

Heavy-Duty Emissions Control: Plasma-Facilitated vs Reformer-Assisted Lean NO_x Catalysis

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Progress has been made in the control of combustion processes to limit the formation of environmentally harmful species, but lean burn vehicles, such as those powered by diesel engines used for the majority of commercial trucking and off-road applications, remain a major source of nitrogen oxides (NO_x) and particulate matter (PM) emissions. Tighter control of the combustion process coupled with exhaust gas recirculation has brought emissions in line with 2004 targets worldwide. Additional modifications to the engine control system, somewhat limited NO_x control, and PM filters will likely allow the 2007 limits to be met for the on-highway regulations for heavy-duty engines in the United States. Concern arises when the NO_x emission limit of 0.2 g/bhphr set for the year 2010 is considered.

One approach being examined is active lean NO_x catalysis [1,2] and modifications thereof. In active lean NO_x catalysis, hydrocarbons react with NO_x on the surface of a catalyst to produce dinitrogen and combustion and partial combustion products of hydrocarbons. It is preferred that these catalysts be active with fuel as the hydrocarbon, but diesel-fuel hydrocarbons, largely longer alkanes, are not as reactive as oxygenated hydrocarbons [3]. Therefore, variants on lean NO_x catalysis such as plasma-facilitated catalysis (PFC) [see *eg.*, 4-6] are being developed that allow conversion of hydrocarbons to oxygenated species to obtain higher NO_x conversion efficiencies. In traditional PFC, the exhaust and added hydrocarbons are treated in an electrical discharge prior to exposure to the lean NO_x catalyst. In this paper, we present a modified approach where the electrical discharge is decoupled from the exhaust gas. Therefore, only the hydrocarbon (*eg.*, fuel) and an oxidant (air) are passed through the plasma. In this arrangement, the plasma is acting as a fuel reformer to provide more active species for NO_x reduction, and the reformed mix is injected into the exhaust. A simple schematic of the approach, termed reformer-assisted catalysis (RAC), is shown in Figure 1. It should be stressed that any sort of reformer that can produce oxygenates or other active species for lean NO_x catalysis can be substituted for plasma in this arrangement.

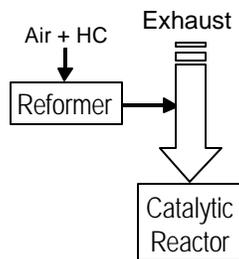


Figure 1. Cartoon showing layout for RAC.

Testing of the RAC system was performed and results were compared to data obtained using traditional PFC to assess whether the plasma could be decoupled from the exhaust and still maintain high NO_x reduction efficiency. Two catalysts were examined: (i) Ag/alumina, and (ii) Ba/zeolite Y

followed by Ag/alumina. The results are shown in Figure 2 where NO_x conversion measured via chemiluminescence is plotted versus catalyst temperature. Significant improvement in NO_x reduction was observed at lower temperature (< 350°C) in case (i), largely due to production of oxygenated hydrocarbons from the propene fed to the reformer. Above 350°C, the propene is reactive on the catalyst and reformation is not required to achieve high conversion. In case (ii), no observable benefit was achieved when adding Ba/zeolite Y to the system. In fact, a slight reduction in activity was observed, which is due to the impact of higher space velocity through the Ag/alumina catalyst [Ag/alumina is half the volume of case (i)]. In PFC, it has been reported [7] that the temperature range can be expanded by utilizing this two-catalyst system.

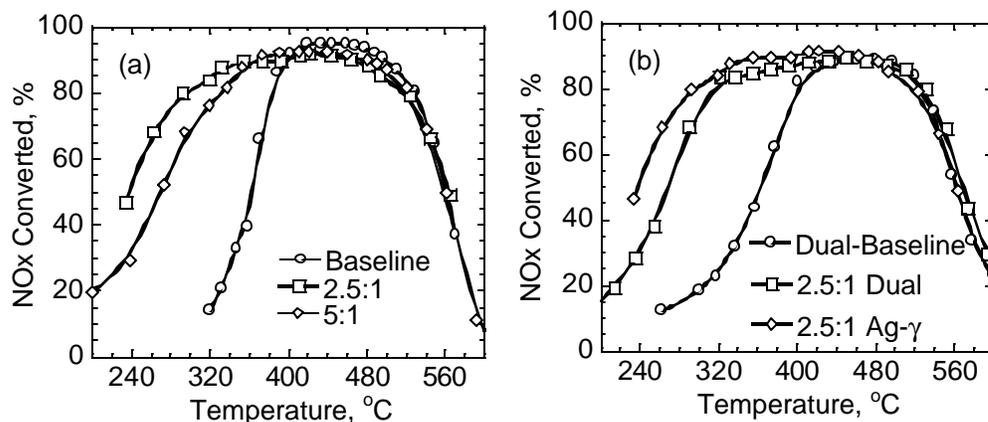


Figure 2. RAC results for (a) Ag/alumina, and (b) Ba/zeolite Y followed by Ag/alumina. 22000-28000 hr⁻¹ space velocity, C₁/NO_x = 12 was based on propene levels fed to the reformer. Exhaust composition was 9% O₂, 8% CO₂, 300 ppm CO, 1.5% H₂O, 500 ppm NO, and balance N₂. Reformer was operated at 500°C and 6 kJ/L of specific plasma energy.

Gas chromatography-mass spectrometry characterization of the reformer effluent showed less than 20% single pass conversion of propene to a variety of oxygenates including aldehydes, alcohols, carboxylic acids as well as other moieties. Notable was the lower level of C₂ and greater aldehydes with respect to PFC. This accounts for the ineffectiveness of Ba/zeolite Y with the RAC approach. Ba/zeolite Y requires C₂ or greater aldehydes to facilitate NO_x reduction. The Ag/alumina catalyst is much less selective in that a wide variety of oxygenates are utilized.

Reformer-assisted catalysis shows promise as an alternative to plasma-facilitated catalysis, particularly at exhaust temperatures for heavier diesel vehicles. Decoupling reformation of hydrocarbon from the exhaust itself will allow smaller and lower cost aftertreatment systems.

References

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