Outline

- DeNOx reaction
- Fuel impact
  - Ash
    - Amount
    - Particle size distribution
    - Minerals
    - Trace Elements
  - % Sulfur
- Catalyst sampling / analysis
- Inspections / housekeeping
- Catalyst replacement / cleaning
- Catalyst on line monitoring
The DeNOx Reaction

4 NO + O₂ + 4 NH₃ ⇌ 4 N₂ + 6 H₂O

First order reaction – mass transport limited

- Catalyst channel
- Laminar gas film
- Catalyst wall surface
- Catalyst pores
- Active site = V₂O₅

DNX Catalyst: V₂O₅ and WO₃ on TiO₂
DeNOx reaction is diffusion limited
more highways = high diffusion rate = higher activity
The DeNOx Reaction

\[ 4 \text{NO} + \text{O}_2 + 4 \text{NH}_3 \Rightarrow 4 \text{N}_2 + 6 \text{H}_2\text{O} \]

First order reaction – mass transport limited

Catalyst deactivation results when one of the above is made less active.
Fossil Fuel Combustion

- Vapors
  - Mg, Na, K, SO₂, As, etc.
- Molten Ash Droplets
- Solid Particles
Fuel Impact - Ash

- Catalyst pitch – amount of ash

![Ash Loading Curve](image)

Coal Heat Value, BTU/Lb.

- DNX-X58
- DNX-X64
- Blackville 2
Fuel Impact - Ash

Catalyst pitch – particle size distribution

- Bituminous Fly Ash
- PRB Fly Ash

Pore Radius (Å)

dV/dlogr (ml/g)

10 100 1,000 10,000 100,000 1,000,000 10,000,000
Fuel Impact – Ash Mineral Analysis

<table>
<thead>
<tr>
<th>Coal Ash Mineral Composition</th>
<th>Bituminous Coal</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Al_2O_3$, %</td>
<td>22</td>
</tr>
<tr>
<td>$Fe_2O_3$, %</td>
<td>20.3</td>
</tr>
<tr>
<td>$SiO_2$, %</td>
<td>42.6</td>
</tr>
<tr>
<td>SO$_3$, %</td>
<td>5</td>
</tr>
<tr>
<td>CaO, %</td>
<td>5.3</td>
</tr>
<tr>
<td>K$_2$O, %</td>
<td>1.4</td>
</tr>
<tr>
<td>MgO, %</td>
<td>0.9</td>
</tr>
<tr>
<td>Na$_2$O, %</td>
<td>0.9</td>
</tr>
</tbody>
</table>

- Al, Fe and Si impacts catalyst erosion
- Fe impacts SO$_2$-oxidation (+ / -)
- Si can also impact catalyst fouling
### Coal Ash Mineral Composition

<table>
<thead>
<tr>
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<th>Coal</th>
<th>Bituminous</th>
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<tbody>
<tr>
<td>Al₂O₃, %</td>
<td>22</td>
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CaSO₄ impacts catalyst fouling

![Image of coal ash mineral composition analysis](image-url)
## Fuel Impact – Ash Mineral Analysis

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</tbody>
</table>

- Ca, Mg, Na and K are all catalyst chemical poisons
- Acid soluble Na in ash > 1.0%
- Acid soluble K in ash > 0.5%
- Risk is considered minor in bituminous coal, as most of the alkaline metals are not water soluble
- PRB, wood, straw fuel the risk is higher
## Fuel Impact – Ash Mineral Analysis

### Coal Ash Mineral Composition

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- SO₃ in the fuel reduces poisoning

### Graph

Graph showing the relationship between (K+Na)/V molar ratio and relative activity, k/k₀, with data points for Coal - USA, AWV, and Wood - Sweden.
### Fuel Impact – Trace Elements

<table>
<thead>
<tr>
<th>Coal Trace Elements</th>
<th>ppm</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Arsenic ppm</strong></td>
<td>13</td>
</tr>
<tr>
<td>Chrome ppm</td>
<td>43</td>
</tr>
<tr>
<td>Vanadium ppm</td>
<td>45</td>
</tr>
<tr>
<td>Copper ppm</td>
<td>21</td>
</tr>
<tr>
<td>Lead ppm</td>
<td>18</td>
</tr>
<tr>
<td>Mercury ppm</td>
<td>413</td>
</tr>
<tr>
<td>Nickel ppm</td>
<td>37</td>
</tr>
</tbody>
</table>

- As is a catalyst poison
- Mechanism – pore blockage
Arsenic poisoning mechanism

As in coal is vaporized to \( \text{As}_2\text{O}_3 \),

if the vapour pressure of \( \text{As}_2\text{O}_3 \) in the gas phase is higher than equilibrium, \( \text{As}_2\text{O}_3 \) will precipitate in the catalyst pores due to capillary forces,

As condenses in the very small micro pores of the catalyst,

As could react with the vanadium in the micro pores to form a vanadia-arsenate compound,

this compound is so stable it inhibits \( \text{V}^{5+}-\text{O}-\text{H} \) reaction cycle.
A catalyst with a diverse and accumulated larger pore volume

![Graph showing cumulative pore volume vs. pore radius. The graph compares a Trimodal (DNX) porous Catalyst and a Micro-porous Catalyst. The y-axis represents cumulative pore volume in ml/kg, and the x-axis represents pore radius in Å. The Trimodal (DNX) porous Catalyst shows a more gradual increase in cumulative pore volume with pore radius compared to the Micro-porous Catalyst, which has a steeper increase at smaller pore radii.]
Fuel Impact – Trace Elements

- is more resistant to As poisoning

**Effect of Arsenic Accumulation on SCR Catalyst**

- Micro-porous catalyst
- Trimodal (DNX) porous catalyst
Fuel Impact – Trace Elements

- As containing fuel
  - Micro-porous catalyst increase CaO.
  - Use a catalyst with a diverse catalyst pore volume, w/ coal levels up to 25 ppm

![Comparing the Effect of Arsenic Accumulation on operating micro-porous and trimodal (DNX) catalyst](image_url)

- Diagram showing the comparison of arsenic accumulation between micro-porous and DNX catalysts.
### Fuel Impact – Trace Elements

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Cr is a catalyst pore blockage poison

![Graph showing catalyst activity versus Chromium content](image)
## Fuel Impact – Trace Elements

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- VOSO$_4$ will accumulate on the catalyst surface
- Can deactivate catalyst, collapses active sites, if accumulation is high enough
- Increases SO$_2^-$ oxidation as V accumulates on the catalyst
- Oil, Pet coke
### Fuel Impact – Trace Elements

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<td>37</td>
</tr>
</tbody>
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- **Heavy and Base metals** – antimony, chrome, copper, lead, mercury, nickel, tin, zinc
- **Alkali Metals** – cesium, lithium, rubidium
- **Alkaline Earth Metals** – barium, strontium
- **Phosphorous**
Fuel Impact - % Sulfur

To prevent operating catalyst below Ammonium Bisulfate (ABS) formation temperature

Ammonium Bisulfate Temperature Curves
Fuel Impact - % Sulfur

- **ABS** – \( \text{NH}_4\text{HSO}_4 \)
  \[
  \text{NH}_3(g) + \text{H}_2\text{SO}_4(g) \rightarrow \text{NH}_4\text{HSO}_4(\text{liq}) \quad (\text{mp.} 147^\circ\text{C})
  \]

- **Mechanism**
  - Pore blockage by capillary condensation.

\[
\begin{align*}
\text{SO}_3 & \quad \text{NH}_3 & \quad \text{NO} \\
\text{SO}_3,\text{eq} & \quad \text{NH}_3,\text{eq}
\end{align*}
\]

- Constant blocked pore volume
- \( \text{NH}_4\text{HSO}_4 \) deposited in micropores
- Catalyst wall
Fuel Impact - % Sulfur

- Mechanism - ABS dew point calculation

  \[ \begin{align*}
  & \text{NH}_3 \text{ concentration} \\
  & \text{SO}_3 + \text{H}_2\text{SO}_4 \text{ concentration} \\
  & \text{H}_2\text{O} \text{ concentration} \\
  & \text{Pore size distribution}
  \end{align*} \]

  \[ \Rightarrow \quad T_{\text{dew}} \]

  Clausius/Clapeyron equation:

  \[
  \ln \left( \frac{P_{\text{NH}_3} \cdot P_{\text{H}_2\text{SO}_4}}{P_{\text{eq, bulk}}} \right) = 27.97 - \frac{26671}{T[K]}
  \]

  Matsuda, S., 1982

  Kelvin equation:

  \[
  \ln \left( \frac{P_{\text{NH}_3} \cdot P_{\text{H}_2\text{SO}_4}}{P_{\text{eq, pore}} / P_{\text{eq, bulk}}} \right) = - \frac{2 \cdot \sigma \cdot M}{\rho \cdot r_{\text{pore}} \cdot R \cdot T}
  \]
ABS dew point temperature depends on the catalyst pore distribution.
A diverse pore volume should allow operation below the ABS dew point.
Fuel Impact - % Sulfur

- Cycle operation below and above the ABS dew point

![Bar chart showing catalyst activity over time with different sulfur impacts.](chart.png)
Fuel Impact - % Sulfur

High dust cycle operation 558°F/630°F
NHSV = 4000h⁻¹, $v_0 = 2$Nm/s, 300 ppmv NOx,
$NH_3/NO_{inlet} = 0.9$, DNX664, 8% $H_2O$

- Five (5) SCR nightly operate below ABS dew point, up to 300 continuous hours, within 10 degrees F of the ABS bulk dew point temperature.
Fuel Impact - % Sulfur

- Managing $\text{SO}_2$-oxidation
  - Fuel
    - $\text{SO}_2$ concentration
    - Ash composition
  - Boiler
    - Excess $\text{O}_2$
  - Catalyst
    - Distribution of vanadium
    - Diverse pore structure

- Current $\text{SO}_2$-oxidation experience
  - High dust guarantees < 0.1%
  - Operating experience < 0.1%

![Graph showing SO$_2$-oxidation vs. Temperature](image-url)
A 2.5 % sulfur coal, SO₂-oxidation of 0.5% increases SO₃ content > 10 ppm thereby increasing the acid dew point temperature.
Catalyst Testing

- Catalyst testing – Elements / Plates
  - When?
    - Annual
    - End of ozone season
  - Where should you sample?
    - 1st layer
    - Look at flow model results
    - Area of highest dust loading
  - What analysis should you request?
    - Activity – STD / Unit Conditions
    - Chemical
    - Surface
Catalyst Testing

- Activity Analysis Results

![Graph showing percent relative activity vs. operating hours. The line graph indicates a decrease in activity as operating hours increase.]
## Catalyst Testing

- **Chemical Analysis**
  - Look at catalyst poisons

<table>
<thead>
<tr>
<th>Sample</th>
<th>Reference</th>
<th>Reactor A</th>
<th>Reactor B</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Original Layer 1</td>
<td>Duplicate Layer 1</td>
</tr>
<tr>
<td>Al, %</td>
<td>1.12</td>
<td>1.52</td>
<td>1.43</td>
</tr>
<tr>
<td>As, ppm</td>
<td>&lt; 10</td>
<td>6,070</td>
<td>6,315</td>
</tr>
<tr>
<td>Ca, %</td>
<td>2.56</td>
<td>2.53</td>
<td>2.43</td>
</tr>
<tr>
<td>Fe, %</td>
<td>0.04</td>
<td>0.68</td>
<td>0.58</td>
</tr>
<tr>
<td>K soluble, ppm</td>
<td>165</td>
<td>1,475</td>
<td>1,600</td>
</tr>
<tr>
<td>Mg, %</td>
<td>0.06</td>
<td>0.08</td>
<td>0.08</td>
</tr>
<tr>
<td>Na soluble, ppm</td>
<td>545</td>
<td>1,380</td>
<td>1,210</td>
</tr>
<tr>
<td>P, ppm</td>
<td>1,680</td>
<td>1,605</td>
<td>2,000</td>
</tr>
<tr>
<td>Si, %</td>
<td>5.24</td>
<td>5.78</td>
<td>5.69</td>
</tr>
<tr>
<td>S, %</td>
<td>0.03</td>
<td>0.55</td>
<td>0.57</td>
</tr>
</tbody>
</table>
Catalyst Testing

- Surface Analysis
  - Internal surface area (BET) pore volume

<table>
<thead>
<tr>
<th></th>
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</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>A</td>
<td>B</td>
<td>Layer 1</td>
</tr>
<tr>
<td>Surface area BET, m²/g</td>
<td>70</td>
<td>68</td>
<td>68</td>
</tr>
</tbody>
</table>
Catalyst Testing

- Reactor specific catalyst management curve
  - To predict future catalyst additions
When / what should you inspect?
- At least annually
  - Inlet duct
  - AIG / Mixer
  - Turning vanes
  - Rectifier
  - SCR catalyst
  - Outlet duct
- Take advantage of short outages
- Take a quick look before start up in the spring
- Remove / prevent ash pile build up on catalyst
Catalyst Replacement – Case 1 (New)

- 600 MWe
- 85% operation, 7500 hours
- SCR layout, 2+1
- DeNOx efficiency, 90%
- Ammonia-slip, 2ppmvdc
- $\text{SO}_2$-oxidation, 0.5%

- New catalyst SOR = 1.0
- New catalyst deactivation = 13% per 10,000 hours
- New catalyst cost based on actual 2005 cost
## Catalyst Replacement – Case 1 (New)

<table>
<thead>
<tr>
<th>Catalyst Layers</th>
<th>New</th>
<th>Hours</th>
<th>New Catalyst Cost</th>
<th>Sealing Cost</th>
<th>Freight Cost</th>
<th>Removal Cost</th>
<th>Installation Cost</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spare</td>
<td>X</td>
<td>32,000</td>
<td>$1,407,375</td>
<td>incl.</td>
<td>$60,000</td>
<td>$0</td>
<td>$250,000</td>
</tr>
<tr>
<td>First</td>
<td>X</td>
<td>73,500</td>
<td>$1,407,375</td>
<td>incl.</td>
<td>$60,000</td>
<td>$250,000</td>
<td>$250,000</td>
</tr>
<tr>
<td>Second</td>
<td>X</td>
<td>102,500</td>
<td>$1,407,375</td>
<td>incl.</td>
<td>$60,000</td>
<td>$250,000</td>
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<tr>
<td>Spare</td>
<td>X</td>
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<td>$1,407,375</td>
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<td>$60,000</td>
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<td>169,500</td>
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<tr>
<td>Second</td>
<td>X</td>
<td>202,000</td>
<td>$1,407,375</td>
<td>incl.</td>
<td>$60,000</td>
<td>$250,000</td>
<td>$250,000</td>
</tr>
</tbody>
</table>

Sub - Total      | $8,444,250 | $0   | $360,000 | $1,250,000 | $1,500,000 |

Total Cost over 27 years | $11,554,250
Catalyst Replacement – Case 2 (New / Clean)

- 600 MWe
- 85% operation, 7500 hours
- SCR layout, 2+1
- DeNOx efficiency, 90%
- Ammonia-slip, 2ppmvdc
- SO$_2$-oxidation, 0.5%

- New catalyst SOR = 1.0
- New catalyst deactivation = 13% per 10,000 hours
- New catalyst cost based on actual 2005 cost

- Clean catalyst SOR = 0.95
- Clean catalyst deactivation = 15 per 10,000 hours
- Damaged catalyst replacement = 10%
- Clean catalyst cost based on actual 2005 cost
## Catalyst Replacement – Case 2 (New / Clean)

<table>
<thead>
<tr>
<th>Catalyst Layers</th>
<th>Hours</th>
<th>Cleaning Catalyst Cost</th>
<th>New Catalyst Cost</th>
<th>Sealing Cost</th>
<th>Freight Cost</th>
<th>Removal Cost</th>
<th>Installation Cost</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spare N</td>
<td>32,000</td>
<td>$0</td>
<td>$1,407,375</td>
<td>incl.</td>
<td>incl.</td>
<td>$0</td>
<td>$250,000</td>
</tr>
<tr>
<td>First C</td>
<td>73,500</td>
<td>$1,367,375</td>
<td>$140,738</td>
<td>$50,000</td>
<td>$100,000</td>
<td>$250,000</td>
<td>$250,000</td>
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<tr>
<td>Second C</td>
<td>125,000</td>
<td>$1,367,375</td>
<td>$140,738</td>
<td>$50,000</td>
<td>$100,000</td>
<td>$250,000</td>
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Sub - Total
- $6,836,875
- $2,111,065
- $250,000
- $500,000
- $1,250,000
- $1,500,000

Total Cost over 27 years
- $12,447,940

Savings over New Catalyst, year = ($34,373)

w/o damaged catalyst
- $11,744,250

Savings over New Catalyst, year = ($7,308)
Catalyst Replacement – Case 3 (Clean)

- 600 MWe
- 85% operation, 7500 hours
- SCR layout, 2+1
- DeNOx efficiency, 90%
- Ammonia-slip, 2ppmvdc
- SO$_2$-oxidation, 0.5%

- Clean catalyst SOR = 0.95
- Clean catalyst deactivation = 15% per 10,000 hours
- Damaged catalyst replacement = 10%
- Clean catalyst cost based on actual 2005 cost quotation
## Catalyst Replacement – Case 3 (Clean)

<table>
<thead>
<tr>
<th>Catalyst Layers</th>
<th>Cleaned</th>
<th>Hours</th>
<th>Cleaning Catalyst Cost</th>
<th>New Catalyst Cost</th>
<th>Sealing Cost</th>
<th>Freight Cost</th>
<th>Removal Cost</th>
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</tr>
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<tbody>
<tr>
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Sub - Total       $8,204,250  $844,428  $300,000  $600,000  $1,500,000  $1,500,000

Total Cost over 27 years $12,948,678

Savings over New Catalyst, year = ($53,632)

w/o damaged catalyst $12,104,250

Savings over New Catalyst, year = ($21,154)
## On Line Catalyst Monitoring

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<th>Units</th>
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<td>Catalyst life</td>
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</table>
On Line Catalyst Monitoring

- NOx conversion graph
  - Data can allow estimate of conversion and slip
On Line Catalyst Monitoring

- Ammonia slip / Ammonia on fly ash
  - How does ammonia get on the ash?
    - Acidic ashes attract ammonia
    - SO$_3$ attracts ammonia
  - Without acid sites, ammonia will remain as gas
  - Other factors that play a role
    - LOI (especially in PRB coal, very highly ammoniated ash)
    - Structure, mineralogy of ash
    - Local concentrations of NH$_3$
  - Rule of Thumb: 2 ppm slip yields 100 ppm on ash
  - *Ammonia levels in ash are known to be quite variable*
On Line Catalyst Monitoring

- Ammonia slip / Ammonia fly ash sampling
  - Where should I measure?
    - Different ammonia levels in fly ash hoppers
    - Pick the hopper with the highest level
  - How often should I measure?
    - For best results sample hopper once a day
    - Combined samples for once a week ammonia analysis
    - Graph results
On Line Catalyst Monitoring

Performance test generated curve

Ammonia ash level versus time

NH3-slip = 2 ppmvdc

Ammonia Concentration, mg/kg

Hours

0 5,000 10,000 15,000 20,000 25,000 30,000
Summary

- Catalyst may be impacted with a fuel switch
  - Ash loading
  - ABS
  - Catalyst poisons
  - SO$_2$-oxidation

- Catalyst sampling / analysis
  - At least annually – activity, chemical analysis, and pore volume

- Inspection
  - At least once per year
  - Quick look in the spring
  - Take opportunities
Summary

- Housekeeping
  - Keep catalyst clean / free of pluggage

- Catalyst replacement / cleaning

- Catalyst On Line Monitoring
  - Monitor catalyst / system performance
  - Monitor NH₃-slip
    - Analyzer
    - Fly ash sampling – daily samples combined for weekly analysis
Summary

- Catalyst may be impacted with a fuel switch
  - Ash loading
  - ABS
  - Catalyst poisons
  - SO$_2$-oxidation

- On line monitoring
  - Monitor catalyst / system performance
  - Ammonia ash sample - daily sample / combined samples for weekly analysis

- Off line monitoring
  - Catalyst sampling / analysis @ least annually – activity, chemical analysis, and pore volume
  - Inspection
    - Complete inspection annually
    - Quick look in the spring
    - Take opportunities

- Housekeeping
  - Keep catalyst clean / free of pluggage
Summary

- Keep care of your investment to prevent this ..................