

Title: DEVELOPMENT OF A NOVEL CATALYST FOR NO DECOMPOSITION

Authors: Ates Akyurtlu and Jale F. Akyurtlu
Hampton University, Chemical Engineering Department,
Hampton, VA 23668

Phone: 757-727-5818, Fax: 757-727-5189

e-mail: ates.akyurtlu@hamptonu.edu

Industrial Collaborator: AirFlow Catalyst Systems, Inc.

Grant Number: DE-FG26-03NT41911

Performance Period: March 15, 2004 to March 14, 2005

ABSTRACT

Objective: The main objective of the proposed research is the evaluation of the Pt/SnO₂ catalysts for the decomposition of NO in simulated power plant stack gases with particular attention to the resistance to deactivation by O₂, CO₂, and elevated temperatures.

Accomplishments to Date: In this reporting period temperature programmed desorption (TPD) and temperature programmed reaction (TPRx) runs (using NO+He and NO+O₂+He reactant gas mixtures) on the 15% Pt and 10% Pt catalysts were performed. The following conclusions summarize the observations during TPD runs with both catalysts:

- During pulse chemisorption, about half of NO is either dissociated into N₂ and O₂ or retained on the catalyst at a steady rate. Based on the N₂/O₂ ratio, the latter is deemed a more probable explanation.
- During TPD, the N₂, N₂O, and NO peaks were separated by about 25 K. There were two O₂ minima, one corresponding to the N₂O peak and the other to the termination of NO peak.
- The observed NO peak could be due to the presence of free surface nitrite or nitrate species, but the presence of N₂O₄ and N₂O₃ is more likely.
- The 25 K separation between the N₂ and N₂O peaks suggests that these two species are produced, at least partially, by different surface reaction schemes.
- During the TPD after successive adsorptions of NO and O₂, the N₂O and NO peaks and the single O₂ minima were all observed around 490 K suggesting that in the presence of excess O₂, both N₂O and NO were produced by parallel surface reactions.
- Presence of excess oxygen increased the production of N₂O.

During the TPRx of NO on the catalyst samples

- There was no NO decomposition below 520 K, but some N₂O formation above 370 K, which peaked at 520 K, but no N₂O was present above 750 K. NO desorption started at 520 K and increased with temperature at almost constant

- rate.
- The N_2O formation on the used catalysts was significantly smaller than that on the fresh catalyst.
 - The start of NO decomposition was pushed to 625 K during subsequent runs with the same catalyst, but the rate was faster so that the NO concentration in the gas at 900 K is the same for fresh and used catalysts, which corresponds roughly to 50% conversion of NO on the catalyst with 15% Pt.

The following results were obtained for the TPRx of $\text{NO} + \text{O}_2$

- The presence of oxygen inhibits the decomposition of NO and promotes the formation of N_2O . Some decomposition activity starts around 425 K on fresh catalyst. During the second run NO decomposition does not start until 575 K, but is faster than that on the fresh catalyst. More N_2O formation was observed during the second run.
- When the feed O_2/NO ratio was cut by one half, the N_2O production on the fresh catalyst was significantly reduced, but this improvement did not continue during the second run. The reduction in the feed oxygen content also appeared to improve the NO decomposition activity.
- During the second run there was no NO decomposition activity up to about 550 K. At higher temperatures the NO decomposition rate increased and this increase was similar to that observed with the feed containing no added oxygen.

The final conversion levels were higher on the 15% Pt catalysts compared to those on the 10% Pt catalysts.

To explain the observed results, two different NO decomposition processes, one occurring on the Pt sites and the other taking place at the Pt- SnO_2 interface and involving the lattice oxygen of SnO_2 and the NO adsorbed on the Pt surface are proposed. The second mechanism involves the formation of surface nitrite/nitrate species as intermediates. There was strong indication of irreversible reduction of SnO_2 upon heating above 600 K under a reactant gas containing no added oxygen.

Future Work:

1. Calculation of the chemical equilibrium for the $\text{N}_2 + \text{O}_2$ system.
2. Testing of the 15% Pt catalyst in the laboratory scale packed bed reactor to determine the optimum reactor temperature and obtain conversion-selectivity data.

Publications:

1. Akyurtlu, J. F., Akyurtlu, A., and Bridges, W. J., "Development of a novel Catalyst for NO Decomposition", presented at the 2004 AIChE Annual Meeting, Austin, TX, Nov. 7-12, 2004.
2. Ates Akyurtlu and Jale F. Akyurtlu, "Pt/ SnO_2 Catalysts for NO Decomposition", to be presented at the 19th North American Catalysis Society meeting, Philadelphia, PA, May 22-27, 2005.