

DATE: May 1998

TITLE: IN SITU INFRARED STUDY OF CATALYTIC
DECOMPOSITION OF NO

PI: Steven S.C. Chuang

STUDENT(S): Mahesh Konduru (Ph.D. Candidate), Ram Krishnamurthy and Scott
A. Hedrick (MS Candidates)

INSTITUTION: The University of Akron
Department of Chemical Engineering
Akron, OH 44325-3906
(330) 972-6993

GRANT NO.: DE-FG22-95PC95224

PERIOD OF PERFORMANCE: August 1, 1995 to July 31, 1998

I. ABSTRACT

OBJECTIVE: The objective of this project is to study the reactivity of adsorbates for the direct NO decomposition and to investigate the feasibility of two novel approaches for improving catalyst activity and resistance to sintering. The first approach is the use of silanation to stabilize metal crystallites and supports for Cu-ZSM-5 and promoted Pt catalysts; the second is utilization of oxygen spillover and desorption to enhance NO decomposition activity. An innovative infrared reactor system is used to observe and determine the dynamic behavior and reactivity of adsorbates during NO decomposition, oxygen spillover, and silanation. The information obtained from this study will provide a scientific basis for developing an effective catalyst for the NO decomposition under practical flue gas conditions.

WORK DONE AND CONCLUSIONS:

- Addition of Tb oxide to Pt/Al₂O₃ allows oxygen from dissociated NO to desorb at 593 K, which is significantly lower than the reported oxygen desorption temperatures for Pt catalysts. The Tb-promoted Pt/Al₂O₃ catalyst which possesses oxygen desorption capability at low temperatures shows activity for decomposition of NO to N₂, N₂O, NO₂, and O₂ under pulse and steady-state reaction conditions at 773-923 K.
- Pd/C exhibited activity towards NO decomposition with N₂, N₂O and O₂ being the main products in the temperature range of 573 K- 773 K. Rh/C showed prolonged activity for NO decomposition to produce N₂, N₂O and CO₂.

SIGNIFICANCE TO FOSSIL ENERGY PROGRAM: Pt/Al₂O₃ based catalysts have shown excellent hydrothermal stability. Promotion of oxygen desorption and spillover holds the key to enhance the activity of Pt/Al₂O₃ for the decomposition of NO to N₂ and O₂. Results of this study provide a better understanding of the NO dissociation and

oxygen desorption mechanism which may serve as a guide for the development of highly active catalysts for NO mechanism. Development of an effective catalyst for the direct decomposition of NO will eliminate the use of reducing reactants, such as H₂, CO, NH₃, and hydrocarbons, significantly simplifying the NO removal process, dramatically decreasing the cost of control of NO emissions from coal-fire power plants.

PLANS FOR THE COMING YEAR:

- Development of catalysts which exhibit high activity under practical flue gas condition.

II. HIGHLIGHT ACCOMPLISHMENTS

- Determination of the role of Tb on the promotion of oxygen desorption from Pt/Al₂O₃
- Determination of the effect of dispersion and partial pressure of reactants on the light-off characteristic of Rh and Pt catalysts
- Identification of activated adsorbate for NO decomposition on Cu-ZSM-5
- Silanation of Cu-ZSM-5 catalyst.

III. ARTICLES AND PRESENTATIONS

“Reactivity of Adsorbates in the Decomposition of Nitric Oxide over Cu-ZSM-5 Catalysts,” S.S.C. Chuang and B. Lopez, Progress in Zeolite and Microporous Materials, The Proceedings of 11th International Zeolite Conferences, 1477-1484, H. Chon, S.-K. Ihm, and Y.S. Uh, Eds. Elsevier, Amsterdam-Oxford-New York-Tokyo, 1997.

"Combined Infrared and Mass Spectrometric Study of NO-CO Adsorption and Reaction on 0.5 wt% Rh/SiO₂ Catalyst," S.S.C. Chuang and C.-D. Tan, Catalysis Today. 35, 369-377, 1997.

"Promotion of Oxygen Desorption to Enhance Direct NO Decomposition over Tb-Pt/Al₂O₃ Catalyst," S.S.C. Chuang and C.-D. Tan, J. of Physical Chemistry, 101(15), 3000-3004, 1997.

“Manganese- and Silane-Modification of Rh Surface states and Adsorbates for NO-CO Reaction,” C.-D. Tan and S.S.C. Chuang, Journal of Catalysis, Submitted.