Application of the AT Research Capabilities:

Investigation of Diesel Soot Oxidation and of the Catalysts Degradation

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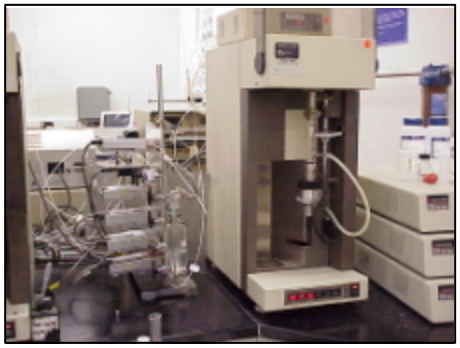
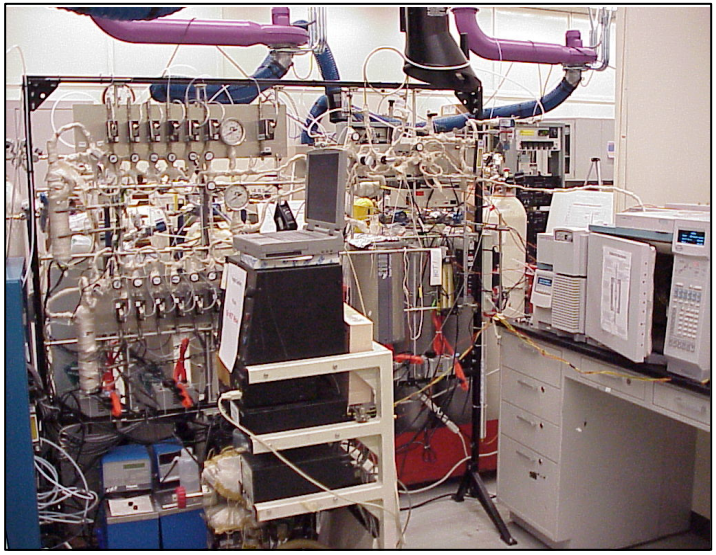
Outline:

- Diesel soot oxidation
  - = f (soot origin/properties)
  - = f (O_2/H_2O)
  - = f (NO_2)

TGA

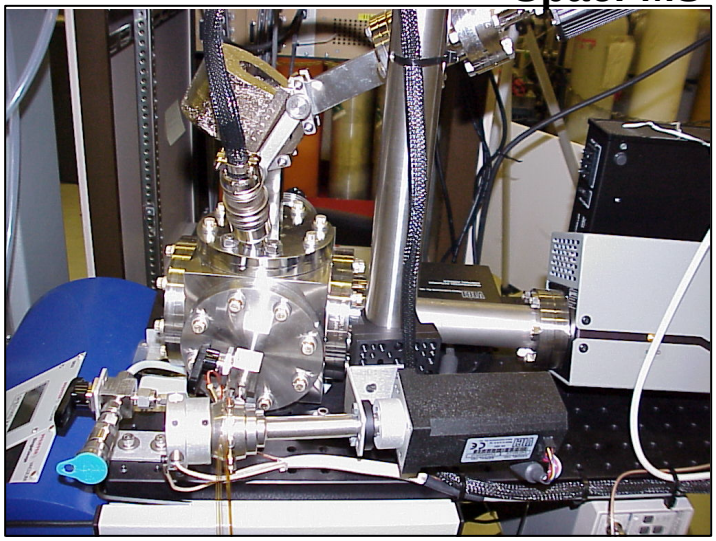


Micro-Reactor



- Catalysts degradation
  - – Micro-reactor probing
  - – Localized heat evolution (Spaci-MS)

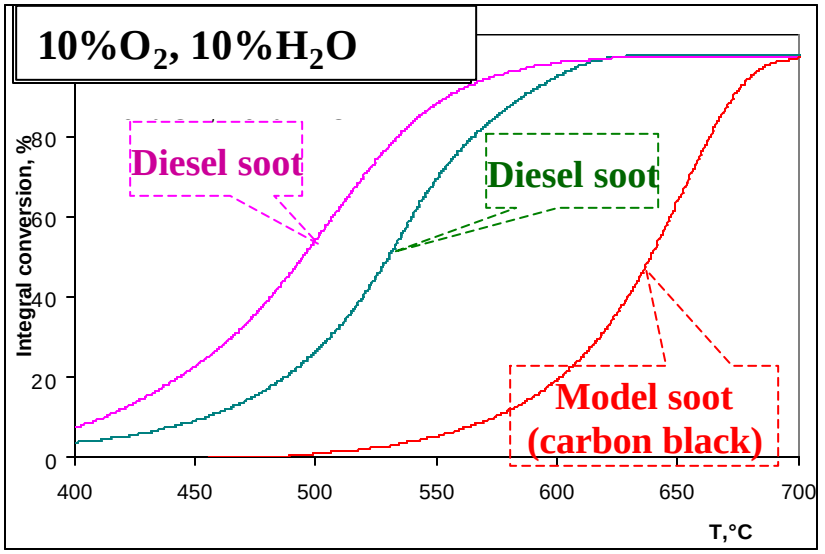
Spaci-MS



Overview of Diesel Soot Oxidation Study (*)

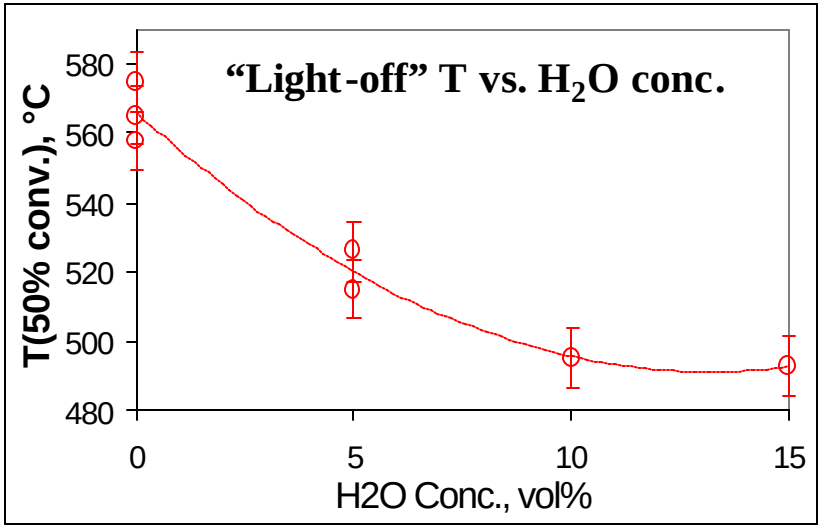
Different samples of soot:

- differences in light-off of diesel soot samples (up to ~40°C)
- large difference with the model soot sample (quantitatively and trends)



Effect of H_2O on oxidation by O_2

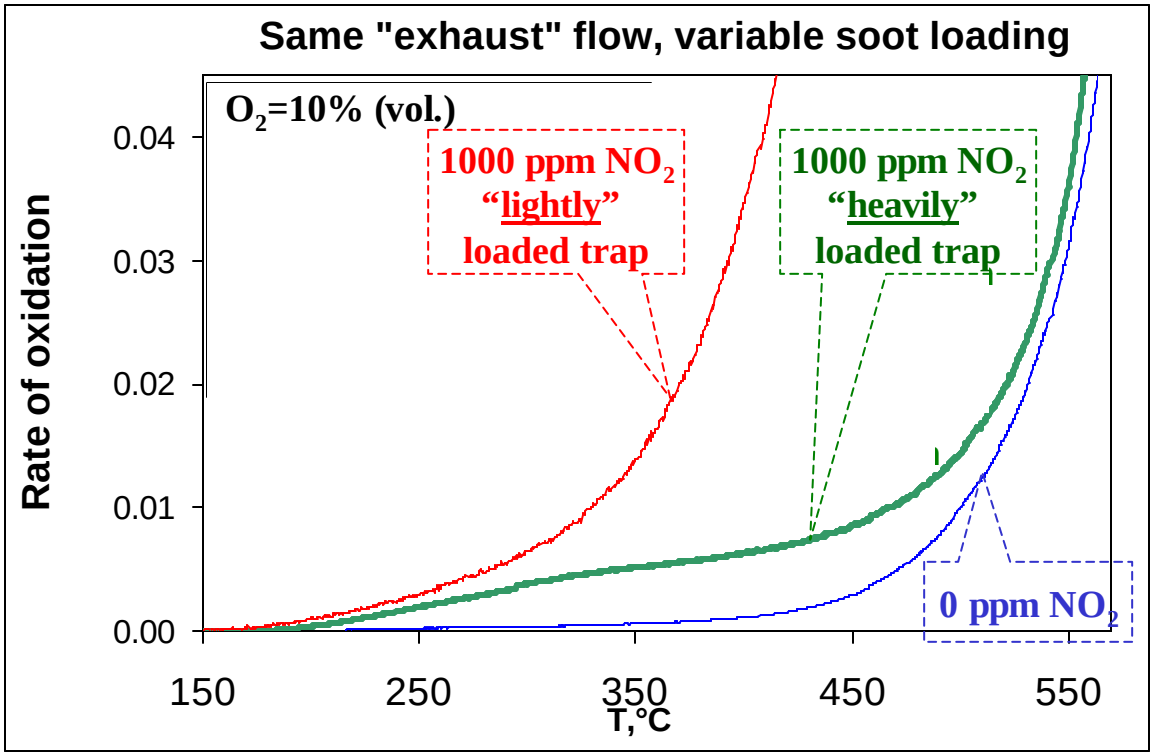
- synergistically boosts soot oxidation (lowered "light-off" by ~40-80°C)
  - diesel exhaust contains enough H_2O
  - dry air (e.g., compressed air for partial flow regen.) requires higher temp.



(*) SAE Paper #2002-01-1684

Overview of Diesel Soot Oxidation Study

- **Effect of NO_2: [NO_2 mass flux]/[soot loading] ratio**
  - at low ratio (e.g., heavily loaded trap), NO_2 is depleted at low T; then active increase in temperature provides only marginal enhancement until "O_2-mechanism" lights off (>500°C).



⇒ Heavily loaded trap or low NO_2: active regen. requires T>500°C

Summary - Soot Oxidation

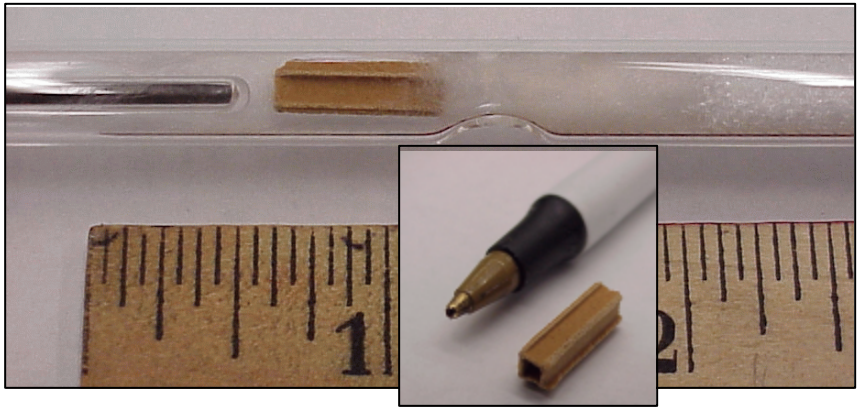
- Equipment and methodology for the quantitative studies of soot oxidation were demonstrated.
  - Cross-validation using multiple *techniques* (temperature-programmed, isothermal, step-response) and *tools* (Micro-Reactor, TGA).
- Initial findings:
  - Complex promotional effect of H_2O on diesel soot oxidation by O_2:
    - effect is substantial, synergistic, non-linear
  - Difference between soot samples:
    - diesel samples: similar trends, but quantitative differences
    - diesel vs. model: vast differences (quantitatively and trends)
  - Also observed: changes in soot reactivity during oxidation process
- Work underway:
  - DPF cores loaded with soot on an engine
  - regenerated under the controlled conditions (pilot reactor)

Probing Catalyst Oxidation Activity

Objectives:

- Measure degradation of *field-tested* catalysts, determine the cause if possible
- Predict degradation at different conditions (e.g., as a f(T), gas composition) - controlled aging of samples *in-situ*.

Experimental:

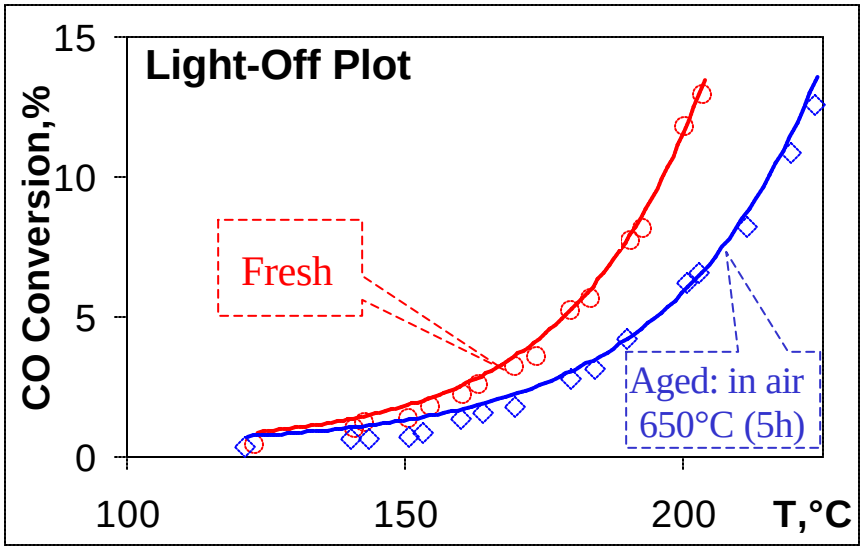


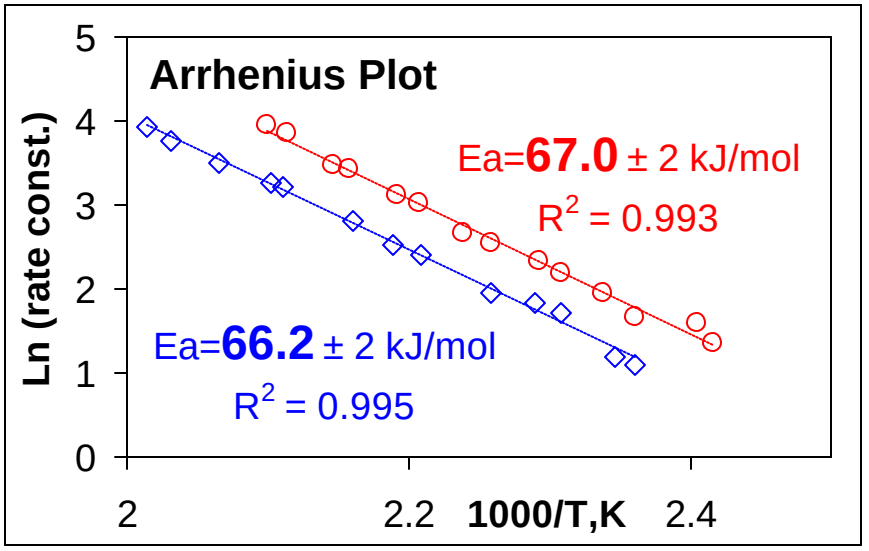
- Probe reaction of CO oxidation
- Single/several channels
  - short (~0.5”) - to minimize T gradient across the sample
  - sufficient to prevent gas “bypass”
    - confirmed the absence of mass-transfer limitations
- Excellent reproducibility of sampling channels from the brick

Probing Catalyst Oxidation Activity

Example:

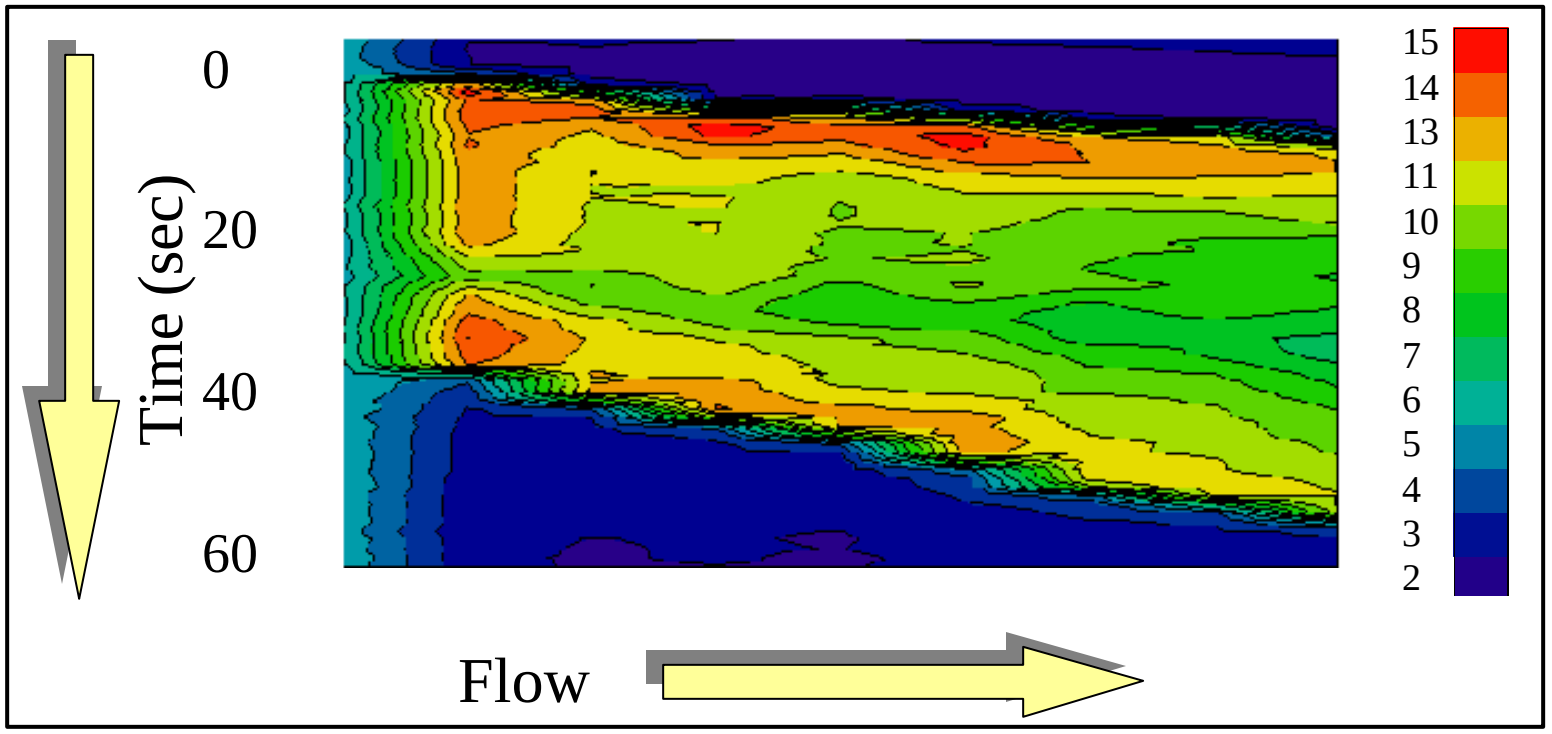
- **Measured degradation**: CO light-off increased by ~20°C
- **Determined the cause**:
  - Activation Energy (E_a) has changed insignificantly: no *chemical* changes to the active sites
  - Hence, the loss is due to the frequency factor decrease, i.e. lost the *number* of exposed sites - ***agglomeration***





Catalysts Deactivation

- Large temperature gradients possible in the operating AT devices
  - cause localized (propagating) deactivation
  - accurate measurements of the T-gradients is difficult
- SPACI-MS(*) can been used to infer location of hot spots from the concentration profiles



(*) *Details about the Spaci-MS technique were published at SAE, CLEERS*

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  - for their support through the Freedom Car program

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