

## Evidence of WGS reaction: Direct H<sub>2</sub> involvement in HC-SCR DeNO<sub>x</sub>:

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**Introduction:** One class of catalysts that has shown promise for hydrocarbon selective catalytic reduction (HC-SCR) of NO<sub>x</sub> to N<sub>2</sub> is metal oxides supported on alumina<sup>1-3</sup>. However, metal oxide systems still suffer from the limitations of activity at low temperature compared to other catalyst DeNO<sub>x</sub> systems. Systemic studies by Hamada have demonstrated that platinum metals at lower temperature can show NO<sub>x</sub> reduction activity using such hydrocarbons as propene<sup>1</sup>. By combining the activity from precious metal systems with the selectivity of metal oxide systems, it is hoped that a catalyst can be developed that will meet both requirements. In an attempt to better understand the reaction pathway for a newly discovered precious metal/metal oxide DeNO<sub>x</sub> catalyst, a study using isotopic gases was conducted.

### Materials and Methods

Catalyst samples were prepared by incipient wetness impregnation techniques. Catalyst samples (~ 150 mg) were loaded into a fixed bed-reactor set at 250 °C. Reaction gases were introduced using mass flow controllers, while water was introduced with a temperature controlled saturator. The outlet gas stream was analyzed by a mass spectrometer (Thermo Prolab). Gas concentrations are 500 ppm NO, 1500 ppm CO, 3.5% H<sub>2</sub>O, 5% O<sub>2</sub>, 0-5000 ppm C<sub>3</sub>H<sub>6</sub>, balance He.

### Results and Discussion

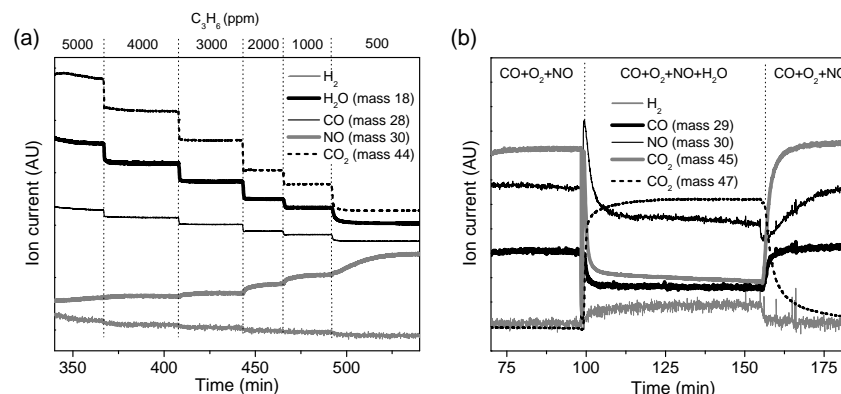
In initial investigations, we observed a large reduction in NO<sub>x</sub> concentration (~ 60 % conversion), when using propene (~ 500 to 5000 ppm) as a reductant gas in combination with carbon monoxide under steady state DeNO<sub>x</sub> conditions (Figure 1a). As expected, the addition of propene produced a mass signal of 18, which is indicative of the formation of water<sup>4</sup>. Additionally, the presence of hydrogen was also detected, corresponding to m/e = 2. The H<sub>2</sub> formation can be explained by the water produced by combustion reacting with carbon monoxide to make hydrogen via the water-gas-shift reaction. It is then necessary to determine if the NO reduction is due to additional hydrocarbon reductant, the addition of water or the generation of hydrogen. To help ascertain this, isotopic labeling studies were conducted.

In order to note the effect of water on NO<sub>x</sub> reduction directly, we added water into the gas stream by use of a saturator instead of *in situ* formation via hydrocarbon combustion. By using H<sub>2</sub><sup>18</sup>O, it allowed for CO<sub>2</sub> produced from WGS (mass 46) to

be distinguished from CO<sub>2</sub> produced from CO oxidation (mass 44). However, the since NO<sub>2</sub> also has a mass of 46, it was necessary to use <sup>13</sup>CO to further distinguish the WGS product CO<sub>2</sub> (mass 47).

Upon addition of water, the level of NO dropped (Figure 1b) as measured as mass 30 in the MS as well as the total NO<sub>x</sub> concentration on the NO<sub>x</sub> detector. This indicates that water is playing a role in the catalytic reaction to favor the NO<sub>x</sub> reduction pathway. More interesting, we see an increase in the level of tagged CO<sub>2</sub> (mass 47), which are formed from the isotopic constituents of WGS. The presence of H<sub>2</sub> is also noted consistent with WGS, as well. Conversely, the level of untagged CO<sub>2</sub> (mass 45) is reduced, indicating a reduction in CO combustion, which allows more CO to be available for NO<sub>x</sub> reduction.

**Conclusions:** With the proper tagging of key constituents involve in HC-NO<sub>x</sub> reduction, we see evidence of WGS as a side reaction occurring and being involved with NO<sub>x</sub> reduction. It is unclear if the decrease in NO<sub>x</sub> levels is due to H<sub>2</sub> formed by WGS, competition for CO, or competition for sites with water. Further labeling studies will be conducted to help further resolve aspects of the DeNO<sub>x</sub> mechanism for our catalyst.



### References

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