation of Ni and Co significantly enhanced the ability to activate CO₂. Doping In(Ga) into the Sn site improved catalyst stability, probably due to the enhanced entropy effect. The combination of these abilities significantly enhances coke resistance and catalyst lifetime. Furthermore, because of the increased thermal stability, multi-metallization also prevents nanoparticle sintering. The HEI/CeO₂ catalyst shows much greater catalyst life and specific activity than Pt–Co–In/CeO₂. In **Chapter 4**, I further applied the Pt–Co–In ternary alloy system to dry reforming of propane (DRP), which has attracted much attention as a syngas production technology for liquid fuels with a low H₂/CO ratio. The PtCo₂In₃/CeO₂ for the DRP reaction with high selectivity and stability even over 100 h. The results show that the catalytic behavior of these catalysts is highly sensitive to the amount of cobalt doping. By controlling the amount of cobalt doping, the catalytic activity of PtCo₂In₃/CeO₂ catalysts can be improved, and the C–H/C–C bond of propane can be selectively cleaved in the presence of CO₂.

In summary, the study in this thesis not only provides excellent catalytic performances in conversion of light alkane and CO₂, but also opens a new window for the design of multi-metallic catalysts for challenging molecular conversions.