Investigation on continuous soot oxidation and NOx reduction by SCR coated DPF

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Outline

• Requirements for combined SCR-DPF catalyst
• Evaluation of CSI catalyst for NO$_x$ removal and soot oxidation
• Engine tests on continuous soot oxidation and NO$_x$ reduction by combined SCR-DPF
• Summary
SCR-DPF. Two Functions:
NO\textsubscript{x} reduction and Soot oxidation

### SCR with ammonia (urea)

- 4NO + 4NH\textsubscript{3} + O\textsubscript{2} → 4N\textsubscript{2} + 6H\textsubscript{2}O
  (standard SCR reaction)
- NO + NO\textsubscript{2} + 2NH\textsubscript{3} → 2N\textsubscript{2} + 3H\textsubscript{2}O
  (fast SCR reaction)
- NO + ½ O\textsubscript{2} → NO\textsubscript{2}
- 6 NO\textsubscript{2} + 8 NH\textsubscript{3} → 7 N\textsubscript{2} + 12 H\textsubscript{2}O
  (slow SCR reaction)

### SOOT OXIDATION

- C + O\textsubscript{2} → CO\textsubscript{2}
- C + ½ O\textsubscript{2} → CO
- C + NO\textsubscript{2} → CO + NO
- C + 2 NO\textsubscript{2} → CO\textsubscript{2} + 2NO
CSI SCR catalysts activity on Flow-Through substrates

More than 90% NOx reduction

Practically 100% efficiency had been measured at every tested point on DOC SCR systems aged on engine for 250 hrs at 500 °C.

Various steady state and transient tests:

> 90% NOx reduction

Re: “Two Catalyst Formulations – One solution for NOx after-treatment systems”
DEER 2008, August 6th.
**Termogravimetric/Derivative Thermal Analysis (TG/DTA) test**

Lab tool to evaluate catalysts soot oxidation quality

Gas composition:
- 0 or 400 ppm NO₂
- 10% O₂
- N₂ – balance

Temperature: 20°C → 500/700°C at 10 °C/min

Soot (from engine) mixed with catalyst at 5/95 wt/wt

**Soot Oxidation without catalyst:** peak of oxidation: 660°C = soot combustion
Soot oxidation by LT-SCR. TG/DTA tests (@TCC)

Catalytic soot oxidation

Catalytic Soot oxidation had been observed with and without NO₂ in feed gas.

Soot Oxidation with LT-SCR catalyst:
- with NO₂ in feedgas peak of oxidation: 425 °C
- without NO₂ in feedgas peak of oxidation: 433 °C
Balance Point Temperature Test (@ ECS)
No urea injection. 5.9L engine. 14L SCR-DPF.

BPT ~ 300 °C
> 90% soot regeneration ~ 440°
SCR-DPF:
NO\textsubscript{x} Reduction and Soot oxidation

- CSI Low-temperature SCR catalyst showed
  - good SCR activity at various conditions and
  - also soot oxidation properties:
    - TG/DTA – lowering peak of soot oxidation by > 200 °C
    - Balance Point Temperature on engine test was found to be around 300 °C (in presence of NO\textsubscript{2})

- Big Question: would LT-SCR catalyst keep SCR and Soot oxidation properties simultaneously?

- Questions addressed in the current study:
  - does presence of soot on filter affect SCR performance
  - does urea-injection affect soot oxidation properties
  - does NO\textsubscript{2} concentration limit SCR or soot performance
Test Set Up (@ Eminox)

Engine: Cummins N14 Eu2 525 hp

SCR-DPF: CS-585 on 9”x12” 300/12 high-porosity cordierite

Set up enables to control:
- temperature,
- flow rate,
- injection stoichiometry
- NO/NO₂ ratio

NOx, NO, NO₂, T, P static (emissions measured with CL)
Adblue injection rate
T, P static, exh flow rate
T, P static, NOx, NO, NO₂, NH3 (emissions measured with FTIR)
Example of a Test on soot loaded SCR-DPF
(340°C; 35Khr⁻¹; NO₂/NOₓ = 0.3, 4.9 g/L soot)
NOₓ conversion; Ammonia slip; ANR

\[ T = 340°C, SV = 35K, \text{NO}_2/\text{NO}_x = 0.3 \]

\[ \text{97.6%} \]
Details of a test on soot loaded SCR-DPF
(340°C; 35Khr⁻¹; NO₂/NOₓ = 0.3, 4.9 g/L soot)
NO,NO₂ – inlet and outlet; ANR

Switched both analysers to downstream location to check correlation.
Details of a test on soot loaded SCR-DPF (@ Eminox)
(340°C; 35Khr⁻¹; NO₂/NOx = 0.3, 4.9 g/L soot)
NO,NO₂ outlet; Backpressure

urea is injected

18% soot removal based on weight of the filter
SCR activity with and without soot

No effect of soot was observed

\[
T = 310 \, ^\circ C, \text{ SV}=32K \, \text{hr}^{-1}, \quad \frac{\text{NO}_2}{\text{NO}_x} = 0.5, \quad 0 \, \text{g soot}
\]

\[
T = 310 \, ^\circ C, \text{ SV}=32K \, \text{hr}^{-1}, \quad \frac{\text{NO}_2}{\text{NO}_x} = 0.5, \quad 5.3 \, \text{g soot}
\]

\[\text{ANR}\]

\[\text{NO}_\text{in} \quad \text{NO}_2\text{in} \quad \text{NO}_\text{out} \quad \text{NO}_2\text{out} \quad \text{NH}_3\text{out} \quad \text{ANR}\]

\[\text{NO}_2\text{ to NO conversion due to soot oxidation did not affect SCR performance – the same high (>97%) NO}_x\text{ conversion at 10 ppm NH}_3\text{ was observed during the tests}\]
SCR activity with and without soot
Monitoring Back Pressure

\[ T = 310 \, ^\circ C, \, SV=32K \, hr^{-1}, \, NO_2/NO_x = 0.5, \, 0 \, g \, soot \]

\[ T = 310 \, ^\circ C, \, SV=32K \, hr^{-1}, \, NO_2/NO_x = 0.5, \, 5.3 \, g \, soot \]

Soot oxidation was relatively low at these conditions – BP lowered about 0.5 kPa BP during 45 min of the test.
Evaluating effect of NO$_2$/NO$_x$ ratio
SCR performance was not influenced by NO$_2$/NO$_x$ ratio at 300°C

T = 310 °C, SV=32K hr$^{-1}$, NO$_2$/NO$_x$ = 0.5, 5.3 g soot

T = 300 °C, SV=32K hr$^{-1}$, NO$_2$/NO$_x$ = 0.3, 4.6 g soot

Competition for NO$_2$ between soot oxidation reaction and SCR reaction did not affect SCR performance – the same high NO$_x$ conversion (>98% at 10 ppm ammonia slip) was observed during the tests.
Evaluating effect of NO$_2$/NO$_x$ ratio
Soot oxidation – no clear effect

T = 310 °C, SV=32K hr$^{-1}$, NO$_2$/NO$_x$ = 0.5, 5.3 g soot
50’ test: % soot removal – n/a

T = 300 °C, SV=32K hr$^{-1}$, NO$_2$/NO$_x$ = 0.3, 4.6 g soot
70’ test: 33.3% soot removal

Based on BP measurements no difference in soot oxidation had been observed at 300 C for both tests.
Based on weight of the filter 33.3% soot was removed during 70 min of the test with 30% NO$_2$. 
Ammonia slip vs. NOx conversion for SCR-DPF
More than 90% NO$_x$ @ 10 ppm NH$_3$ slip

More than 90% of NO$_x$ reduction at 10 ppm NH$_3$ slip had been observed for all tests regardless soot loading, SV, NO$_2$/NO$_x$ ratio and temperatures within evaluated range of conditions.

Significant soot removal at T> 300 C.
Summary of Observations
Simultaneous NO$_x$ and Soot removal

- No negative effect of soot loading (up to 6 g/L) was observed on NO$_x$ oxidation function of SCR-DPF
- SCR performance was not affected by concentration of NO$_2$ in feed gas (within 0.3-0.5 NO$_2$/NO$_x$ range)
- Competition for NO$_2$ between SCR and passive soot oxidation did not affect rate of soot removal (within 0.3-0.5 NO$_2$/NO$_x$ range)
- We did not observe slowing down in rate of soot oxidation due to urea injection.
- Soot removal was continued when NO$_x$ reduction was higher than 90% and no NO$_2$ was available for soot oxidation

- Overall: simultaneous NO$_x$ and soot removal had been confirmed.
People working on the project

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Santa Monica Mtns. Pt. Diesel.

DEER. Emission Control Technologies
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