Nitrogen oxide chemistry in combustion processes

Based on material originally by Prof. Mikko Hupa
Background - NOx

- Nitrogen oxides, "NO\textsubscript{x}\" = NO + NO\textsubscript{2}
- In combustion flue gases >95% NO and <5% NO\textsubscript{2}
- NO\textsubscript{2} formed via NO
- In the atmosphere NO reacts to NO\textsubscript{2}, and further to compounds such as HNO\textsubscript{3} and nitrates (acidification)
- NO\textsubscript{2} + hydrocarbons $\rightarrow$ photochemical "smog"
- In some special cases also emissions of N\textsubscript{2}O
Background – NOx (ii)

• In some special cases also emissions of N\textsubscript{2}O
• N\textsubscript{2}O stable in the atmosphere – not part of the acidification
• However, N\textsubscript{2}O is green-house gas and upper atmosphere ozone destructor

• Nitrogen oxide chemistry in combustion complicated – and fascinating!
Emissions are expressed in a variety of units – be aware!

100 ppm (3 % O₂)

= 72 ppm (8 % O₂)
= 83 ppm (8 % O₂, dry gas)
= 237 mg NO₂/m³ (3 % O₂, dry)
= 71 mg NO₂/MJ (LHV)
= 0.104 gr/DSCF (3 % O₂, dry)
= 0.00116 gr/BTU (LHV)
Background

Nitrogen oxide emissions from various combustion processes without reduction measures

(Hupa et al.)
Nitrogen oxide chemistry in combustion processes – Terms and concepts

- Chemical equilibrium and NO
- Thermal-NO (Zeldowich NO) and kinetics
- Fuel-NO
- Volatile-NO – HCN, NH₃
- Char-NO
- Staged Combustion: Low Nox Technologies
- NO formation tendency characterization
- (NO chemistry in fluidised bed combustion)
Nitrogen species in a typical flue gas under chemical equilibrium conditions
Nitrogen species in a typical flue gas under chemical equilibrium conditions

• Most abundant ones
  \( \lambda = 1.1: \) \( \text{N}_2, \text{NO}, \text{NO}_2, \text{N}_2\text{O} \)
  \( \lambda = 0.9: \) \( \text{N}_2, \text{HCN}, \text{NH}_3 \)

• Equilibrium conc. of \( \text{NO}_x \) decreases with temp.:
  data from flue gas with \( \lambda = 1.1: \)
  \( T = 1800 \text{ K} \) \( 1200 \text{ ppm} \)
  \( T = 500 \text{ K} \) \( <1 \text{ ppm} \)
NO – Chemical equilibrium vs. Kinetics

Schematic view of NO formation in a boiler

Equilibrium concentration ($\lambda=1.1$)

Typical measured concentration

Time

NO concentration

Temperature

Furnace  Superheaters
CO Vs. NO kinetics

1000 °C

Mole fraction

Time (s)

O2
CO
CO2
H2O
NO
NO2
N2O
H
O
OH
H2
HO2
H2O2
HCO

Kinetic calculation, plug flow reactor
CO Vs. NO kinetics

1000 °C

Mole fraction (-)

Time (s)

O2

CO

CO2

H2O

NO

Equilibrium NO

Equilibrium CO

Kinetic calculation, plug flow reactor
CO Vs. NO kinetics

1500 °C

Mole fraction (-)

Time (s)

Kinetic calculation, plug flow reactor
CO Vs. NO kinetics

2000 °C

- Mole fraction (\( \cdot \))
- Time (s)

Kinetic calculation, plug flow reactor
NO – Chemical equilibrium vs. Kinetics

Schematic view of NO formation in a boiler

Equilibrium concentration ($\lambda=1.1$)

Typical measured concentration

Time

NO concentration

Temperature

Furnace  Superheaters
Nitrogen chemistry in boilers

- At equilibrium: high temperatures -> high NO, low temperatures -> no NO
- Formation of NO determined by kinetics – not equilibrium
- Decomposition at lower temperatures kinetically ”frozen”
- Great variations in NO emissions. Which factors influence NO-formation kinetics?
- Major research activities to understand mechanisms of NO formation since 1970’s
- NO emissions brought detailed chemistry into combustion engineering
NO formation mechanisms
Thermal-NO

- NO formed from \( \text{N}_2 \) in combustion air
- Direct reaction \( \text{N}_2 + \text{O}_2 \rightarrow 2\text{NO} \)
  very slow even at high temperatures (\( \text{N}_2 \))
- Zeldowich (1946): *atomic oxygen* needed to react with \( \text{N}_2 \):
  \[
  \text{O} + \text{N}_2 \rightarrow \text{NO} + \text{N} \quad (1)
  \]
- The N atom reacts further to yield a second NO
  \[
  \text{N} + \text{O}_2 \rightarrow \text{NO} + \text{O} \quad (2)
  \]
- \( \frac{d(\text{NO})}{dt} = k_1 \cdot (\text{O}) \cdot (\text{N}_2) \)
Estimated rates of thermal-NO formation

![Graph showing estimated rates of thermal-NO formation with temperature on the x-axis and rates on the y-axis. The graph includes lines for different oxygen concentrations: O₂ 4%, O₂ 1%, and O₂ 0.25%.](image)
Thermal NOx - Summary

- Molecular nitrogen in air reacts to form NO
- Reaction kinetically possible with atomic oxygen
- Significant only if T > 1400 C
- Formation steeply dependent on temperature
- Main contributor to NO formation in gasoline engines and gas combustion systems
Fuel-NO:

Gradual Exploration of the Mechanisms
Fuel NOx – Fuel Oil with Organic Nitrogen Addition

Based on Martin and Berkan, Air pollution and its control, AICHE symp. Series 68, Nr 126, 45, 1972
Fuel-NO

- NO formed from N in fuel
- Fuel-N organically bound - reactive
- Until the 1970’s fuel-N was not considered a source of NO

<table>
<thead>
<tr>
<th>Amines</th>
<th>Pyridine</th>
<th>Pyrrole</th>
<th>Quinoline</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td><img src="image1.png" alt="Pyridine" /></td>
<td><img src="image2.png" alt="Pyrrole" /></td>
<td><img src="image3.png" alt="Quinoline" /></td>
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<table>
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<tr>
<th>Calculated as CaO</th>
<th>Calculated as Na</th>
<th>Not organic-N, but molecular-N, N₂</th>
</tr>
</thead>
</table>

### Table: Fuel-N Distribution and Spent Pulping Liquors

<table>
<thead>
<tr>
<th></th>
<th>Natural gas</th>
<th>Light</th>
<th>Heavy</th>
<th>Coal (Polish)</th>
<th>Coal (Petshora)</th>
<th>Peat</th>
<th>Wood</th>
<th>Spent pulping liquors</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Sulfite</td>
</tr>
<tr>
<td>wt-%</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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</tr>
<tr>
<td>ash</td>
<td>0.0</td>
<td>0.0</td>
<td>0.1</td>
<td>11.0</td>
<td>15.0</td>
<td>5.0</td>
<td>0.4</td>
<td>12.0 (¹)</td>
</tr>
<tr>
<td>C</td>
<td>72.0</td>
<td>85.8</td>
<td>85.5</td>
<td>73.2</td>
<td>71.2</td>
<td>55.0</td>
<td>50.4</td>
<td>45.0</td>
</tr>
<tr>
<td>H</td>
<td>22.8</td>
<td>13.2</td>
<td>11.2</td>
<td>4.7</td>
<td>4.4</td>
<td>5.5</td>
<td>6.2</td>
<td>4.9</td>
</tr>
<tr>
<td>S</td>
<td>0.0</td>
<td>0.5</td>
<td>2.5</td>
<td>1.0</td>
<td>0.6</td>
<td>0.2</td>
<td>0.0</td>
<td>5.0</td>
</tr>
<tr>
<td>O</td>
<td>0.2</td>
<td>0.3</td>
<td>0.4</td>
<td>9.1</td>
<td>6.6</td>
<td>32.6</td>
<td>42.5</td>
<td>33.0</td>
</tr>
<tr>
<td>N</td>
<td>5.0 (³)</td>
<td>0.2</td>
<td>0.3</td>
<td>1.0</td>
<td>2.2</td>
<td>1.7</td>
<td>0.5</td>
<td>0.1</td>
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<tr>
<td>sum</td>
<td>100.0</td>
<td>100.0</td>
<td>100.0</td>
<td>100.0</td>
<td>100.0</td>
<td>100.0</td>
<td>100.0</td>
<td>100.0</td>
</tr>
</tbody>
</table>

Dry flue gas with 3 vol-% oxygen, assuming all fuel-N forms NO

<table>
<thead>
<tr>
<th>NO (ppm)</th>
<th>Sulfite</th>
<th>Sulfate</th>
</tr>
</thead>
<tbody>
<tr>
<td>264</td>
<td>1465</td>
<td>317</td>
</tr>
<tr>
<td>3177</td>
<td>543</td>
<td>2512</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>N₂/NO</th>
<th>383</th>
<th>2054</th>
</tr>
</thead>
</table>

1) Calculated as CaO
2) Calculated as Na
3) Not organic-N, but molecular-N, N₂
NO formation – Fuel Oil Containing N

\[ \text{N}_2 \xrightarrow{\text{Thermal}} \text{NO} \]

\[ \text{N}_{\text{fuel}} \rightarrow \text{N}_{\text{vol}} \xrightarrow{\text{N}_{\text{tar}}} \text{N}_2, \text{HCN}, \text{NH}_3 \]

\[ \text{N}_2 \xrightarrow{\text{OX.}} \text{NO} \]

\[ \text{NO} \xrightarrow{\text{Red.}} \text{N}_2 \]
NO formation

\[ \text{N}_2 \xrightarrow{\text{Thermal}} \text{NO} \]

\[ \text{N}_\text{fuel} \xrightarrow{} \text{N}_{\text{vol}} \]

\[ \text{N}_{\text{vol}} \xrightarrow{} \text{N}_2 \]

\[ \text{N}_2 \xrightarrow{} \text{HCN, NH}_3 \]

\[ \text{HCN, NH}_3 \xrightarrow{\text{Ox.}} \text{NO} \]

\[ \text{NO} \xrightarrow{\text{Red.}} \text{N}_2 \]

\[ \text{N}_2 \xrightarrow{100-y} \text{N}_2 \]

\[ \text{N}_2 \xrightarrow{y} \text{NO} \]
NO formation – volatiles and char
How to Characterize Fuels with Respect to their NO Formation Tendency?

\[ \text{N}_2 \xrightarrow{\text{Thermal}} \text{NO} \]

\[ \text{N}_{\text{fuel}} \xrightarrow{} \text{N}_{\text{vol}} \]

\[ \text{N}_{\text{fuel}} \xrightarrow{} \text{N}_{\text{char}} \]

\[ \text{N}_{\text{vol}} \xrightarrow{} \text{N}_2 \xrightarrow{} \text{HCN, NH}_3 \xrightarrow{\text{Ox}} \text{NO} \xrightarrow{\text{Red}} \text{N}_2 \]

\[ \text{N}_{\text{char}} \xrightarrow{} 100\text{-y} \xrightarrow{\text{Y}} \text{N}_2 \xrightarrow{\text{Y}} \text{NO} \]
Experimental procedure #1

- 2-step burning of fuel in 5 vol-% O$_2$ + Ar
  - Pyrolysis: Ar through bed + secondary O$_2$
  - Char ox: Ar + O$_2$ through bed
- Release curves integrated to obtain cumulative CO$_2$ and NO (NO I + NO II)

Temperature: $T_{\text{bed}}=850^\circ C$
Volatile NO and Char NO – Laboratory Tests
(NO results given as weight-% N per original fuel)

Graph showing nitrogen content (% by dry fuel) for different samples:
- Bit. coal I
- Bit. coal II
- Brown coal
- Brown coal II
- Peat I
- Peat II
- Peat III
- Peat IV
- Peat V

Legend:
- Fuel-N
- Vol NO (NO I)
- Total NO (NO I + NO II)
Volatile NO and Char NO – Laboratory Tests
(NO results given as weight-% N per original fuel)
Fuel NOx Summary

- Fuel-N very reactive: 20-60 % conversion to NO
- Not very sensitive to temperature (cf thermal NO)
- Main contributor to NO in solid fuel combustion
- Different fuels have different fuel-N behavior
Schematic view of NO emission distribution by source

![Diagram showing NO emissions distribution by source](image_url)

- **Thermal-NO**
- **Fuel-NO**
- **Prompt-NO**

**NO\textsubscript{x} emissions, mg NO\textsubscript{2}/MJ**

**°C**
Fuel NOx – How to Control?

• Traditional efficient combustion: Maximize the three T’s: Temperature, Turbulence, Time
• Gives low CO and CxHy
• ...but effective conversion of Fuel-N to NO!
• ”Low NOx” concept: staged combustion to convert Fuel-N to N$_2$
• A variety of novel burner and furnace technologies with Low-NOx
Conventional Burner for Solid Fuels

Fuel powder + Transport air

Combustion air

Devolutilization and gas combustion

Char combustion

Ash
Low-NO\textsubscript{x} Burner for Solid Fuels

Fuel powder + Transport air

Primary air

Primary zone $\lambda < 1$

Secondary air

Secondary zone, "Burn-out zone" $\lambda > 1$
Examples of NO$_x$ chemistry in a BFB and in a Kraft recovery boiler
Bubbling fluidized bed (BFB) boiler
- $107 \text{ MW}_{\text{th}}$, 118 bar, 535 °C
Boiler parameters during the campaign

- Air distribution
  - Fluidizing air: 50%
  - Secondary air: 25%
  - Tertiary air: 25%
- $\lambda = 1.3$
- Bark / sludge / SRF
  - 74 / 16 / 10 wt-%
- Temperatures
  - Bed: 800°C
  - Peak temp. 1100°C
  - At bull nose level 950°C
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### Gas composition from bed

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<td>NH$_3$</td>
<td>640</td>
<td>ppm</td>
</tr>
<tr>
<td>HCN</td>
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<td>ppm</td>
</tr>
<tr>
<td>NO</td>
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<td>H$_2$O</td>
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<td>C$_2$H$_4$</td>
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BFB – main nitrogen species

Measured

% of fuel-N

Height (m)

HCN  NH₃  NO

Tert. air  Sec. air  Fuel

10%  27%  95%
BFB simulations to understand NO chemistry

- Prim 50%, sec 25%, tert 25% of total
- Furnace temperature 1000 °C
- Furnace gas below sec. air as input

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BFB simulations

Secondary air jet

TFN = all nitrogen species except $N_2$

Entrainment, no chemistry
BFB simulations

Secondary air jet

\[ \text{NH}_3 + \text{O}_2 \rightarrow \text{NO} \]
\[ \text{NO} + \text{NH}_3 \rightarrow \text{N}_2 \]
BFB simulations

Secondary air jet

Temperature vs Position along jet axis (m)

- $T < 830 \, ^\circ C$
- $T > 830 \, ^\circ C$
- $\lambda > 1$
- $\lambda < 1$

Concentration (ppmv)

- NO$_2$
- N$_2$O
- NO
- NH$_3$
- TFN
- HCN

Entrainment, no O$_2$, "no chemistry"
BFB simulations

Measured gas composition

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<td>0.28</td>
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BFB simulations

\( \lambda = 1.3 \)

\( \lambda < 1 \)
BFB simulations

Modeled

Height (m)

Air

Air

Fuel

% of fuel-N

HCN

NH₃

NO

Tert. air

Sec. air

Fuel
BFB simulations

Tertiary air jet

Temperature (°C)

Concentration (ppmv)

Position along jet axis (m)

- T<830 °C
- T>830 °C

TFN

Temp.

NH₃

NO

NO₂

HCN

N₂O
BFB simulations

\[ \lambda = 1.3 \]

\[ \lambda < 1 \]

Air

Fuel
BFB simulations

Measured

Modeled

Height (m)

% of fuel-N

HCN  NH₃  NO

Sec. air  Tert. air

Fuel

10%  27%  95%

9%  30%  95%
BFB – NOx chemistry

Efficient NOx reduction

- NH₃ in furnace gas up to tert air
- Air jet NO, NH₃, O₂, T
Kraft recovery boiler

- 3000 tds/day
- DS 80%
- Air distribution
  - Primary 32 %
  - Lower secondary 32 %
  - Upper secondary 22 %
  - Tertiary 14 %
- $\lambda = 1.2$

(Valmet)
Kraft recovery boiler

- Combustion air jet
- Fuel particle release profiles:
  - Moisture
  - Volatiles
  - Char-C
  - Char-N
- Entrainment of furnace gas into air jet
- Furnace gas below first jet
- Furnace section
- To next air jet / exit
Kraft recovery boiler

- CFD for profiles
- Temperature, moisture, volatiles, char-C, char-NO

Main difference to BFB is that fuel-N release is not confined to bed
Fuel-N release is not confined to bed and there is oxidizing conditions in the furnace.
Kraft recovery boiler

- Tertiary
- Upper secondary
- Lower secondary
Kraft recovery boiler

- NH₃ reacts between air levels

**Graph:**

- **NH₃, NO₂, and N₂O**
- **TFN**
- **NO**

**Axes:**
- **Temperature (°C)**
- **Concentration (ppmv)**

**Legend:**
- **Temp.**
- **Normalized vertical position between lower and upper secondary air (-)**
Kraft recovery boiler

- Tertiary
- Upper secondary
- Lower secondary
Kraft recovery boiler

Upper secondary air jet

- Concentration (ppmv)
- Temperature (°C)
- Position along jet axis (m)

- Temp.
- TFN
- NO
- NO₂
- NH₃
Kraft recovery boiler

This would be the NO profile if all released fuel-N was fully oxidized to NO.

In-furnace NO reduction

Cumulative NO+NH₃ release

Predicted final NO emission

26% NO in stack

25%
BFB and Kraft recovery boiler
NOx chemistry

- BFB - NOx chemistry takes place in the air jets (more generally, where furnace gas and combustion air mix)

- Recovery boiler - NOx chemistry between air levels
  - Compared to BFB, fuel nitrogen released in a larger volume of the furnace, not confined to the bed, and in-furnace conditions are more oxidizing

- Efficient in-furnace NOx reduction: NH$_3$, NO, and O$_2$ simultaneously present at temperatures favoring reduction
  - Air staging
NO chemistry in CFBC
Coal vs Wood
## Nitrogen Oxides with Coal and Wood

*(Leckner et al.)*

<table>
<thead>
<tr>
<th></th>
<th>Coal</th>
<th>Wood</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volatile, % db</td>
<td>40</td>
<td>80</td>
</tr>
<tr>
<td>N(fuel), % db</td>
<td>1.6</td>
<td>0.14</td>
</tr>
<tr>
<td>CFBC NO, ppm</td>
<td>50</td>
<td>60</td>
</tr>
</tbody>
</table>
NO Profiles in CFBC

(Leckner et al.)

![Graph showing NO profiles in CFBC](image)

- Secondary air

(NO ppm vs Reactor height, m)

- Wood
NO Profiles in CFBC
(Leckner et al.)

![Graph showing NO profiles in CFBC with reactor height and NOx ppm on the y-axis. Points for wood and coal are marked with different colors and symbols, indicating variation at different reactor heights.](image.png)
NO formation for solid fuels

N\textsubscript{2} \rightarrow \text{Thermal} \rightarrow \text{NO}

N\textsubscript{2} \rightarrow N\textsubscript{vol} \rightarrow HCN, NH\textsubscript{3} \rightarrow \text{Red} \rightarrow N\textsubscript{2} \rightarrow \text{100-Y} \rightarrow \text{NO}

N\textsubscript{fuel} \rightarrow N\textsubscript{char}
NO formation for solid fuels in CFBC

\[ (N_2 \xrightarrow{\text{Thermal}} NO) \]

\[ \text{NH}_3, \text{HCN} \]

\[ N_{\text{fuel}} \xrightarrow{\text{N}_2} N_{\text{vol}} \]

\[ N_{\text{char}} \xrightarrow{\text{N}_2} HCN, NH_3 \]

\[ N_2 \xrightarrow{\text{Red.}} NO \quad \xrightarrow{\text{100-y}} N_2 \quad \xrightarrow{\text{Red.}} NO + C(s) \]
NO Profiles in CFBC (Leckner et al.)
Modelling Attempts (Goel et al.)

![Graph showing NO profiles in CFBC with secondary air, wood, and coal fuel sources.](image-url)
NO Emission with Wood-Coal
(Leckner et al.)

Nitric oxide ppm (6% O₂)

Wood

50%
(Energy)

Coal
NO Emission with Wood-Coal
(Leckner et al.)

Nitric oxide ppm (6% O₂)

Wood
50%
50%
(50% Energy)
Coal
With increasing share of coal as fuel, the NO emission initially increases, but with higher share coal, the NO reduction by char carbon becomes more effective.
NO Emission with W&C (Leckner et al.) and Modelling Attempts (Engblom et al.)

Nitric oxide ppm (6% O₂)

![Graph showing NO emission with Wood, 50% (Energy) and Coal. Case 1 and Case 2 are compared with Measured data.]
Nitrogen oxide chemistry in combustion processes – Terms and concepts

- Chemical equilibrium and NO
- Thermal-NO (Zeldowich NO) and kinetics
- Fuel-NO
- Volatile-NO – HCN, NH3
- Char-NO
- Staged Combustion: Low Nox Technologies
- NO formation tendency characterization
- (NO chemistry in fluidised bed combustion)