165a Oxidation of No and Co over Cobalt Based Metal Oxide Supported Catalysts

Matthew M. Yung, Erik M. Holmgreen, and Umit S. Ozkan

Combustion processes provide the bulk of the world's energy demands, but the emission of CO and NOx associated with these processes pose environmental concerns. In order to address these pollutants, a series of cobalt-based oxidation catalysts supported on zirconia and titania were synthesized and found to be effective for oxidation of NO and CO. NO oxidation has been examined as part of a two-stage lean NOx reduction system. NO2 is a more easily reduced species than NO, and additionally has been proposed as a reaction intermediate in the selective catalytic reduction of NO. It was thus proposed to develop a highly active NO oxidation catalyst. The synthesized cobalt-based catalysts were found to be active at low temperatures and showed high hydrothermal stability. Equilibrium conversions of NO to NO2 were reached at temperatures below 300°C. Additionally, tests of the oxidation catalyst as part of a mixed-bed that also contained a NO2 reduction catalyst have shown very promising results as a hydrocarbon-based NOx reduction system. The Co-based catalysts were additionally shown to be highly active for CO oxidation at relatively low temperatures. Complete oxidation of CO was observed at 135°C, and was steady over more than 180 hours on-stream.

Steady-state kinetic experiments were conducted to determine catalyst activity, and were done under both model and simulated lean exhaust conditions. Additionally, deactivation and regeneration steps were examined. Synthesis parameters including composition, synthesis technique, metal doping, and calcination temperature were explored. Structural and chemical characterization is performed using BET surface area measurements, in-situ X-ray diffraction (XRD), X-ray photoelectron spectroscopy (XPS), and thermogravimetric analysis combined with differential scanning calorimetry. Mechanistic studies by temperature programmed desorption (TPD), laser Raman spectroscopy (LRS), and diffuse reflectance infrared Fourier transform spectroscopy (DRIFTS) are also presented.