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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
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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 $\text{cm}^3/(\text{cm}^3\cdot\text{h})$ 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 $\text{cm}^3/(\text{cm}^3\cdot\text{h})$. 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 $\text{cm}^3/(\text{cm}^3\cdot\text{h})$. 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 $\text{cm}^3/(\text{cm}^3\cdot\text{h})$ and 88 percent at 5,000 std $\text{cm}^3/(\text{cm}^3\cdot\text{h})$.

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