NO_x emissions from biomass combustion plants DI Dr. Ingwald Obernberger





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- N content of different biomass fuels
- Mechanisms of NO_x formation
- NO_x emission reduction potential of different technologies
- NO_x emissions and N conversion related to the N content of the fuel
- Investigation of release profiles based on lab-scale reactor tests
- CFD simulations of NO_x emissions
- NO_x reduction by primary measures
- NO_x reduction by secondary measures
- Summary and conclusions



N content of different biomass fuels

Mechanisms of NO_x formation

NO_x emission reduction potential of different technologies

NO_x emissions and N conversion related to the N content of the fuel









Mechanisms of NO_x formation:

≻ thermal NO_x

> prompt NO_x

≻ fuel NO_x

(NO_x is the sum of the nitric oxides NO and NO₂)



NO_x-emissions - dependence on the fuel and the furnace temperature





NO_x-emissions reaction paths of fuel-N





NO_x emission reduction potential of different technologies





NO_x emissions and N-conversion related to the N content of the fuel



NO_x calculated as NO₂ Results of test runs at real-scale plants equipped with air staging technology



Investigation of release profiles based on lab-scale reactor tests



Due to wide variations in fuel composition, the combustion behaviour of different biomass fuels cannot be predicted only on a theoretical basis.

In order to examine relevant combustion characteristics of biomass fuels in grate combustion systems a specially designed lab-scale reactor was developed.

The purpose of this reactor is to gain information regarding

- fuel decomposition behaviour
- release behaviour of NO_x precursors
- release behaviour of aerosol forming elements (K, Na, Zn, S and Cl)
- 11 > slagging behaviour (first indications)



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Description of the lab-scale reactor – general aspects

Constraints for the reactor design

- Simulation of the fuel behaviour in grate combustion systems (packed bed)
- Reasonable sample intake in order to simulate secondary reactions in the fuel bed appropriately
- High heating rates of the fuel comparable with real-scale grate furnaces
- Inert reactor material in order to avoid reactions of the gases with the reactor
- High flexibility regarding analytical equipment connected with the reactor
- Online recording of relevant operation data and emissions



Description of the lab-scale reactor

- Batch type reactor
- Electrically heated retort made of fibre reinforced silica carbide (SiC)
- Two independently controllable heating zones (the fuel layer is heated by radiation from above and beside the fuel bed)
- Sample holder (also made of SiC) is placed on a balance for on-line mass loss detection
- The reaction medium (e.g. air, N₂ or mixtures of N₂ and O₂) flows through a perforated bottom plate
- Sample holder sealing is realised with thermal oil





Experimental approach

- Energy input via radiation from the upper heating element (radiation section)
- Energy input into the bed zone via the lower heating element (bed section)
- Primary combustion air is supplied from below the grate
- Under consideration that the fuel transport along the grate can be compared with a plug flow, the time dependent results of the lab-scale reactor can be correlated to the local burning conditions on a grate
 - drying phase
 - pyrolysis/gasification
 - charcoal burnout





Measurements, sampling and analyses – on-line measurements

- Mass reduction of the sample (scale)
- Flue gas composition

 - Emerson NGA 2000: Paramagnetism: O₂; NDIR: CO₂, CO; heat conductivity: H₂
 - **FID (Bernath Atomic 3005): OGC (C_xH_y)**
 - CLD (ECO Physics CLD 700 EL ht): NO, NO_x
 - Lambda-sensor: O₂
- > Temperature measurements
 - 5 thermocouples in the fuel bed (at three different vertical positions)
 - Thermocouples in the flue gas above the fuel bed
- Combustion air flow
- Pressure in the reactor



Measurements, sampling and analyses – sampling and analyses

Fuel sampling and analyses(C, H, N, ash content, moisture content,S, CI, major and minor ash forming elements)

Ash sampling and analyses

- optical evaluation
- chemical analyses
 (TOC, TIC, major and minor elements)







Evaluations

- Evaluation of characteristic data describing the thermal fuel decomposition
 - start and duration of the volatilisation and charcoal gasification phase
 - start and duration of the main charcoal combustion phase
 - maximum fuel bed temperatures measured during these phases
 - decomposition rates determined during these phases
- Evaluation of the gas phase measurements
 - trend of the main flue gas species and NO_x precursors
 - trend of the excess air ratio over the whole test run
- Evaluations based on the results from the fuel and ash analyses
 - release of ash forming elements



Beech woodchips (example) – mass loss over time



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dm/dt [%/min]

— pressure drop reactor [Pa]

– mass decrease (scale) [g]



Beech woodchips (example) – flue gas species



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Beech woodchips (example) – bed and gas temperatures



Average flue gas temperature: measured above the fuel bed before reactor outlet



Beech woodchips (example) – NO_x precursors



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Beech woodchips (example) – conversion of fuel N to TFN



TFN ... Total Fixed Nitrogen (sum of: NO, NH₃, HCN, NO₂, N₂O)



Release of NOx precursors (I) share of N species on TFN





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Release of NO_x precursors (II)

Share of NH₃ on total fixed nitrogen (TFN) [%]:

high:	70.2 – 87.7	waste wood pellets, straw pellets,		
		Miscanthus pellets, sewage sludge		
medium	37.7 – 51.9	torrefied softwood. SRC poplar, spruce bark		

Iow: 24.2 – 30.7 softwood pellets, spruce woodchips, beech woodchips

Share of HCN on total fixed nitrogen (TFN) [%]:

- high: 47.6 51.1 softwood pellets, spruce woodchips, beech woodchips
- medium: 24.1 39.6 torrefied softwood, SRC poplar, spruce bark
- low: 1.8 10.7 waste wood pellets, straw pellets, Miscanthus pellets, sewage sludge

Share of NO on total fixed nitrogen (TFN) [%]:

- medium: 16.1 22.7 wood fuels
 low: 3.5 6.9 waste wood pellets, straw pellets, Miscanthus pellets, sewage sludge, torrefied softwood
- During charcoal combustion NO formation clearly dominates



Conversion rate of N to TFN versus N content of the fuel





CFD simulations of NO_x emissions



CFD simulation of NO_x formation – model overview

- Extension of an empirical fixed bed model for solid biomass conversion on the grate by the relevant NO_x precursor species NO, NH₃ und HCN or utilisation of the tar conversion model in product gas burners
- Eddy Dissipation Concept
- Detailed kinetics (Kilpinen 92) and reduced kinetics mechanism (Kilpinen 97-Skeletal)
- ISAT (In-Situ Adaptive Tabulation) algorithm for the tabulation of reaction kinetics during run-time (for the reduction of calculation time)
- Cell clustering algorithm for further calculation time reduction
- Appropriate CFD-based NOx formation model developed by BIOS in cooperation with the Graz University of Technology



CFD simulation of NO_x formation – global kinetics vs. detailled kinetics

- Global reaction kinetics coupled with EDM
 unrealistic results
- Detailed reaction kinetics coupled with EDC

 → good results but long calculation time
 Usage of reduced reaction mechanisms is necessary

Mechanism	Calculation model	Calculation time	Concentrations in flue gas at boiler exit		
				NO _x	NH ₃
			[h]	[ppmv]	[ppmv]
Measurement				264	0
De Soete	EDM		1-2	203	1977
Brink / Kilpinen	EDM		1-2	637	0
Kilpinen92	EDC	3	350	286	0



CFD simulation of NO_x formation – Scope of work

- 3D CFD NO_x formation model (postprocessor) including detailed reaction kinetics for biomass grate furnaces
 - must be applicable to engineering problems
 - with reasonable accuracy
 - with reasonable calculation time
- Application of the CFD NO_x postprocessor
 - Simulation of a pilot-scale biomass grate furnace and comparison with measurement data taken during test runs





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Modelling of turbulent reactive flow – basic combustion simulation

- Turbulence
 - Gas phase combustion

Eddy Dissipation model $(A_{mag} = 0.6) /$ global methane 3-step mechanism $(CH_4, CO, CO_2, H_2, H_2O und O_2)$

Radiation

Discrete Ordinates model

Realizable k-*e* model

Modelling of NO_x formation – postprocessing mode

- Eddy Dissipation Concept (EDC)
- reduced "skeletal Kilpinen97" reaction mechanism (28 species, 104 reactions)
- ISAT (In-Situ Adaptive Tabulation) algorithm for reaction kinetics

ZAHIROVIĆ, 2008: CFD analysis of gas phase combustion and NO_x formation in biomass packed-bed furnances, PhD Thesis ZAHIROVIĆ et al., 2011: Validation of flow simulation and gas combustion sub-models for CFD-based prediction of NO_x formation in biomass grate furnaces. In: Combustion Theory and Modelling (2011), Vol. No. 15, Issue No. 1, pp. 61-87



CFD model geometry basic variant





Basic operating conditions – chemical composition of grass pellets

Parameter	Unit	Grass pellets basic	
C	[wt.% (d.b.)]	48.17	
н	[wt.% (d.b.)]	6.82	
0	[wt.% (d.b.)]	31.83	
Ν	[wt.% (d.b.)]	5.77	
S	[wt.% (d.b.)]	0.69	
ash	[wt.% (d.b.)]	6.72	
moisture	[wt.% (w.b.)]	10.81	
GCV (analysed)	[MJ/kg (d.b.)]	21.20	
GCV (Gaur)	[MJ/kg (d.b.)]	21.40	
NCV	[MJ/kg (w.b.)]	17.30	



Basic operating data

Parameter	Unit	Grass pellets basic
Adiabatic flue gas temperature	[°C]	1,361
Fuel power (related to NCV)	[kW]	432
Flue gas in combustion chamber - total	[kg/h]	949
- Flue gas release from fuel	[kg/h]	85
- Combustion air - total	[kg/h]	864
Primary air (below grate)	[kg/h]	403
Secondary air (nozzles)	[kg/h]	461
recirculated flue gas	[kg/h]	-
Stoichiometric air ratio – fuel bed	[-]	0.84
Total stoichiometric air ratio	[-]	1.67
O2 fraction at combustion chamber outlet, dry	[Vol% (d.b.)]	8.4



Results of basic analysis – temperatures and O₂ concentrations



Iso-surfaces of temperatures [°C] (left) and O₂ concentrations [m³ O₂/ m³ wet flue gas] (right) in the symmetry plane of the combustion chamber and the boiler



Results of basic analysis – NH₃ and HCN concentrations



Iso-surfaces of NH₃ concentrations [ppmv w.b.] (left) and HCN concentrations [ppmv w.b.] (right) in the symmetry plane of the combustion chamber and the boiler 36





Iso-surfaces of the reaction rates [kmol/(m³*s)] of the reaction N + NO \rightarrow N₂ + O for the reduction to N₂ (left) and of the reaction N + NO \leftarrow N₂ + O for the formation of NO from N₂ (right) in the symmetry plane of the combustion chamber and the boiler ³⁷



Results of basic analysis – NOx concentrations and TFN/TFN_{in} ratio

0.19

1.00

0.90

0.80

0.70

0.60

0.50

0.40

0.30

0.20

0.10

0.00



Iso-surfaces of NO_x concentrations [ppmv w. b.] in the symmetry plane of the combustion chamber and the boiler

<u>explanations</u>: NO_x concentrations as sum of NO, NO_2 and N_2O concentrations, all in [ppmv w. b.]

Iso-surfaces of local TFN/TFN_{in} ratios in the symmetry plane of the combustion chamber and the boiler

explanation: all data taken from reactor experiments with lab-scale pot furnace;

TFN ... mass of all N-moles contained in NO, NH_3 , NO_2 , HCN und N_2O , released from the fuel bed 38



Evaluation of basic analysis:

- small primary combustion zone (small flue gas residence time for reduction)
- thermal NO_x (high local flue gas temperatures)

Measures taken for optimization:

- new position of secondary air nozzles
- flue gas recirculation (temperature control)





Parameter	Unit	Grass pellets	Grass pellets
		basic	optimised
Adiabatic flue gas temperature	[°C]	1,361	1,042
Fuel power (related to NCV)	[kW]	432	370
Flue gas in combustion chamber - total	[kg/h]	949	1,126
- Flue gas release from fuel	[kg/h]	85	72
- Combustion air - total	[kg/h]	864	753
Primary air (below grate)	[kg/h]	403	362
Secondary air (nozzles)	[kg/h]	461	391
recirculated flue gas	[kg/h]	-	301
Stoichiometric air ratio – fuel bed	[-]	0.84	0.79
Total stoichiometric air ratio	[-]	1.67	1.64
Effective stoichiometric air ratio on grate ⁽¹⁾	[-]		0.91
Effective stoichiometric air ratio in primary combustion zone ⁽²⁾	[-]		1.03
Ratio of recirculated flue gas below grate	[-]	-	0.52
Flue gas recirculation ratio	[-]	-	0.27
O2 fraction at combustion chamber outlet, dry	[Vol% (d.b.)]	8.4	8.3

⁽¹⁾stoichiometric ratio including primary air and recirculated flue gas below the grate

⁽²⁾stoichiometric ratio including primary air and recirculated flue gas below the grate and through the nozzles



CFD model geometry optimised variant





Results of optimization – NO_x concentrations

grass pellets grass pellets basic geometry optimized geometry basic operating conditions optimized operating conditions 800 525 mg/Nm³ dry flue gas; 13 Vol% O₂ 837 mg/Nm³ dry flue gas; 13 Vol% O₂ 640 480 320 160

0

Iso-surfaces of NO_x concentrations [ppmv w. b.] in the symmetry plane of the combustion chamber and the boiler

explanations: NO_x concentrations as sum of NO, NO₂ and N₂O concentrations, all in [ppmv w. b.]





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grass pellets basic geometry basic operating conditions



Results of optimization – TFN/TFN_{in} ratios

grass pellets optimized geometry optimized operating conditions



Iso-surfaces of local TFN/TFN_{in} ratios in the symmetry plane of the combustion chamber and the boiler <u>explanation</u>: TFN ... mass of all N-moles contained in NO, NH₃, NO₂, HCN und N₂O, released from the fuel bed

Results of optimization – measurement compared to simulation results



	Unit	Grass pellets optimised
simulated NO _x -emissions (calculated as NO ₂) at boiler exit	mg NO _x /Nm³ dry fuel gas; 13 Vol.% O2	525
measured NO _x -emissions	mg NO _x /Nm³ dry fuel gas; 13 Vol.% O2	572



NO_x reduction by primary measures



Project ERANET-FutureBiotec -Measurement set-up





Project ERANET-FutureBiotec – operating data - fuel chipboard

General test run data

- Average amount of false air (related to the total combustion air input): 10.1%
- Average boiler capacity for the test runs performed: 150 kW at full load and 75 kW at partial load
- Average O₂ content in the flue gas:
 6.2 Vol% (dry flue gas)
 corresponding to a total air ratio of 1.4
- Average CO emissions:
 19 mg/Nm³ (dry flue gas, 13% Vol% O₂





Project ERANET-FutureBiotec – test programme

Test series 1, 2, 3:

Side constraints: full load (150 kW), fg recirculation above grate and large PCC Varied parameter: primary air ratio Fixed parameter: temperature at PCC outlet: 900; 1,000 and 1,100 °C

Test series 4, 5, 6: Side constraints:

Side constraints: full load (150 kW), fg recirculation below grate and large PCC Varied parameter: primary air ratio Fixed parameter: temperature at PCC outlet: 900; 1,000 and 1,100 °C

Test series 7:

Side constraints: full load (150 kW), fg recirculation above grate and small PCC Varied parameter: primary air ratio Fixed parameter: temperature at PCC outlet: 1,000 °C

Test series 8:

Side constraints: full load (150 kW), fg recirculation below grate and small PCC Varied parameter: primary air ratio Fixed parameter: temperature at PCC outlet: 1,000 °C

Test series 9:

Side constraints: partial load (75 kW), fg recirculation above grate and large PCC Varied parameter: primary air ratio Fixed parameter: temperature at PCC outlet: 1,000 °C

fg ... flue gas; PCC ... primary combustion chamber



Project ERANET-FutureBiotec – analysis of the chipboard used

fuel		chipboard	chipboard	chipb	oard - data	base values
analysis-no.		8,673	9,131	min.	max.	no. of samples
mc	wt% w.b.	9.1	6.9			
gross calorific value	<mark>kJ/kg</mark> d.b.	19,500	19,600	19,600	19,600	1
ac	wt% d.b.	0.88	1.20	0.52	1.49	6
ac (without TIC)	wt% d.b.	0.75	1.01			
C	wt% d.b.	47.4	48.0	47.7	51.2	17
Н	wt% d.b.	6.2	6.0	5.4	7.3	17
Ν	wt% d.b.	3.6	3.8	2.4	3.6	17
S	mg/kg d.b.	322.0	179.0	151.0	630	5
CI	mg/kg d.b.	285.0	191.0	123.0	1,250	5
Si	mg/kg d.b.	788	1,240	36.3	1,360	8
Ca	mg/kg d.b.	1,770	2,190	1,197	3,001	8
Mg	mg/kg d.b.	333.0	434.0	176.0	310.0	8
K	mg/kg d.b.	600	698	527	980	8
Na	mg/kg d.b.	192.0	232.0	90.9	301.0	8
P	mg/kg d.b.	73.1	76.0	142.0	149.0	2
Zn	mg/kg d.b.	60.7	42.0	15.6	64.3	8
Pb	mg/kg d.b.	9.4	5.0	1.73	61.0	5



Test runs performed with chipboard – air ratio in the PCC versus NO_x emissions for varying temperatures in the PCC



Explanations: Rec ... flue gas recirculation; PCC ... primary combustion chamber; T_PCC ... temperature at outlet of the primary combustion chamber;



Test runs performed with chipboard – air ratio in the PCC versus NO_x emissions for varying residence time



Explanations: Rec ... flue gas recirculation; PCC ... primary combustion chamber; T_PCC ... temperature at outlet of the primary combustion chamber;



Test runs performed with chipboard – air ratio in the PCC versus NO_x emissions for different type of flue gas recirculation



Explanations: Rec ... flue gas recirculation; PCC ... primary combustion chamber; T_PCC ... temperature at outlet of the primary combustion chamber;



Project ERANET-FutureBiotec – influence of air staging on NO_x emissions for different fuels



 NO_x as NO_2 related to dry flue gas and 13vol% O_2 ; all measurements at full load; PCC ... primary combustion chamber Side constraints: Rec. above grate, T_PCC = 1,000°C, large_PCC, full load



- λ_{PCC} is the most relevant influencing parameter
- technology dependent optimum of λ_{PCC} at λ_{PCC} <1</p>
- the optimum λ_{PCC} seems to be fuel independent for a certain combustion technology
- a second relevant influencing parameter is the residence time in the PCC (increasing NO_x emissions with decreasing residence time)
- air staging shows a great NO_x reduction potential
- The amount of false air should be minimized



Range of NO_x emissions at different operation conditions Data taken from test runs with wood chips, willow and chipboard



NO_x reduction by secondary measures



If secondary measures are necessary, they should always be combined with primary measures.

Possible secondary measures:

SNCR: selective non catalytic reduction

- Injection of a reducing agent (ammonia or urea) into the hot furnace
- Temperature window: between 850 and 950°C

SCR: selective catalytic reduction

- Injection of a reducing agent into the flue gas (downstream the boiler)
- Reducing agents:
 - ammonia (at temperatures between 220 and 270°C)
 - urea (at temperatures between 400 and 450°C)
- Platinum, titanium or vanadium oxide based catalysts are used for SCR

Reduction reactions

 $4NO + 4NH_3 + O_2 => 4N_2 + 6H_2O$ $3NO_2 + 4NH_3 + O_2 => 3N_2 + 6H_2O$



NO_x-reduction by secondary measures



Urea based SNCR usually increases the CO emissions by about 10 to 20 ppm stoichiometric coefficient: NH₃/NO_x respectively CO(NH₂)₂/2NO_x



SNCR process: costs (example):

- Investment costs of a SNCR unit for a biomass boiler system (nominal thermal capacity of 5.2 MW): 270,000 € (excl. VAT) (including filling device, storage tank (22 m³), nozzels, dosing and control unit)
- Operating cost SNCR unit : 5,000 - 20,000 €/a (depending on the N content in the fuel)



NO_x-reduction by secondary measures



Constraints:

- reducing agent: ammonia or urea
- stoichiometric coefficient ~ 1.0
- operation temperature: 220-270°C (ammonia) respectively 400-450°C (urea)
- > average NO_x-reduction: 80 to 95%
- considerably higher investment costs than SNCR
- Iower operating costs concerning reducing agent
- catalyst deactivation by alkali metals represents a major problem



Inhalt

Summary and conclusions



Conclusions (I) – Investigation of release profiles based on lab-scale reactor tests

The results of the lab-scale reactor tests provide valuable information about different aspects of fixed-bed combustion

Thermal decomposition behaviour as well as data concerning the main flue gas species released from the fuel during devolatilisation and charcoal combustion

Adata can further be used for the development, validation and calibration of models for fixed-bed combustion

- Comprehensive data on the release of NO_x precursors
 can be applied as input data for NO_x post-processing in CFD simulations
- Data regarding ash related problems
 - indications regarding the ash melting behaviour
 - estimation of aerosol and deposit formation tendencies

release data can be used for subsequent CFD-based aerosol and deposit formation modelling



Conclusions (II) – Investigation of release profiles based on lab-scale reactor tests

N-species release from the fuel bed

- Major NO_x precursors released are HCN and NH₃
 - Clearly NH₃-dominated: waste wood pellets, straw pellets, Miscanthus pellets, sewage sludge
 - HCN-dominated: softwood pellets, spruce woodchips, beech woodchips
 - Similar NH₃ and HCN release was observed for: torrefied softwood, SRC poplar
 - Wood fuels except torrefied softwood show higher NO fraction
- \blacktriangleright NO and NO₂ are also released but to a smaller extent than NH₃ and HCN
- N-species release is relevant for NO_x formation modelling



- 3D simulations of biomass grate furnaces with a CFD NO_x post-processor including detailed chemistry are a valuable tool to predict NO_x emissions.
- Detailed information of NO_x formation and reduction in grate combustion plants as well as relevant influencing parameters can be gained.
- Good qualitative and semi-qualitative agreement of simulation results with measurements achieved for different biomass fuels.
- The NO_x postprocessor for biomass grate furnaces is a powerful tool for the design and optimisation of furnace geometries and process control in order to optimize NO_x reduction by primary measures but also to design SNCR system integration



Conclusions (IV) – NO_x reduction by primary measures

- The air ratio in the primary combustion chamber is the most important influencing parameter regarding NO_x emissions
- The lowest NO_x emissions were detected at an air ratio in the primary combustion chamber of 0.9 1.0
- False air input should be minimised because it cannot be controlled
- The results also show that the residence time (volume of the PCC) has a relevant influence on NO_x emissions (this influence seems to get of rising importance the smaller the residence time available in the PCC is). A residence time of at least 0.5 s can be recommended for low-NO_x operation.
- The temperature in the PCC has a minor influence on NO_x emissions as long as it is in the target range of 900 – 1,100°C.
- Flue gas recirculation above the grate seems to be slightly more efficient regarding NO_x reduction than flue gas recirculation below the grate (at same temperature conditions)
- → The potential to reduce NO_x emissions by primary measures is considerable (typically in the range of 30 50%)



Conclusions (V) – NO_x reduction by secondary measures

SNCR process:

- Average NO_x-reduction: 60 to 70%
- Moderate investment costs but higher operating costs
- SNCR process:
 - Average NO_x-reduction: 80 to 95%
 - Higher investment costs but lower operating cost
- → Secondary measures should always be combined with primary measures



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Thank you for your attention

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