

## Transient Infrared Studies of NO Decomposition and Reduction with H<sub>2</sub> on Ag-Pd/Al<sub>2</sub>O<sub>3</sub>

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Catalytic decomposition of nitric oxide (NO) over Pd/Al<sub>2</sub>O<sub>3</sub> and Ag-Pd/Al<sub>2</sub>O<sub>3</sub> has been studied using the pulse transient response technique coupled with *in situ* infrared (IR) and mass spectrometry (MS) at 723-823 K. In the absence of H<sub>2</sub>, the pulsing NO reaction on the Pd/Al<sub>2</sub>O<sub>3</sub> catalyst produces adsorbed NO species (i.e., Pd<sup>+</sup>-NO, Pd<sup>0</sup>-NO, Pd-NO<sup>-</sup>) as well as gaseous N<sub>2</sub>, O<sub>2</sub>, and N<sub>2</sub>O products. Transient responses of the N<sub>2</sub> and O<sub>2</sub> profiles show that the addition of Ag onto Pd/Al<sub>2</sub>O<sub>3</sub> catalyst shifts the O<sub>2</sub> profile forward, increases oxygen formation and the oxidation resistance of Pd, but did not decrease the amount of retained oxygen (O<sub>ret</sub>) and did not improve the catalytic cycle for NO decomposition. O<sub>ret</sub> on the Pd surface is not able to desorb in the temperature range of this study; however, O<sub>ret</sub> on Ag-Pd/Al<sub>2</sub>O<sub>3</sub> can be desorbed at higher temperatures than its formation and adsorption temperature. The presence of H<sub>2</sub> during the NO pulse allowed NO reduction to occur, producing N<sub>2</sub>, N<sub>2</sub>O, O<sub>2</sub>, NH<sub>3</sub>, and H<sub>2</sub>O. Pd/Al<sub>2</sub>O<sub>3</sub> is a more active catalyst for the formation of NH<sub>3</sub> and H<sub>2</sub>O than Ag-Pd/Al<sub>2</sub>O<sub>3</sub>. Comparison of the transient gaseous product responses over Pd/Al<sub>2</sub>O<sub>3</sub> and Ag-Pd/Al<sub>2</sub>O<sub>3</sub> catalysts show that Ag (i) promotes the formation of N<sub>2</sub>, shifting its profile forward and (ii) suppresses the formation of NH<sub>3</sub> and H<sub>2</sub>O, delaying their formation. The lack of the initial activity of Ag-Pd/Al<sub>2</sub>O<sub>3</sub> for NH<sub>3</sub>/H<sub>2</sub>O formation can be attributed to the alloy state of Ag-Pd on Al<sub>2</sub>O<sub>3</sub>. As the NO reduction proceeds in the presence of H<sub>2</sub>, adsorbed oxygen produced from N-O dissociation could cause the dealloying of Ag-Pd, producing Pd sites, which exhibited high selectivity for NH<sub>3</sub>/H<sub>2</sub>O formation.

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