Emissions Control for Lean Gasoline Engines

Todd J. Toops, James E. Parks (PI), Josh A. Pihl, Shean Huff, Vitaly Prikhodko
Oak Ridge National Laboratory

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Gurpreet Singh and Ken Howden
Advanced Combustion Engines Program
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Project Overview

Timeline

- New project:
  - Combination of two related efforts
  - Refocused to address current DOE and industry needs
- Lean Gasoline: Started in FY10
- Fundamental Catalysis: Started in FY02

Barriers Addressed

- Barriers listed in VT Program Multi-Year Program Plan 2011-2015:
  - 2.3.1B: *Lack of cost-effective emission control*
  - 2.3.1C: *Lack of modeling capability for combustion and emission control*
  - 2.3.1.D: *Durability*

Budget

- Lean Gasoline
  - FY11: $200k
  - FY12: $400k
- Fundamental Catalysis
  - FY11: $200k
  - FY12: $0k

Collaborators & Partners

- Umicore
- General Motors
- University of South Carolina
- University of Wisconsin
- Cross-Cut Lean Exhaust Emissions Reduction Simulations (CLEERS)

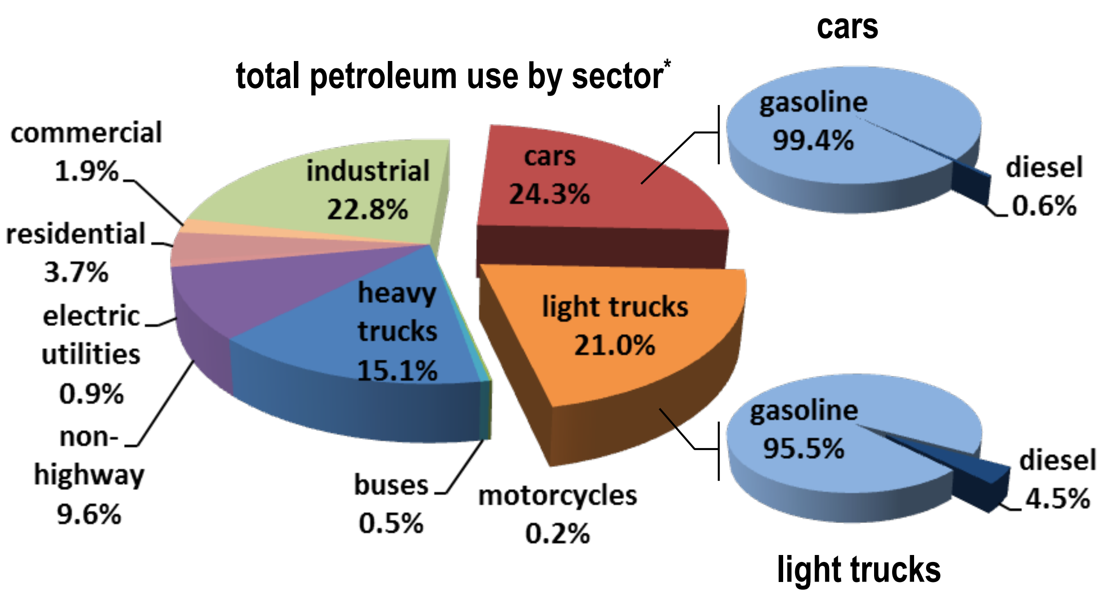
Objectives and Relevance

Enabling lean-gasoline vehicles to meet emissions regulations will achieve significant reduction in petroleum use

- **Objective:**
  - Address technical challenges to meeting emissions regulations
  - Investigate strategies to achieve cost-effective compliance
    - minimize precious metal content while maximizing fuel economy

- **Relevance:**
  - U.S. passenger car fleet is dominated by gasoline-fueled vehicles.
  - Enabling introduction of more efficient lean gasoline engines can provide significant reductions in overall petroleum use
    - thereby lowering dependence on foreign oil and reducing greenhouse gases

Relevance: small improvements in gasoline fuel economy significantly decreases fuel consumption



Lean gasoline vehicles can decrease US gasoline consumption by ~30 million gal/day

- **132,000 million gallons of fuel used by cars and light trucks annually****
- **New car and light-truck sales dominated by gasoline engines**
- **10% fuel economy benefit † from base case of 22.6/18.1 mpg** has big impact**
  - Saves >200,000,000 barrels gasoline annually
  - 5% of overall petroleum used
- **HOWEVER…emissions compliance needed!!!**

*, ** - *References: Transportation Energy Data Book, Ed. 29; *2009 data; **2008 data.*

† - *data from chassis dynamometer data at ORNL; see technical backup slides*

Current and Future Milestones

- **FY2011: Investigate performance, sulfation and desulfation of Ca-doped LNTs with a Ca-level between 3 and 9% (9/30/2011).**
  - Complete; project merged with Lean Gasoline project
- **FY2011: Analysis of lean-rich period modification for enabling lower volume LNTs to reduce the higher concentrations of NOx associated with lean gasoline engines (9/30/2011).**
  - Complete
- **FY2012: Measure the effect of oxygen storage capacity on NH_3 formation by three way catalyst for use in passive SCR emission control strategy. (9/30/2012)**
  - On track

- **Future years will adopt a quantitative milestone approach to ensure progression towards goal of low-cost emissions control solution for fuel efficient lean-burn gasoline vehicles**

	FY13	*FY14*	*FY15*	*FY16*	*FY17*
Fuel economy gain over stoichiometric	7%	10%	10%	12%	15%
Total emissions control devices Pt* (g/L_{engine})	8	7	6	5	4

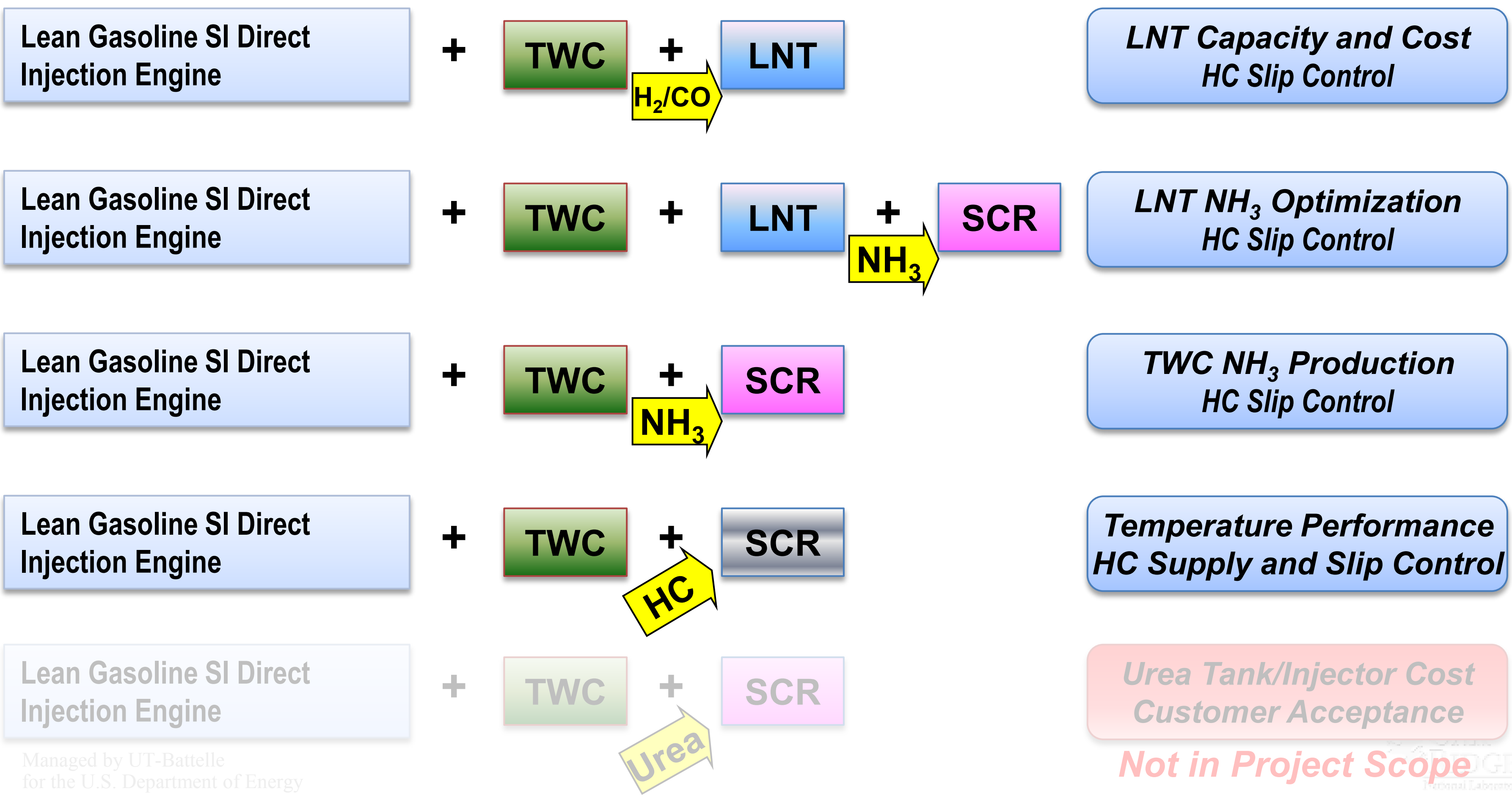
	5-year Average ($/troy oz.)	Pt-equivalent
Platinum	$ 1,504 /troy oz.	1.0
Palladium	$ 463 /troy oz.	0.3
Rhodium	$ 3,582 /troy oz.	2.4
Gold	$ 989 /troy oz.	0.7

*** - will use Pt equivalent to account for different costs of Pt, Pd and Rh; 5-year average value fixed at beginning of project**

Approach: Technology Options and Critical Issues Related to Cost and Performance

- Goal: Enable Tier II Bin 2 Emission Compliance for Lean Gasoline Engine
- Focus on NOx, CO, HC (PM may be issue for DI engines, but outside of project scope; new project starting)
- Technologies: TWC = Three-Way Catalyst
  LNT = Lean NOx Trap
  SCR = Selective Catalytic Reduction

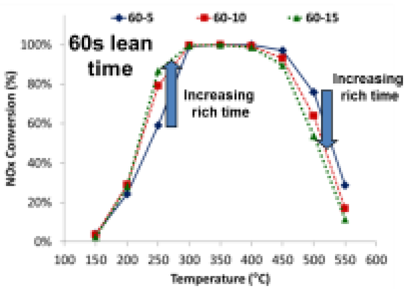
Specific Key Issues:
Cost, Durability, Fuel Penalty, Operating Temp., etc…

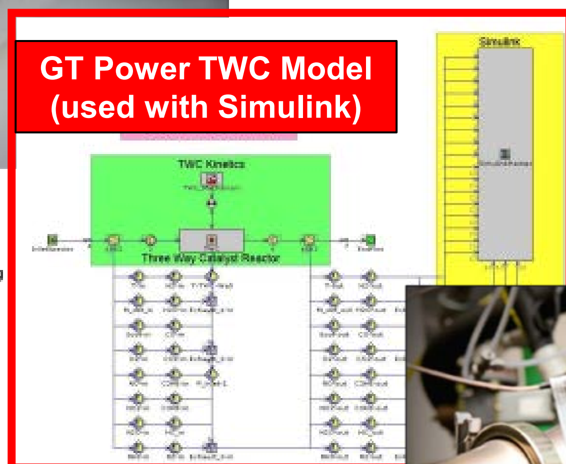


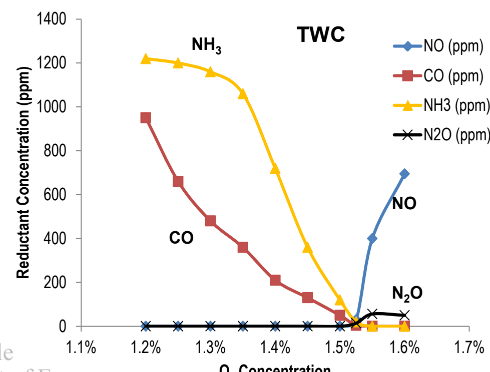
Overall approach to meeting long term milestones

- **Improve catalyst understanding and optimize emissions control**
  - Catalyst technology focus → Efficiency gains from lean vs. stoichiometric operation

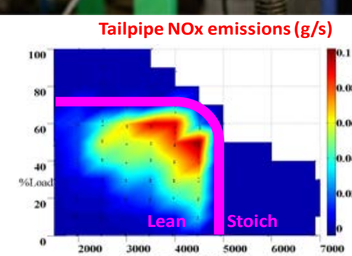
- **Understanding from bench reactor catalyst data guides engine strategy**
  - Supply data and kinetics to modeling collaborators







- **Engine platform to study catalyst system under real exhaust conditions**
  - Steady state modes employed
  - Drivven developed engine control for system optimization



- **Vehicle platform for validation**
  - Drive cycles

Collaborations and partners

- **Currently all collaborations are based on mutual interest**
  - No subcontracts between partners
- **General Motors**
  - Monthly teleconferences
- **Umicore**
  - Catalyst supplier for the commercial LNT and TWCs
  - Facilitating range of catalysts with varying PGM and functionality
  - Monthly teleconferences
- **University of South Carolina**
  - Visiting graduate student operating and analyzing bench reactor data
- **University of Wisconsin**
  - Modeling components; based on global kinetics
- **CLEERS**
  - share results and identify research needs

Summary of Technical Accomplishments

- **Progressed towards lean gasoline engine dynamometer platform**
  - Obtained two lean gasoline vehicles (BMW 120i)
    - Engine to be removed from one for full control in test cell
    - Other vehicle to be kept intact for chassis-studies
  - Subcontract to Drivven to develop full pass controller in progress
  - Transient drive cycle data uploaded to CLEERS website
- **Measured impact of lean/rich duration on NOx reduction and product yields**
  - Maximized NOx conversion with constant fuel penalty with:
    - short, concentrated rich dose for high temperatures T>450°C
    - long, low concentration rich dose for T<300°C
  - Identified peak NH_3 and N_2O formation at 250°C
  - Established desulfation temperature: Peak desulfation at 550-580°C (sulfated to 3.4 g/L)
- **Identified TWC NH_3 production conditions for passive SCR approach**
  - Two TWC technologies studied
    - High PGM, Pd-based technology with <u>no</u> OSC (oxygen storage capacity)
    - Low PGM, Pd/Rh-based technology <u>with</u> OSC
  - Demonstrated controlled NH_3 production while rich with >98% CO/HC conversion

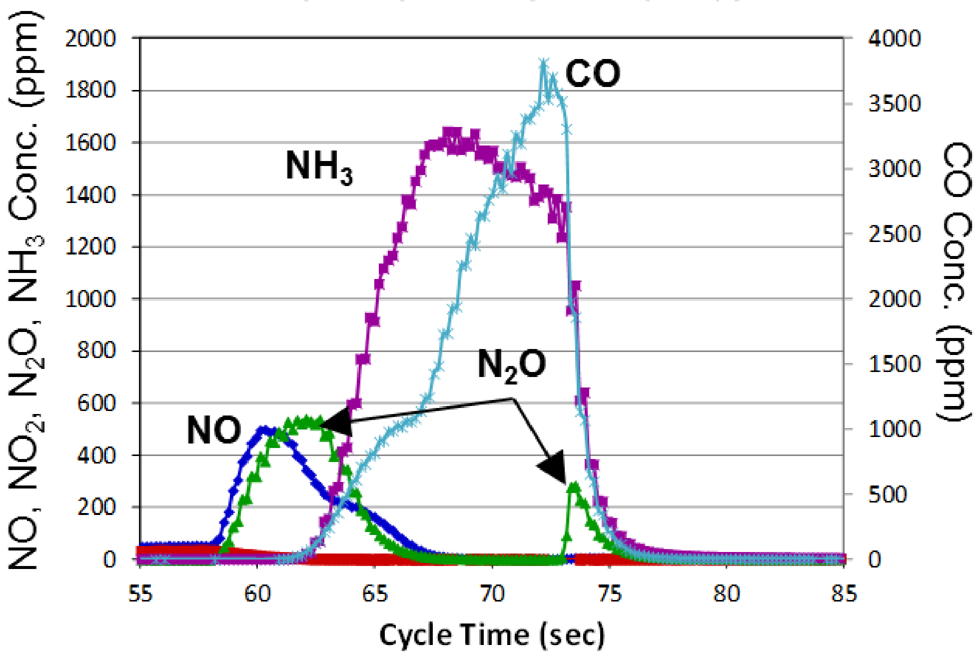
Engine and vehicle platform is the lean burn gasoline version of the BMW120i – 2.0L L4

- **Vehicle based on industry suggestion**
  - BMW lean-burn gasoline vehicle
  - Close-coupled Three-way Catalyst (TWC)
  - Underfloor Lean NOx Trap (LNT)
    - New CLEERS reference catalyst
  - Total PGM = 7.5 g/L-engine
  - As calibrated, does not meet current US emissions standards

- **Bench reactor studies focused on catalyst technologies relevant to this application**
  - Umicore recommended TWC
    - Pd-only with no oxygen storage component (OSC) in front zone
    - Pd/Rh with OSC in rear zone
  - Identical LNT to BMW

LNT: Lean/rich durations varied to establish impact on NOx reduction and NH_3 formation

- **Investigate 3 lean times and 3 rich times for a total of nine conditions**
  - Hold rich dose/fuel penalty constant for all cases
  - Stoichiometric reductant concentrations for nitrate reduction to carbonate + OSC reduction
- **Develop understanding on tradeoffs of long and short durations**
  - *Is it better to have a short rich time with high reductant concentrations?*
  - *What is the impact on conversion and NH_3 and N_2O formation?*

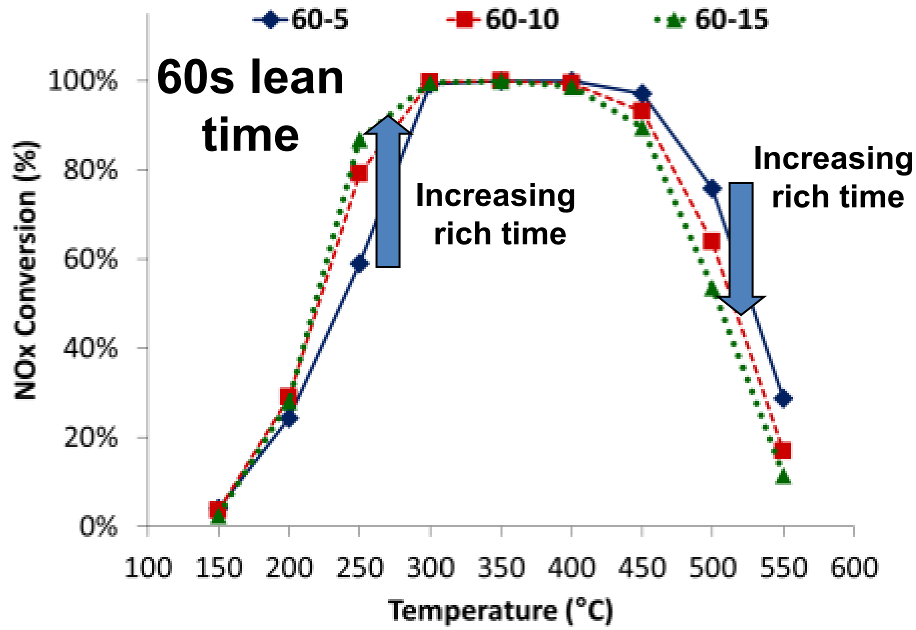


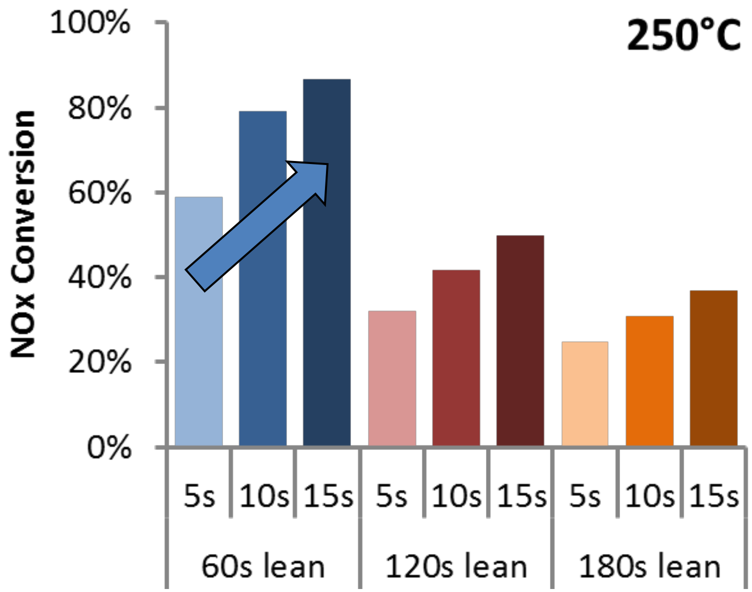
Example products during rich portion of 60s lean and 15s rich at 250ºC

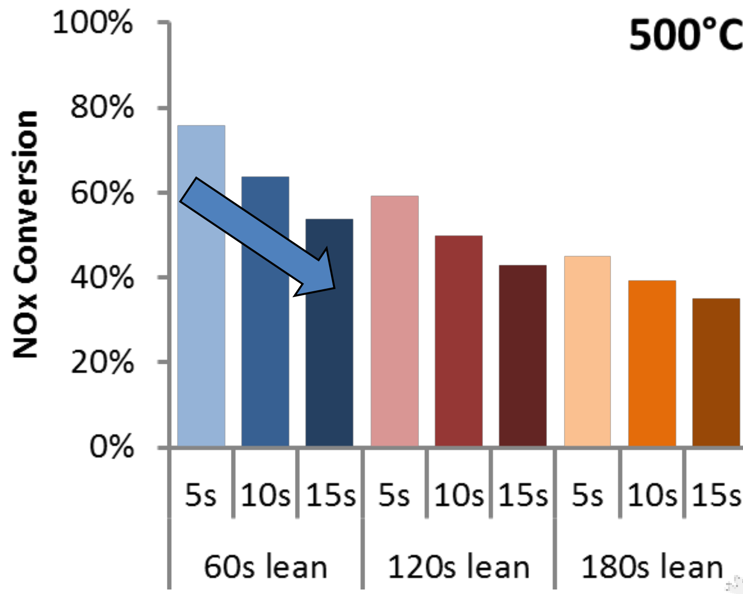
Lean / Rich times	CO	H_2	C_3H_6
60 s lean/5 s rich	2.01%	0.67%	0.112%
60 s lean/10 s rich	1.01%	0.34%	0.056%
60 s lean/15 s rich	0.67%	0.22%	0.037%
120 s lean/5 s rich	2.83%	0.94%	0.157%
120 s lean/10 s rich	1.42%	0.47%	0.079%
120 s lean/15 s rich	0.94%	0.31%	0.052%
180 s lean/5 s rich	3.65%	1.22%	0.203%
180 s lean/10 s rich	1.82%	0.61%	0.101%
180 s lean/15 s rich	1.22%	0.41%	0.068%

Practical lean/rich timing variation impacts NOx conversion performance

- **Short lean times and long, low concentration rich dose favored at T<350°C**
  - At 250°C, NOx conversion varies from 25% to 85%...fuel penalty is constant
  - Slow NOx release and reduction kinetics
- **Need fast, high concentration rich dose to convert stored NOx at T>400°C**
  - Fast NOx release and reduction kinetics



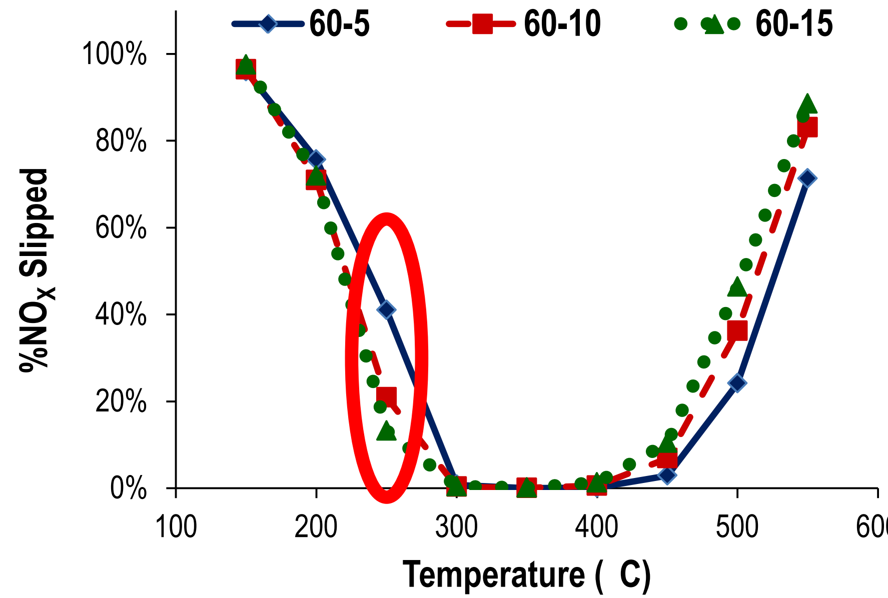


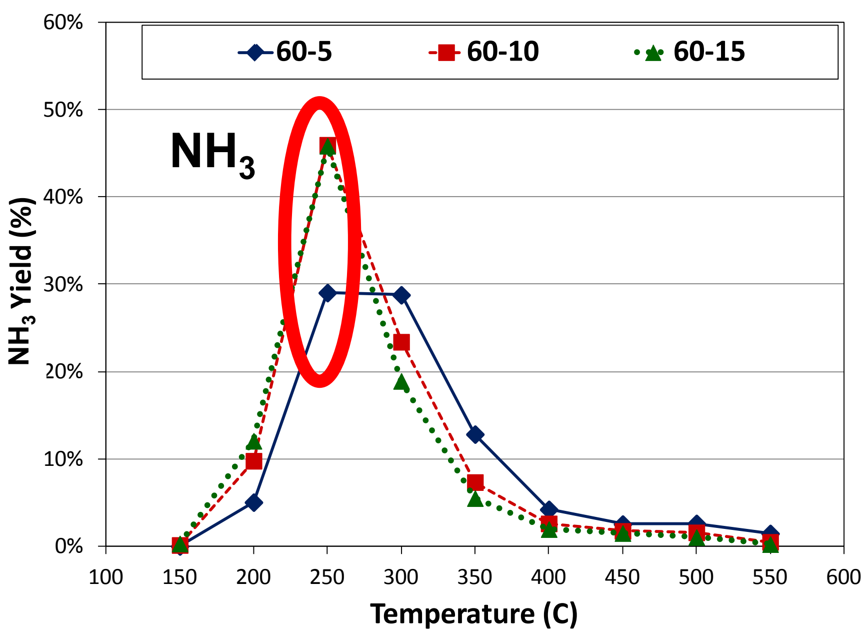


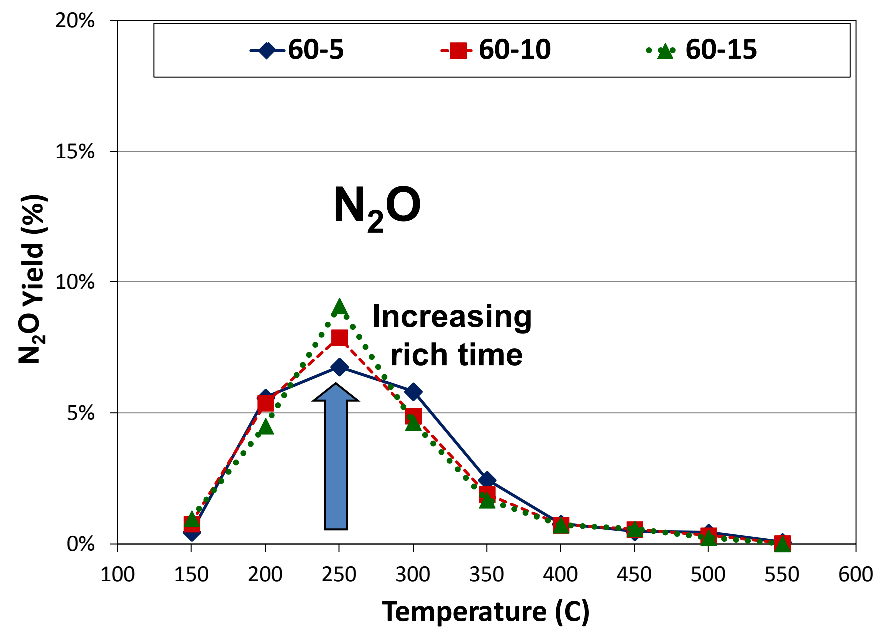
NH_3 generation occurs between 200 and 350°C; potential of LNT+SCR

$$Yield_{NH_3} = \left(\frac{NH_3(produced)}{NO_X(fed)} \right)$$

- Up to 45% NO fed goes to NH_3
- Comparing NOx and NH_3 slip at 250ºC illustrates potential LNT+SCR system
- Unfortunately, N_2O yield also peaks at these conditions

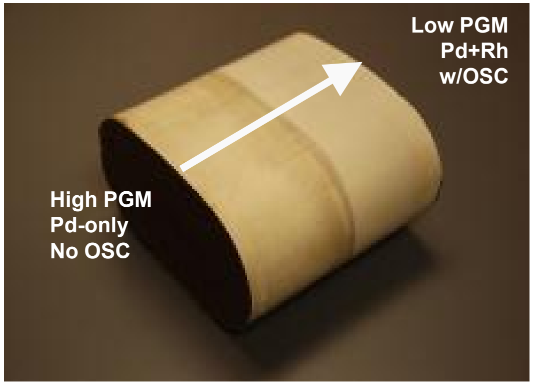


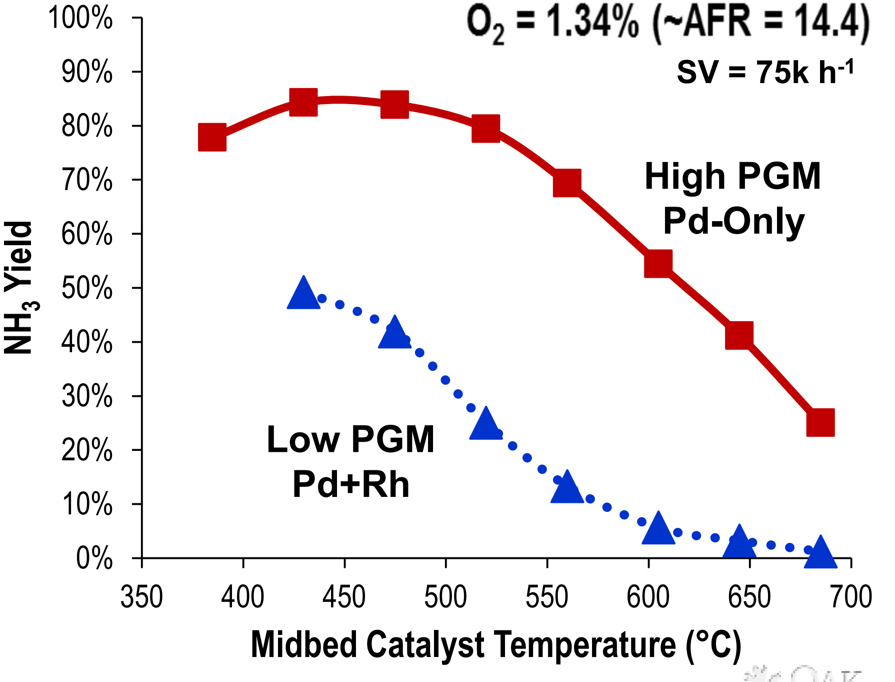




Understanding NH_3 generation over TWC may enable TWC+SCR system without Urea

- **Measure performance of TWCs as a function of temperature and components**
  - Overall NO, CO, C_3H_6 conversion
  - N_2O and NH_3 formation
- **TWC sections studied independently**
  - High PGM Pd-only with no oxygen storage component (OSC)
    - 0/4.4/0 g Pt/Pd/Rh
  - Low PGM Pd+Rh with OSC in rear zone
    - 0/0.8/0.21g Pt/Pd/Rh
- **Including LNT into this study will give the following catalyst types:**
  - PGM-only
  - PGM + OSC
  - PGM + OSC + NOx storage

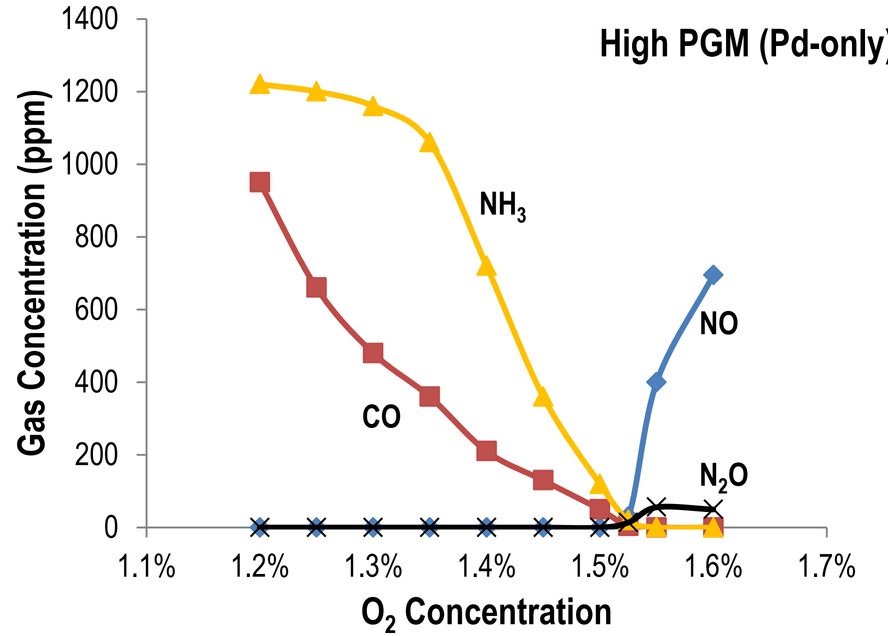


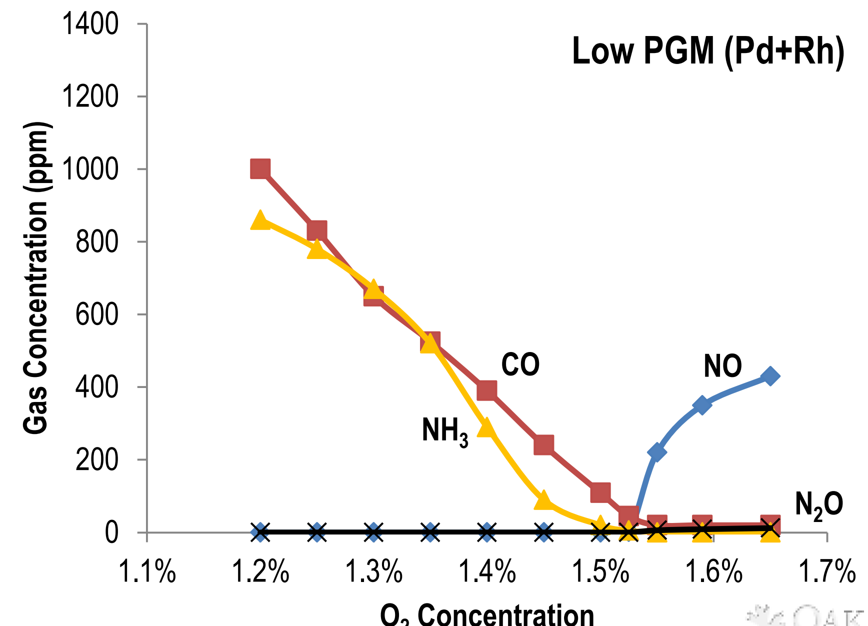


TWC is effective and tunable NH_3 generator

- **NH_3 readily generated; varies with PGM**
  - For high PGM (Pd-only) TWC:
    - All NO fed converted to NH_3 when very rich
  - For low PGM (Pd+Rh) TWC:
    - NH_3 production is still significant but reduced
- **At all conditions, >95% CO conversion**
  - C_3H_6 not observed in effluent
  - >98% CO conversion for O_2>1.35% (AFR ~14.4)
- **N_2O formation observed under lean conditions and varies with PGM content**
  - Up to 56 ppm with high PGM (Pd-only) TWC
  - Less than 10 ppm with low PGM (Pd+Rh) TWC

~AFR	O_2	NO	CO	H_2	C_3H_6
14.6	1.59%	0.12%	1.80%	0.60%	0.10%
14.4	1.34%	0.12%	1.80%	0.60%	0.10%
14.2	1.06%	0.12%	1.80%	0.60%	0.10%

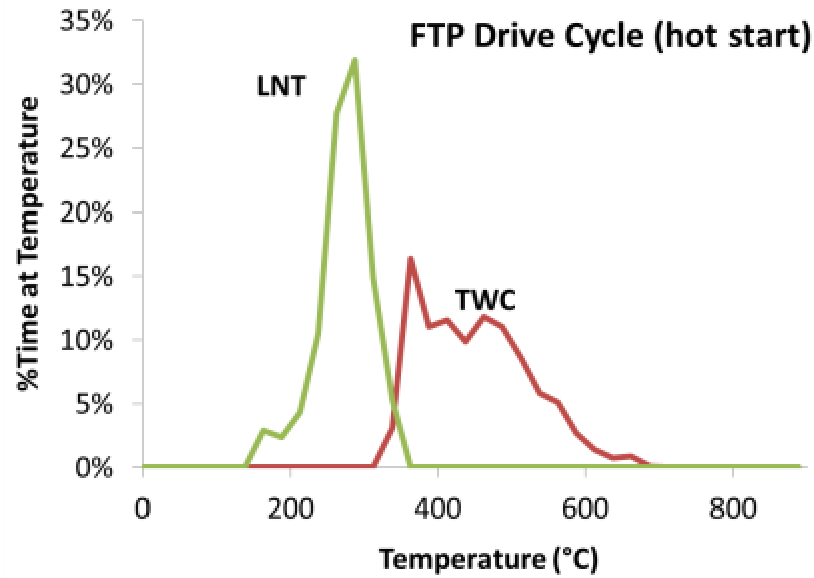




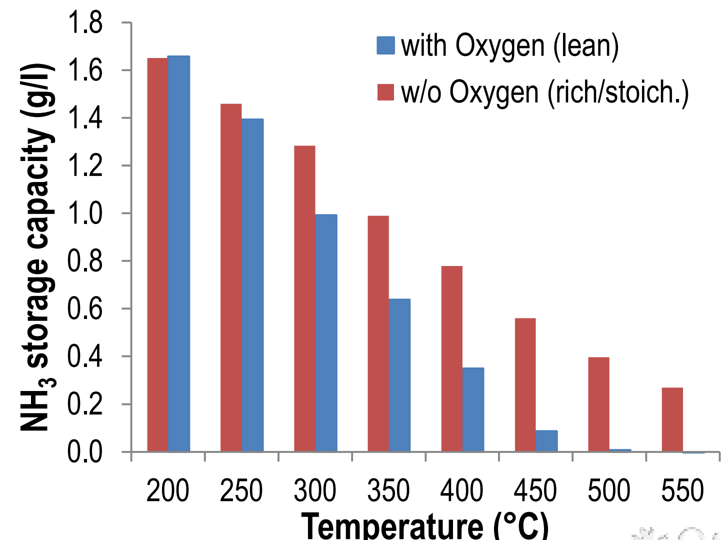
Midbed Temperatures: 460-500°C

NH_3 production over LNT and TWC occurs at temperatures relevant to vehicle operation and NH_3 storage on SCR

- **Histogram of catalyst temperatures during drive cycle with BMW 120i**
  - FTP (hot start)
  - 200-350ºC for LNT
  - 350-600ºC for TWC
- **LNT: max NH_3 yield at 250-300ºC**
- **TWC: tunable NH_3 production 350-600ºC**



- **NH_3 production temperatures mesh well with NH_3 storage temperatures on SCR**
  - More NH_3 storage occurs under rich/stoichiometric conditions
  - However switching from rich to lean will result in NH_3 release if over-saturated



Lean Gasoline Engine Research Platform Development In Progress

- **Two BMW 120i lean gasoline engine vehicles were procured from Europe and are being mapped for engine controller development**
  - One vehicle's engine will be pulled for installation on engine dynamometer
- **Subcontract to Drivven for controller development in progress**
  - End controller will have full control capability for research specific engine operation and OEM map operation for general catalyst and reference studies

Tasks in Controller Development:

1. **Procure Vehicles** (complete)
2. **Verify lean operation** (complete)
3. **Drivven visit for electronic signal probing** (complete)
4. **Shipment of one vehicle to Drivven** (complete)
5. **Calibration of sensors, injectors, etc.** (in progress)
6. **Harness wiring and mapping preparation** (in progress)
7. **Full mapping exercise on vehicle at ORNL** (May-June 2012)
8. **Control software development** (June 2012)
9. **Engine installation at ORNL** (July 2012)
10. **Controller installation at ORNL** (July 2012)
11. **Controller operation and verification** (August 2012)

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Additional future work

- **Study effects of catalyst composition by varying:**
  - PGM content, OSC content
  - Adding NOx storage capacity to TWC
  - Low temperature CO/HC oxidation
- **Impact of lean/rich cycling on TWC**
  - Investigating strategies for minimizing N_2O formation
- **Age catalysts and evaluate end of life performance**
  - Define durability requirements, time and temperature as related to sulfur exposure and desulfation requirements
- **Improve modeling capability to better guide research and meet milestones**

- **Finalize milestones and achieve year one target**

	FY13	*FY14*	*FY15*	*FY16*	*FY17*
Fuel economy gain over stoichiometric	**7%**	**10%**	**10%**	**12%**	**15%**
Total emissions control devices Pt* (g/L_{engine})	**8**	**7**	**6**	**5**	**4**

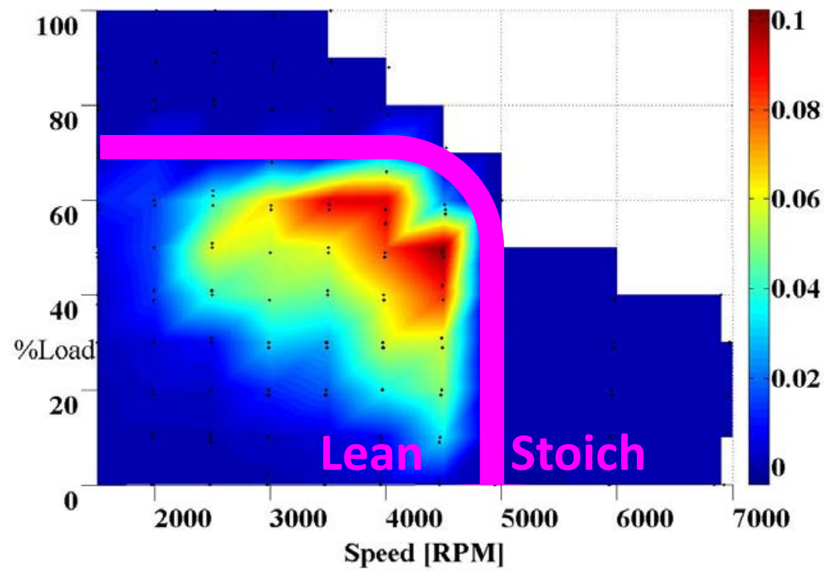
Summary

- **Relevance**: Enabling lean gasoline vehicles will impact significantly on US petroleum use
- **Approach**: Cost-effective emissions control facilitates introduction of lean-gasoline engines
  - Employ multi-platform approach to optimize catalysts and lean engine operation
  - Employ annual cost and efficiency milestones to push technology
- **Collaborations**: Industrial (GM and Umicore) and Academic (U-So. Carolina and U-Wisconsin)
- **Technical Accomplishments**:
  - Progressed towards lean gasoline engine dynamometer platform
  - Measured impact of LNT lean/rich duration on NOx reduction and product yields
  - Identified TWC NH_3 production conditions for passive SCR approach
- **Future Work**:
  - Complete and install DRIVVEN-developed engine controller for engine/chassis operation
  - Vary OSC and NOx storage component on LNT and TWCs under cycling conditions
  - Develop and employ aging routine to rapidly age catalysts to end of life
  - Meet first cost and efficiency milestone of the multi-year program

Technical back-up slides

4-15% Fuel Economy Benefit *but* Challenge of Emissions Exceeding U.S. Regulation Levels

- Vehicle designed to meet emissions levels required by European regulations
- NOx emission levels exceed U.S. Tier II Bin 5, 0.05 g/mile at 50k miles
  - Bin 2 → 0.02 g/mile
- NOx emissions during lean operation are problematic
  - Data shown is from engine vehicle evaluation at ORNL



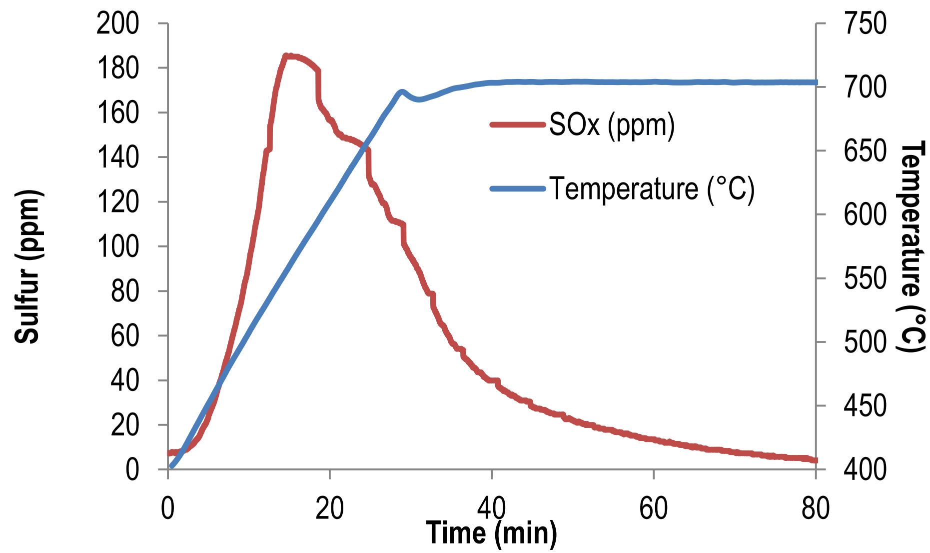
Improved Lean NOx catalysis required for deployment of lean gasoline vehicles

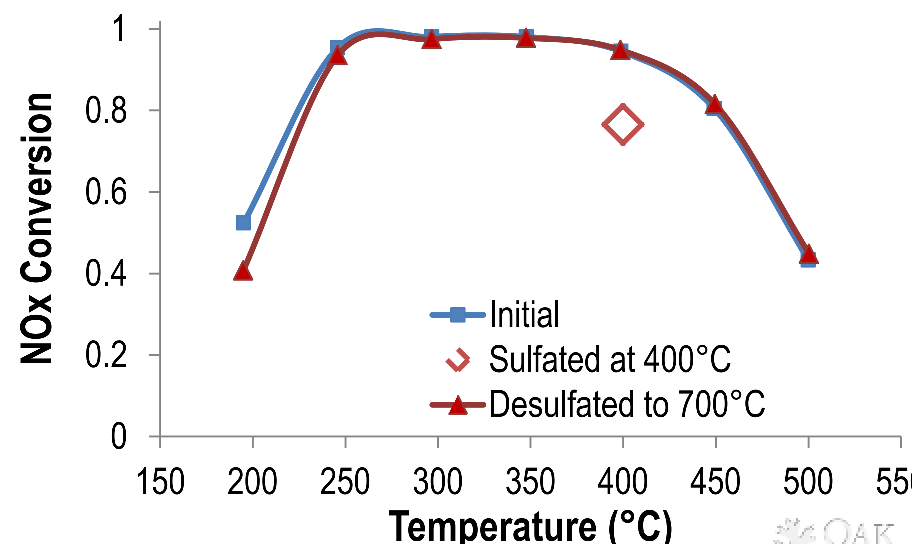
Drive Cycle	Fuel Economy Improvement*	NOx Emissions (g/mile)
FTP	10.0%	0.11
HFET	14.6%	0.11
US06	4.4%	0.35

*comparing stoichiometric operation to lean

Peak LNT desulfation at 550-580°C and full performance recovery after desulfating to 700°C

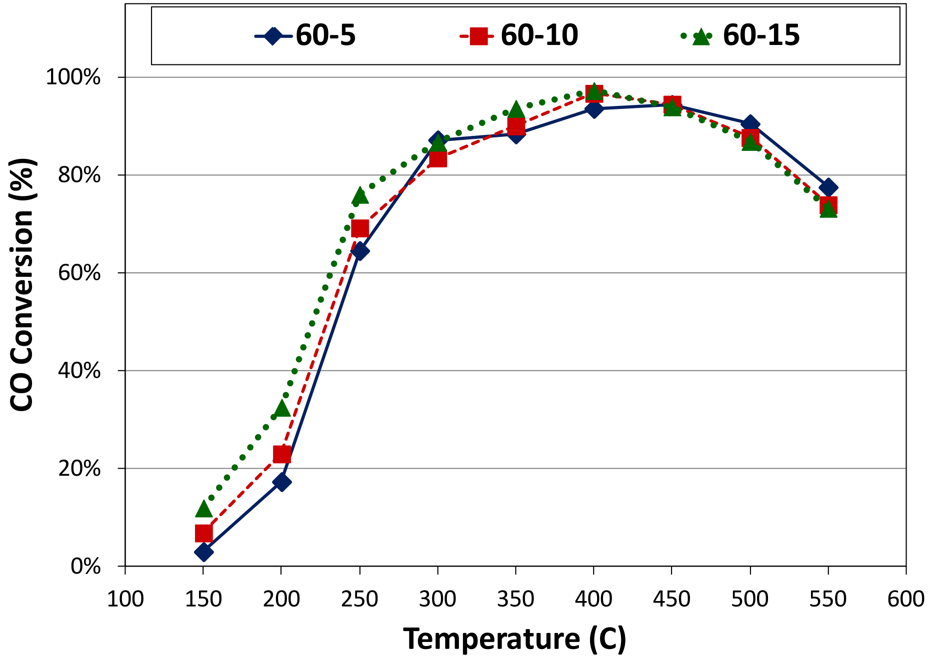
- Sulfation to 3.4 g/L_{cat} at 400°C
  - 45 ppm SO_2 accelerated
  - NOx conversion drops from 95% to 77%
- LNT shows low desulfation temperature
  - Sulfur removal occurs readily above 400°C
  - Peaks at 550-580°C
  - Slow release up to 700°C
- Desulfation to 700°C results in complete performance recovery
  - Suggests targeting 700-750°C for aging conditions

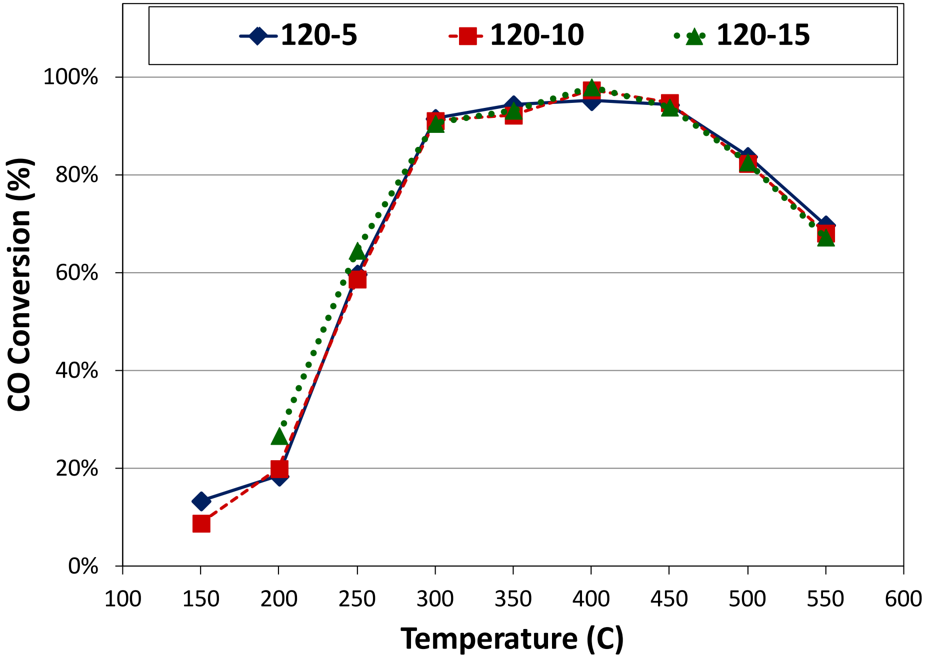


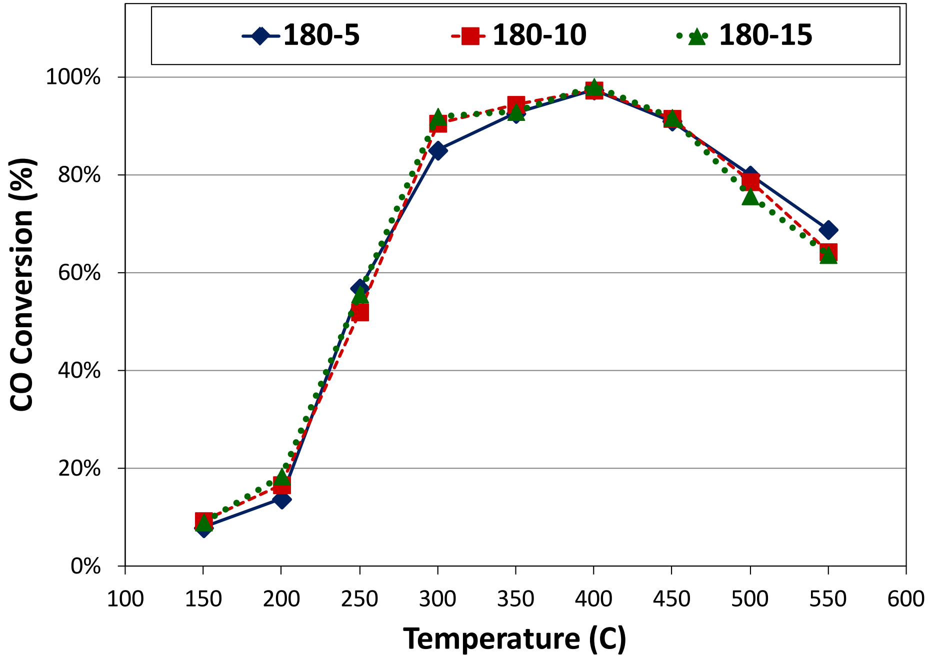


Cycle averaged CO conversions

- Essentially independent of lean rich timing
  - at 300-500C, >80% conversion all conditions
- CO utilization decreases at higher temperatures
  - Less NOx stored and fast release
  - Mismatch in reductant feed and NOx release

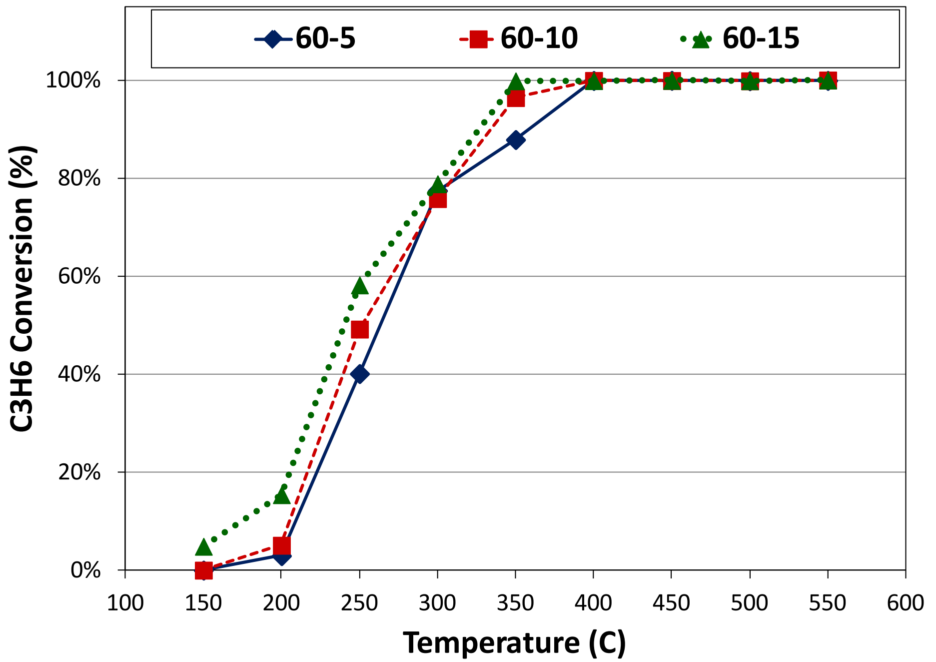


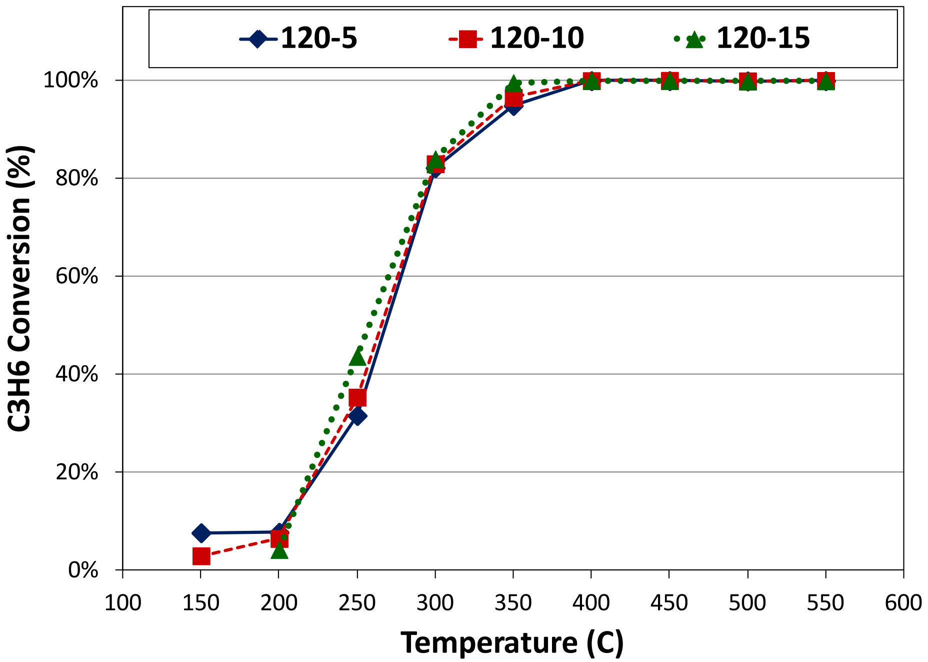


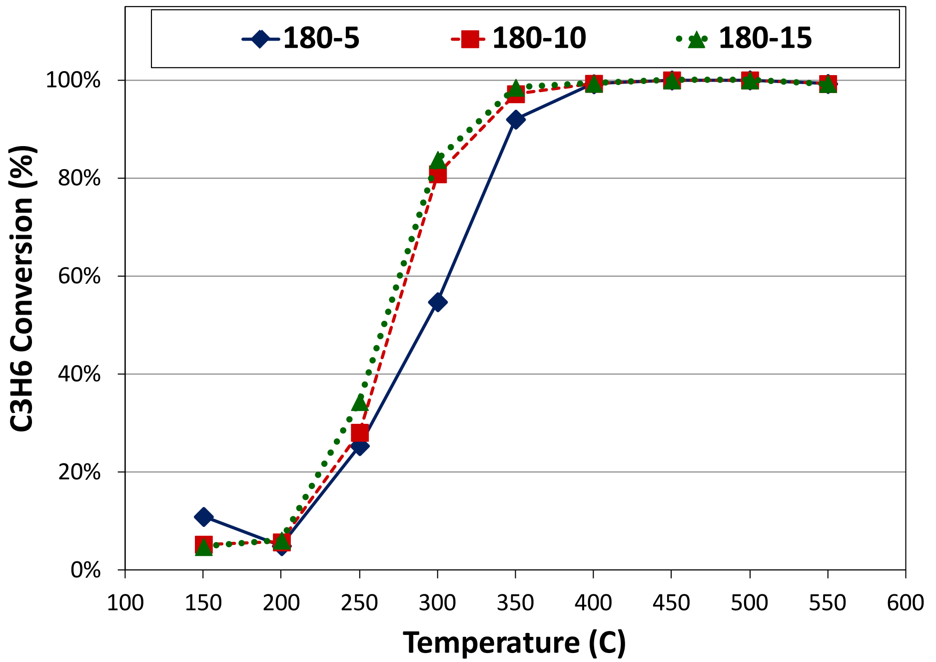


Cycle averaged C_3H_6 conversions

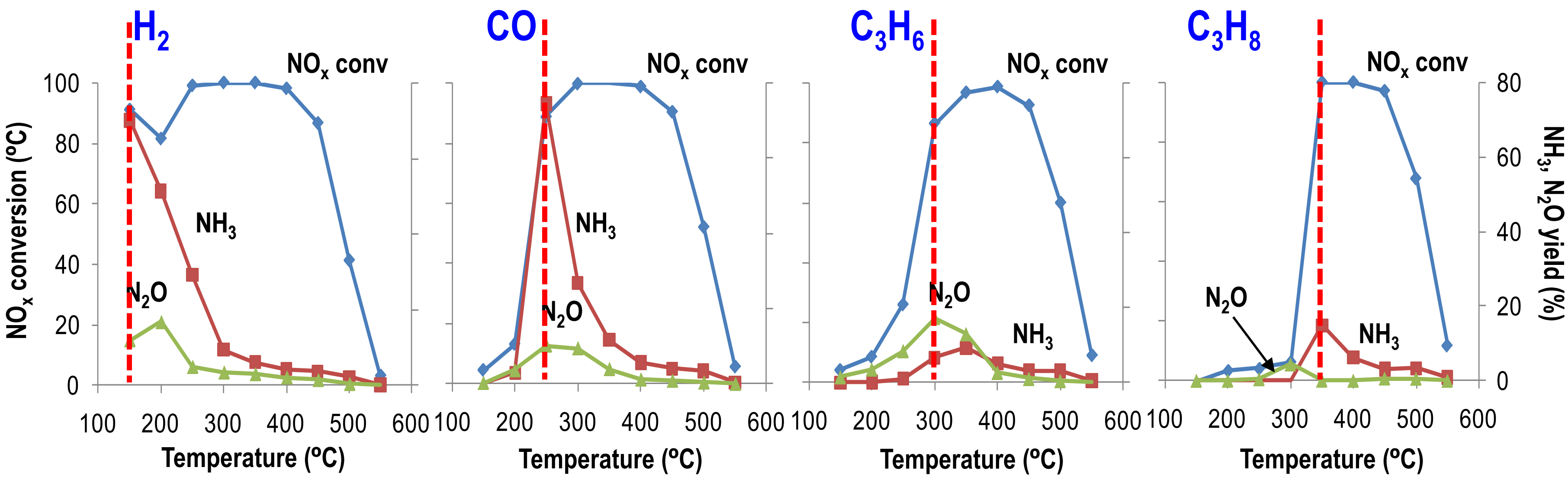
- Similarly independent of lean/rich timing
  - at T>300, >80% conversion all conditions
    - Except 180s lean / 5s rich
- Looks like a typical light off curve
- Represents propylene disappearance rather than complete oxidation



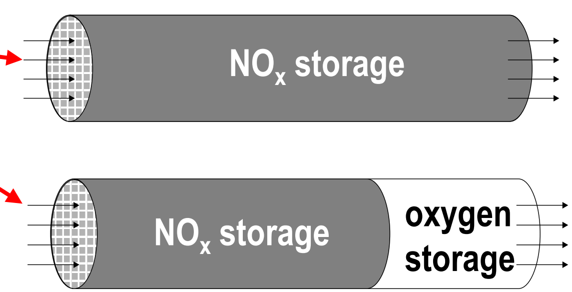




Reductant type affects spatial distribution of NH_3 chemistry controlling NH_3 & N_2O yields



- **Light-off temperature is highly dependent on reductant type: $H_2 < CO < C_3H_6 < C_3H_8$**
  - At light-off: near max conv. reached using whole LNT
  - Above light-off: max conv. using partial length of LNT



- **NH_3 & N_2O yields peak at light-off T**
  - Significant slip possible: NH_3 ~70%; N_2O ~20%
  - At light-off: reactions forming NH_3 (reductant + stored NO_x) & N_2O (NH_3 + stored NO_x) maximized
  - Above light-off: NH_3-consuming reactions without generating N_2O increase (NH_3 + CeO_2; decomposition)