

研 究 主 論 文 抄 録

論文題目 Synthesis of Precious Metal Catalysts Supported on Porous Oxides and Phosphates
 for NO_x Reduction
 (多孔性酸化物およびリン酸塩に担持した NO_x 還元用貴金属触媒の合成)

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主論文要旨

Emission of nitrogen oxides (NO_x) from stationary and automobile sources is extremely deleterious to the environment and represents a serious hazard to the health. The most efficient solution for the issue is to use catalysts that can reduce NO_x to N₂ in the presence of H₂, CO and/or hydrocarbons. Catalytic NO_x reduction use precious metals including Pt, Pd and Rh as active components. Because of rapidly increasing use as catalysts in automobile exhaust gas after-treatment, these are currently among the scarcest of precious metals that need a substantial thrift. In this regard, material chemistry to design support materials that can stabilize precious metal nanoparticles is strongly requested in response to such a serious situation. The combination of low-temperature catalytic activity, high-temperature thermal stability, and tolerance to poisoning of catalysts with minimum threshold loading of precious metals can be broadly extended to the practical high-temperature applications in the area of environmental protection. In the present work, various porous metal oxides and phosphates have been studied in connection with their use as support materials for precious metals.

In Chapter 1, the background of this study is discussed. Air pollution phenomena concerning NO_x related to fossil fuel combustion are discussed elaborately. Specifically hassles related to NO_x emission, sources and potentialities for their control are highlighted. After a brief introduction, environmental and health hazards related to this NO_x pollution are presented. Subsequently the history, the regulations and the emission trends are summarized. To find out the suitable remedial measure, the approaches for NO_x removal are thoroughly reviewed. Finally the state of the art with respect to the catalytic solutions to the problem is discussed with emphasis on the precious metal supported catalytic reduction of NO_x.

In Chapter 2, catalytic NO-H₂-CO-O₂ reactions over Pt-supported mesoporous yttrium oxide are described. Catalytic light-off of a stream of NO, H₂, CO in an excess O₂ has been studied over various metal oxides loading Pt. Because a low-surface area Y₂O₃ (<5 m² g⁻¹) was found to exhibit the highest de-NO_x activity, a mesoporous Y₂O₃ was then synthesized from an yttrium-based mesophase templated by dodecyl sulfate (DS = C₁₂H₂₅OSO₃⁻), which was anion-exchanged by acetate (ACO = CH₃COO⁻). The product showed a 3D mesoporosity with a large surface area (~400 m² g⁻¹) and the Pt-supported catalyst achieved much improved light-off characteristics suitable for the low-temperature de-NO_x in the presence of CO and excess O₂.

In Chapter 3, NO-H₂-CO-O₂ reactions over Pt catalysts supported on Ln-incorporated FSM-16 (Ln=La, Ce and Pr) have been investigated. Ln incorporated into FSM-16 (Ln = La, Ce and Pr) materials, which were synthesized by a modified sheet folding process of kanemite-CTAB system. The Ln-FSM-16 products showed a mesoporosity with the specific surface areas of 700-750 m² g⁻¹, which were less than 1100 m² g⁻¹ of the pristine FSM-16 while pore size and pore volume remained unchanged. Local structure of Ln and porous structure of Ln-FSM-16 were found thermostable up to 800 °C. Among the series of Ln-FSM-16, Pt loaded on Pr-FSM-16 exhibited the highest catalytic activity for NO reduction by H₂ at the low temperature (≤ 200 °C) in presence of CO and excess O₂.

In Chapter 4, Catalytic NO-CO-C₃H₆-O₂ reactions over Rh catalysts supported on hydrothermally synthesized ZrP₂O₇ have been studied. A novel and facile hydrothermal synthetic route for ZrP₂O₇ from ZrO(OH)₂ and H₃PO₄ has been developed to use this as an active support for Rh catalyst. Temperature-programmed reactions of mixtures of NO, CO, C₃H₆ and O₂ with a stoichiometric air-to-fuel (A/F) ratio were investigated over the supported Rh catalyst after thermal ageing at 900 °C in the stream of 10% H₂O/air. It was found that Rh nanoparticles supported on ZrP₂O₇ were stable even after aging up to 1200 °C without serious deactivation.

In Chapter 5, the results concluded in the previous chapters are summarized briefly.