Emissions Control for Lean Gasoline Engines

Jim Parks (PI), Todd Toops, Josh Pihl, Vitaly Prikhodko

Oak Ridge National Laboratory

Sponsors: Gurpreet Singh, Ken Howden, and Leo Breton

Advanced Combustion Engines Program
U.S. Department of Energy

ACE033
June 9, 2016

This presentation does not contain any proprietary, confidential, or otherwise restricted information
Project Overview

**Timeline**
- Year 1 of 3-year program
- Builds on previous R&D in FY13-FY15

**Budget**
- FY16: $400k (Task 2*)

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**Barriers Addressed**
- Barriers listed in VT Program Multi-Year Program Plan:
  - 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: Emissions control durability

**Collaborators & Partners**
- General Motors
- Umicore
- University of South Carolina
- Cross-Cut Lean Exhaust Emissions Reduction Simulations (CLEERS)

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*Task 2: Lean Gasoline Emissions Control

Part of large ORNL project “Enabling Fuel Efficient Engines by Controlling Emissions” (2015 VTO AOP Lab Call)
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

54.5 mpg CAFE by 2025
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
    - Compliance required: U.S. EPA Tier 3
  - Investigate strategies for 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 has significant impact
    - Potential to save 13 billion gallons gasoline annually
- HOWEVER…emissions compliance needed!!!

References: Transportation Energy Data Book, Ed. 34 (2013 petroleum/fuel use data)
Milestones

Quarterly Milestones

- FY2015, Q4: Simulate transient load/speed operation of passive SCR on BMW lean gasoline engine platform. **Complete**
- FY2016, Q1: Complete bench flow reactor assessment of Pd-only and TWC/NSC formulations for NH₃ production during Passive SCR. **Further studies ongoing**

Annual SMART Milestones

- FY2015: Determine effect of aging and/or poisoning on TWC NH₃ formation through flow reactor experiments. **Complete**
- FY2017: Achieve EPA Tier 3 level emissions with 15% fuel economy gain vs. stoichiometric operation and less than 4 g Pt-equivalent per liter engine with commercial feasibility assessment including material costs. **Further studies ongoing**

GO/NO-GO Decisions

- FY2017, Q2: criteria based on FY2017 SMART Milestone
Approach focuses on catalyst and system optimization of Passive SCR (and LNT+SCR)

**Key Principle:** system fuel efficiency gain depends on optimizing NH$_3$ production during rich operation and NOx reduction during lean operation

**Other Core Principles:**
- Expand range of temperature operation
- Materials must be durable to temperature and poisons (S)
- Understand Pt group metals utilization to minimize cost

Lean
- Minimize engine out Lean NOx emissions

**Lean Gasoline Engine**
- Minimize engine out Rich NOx emissions
- Maximize NOx to NH$_3$ conversion efficiency
- Maximize CO and HC oxidation efficiency

TWC

SCR
- Add NOx storage for lean NOx storage and rich phase NH$_3$ production
- Control emissions during lean-rich transitions
- Minimize NH$_3$ oxidation (keep NH$_3$ stored)
- Maintain high NOx conversion over temperature range
- Minimize engine out Rich CO/HC emissions
- Clean up CO/HC emissions (if needed)
Iterative Bench Reactor + Engine Study Approach

Bench Flow Reactor with cycling and multi-catalyst (close-coupled and underfloor) capabilities

BMW 120i lean gasoline engine platform with National Instruments (Drivven) open controller

Formulation-dependent TWC performance

Define exhaust conditions

Controlled TWC+SCR system performance

TWC performance vs. AFR with realistic HC and H₂ characterization

TWC+SCR performance with refined load step cycle

Realistic TWC+SCR system performance with fuel efficiency measurement

Combustion parameter optimization to maximize NH₃ production

Collaborations with modeling community and CLEERS
Collaborations and Partners

Primary Project Partners

• **GM**
  - guidance and advice on lean gasoline systems via monthly teleconferences

• **Umicore**
  - guidance (via monthly teleconferences) and catalysts for studies (both commercial and prototype formulations)

• **University of South Carolina (Jochen Lauterbach)**
  - Catalyst aging studies with student Calvin Thomas

Additional Collaborators

• **CDTi**: catalysts for studies

• **CLEERS**: Share results/data and identify research needs

• **LANL**: Engine platform used for NH₃ sensor study (M. Mukundan, E. Brosha, C. Kreller)

• **MECA**: GPF studies via Work For Others contract

• **University of Minnesota**: Collaboration on DOE funded project at U of Minn. related to lean GDI PM (PI: Will Northrop)

• **CTS (Filter Sensing Technologies)**: Small business technical assistance on RF sensors for GPF on-board diagnostics

• **DOE VTO Fuel and Lubricant Technology Program**: Engine platform used for ethanol-based HC-SCR studies

(3) Lean GDI PM Projects
Responses to 2015 Reviewers

FY2015 AMR Review
(5 Reviewers)
[scores: 1 (min) to 4 (max)]

<table>
<thead>
<tr>
<th>Category</th>
<th>Score</th>
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<tr>
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Summary of Reviewers’ Feedback:

- Generally positive feedback on:
  - Approach (bench+engine)
  - Collaborations with industry
  - Project design, relevance, future plans
  - Inclusion of S, aging effects

- More interest in OEM perspectives
- Consider deS and desoot fuel penalties and CO/HC emissions
- Consider N₂O (relative to GHG* standard)
- Fundamental questions on ceria, Rh effects
- Want more soot and modeling R&D

Relevant to DOE Objectives?
YES (100%)

Sufficiency of Resources

| Insufficient (40%) | Sufficient (60%) |

*N₂O Greenhouse Gas (GHG) cap is 10 mg/mile
Responses to 2015 Reviewers

Summary of Reviewers’ Feedback:

“…feedback from OEMs on value of passive systems, lessons learned and technical challenges would improve rating to outstanding. Several OEMs indicate passive system challenges are constraining use especially predictability of efficiently producing NH₃.”

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Project Adjustments/Responses:

- Project directly addressing OEM concerns

Project results shared with Ford and FCA. Lots of questions on feasibility, transient control, and net fuel efficiency gain.


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- N₂O data collected; more analysis occurring

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- Surface science studies (DRIFTS/XPS)

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- Will calculate deS/desoot effects
- N₂O data collected; more analysis occurring
- Surface science studies (DRIFTS/XPS)
- Resource limited, but project leveraged for (3) PM/GPF projects & CLEERS database

*N₂O Greenhouse Gas (GHG) cap is 10 mg/mile
Summary of Technical Accomplishments

- **Bench Reactor Studies:**
  - Cold start selectivity as a function of $\lambda$ defined for catalyst sample matrix

- **Engine Studies:**
  - Completed study of effects of NH$_3$:NOx on fuel efficiency and NOx/NH$_3$/N$_2$O slip

- **Aging and S Effects:**
  - Materials characterization conducted on Malibu-1 catalyst (PGM size) after aging
  - S exposure study during lean-rich cycling completed (data analysis ongoing)

### Catalyst Sample Matrix [OSC=oxygen storage capacity; NSC=NOx storage capacity]

<table>
<thead>
<tr>
<th>sample ID</th>
<th>Description</th>
<th>Pt (g/l)</th>
<th>Pd (g/l)</th>
<th>Rh (g/l)</th>
<th>OSC</th>
<th>NSC</th>
</tr>
</thead>
<tbody>
<tr>
<td>Malibu-1</td>
<td>Front half of TWC</td>
<td>0</td>
<td>7.3</td>
<td>0</td>
<td>N</td>
<td>N</td>
</tr>
<tr>
<td>Malibu-2</td>
<td>Rear half of TWC</td>
<td>0</td>
<td>1.1</td>
<td>0.3</td>
<td>Y</td>
<td>N</td>
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<tr>
<td>Malibu-combo</td>
<td>Full TWC</td>
<td>0</td>
<td>4.0</td>
<td>0.16</td>
<td>Y</td>
<td>N</td>
</tr>
<tr>
<td>ORNL-1</td>
<td>Pt + Pd + Rh</td>
<td>2.47</td>
<td>4.17</td>
<td>0.05</td>
<td>Y</td>
<td>Y</td>
</tr>
<tr>
<td>ORNL-2</td>
<td>Pd + Rh</td>
<td>0</td>
<td>6.36</td>
<td>0.14</td>
<td>N</td>
<td>N</td>
</tr>
<tr>
<td>ORNL-6</td>
<td>Pd</td>
<td>0</td>
<td>6.50</td>
<td>0</td>
<td>N</td>
<td>N</td>
</tr>
<tr>
<td>ORNL-5</td>
<td>Pd + OSC high</td>
<td>0</td>
<td>6.50</td>
<td>0</td>
<td>H</td>
<td>N</td>
</tr>
<tr>
<td>ORNL-4</td>
<td>Pd + OSC med</td>
<td>0</td>
<td>4.06</td>
<td>0</td>
<td>M</td>
<td>N</td>
</tr>
<tr>
<td>ORNL-3</td>
<td>Pd + OSC low</td>
<td>0</td>
<td>1.41</td>
<td>0</td>
<td>L</td>
<td>N</td>
</tr>
</tbody>
</table>
Extensive flow reactor investigations show impact of TWC formulation, $\lambda$ on light-off performance for passive SCR

- Gas composition has a large effect on light-off performance
  - best combination of NOx reduction, NH$_3$ production, and HC conversion achieved at $\lambda$ 0.97 for all 5 TWCs evaluated
  - same as optimum rich $\lambda$ for passive SCR operation

- Conversion of NO and CO, production of NH$_3$ all light-off at around 150°C

- Formulation has only a minor influence on light-off performance among TWCs tested
  - T50 (Temperature at >50% conversion) and T90 (Temperature at >90% conversion) shown at right (dark bars=T50; light bars=T90)
\( \lambda \) affects NH\(_3\) and N\(_2\)O formation during light-off as well

- NH\(_3\) formed at >80% yield starting at \( \sim 150^\circ\text{C} \) for Malibu-1 catalyst (Pd)
- Temperature for N\(_2\)O to decrease to <10% yield a function of \( \lambda \)
- Note: *inlet temperature* shown
\( \lambda \) affects NH\(_3\) and N\(_2\)O formation during light-off as well

- NH\(_3\) formed at >80% yield starting at \( \sim 150^\circ\)C for Malibu-1 catalyst (Pd)
- Temperature for N\(_2\)O to decrease to <10% yield a function of \( \lambda \)
- Note: inlet temperature shown

\( \lambda = 0.97 \) during cold start gives best NH\(_3\) production with least N\(_2\)O

\[ \text{Temperature for NH}_3 > 80\% \text{ yield (C)} \]

\[ \text{Temperature for N}_2\text{O} < 10\% \text{ yield (C)} \]
Engine-based studies show NH$_3$:NOx>1 critical to achieving highest NOx conversion

- NOx conversion reaches >99% at NH$_3$:NOx=1.13
- Increasing NH$_3$:NOx to 1.20 results in NH$_3$ slip
- N$_2$O primarily forms over SCR during lean phase (~2ppm) and not affected by NH$_3$:NOx
- N$_2$O spike over TWC observed at lean to rich transition (~8ppm)
Reducing cycle timing enables >99% NOx conversion at NH$_3$:NOx=1 (engine studies)
PGM sintering occurs mostly in early aging hours but continues to 100 hours.

Recall FY15 Aging Data

Pd map and HAADF for 50hr aged Malibu-1

This research was performed, in part, using instrumentation (FEI Talos F200X S/TEM) provided by the Department of Energy, Office of Nuclear Energy, Fuel Cycle R&D Program and the Nuclear Science User Facilities. STEM images collected by Michael Lance (ORNL).
Sulfur impact evaluated on hydrothermally aged TWC while cycling; some effects observed but recoverable

1. Began with Hydrothermally aged Pd TWC (Malibu-1)

Note: Preliminary Results Shown
Sulfur impact evaluated on hydrothermally aged TWC while cycling; some effects observed but recoverable.

1. Began with hydrothermally aged Pd TWC (Malibu-1)
2. S aged TWC while lean-rich cycling (fixed load shown)

Note: Preliminary Results Shown
Sulfur impact evaluated on hydrothermally aged TWC while cycling; some effects observed but recoverable

1. Began with Hydrothermally aged Pd TWC (Malibu-1)
2. S aged TWC while lean-rich cycling (fixed load shown)
3. After S aging, deS at 650°C and λ=0.97 while lean-rich cycling
4. After deS: 350°C and 650°C data show good performance recovery

Note: Preliminary Results Shown
Remaining Challenges

- Improve system level fuel economy (reduce NH₃ production fuel penalty)
- Address catalyst performance during transients and rich-lean transitions
- Determine technique to enable NSC functionality over temperature range
- Broaden aging studies to include SCR

Fuel Consumption Benefit (vs. Stoich.):

**Fuel Economy**

- FY13
- FY14
- FY15
- FY16
- FY17

**PGM [Cost]**

-更好地

**Size**

- 蓝色
- 金色
- 绿色

**Bench Flow Reactor Status**

**Engine-Based Study Status**

**Bench Flow Reactor Status**

- FY13
- FY14
- FY15
- FY16
- FY17

**Engine-Based Study Status**

- FY13
- FY14
- FY15
- FY16
- FY17

**Better**

**Fuel Consumption Benefit (vs. Stoich.)**

- max
- calc

**Pt-Equivalent (g/Liter engine)**

- BMW 120i
- TWC+LNT

**Catalyst Volume (liters)**

- BMW 120i
- Passive SCR

**acağız**
Future Work: Addressing Remaining Challenges

• Catalyst formulation studies on bench flow reactor
  - Examine SCR formulations for NH$_3$ oxidation and compare resulting data with engine-based data

• Continue aging studies including studying select prototype formulations
  - Perform materials characterization [TEM, BET/Chemi, XRD]
  - Continue aging of TWC formulations with sulfur while cycling
  - Characterize aged SCR (aging complete)

• Continue engine-based studies to maximize system fuel efficiency
  - Repeat existing studies with TWC formulation that contains NSC
  - Understand transient and switching effects on Passive SCR/LNT+SCR
  - Define method to predict transient emissions and fuel efficiency
## Summary

<table>
<thead>
<tr>
<th>Relevance</th>
<th>Enabling lean gasoline vehicles will significantly reduce US petroleum use</th>
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</table>
| **Approach** | Focus on non-urea Passive SCR and LNT+SCR  
Evaluate catalyst formulations on bench flow reactor for cost-effective emissions control  
Study fuel penalty and realistic performance on lean gasoline engine research platform |
| **Collaborations** | Primary: GM, Umicore, and Univ. of South Carolina  
Additional: CDTi, LANL, MECA, Univ. of Minnesota, CTS(FST) |
| **Technical Accomplishments** | Characterized NOx/HC/CO light-off and NH$_3$/N$_2$O formation vs. temperature on bench flow reactor of Umicore catalyst matrix  
Determined optimal NH$_3$:NOx of 1.13 for fuel efficiency and NOx/NH$_3$/N$_2$O slip performance  
Measured PGM peak size shift from 3.5 nm to ~37 nm with aging time  
99% NH$_3$ yield at 350°C after S during lean-rich cycling + deS |
| **Future Work** | Bench reactor, aging, and engine studies ongoing toward project goals of fuel efficiency and cost (Pt-equivalent) |
Technical Backup slides
## Project Goals Defined by Industry

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.

<table>
<thead>
<tr>
<th></th>
<th>FY13</th>
<th>FY14</th>
<th>FY15</th>
<th>FY16</th>
<th>FY17</th>
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<tbody>
<tr>
<td>Fuel economy gain over stoichiometric</td>
<td>7%</td>
<td>10%</td>
<td>10%</td>
<td>12%</td>
<td>15%</td>
</tr>
<tr>
<td>Total emissions control devices Pt* (g/L_engine)</td>
<td>8</td>
<td>7</td>
<td>6</td>
<td>5</td>
<td>4</td>
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<table>
<thead>
<tr>
<th></th>
<th>5-year Average ($/troy oz.)</th>
<th>Pt-equivalent</th>
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<tbody>
<tr>
<td>Platinum</td>
<td>$ 1,504/troy oz.</td>
<td>1.0</td>
</tr>
<tr>
<td>Palladium</td>
<td>$ 463/troy oz.</td>
<td>0.3</td>
</tr>
<tr>
<td>Rhodium</td>
<td>$ 3,582/troy oz.</td>
<td>2.4</td>
</tr>
<tr>
<td>Gold</td>
<td>$ 989/troy oz.</td>
<td>0.7</td>
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* - will use Pt equivalent cost to account for different costs of Pt, Pd and Rh; 5-year average value fixed at beginning of project.
BMW 120i engine features three main combustion modes

- Spray guided combustion system design
- Piezoelectric injectors operate at different voltages as well as different durations
- Multiple sparks enable ignition under lean operation
- In addition to three main combustions modes, there is also an OEM rich homogeneous mode for LNT control of NO\textsubscript{X} emissions to meet EURO V NO\textsubscript{X} emission standards
Conducted transient flow reactor experiments to estimate TWC effects on fuel consumption

- Used feedback-controlled cycles on flow reactor to evaluate dynamic TWC response in context of passive SCR
- Evaluated two different simulated engine cycles (fixed load, load step)

<table>
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<tr>
<th>Rich</th>
<th>Lean</th>
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<tbody>
<tr>
<td>λ</td>
<td>0.95 0.96 0.97 0.98 0.99 1.00</td>
</tr>
<tr>
<td>O₂ (%)</td>
<td>0.96 1.02 1.07 1.13 1.17 1.22</td>
</tr>
<tr>
<td>CO (%)</td>
<td>2.0 1.8 1.6 1.4 1.2 1.0</td>
</tr>
<tr>
<td>H₂ (%)</td>
<td>1.0 0.9 0.8 0.7 0.6 0.5</td>
</tr>
<tr>
<td>NO (ppm)</td>
<td>600 (or 1200)</td>
</tr>
<tr>
<td>C₃H₈ (ppm C₁)</td>
<td>3000</td>
</tr>
<tr>
<td>H₂O (%)</td>
<td>11</td>
</tr>
<tr>
<td>CO₂ (%)</td>
<td>11</td>
</tr>
<tr>
<td>TWC SV (hr⁻¹)</td>
<td>27000 (or 60000)</td>
</tr>
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- Compositions & flows selected to mimic BMW GDI engine exhaust
- Space velocity changed with λ and load
- C₃H₈ chosen as challenging HC