Emissions Control for Lean Gasoline Engines

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Sponsors: Gurpreet Singh and Ken Howden
Advanced Combustion Engines Program
U.S. Department of Energy

Project Overview

Timeline

- Project began in FY12
- Project Ongoing

Budget

- FY12 $400k
- FY13 $500k

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*

Collaborators & Partners

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

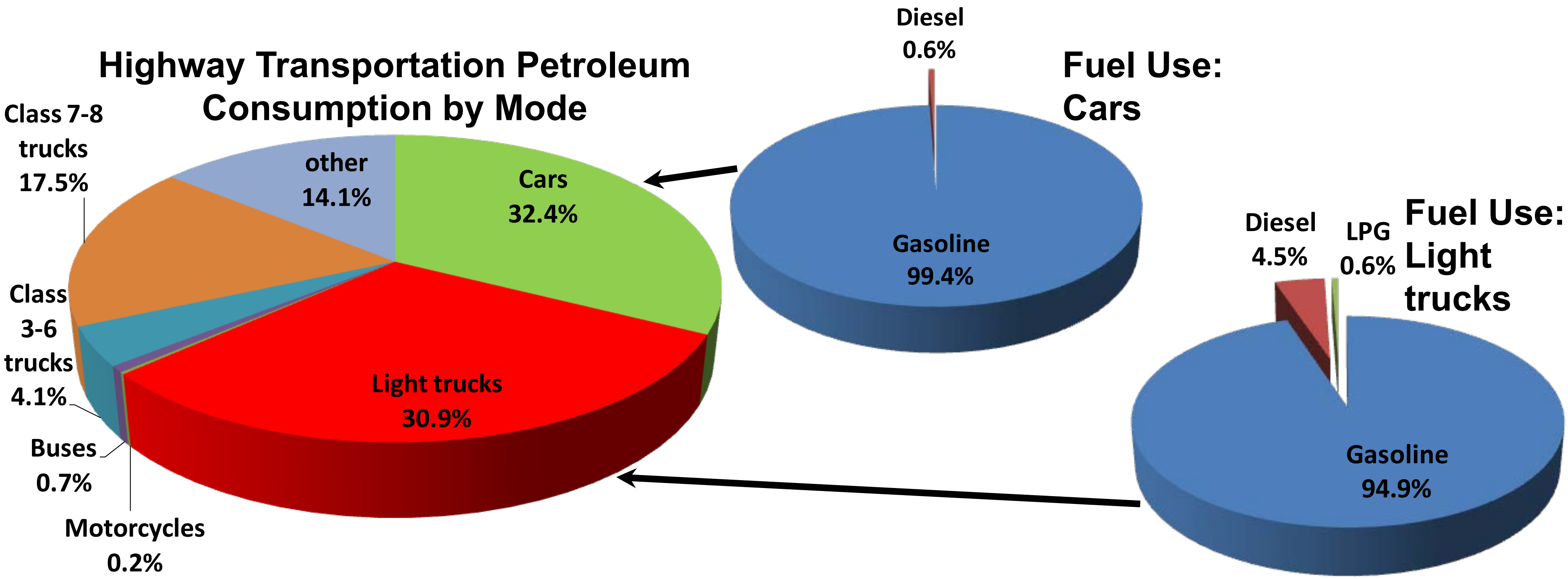
Objectives and Relevance

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

- Objective:
  - Demonstrate technical path to emission compliance that would allow the implementation of lean gasoline vehicles in the U.S. market.
    - Lean vehicles offer 5–15% increased efficiency over stoichiometric-operated gasoline vehicles.
  - 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



- US car and light-truck fleet dominated by gasoline engines
- 10% fuel economy benefit from base case of 23.0/17.1 mpg has significant impact
  - Saves 12.8 billion gallons gasoline annually
  - Or, save $47 billion/year (at $3.68/gallon 2012 US price)
- HOWEVER…emissions compliance needed!!!

Lean gasoline vehicles can decrease US gasoline consumption by ~12 billion gal/year

References: Transportation Energy Data Book, Ed. 31 (2010 petroleum/fuel use data); www.eia.gov (2012 US gasoline price)

Current and Future Milestones

- **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)
  – Complete
- **FY2013:** Commission lean gasoline direct injection engine platform (September 30, 2013).
  – Complete
- **FY2013:** Characterize the fuel efficiency and emission performance of a TWC+SCR system on the engine dynamometer platform as a function of the ratio of lean to rich periods (September 30, 2013).
  – In Progress (On Track)
- In addition to milestones, a set of project goals has been adopted 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

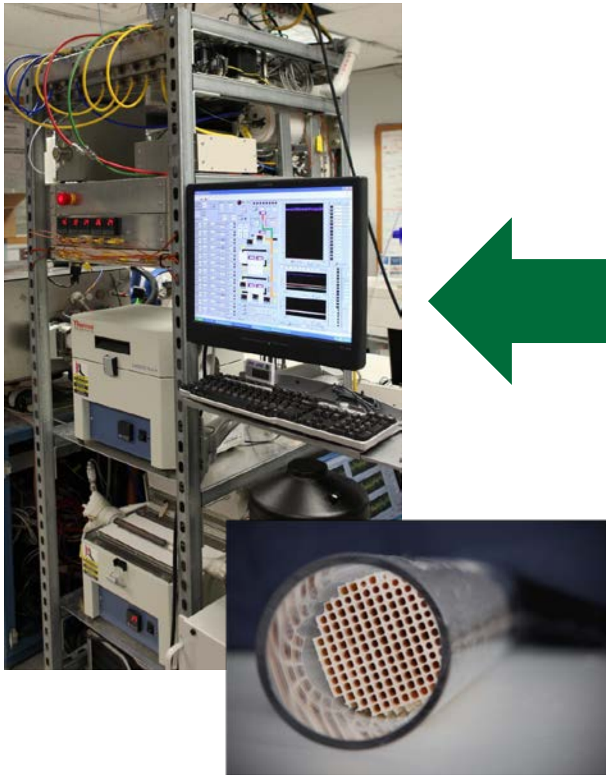
Approach: Technology Options and Critical Issues Related to Cost and Performance

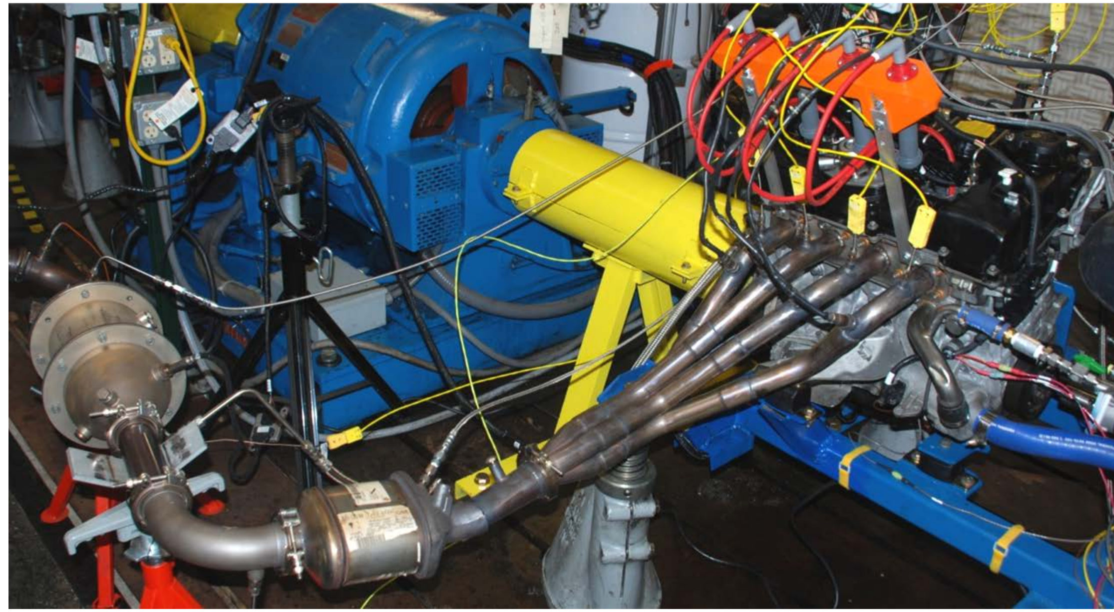
- **Goal: Enable Tier 2 Bin 2 Emission Compliance for Lean Gasoline Engine Vehicle**
- Focus on NOx, CO, HC (PM may be issue for DI engines, but outside of project scope; new project starting)
- Technologies: **TWC** = Three-Way Catalyst
  LNT = Lean NOx Trap
  SCR = Selective Catalytic Reduction

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

	Specific Key Issues
Lean Gasoline SI Direct Injection Engine + TWC + LNT (H_2/CO → LNT)	*LNT Capacity and Cost* *HC Slip Control*
Lean Gasoline SI Direct Injection Engine + TWC + LNT + SCR (NH_3 → SCR)	*LNT NH_3 Optimization* *HC Slip Control*
Lean Gasoline SI Direct Injection Engine + TWC + SCR (NH_3 → SCR)	*TWC NH_3 Production* *HC Slip Control*
Lean Gasoline SI Direct Injection Engine + TWC + SCR (HC → SCR)	*Temperature Performance* *HC Supply and Slip Control*
Lean Gasoline SI Direct Injection Engine + TWC + SCR (Urea → SCR) — *Not in Project Scope*	*Urea Tank/Injector Cost* *Customer Acceptance*

Approach: Studies on Bench Reactor and Engine





- **Studies on BMW 120i lean gasoline engine platform with Drivven open controller**
  - Realistic exhaust conditions
  - Full control of rich AFR for catalyst regeneration and reductant production/control
  - Scope does not include lean combustion optimization

- **Studies on Bench Flow Reactor**
  - Commercial, prototype, and model catalysts
  - Study of chemistry and mechanisms under simulated exhaust conditions
  - Two reactors simulate two catalysts in close coupled and underfloor positions

Data supplied to modeling community via CLEERS

Collaborations and partners

- **General Motors, Ford, Chrysler**
  - Teleconferences to share and discuss results
- **Umicore**
  - Catalyst supplier for the commercial LNT and TWCs
  - Facilitating range of catalysts with varying PGM and functionality
- **University of South Carolina (Michael Amiridis)**
  - Visiting graduate student Chris DiGiulio collaborated on bench reactor studies (Dr. Chris DiGiulio received his Ph.D. in Dec. 2012 and is now employed with UOP)
- **University of Wisconsin (Chris Rutland)**
  - Monthly teleconferences focused on sharing data for modeling of lean emission control systems (with Ph.D. candidate Jian Gong)
- **CLEERS**
  - share results/data and identify research needs

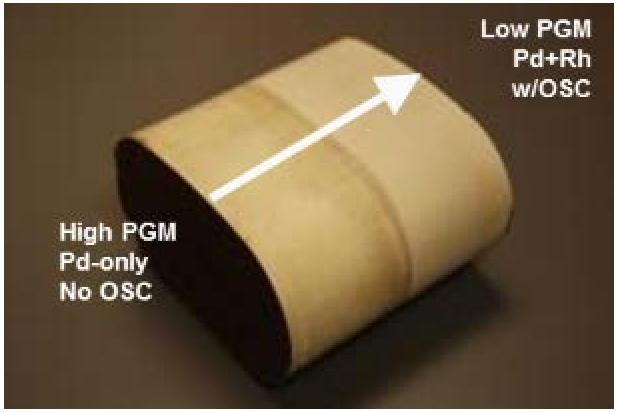
Related DOE VTP Projects of note:
ACE084; Thomas Wallner, ANL: High Efficiency GDI Engine Research, with Emphasis on Ignition Systems
FT007; Scott Sluder, ORNL: Fuel Effects on Emissions Control Technologies
ACE063; Halim Santoso, General Motors: Lean Gasoline System Development for Fuel Efficient Small Car

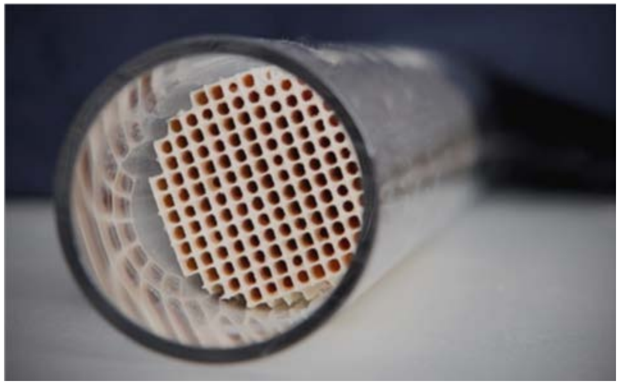
Summary of Technical Accomplishments

- **Completed characterization of NH_3 production from TWC matrix**
  - Catalysts studied included:
    - Commercial State-of-the-Art TWC (Umicore recommended SULEV TWC)
      - Front and rear catalyst formulations differ (studied individually and as combination)
    - Commercial LNT (BMW 120i OEM)
- **Demonstrated >99% NOx reduction efficiency with TWC+SCR approach on bench flow reactor under simulated exhaust conditions**
  - Catalyst combination and operating parameters based on TWC matrix study
  - TWC and SCR operated at separate temperatures consistent with BMW 120i close-coupled and underfloor catalyst positions
  - Cu-based and Fe-based SCR formulations compared for system performance
- **Completed development of lean gasoline engine research platform with full control capability**
  - Drivven-based controller with full OEM-based map plus ability for full-pass control of all engine parameters and actuators
    - close collaboration with Drivven staff Kris Quillen and Matt Viele
  - Engine now operational on engine dyno; studies commencing

TWC and LNT studied in bench-core reactor with varying PGM content

- For bench reactor, focusing on modern TWC technology (Umicore recommended formulations representative of SULEV emission level technology)
- All catalysts degreened for 16 hr at 700ºC in humidified air (2.7% H_2O)





Catalyst Matrix

Catalyst	Description	Pt/Pd/Rh (g/L)
Pd-only	High Pd-only	0/6.7/0
Pd/Rh+Ce	Pd/Rh with O_2 storage	0/1.1/0.3
Combo	Combination of 2 above (as designed for SULEV vehicle, Pd-only upstream)	0/4.0/0.16
Pt/Pd/Rh+Ce+Ba	BMW LNT formulation (with NOx storage)	7/3/1

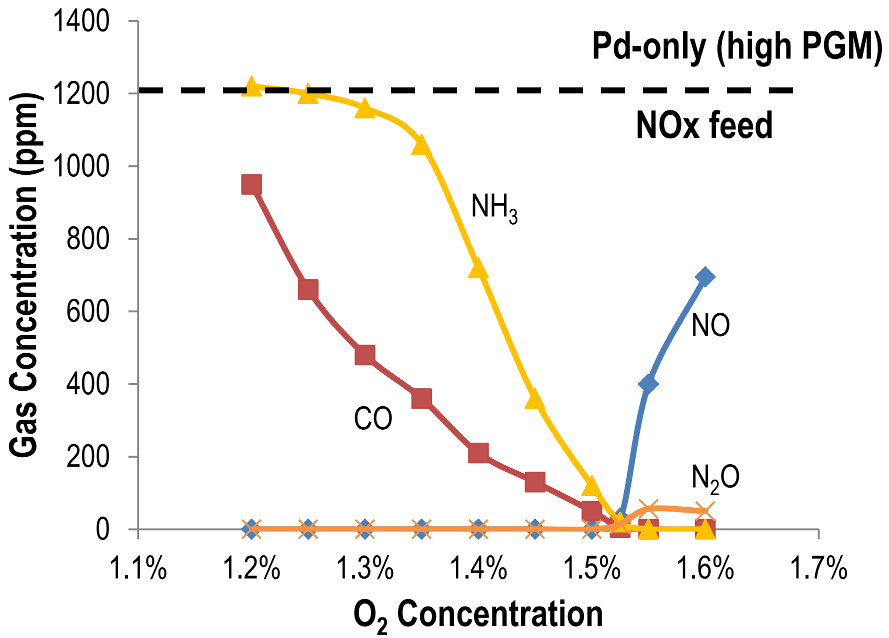
**See extra slides section for more detail on catalyst matrix*

TWC is effective and tunable NH_3 generator for "Passive SCR"

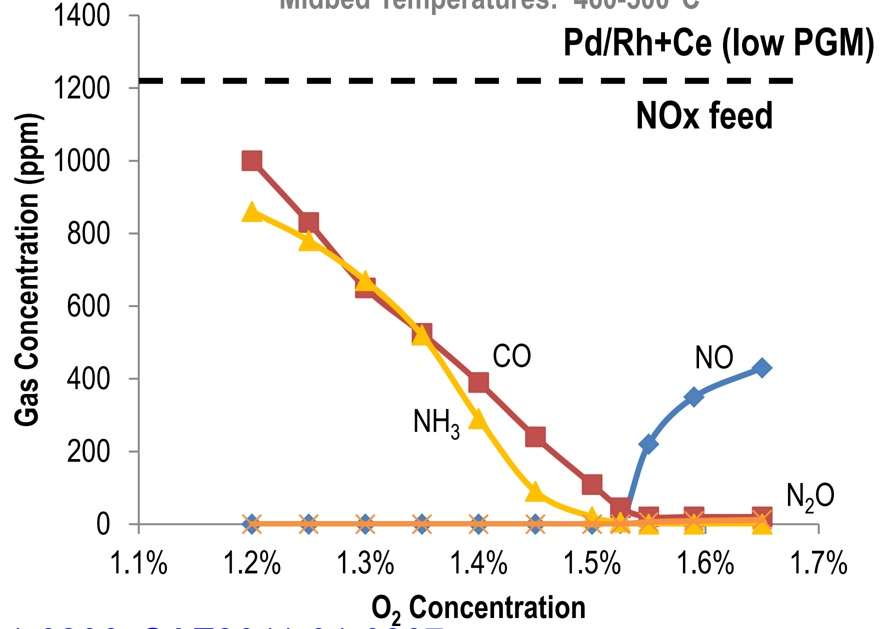
- **Example feed conditions:**

~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%

- NH_3 readily generated; varies with PGM
  - For Pd-only TWC with high PGM:
    - All NO converted to NH_3 when very rich
  - For Pd/Rh+Ce (low PGM) TWC:
    - NH_3 production is still significant but reduced
- At all conditions, >95% CO conversion
  - C_3H_6 not observed in effluent
- 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



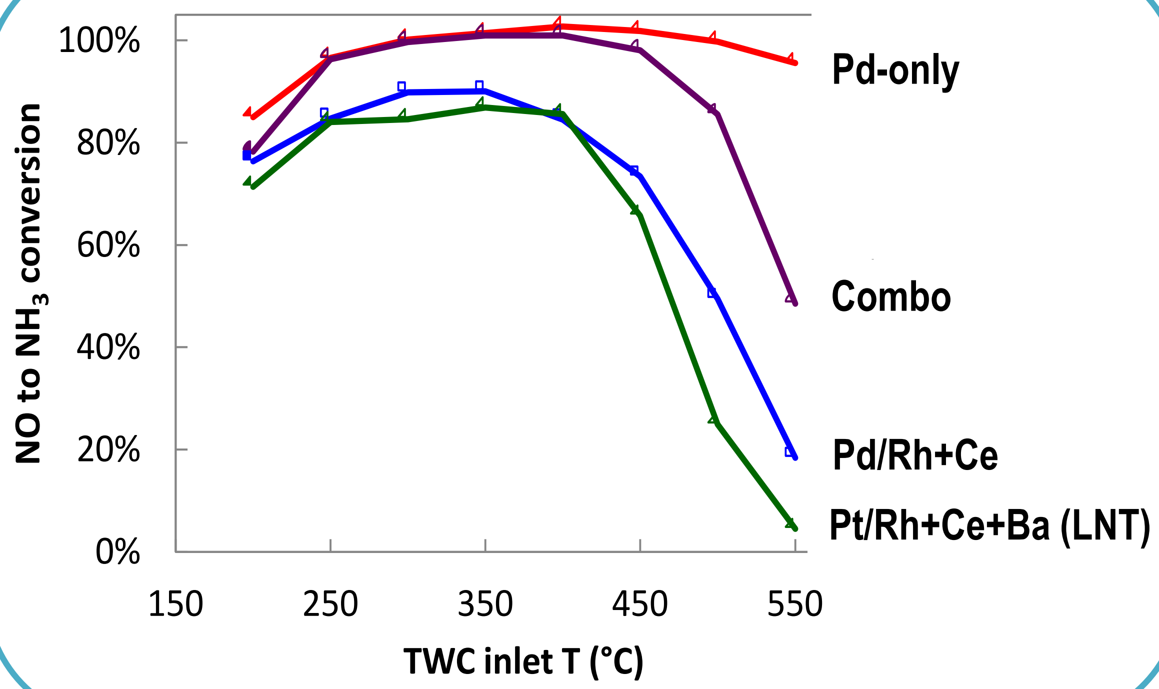
Midbed Temperatures: 460-500ºC

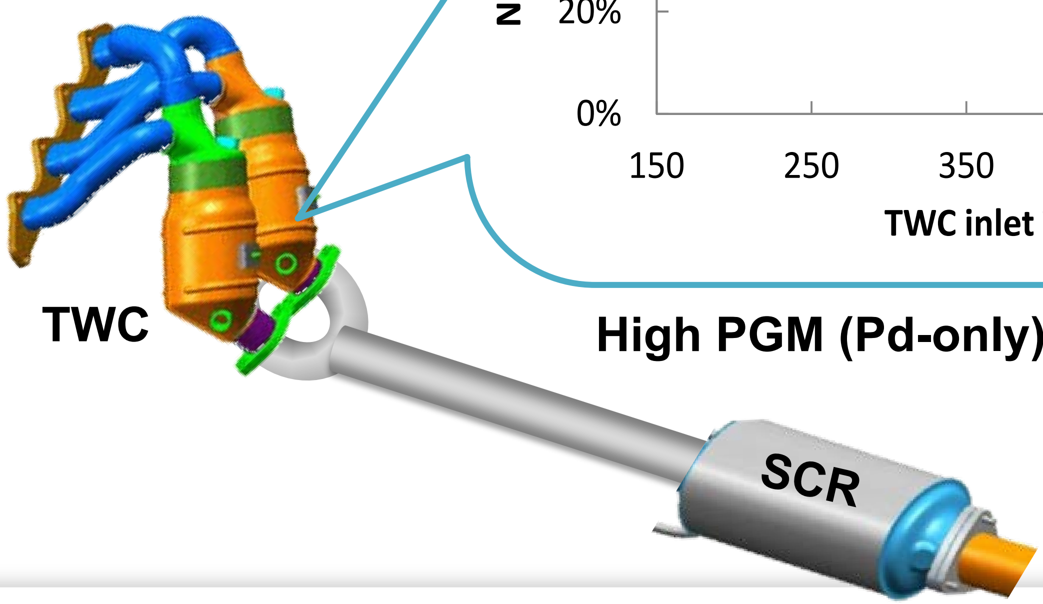


Passive SCR References: SAE2010-01-0366, SAE2011-01-0306, SAE2011-01-0307

PGM content and Pt/Pd/Rh ratios impact NH_3 production

Evaluated multiple upstream catalyst formulations for NH_3 generation

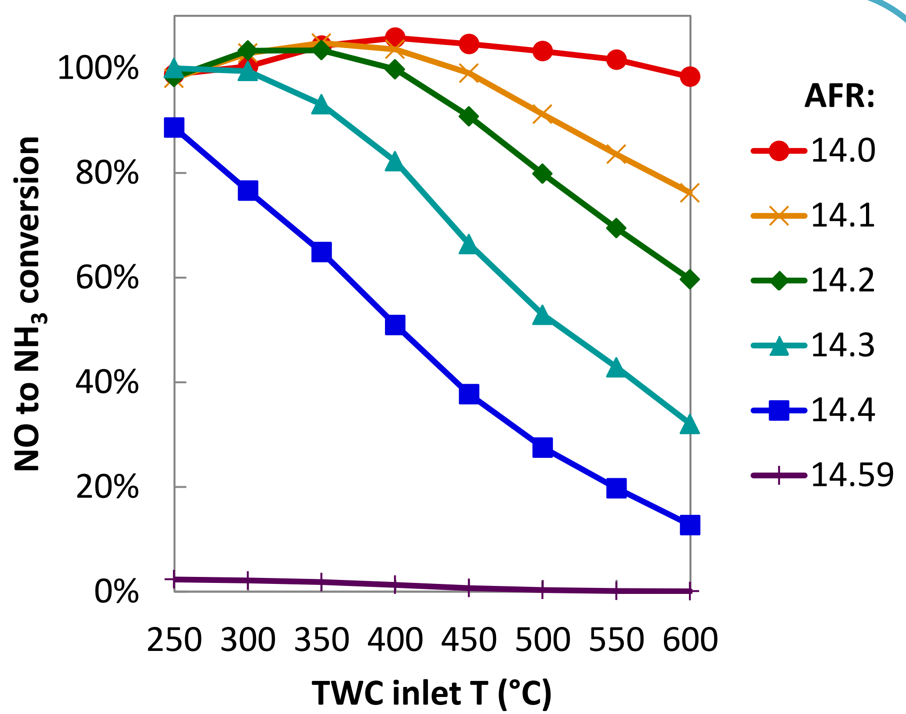




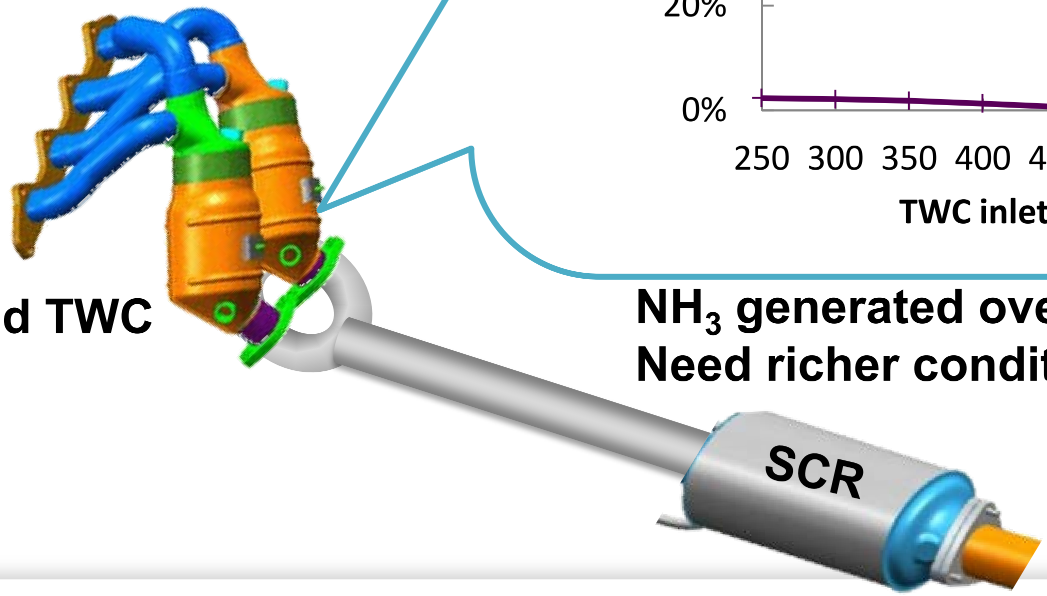
High PGM (Pd-only) best for NH_3 generation

AFR and temperature dictate NH_3 production

Quantified NH_3 generation over Pd-only TWC



Pd TWC

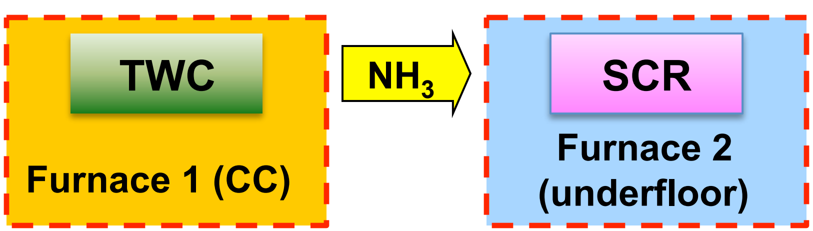


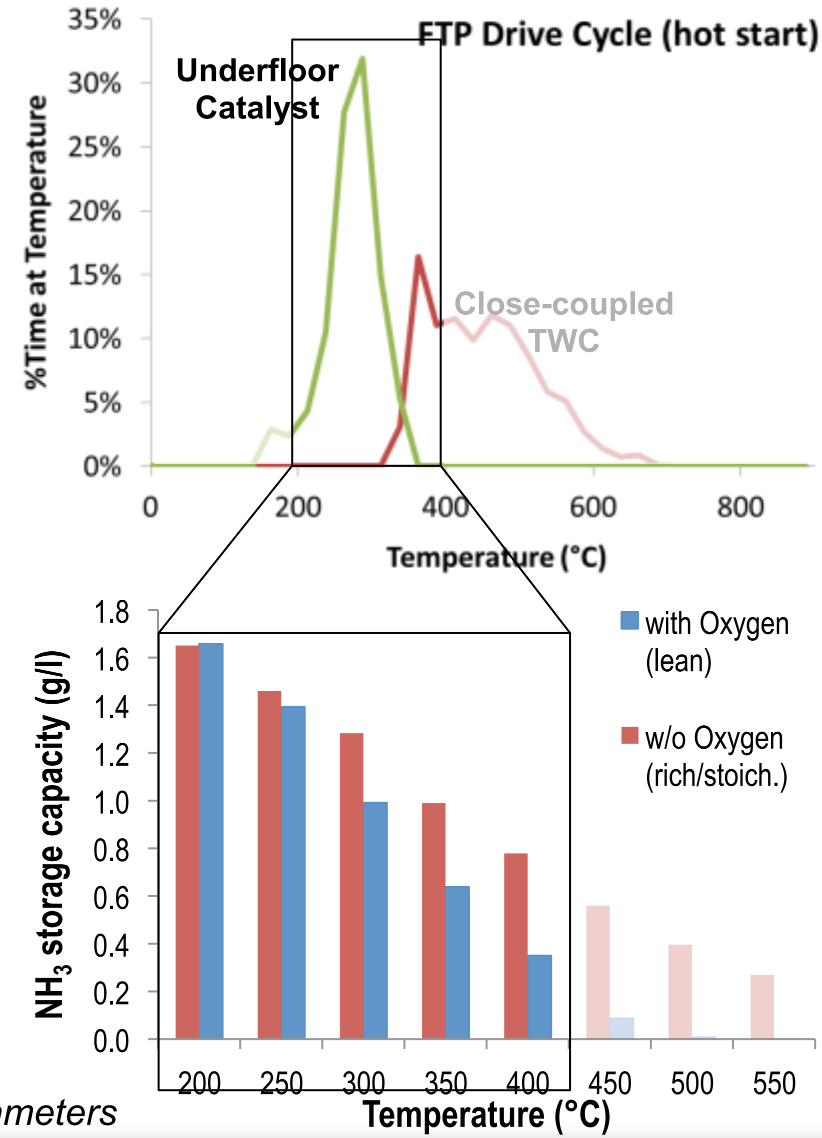
NH_3 generated over wide T window
Need richer conditions at higher T

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 (Hot LA4) with BMW 120i
  - 200-350°C for underfloor catalyst
  - 350-600°C for close-coupled (CC) TWC
- TWC: tunable NH_3 production 250-600°C
- NH_3 production temperatures over CC-TWC mesh well with NH_3 storage temperatures on underfloor 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

Separate furnaces on bench flow reactor mimic CC and underfloor locations



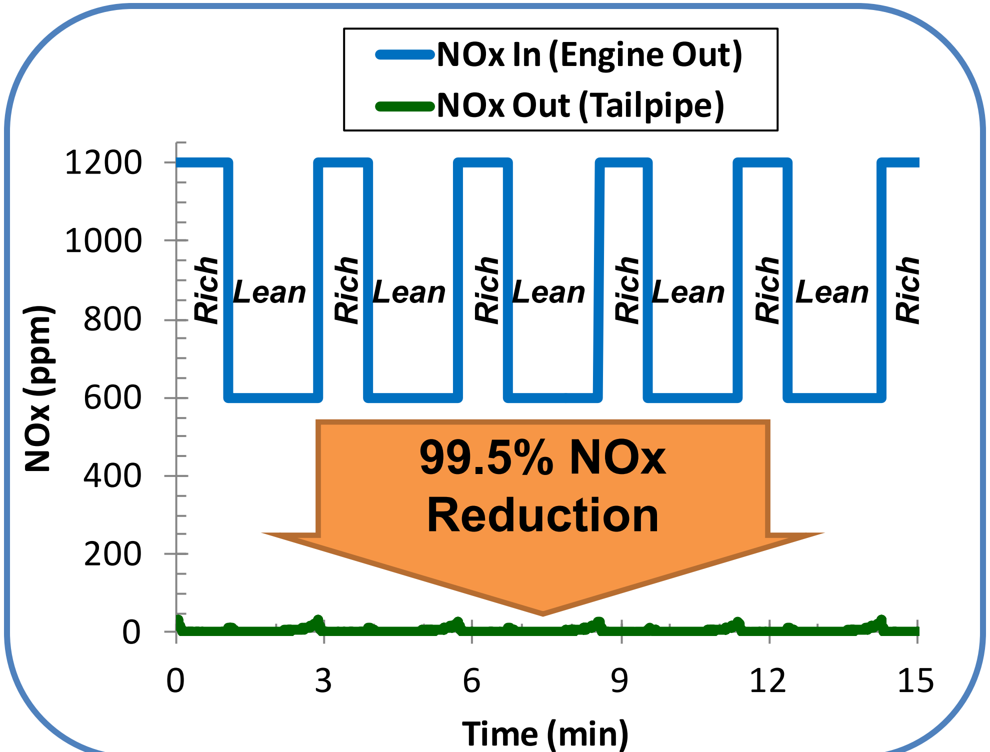


**See extra slides section for more detail on bench flow parameters*

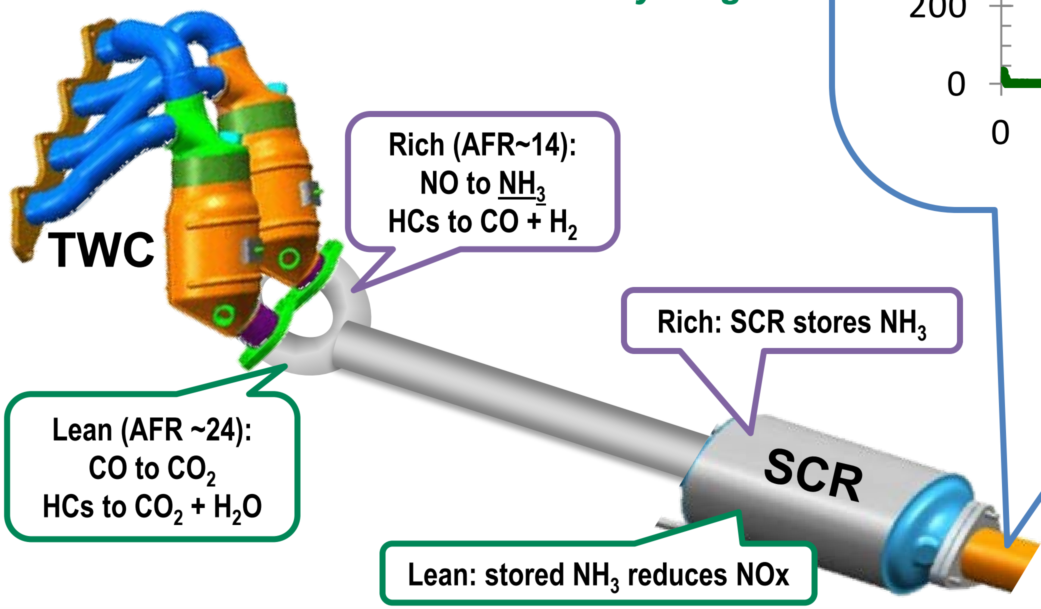
OAK RIDGE NATIONAL LABORATORY
MANAGED BY UT-BATTELLE FOR THE U.S. DEPARTMENT OF ENERGY

Lean gasoline passive-NH_3 SCR (TWC+SCR) demonstrates >99% NOx reduction efficiency on bench flow reactor

- ***>99% NOx reduction efficiency*** achieved by cycling between **lean** (NH_3 production) and **rich** (NH_3 consumption for NOx reduction) modes
- ***Net fuel economy benefit of 5-6%*** over comparable stoich. operation (*estimated* from AFR and 10% lean improvement level)
- CO slip an issue during rich mode



Flow reactor proof of concept: TWC+SCR with lean-rich cycling



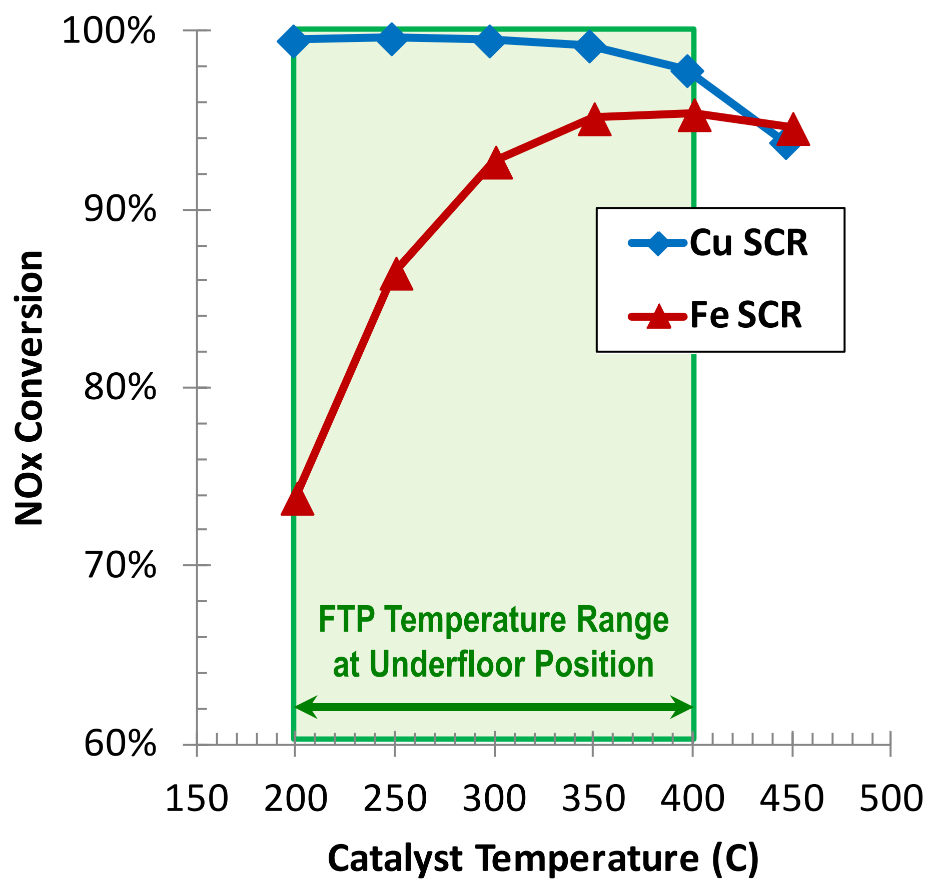
Tailpipe	Avg (ppm)	Max (ppm)
NOx	4	31
NH_3	3	4
N_2O	2	32*
CO	1200	3900

**N_2O made during lean-rich transition*

Cu-SCR gives better NOx conversion than Fe-SCR

- Fe zeolite has much lower NH_3 storage capacity than Cu, limiting lean operating time and high temperature NOx conversion
- Fe zeolite is less active in SCR reactions, resulting in much lower NOx conversions, particularly at low temperatures
- Relation of NH_3 storage and oxidation temperature profiles to NOx conversion activity critical for achieving high NOx conversion

TWC (CC Position)=450°C
Rich AFR=14.0
Rich time varies



Lean Gasoline Engine Research Platform Operational

- Platform based on BMW 120i lean gasoline engine vehicle commercialized in Europe
- Drivven based system allows OEM map operation as well as full control of engine for custom control (Emphasis is chemistry and AFR control, not driveability)

Timeline (since last AMR)

July 2012: Chassis dyno mapping

Aug. 2012: Analysis of mapping data complete

Sept. 2012: installation of engine on dyno and first controller installation (first burn) and sensor/actuator checks

Oct. 2012: map development and controller programming

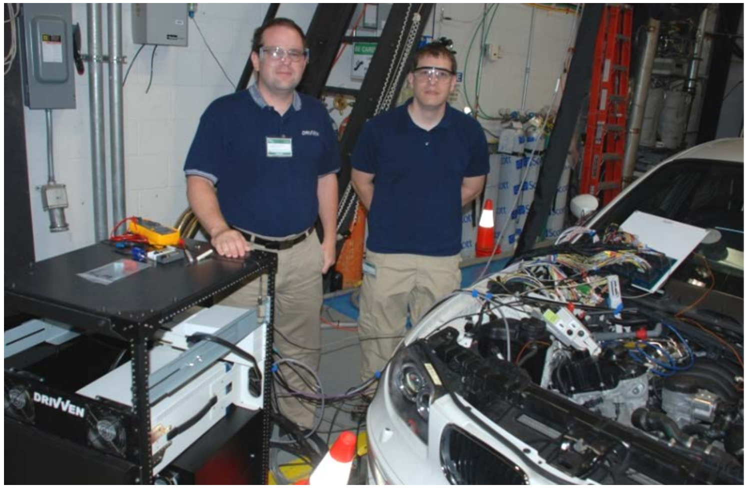
Nov. 2012: controller tuning and implementation

Found difficulty in controlling AFR during lean stratified operation off of OEM map settings (poor cylinder balance and misfires)

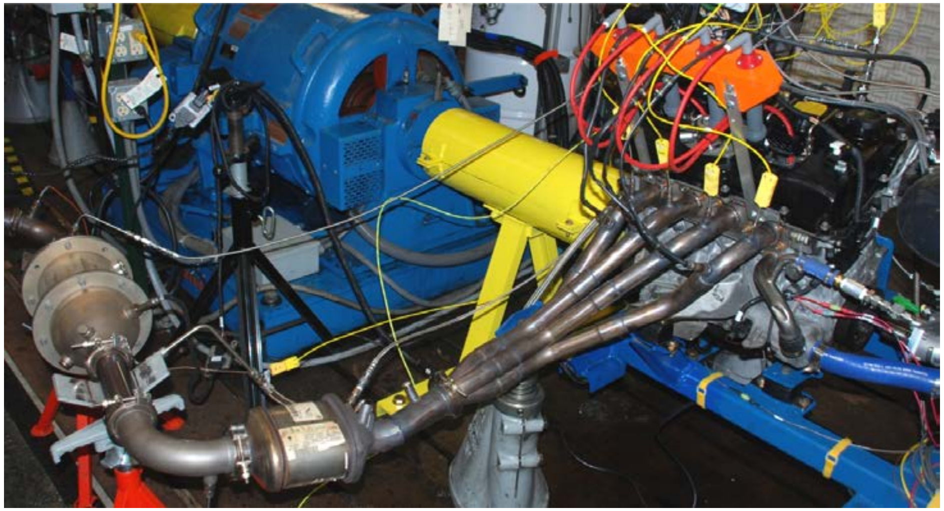
Jan. 2012: re-programming and implementation of UEGO per cylinder AFR control

Feb. 2012: final controller tuning and check of UEGO per cylinder AFR control

Engine Fully Operational!



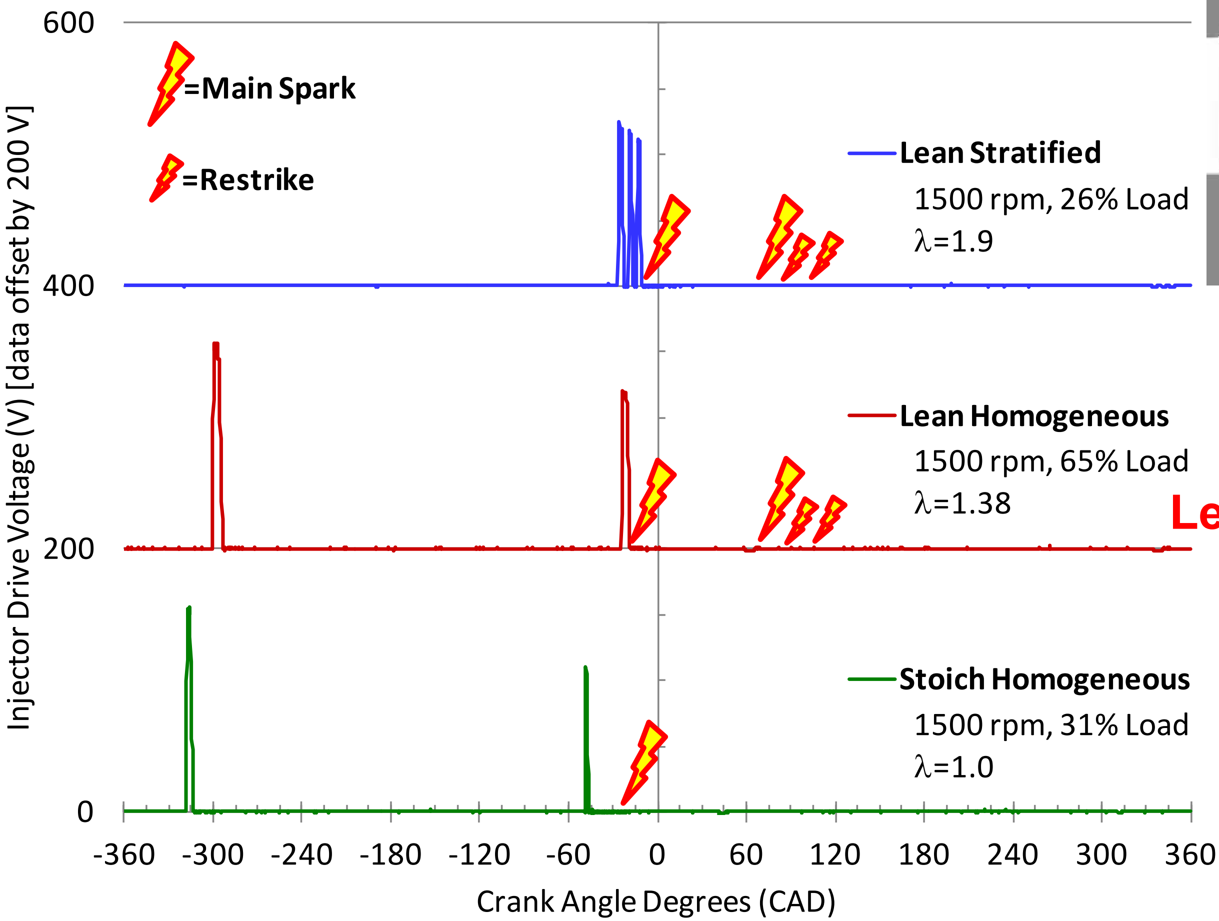
Engine mapping of BMW 120i in ORNL chassis dyno lab with Drivven staff (via subcontract)



Final BMW 120i engine setup installed in ORNL engine dyno lab

BMW 120i Engine Features Three Main Combustion Modes

- Piezoelectric injectors operate at different voltages as well as different duration
- Multiple sparks enable ignition under lean operation



Lean Stratified (λ~1.6-2.2)

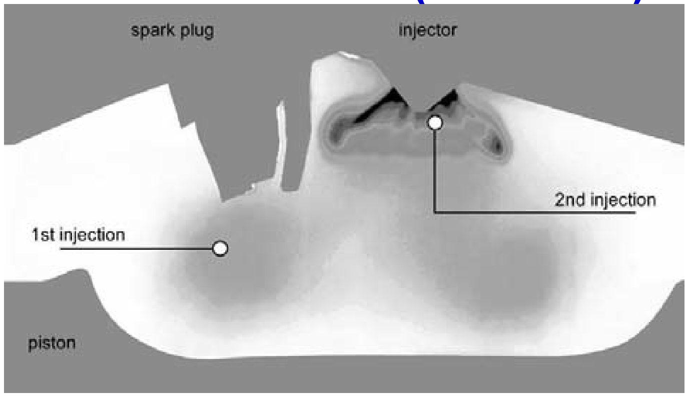


Fig. 5 from SAE 2006-01-1265 (Schwarz, et.al., BMW Group)

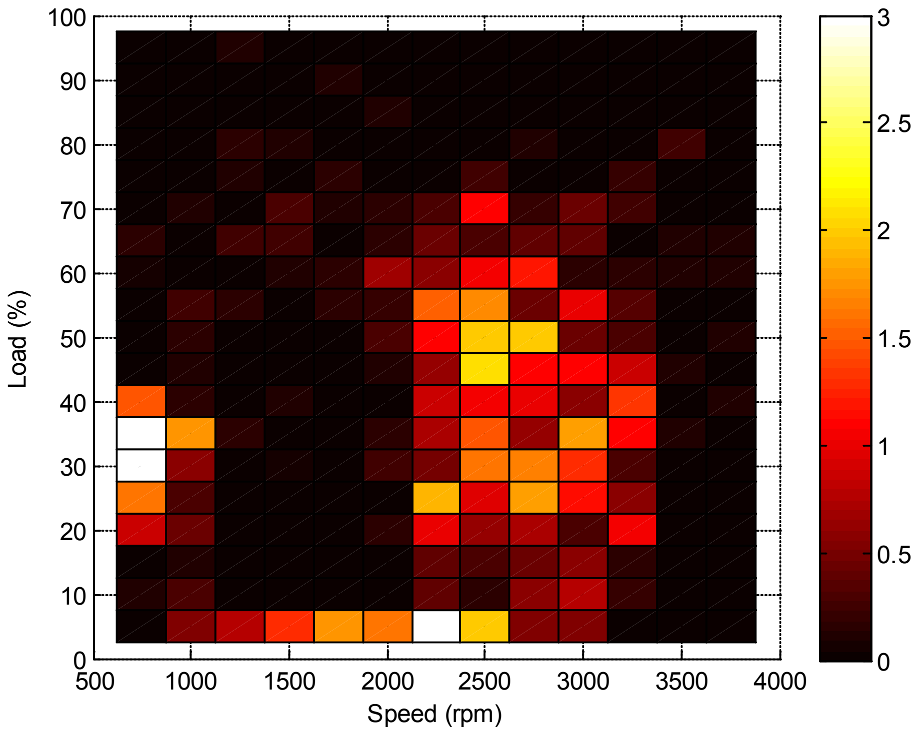
Lean Homogeneous (λ~1.3-1.6)

Stoich Homogeneous (λ=1)

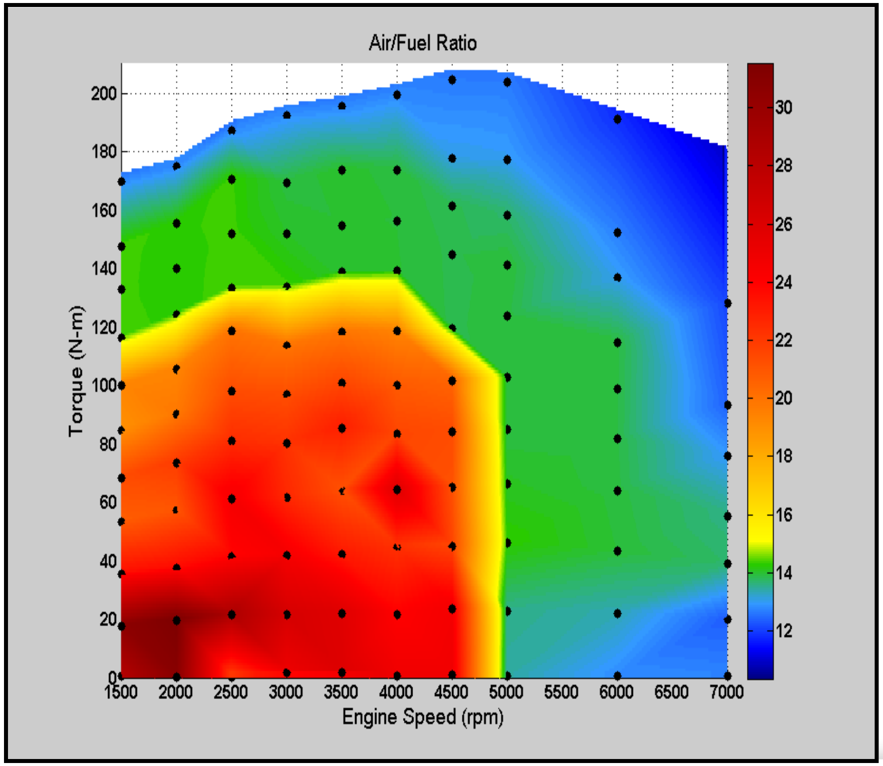
Mode of Operation Depends on Speed and Load

- Lean operation occurs at low loads and speeds
- Hot FTP drive cycle analysis shows a high percentage of operation under low speed, low load
  - Over Hot FTP, 34% of time in stoichiometric or rich modes and 66% time in lean mode
- Load/Speed points for engine dynamometer studies will be based on FTP analysis and recommended points by OEM partners

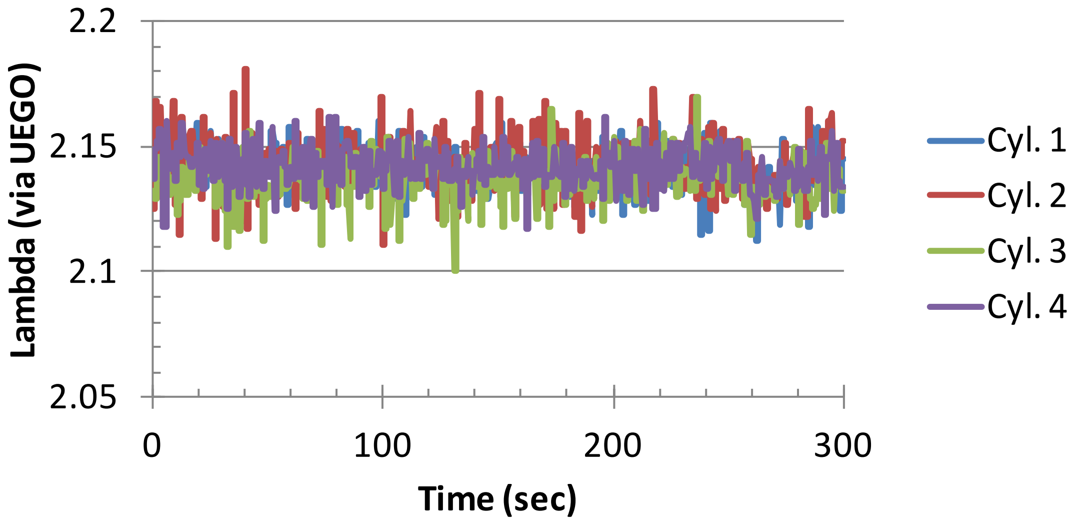
<u>Histogram of operation over FTP drive-cycle</u>
Map shows regions during FTP operation are primarily <3500 rpm and <70% load



<u>AFR as function of load and speed:</u>
Map shows regions of lean operation as well as regions of rich operation for catalyst protection



Example AFR control and LNT cycling

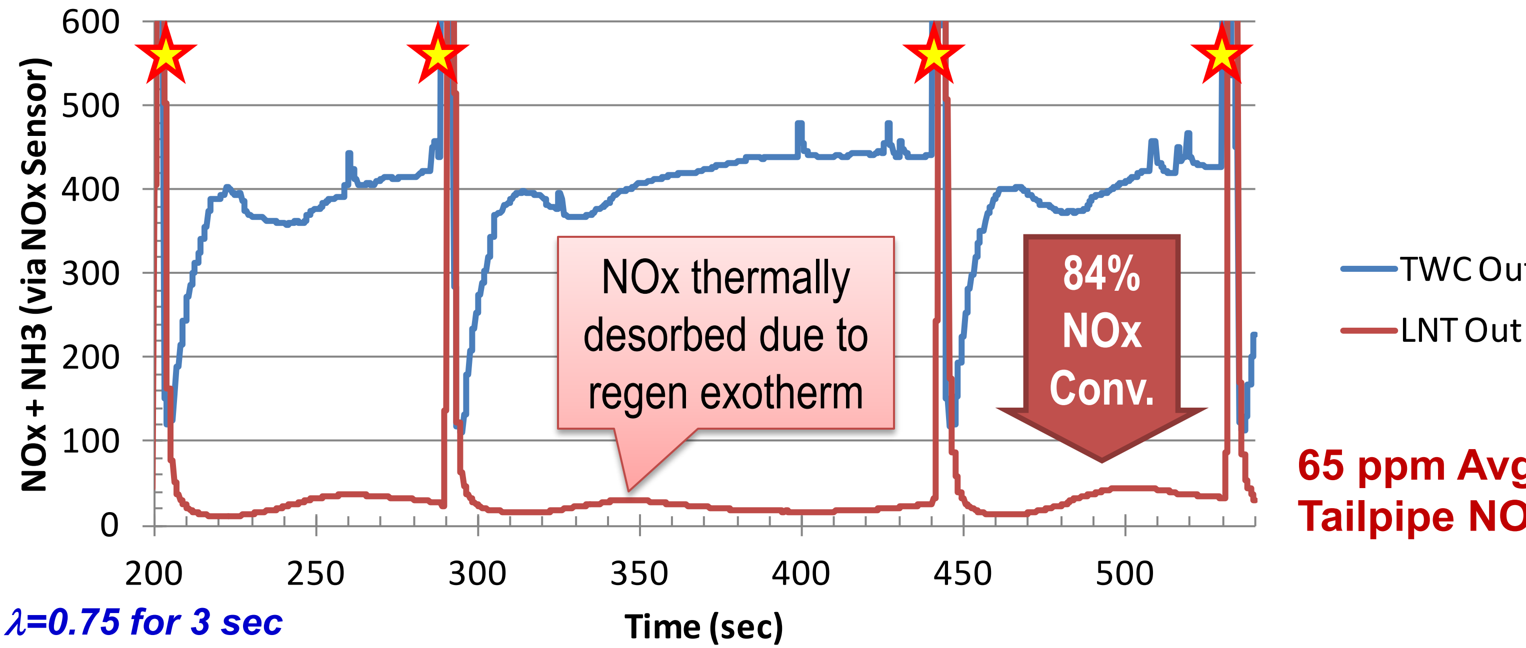


2000 rpm, 25% load, λ=2.14

	Cylinder λ (via UEGO)			
	1	**2**	**3**	**4**
Mean	2.142	2.143	2.137	2.142
Std. Dev.	0.009	0.012	0.010	0.008
COV	0.41%	0.55%	0.45%	0.38%

Engine Out NOx=287 ppm

2000 rpm
35% load
λ=1.72

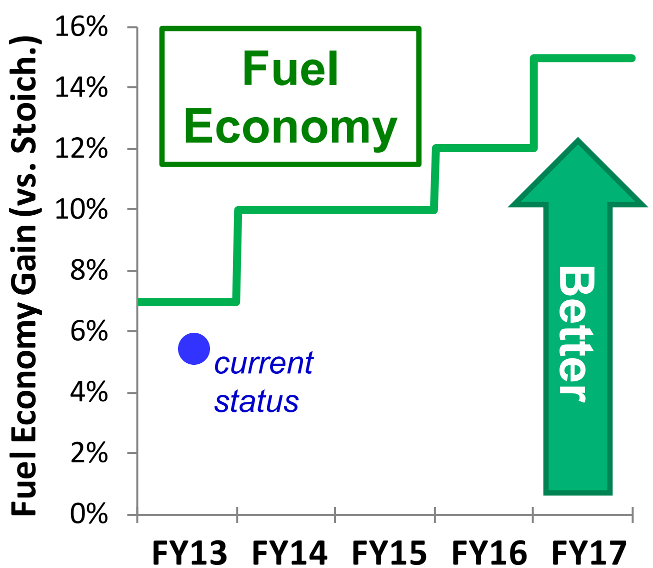


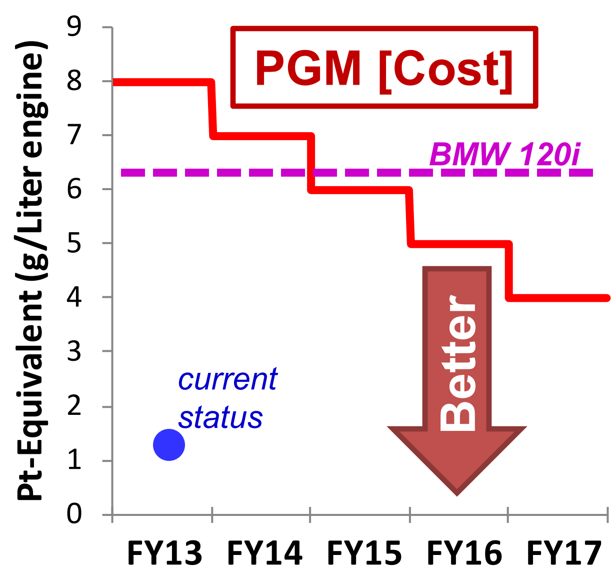
65 ppm Avg. Tailpipe NOx

LNT Regens at λ=0.75 for 3 sec

Future Work

- Continue bench flow reactor studies of catalyst formulation effects (focus on NOx, NH_3, N_2O)
  - Role of NOx storage component on TWC
  - Combination of PGMs, oxygen storage, and NOx storage components
  - TWC+LNT+SCR geometry (LNT at underfloor position/temperature)
  - Effect of S on NH_3 production by TWC
- Conduct studies of TWC+SCR system on engine
  - Investigate role of rich AFR profile on emissions
  - Characterize fuel penalty for passive SCR at representative speed and load points





Estimated from bench reactor data with AFR calculation

Note: CO emissions excessive to Tier 2 Bin 2 goal

For reference, BMW 120i TWC+LNT system=6.3 g/L_{engine}

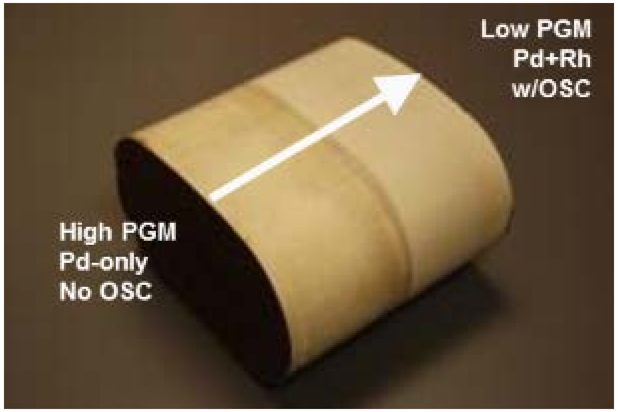
Summary

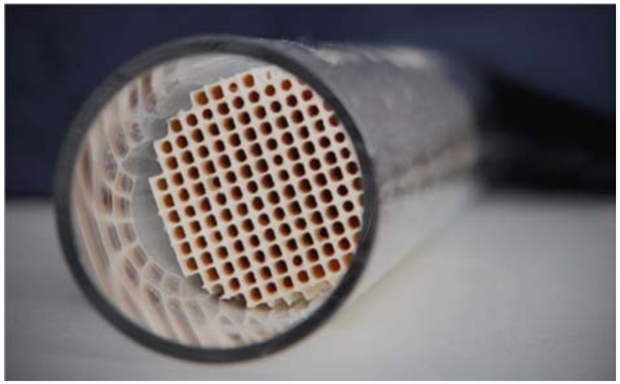
- **Relevance:**
  - Enabling lean gasoline vehicles will significantly impact US petroleum use
- **Approach:**
  - Evaluate catalyst formulations and system geometries on bench flow reactor for cost-effective emissions control (focus on non-urea systems)
  - Study fuel penalty and realistic performance on lean gasoline engine dynamometer research platform
- **Collaborations:**
  - OEMs (GM, Ford, Chrysler) and catalyst supplier Umicore
  - University of South Carolina and the University of Wisconsin
- **Technical Accomplishments:**
  - Completed characterization of NH_3 production from TWC matrix
  - Demonstrated >99% NOx reduction efficiency with TWC+SCR approach on bench flow reactor under simulated exhaust conditions
  - Completed development of lean gasoline engine research platform with full control capability (Drivven system)
- **Future Work:**
  - Further investigation of formulation and S effects for passive SCR and LNT+SCR approach
  - Engine-based studies for fuel penalty assessment and realistic exhaust conditions

Technical Support Slides

Full detail on matrix of TWC formulations for NH_3 production studies

- For bench reactor, focusing on modern TWC technology (Umicore recommended formulations)
  - 1.3L TWC is a 2 formulation combination (<u>combo</u>)
    - Total PGM: 0/4.0/0.16 g/L Pt/Pd/Rh (118 g/ft^3 total PGM)
  - Front 0.6L of TWC is <u>Pd-only</u> no Ce
    - High PGM: 0/6.7/0 g/L Pt/Pd/Rh (190 g/ft^3 total PGM)
    - No ceria-based OSC, but oxygen storage measured
      - Expected to proceed via Pd-O formation
  - Rear 0.7L of TWC is <u>Pd/Rh+Ce</u> w/ Ceria
    - Low PGM: 0/1.1/0.3 g/L Pt/Pd/Rh (40 g/ft^3 total PGM)
  - Investigating each portion individually and in combined form
    - Degreened at 16h at 700C in humidified air (2.7% H_2O)





- <u>LNT</u> is commercial formulation from lean gasoline BMW
  - 2.6L Pt/Pd/Rh = 7/3/1, 3.3 g/L-cat (94 g/ft^3); Ba loading: 20 g/L (560 g/ft^3); Ce: 56 g/L (1600 g/ft^3)
  - Degreened at 16h at 700 C in humidified air (2.7% H_2O)

Full detail on bench flow experiments with TWC and SCR in separate furnaces for temp. control

- Catalysts:

	TWC	SCR
formulation	high Pd	Cu or Fe zeolite
SV (hr^{-1})	70k	28k
T (°C)	300, 450, 600 ***(close coupled)***	200, 250, 300, 350, 400, 450 ***(underfloor)***

- Lean-Rich Cycle Switch Conditions:
  - lean to rich: >20 ppm NOx at SCR out
    - had to increase threshold for Fe zeolite
  - rich to lean: fixed rich time based on empirical optimization to achieve ~ 10 ppm NH_3 slip at SCR out

- Gas compositions:

	Lean	Rich			
AFR	24	14.0	14.1	14.2	14.3
O_2 (%)	8	0.79	0.98	1.08	1.20
NO (ppm)	600	1200			
CO (%)	0	1.8			
H_2 (%)	0	0.6			
C_3H_6 (%)	0	0.1			
H_2O (%)	5	5			
CO_2 (%)	5	5