July 1997

Catalytic Ammonia Decomposition for Coal-Derived Fuel Gases

Topical Report

Work performed under
Contract No. DE-AC21-92MC29011

for
U.S. Department of Energy
Federal Energy Technology Center
3610 Collins Ferry Road
Morgantown, WV 26505

by
S.K. Gangwal
R. Gupta
J.W. Portzer
B.S. Turk
Research Triangle Institute

Catalytic Ammonia Decomposition for Coal-Derived Fuel Gases

Topical Report

Work performed under
Contract No. DE-AC21-92MC29011

for
U.S. Department of Energy
Federal Energy Technology Center
3610 Collins Ferry Road
Morgantown, WV 26505

by
S.K. Gangwal
R. Gupta
J.W. Portzer
B.S.Turk
Research Triangle Insitute
3040 Cornwallis Road
Research Triangle Park, NC 27709

July 1997

ABSTRACT

When coal is gasified, fuel bound nitrogen compounds are principally released as ammonia (NH_3) which would readily oxidize to NO_x during combustion of the fuel gas in the turbine. The objective of this project, a joint effort between Research Triangle Institute, SRI International, and General Electric, is to develop catalytic approaches for decomposing a significant percentage (up to 90 percent) of the NH_3 present in fuel gas to N_2 and H_2 at elevated temperatures (550 to 900 °C) and pressures.

Two catalytic approaches for decomposing NH_3 have been experimentally evaluated. The first approach, evaluated during the early phases of this project, involved the screening of catalysts that could be combined with hot-gas desulfurization sorbents (e.g., zinc titanate) for simultaneous NH_3 and hydrogen sulfide (H_2S) removal. In a commercial system, this approach would reduce capital costs by eliminating a process step. Numerous catalysts and catalyst-sorbent combinations were prepared and tested for their NH_3 decomposition activity. These included catalysts based on Ni, Co, Mo, and W in various proportions combined with zinc titanate, titania, or refractory supports. Although some of the catalysts tested had high initial decomposition activity, they quickly deactivated in the presence of 2,000 ppmv H_2S and achieved only 10 to 40 percent NH_3 decomposition at 700 to 750 °C and 1 to 15 atm. Thus, this approach was deemphasized in favor of a stand-alone high-temperature (800 to 900 °C) catalytic decomposition approach.

Four nickel-based catalysts were selected for evaluation as high-temperature NH_3 decomposition catalysts. These included HTSR-1, G-47 (3 percent Fe_2O_3 on silica) from United Catalysts, Inc. (UCI), C11-9 (12 percent Ni on a ceramic support) from UCI, and C100N (proprietary Ni on stabilized alumina) from Contract Materials Processing, Inc. (CMP). Screening tests were conducted with these catalysts at 850 °C, 10,000 std $cm^3/(cm^3 \cdot h)$ in the presence of Texaco coal gas containing 1,800 ppmv NH_3 and 7,500 ppmv H_2S. The HTSR-1 demonstrated a steady 90 percent ammonia decomposition while the other catalysts showed <50 percent decomposition at equivalent test conditions. Based on these screening tests, HTSR-1 was selected for long-term (100-h) durability testing.

Two 100-h tests were conducted using HTSR-1. The first test was conducted using a mobile reactor facility at U.S. Department of Energy/Federal Energy Technology Center (DOE/FETC) with a hot slipstream of actual coal gas from the FETC 10-in. diameter fluidized-bed coal gasifier. The 3-in. diameter NH_3 decomposition reactor in the mobile reactor facility was operated at 11 atm, 780 °C, and 5,000 std $cm^3/(cm^3 \cdot h)$. The FETC coal gas had average NH_3 and H_2S concentrations of 3,000 ppmv and 1,300, respectively. Over the 100-h test duration, NH_3 decomposition averaged 90 percent with no catalyst deactivation apparent due to trace contaminants in coal gas. The second test was conducted using a simulated Texaco coal gas nominally containing 7,500 ppmv H_2S and 1,800 ppmv NH_3 at 8.5 atm, 900 °C, and 5,000 to 10,000 std $cm^3/(cm^3 \cdot h)$. The catalyst showed a small amount of deactivation over the first 40 h of operation and then a stable catalytic activity level was achieved. The average NH_3 decomposition was 80 percent at 10,000 std $cm^3/(cm^3 \cdot h)$ and 88 percent at 5,000 std $cm^3/(cm^3 \cdot h)$.

TABLE OF CONTENTS

Section **Page**

TABLE OF CONTENTS (continued)

Appendices

LIST OF FIGURES

LIST OF TABLES