MODELING OF SOLID FUELS COMBUSTION IN A COMPLEX GEOMETRY CLC EQUIPMENT

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Abstract – Since chemical looping combustion (CLC) of solid fuels it is still an emerging technology and the complexity of a process is not sufficiently recognized, the development of models of CLC equipment, especially gaseous pollutants emissions, is of practical significance.
The paper presents an empirical model of NO_x ($NO + NO_2$) and N_2O emissions from coal combustion in a complex geometry of fluidized bed Chemical-Looping-Combustion of Solid-Fuels (FB-CLC-SF) facility. The study was conducted for three hot tests (test 1 – 3), at which the unit operated stably and smoothly. The tests 1 and 2 were carried out in air, and gas mixture composed of 21% O_2 and 79 % CO_2, respectively. During test 3 CO_2 and ilmenite were fed into the fuel reactor as a fluidizing gas. The validity of the model was successfully performed on a 5-7 kW_{th} hot FB-CLC-SF equipment.
The presented method constitutes a non-iterative and easy to use procedure allowing to predict gaseous pollutants emissions.
Such approach, often applied by commercial manufactures of industrial-scale boilers is a non-iterative and easy to use technique in prediction of NOx and N_2O emissions from a CLC unit.

INTRODUCTION

There are many different approaches in modeling of gaseous emissions during solid fuel combustion in fluidized bed systems. A wide-range review was provided by Basu (1999). The author distinguished three levels of details or/and sophistication: level I: 1-D, plug flow/stirred tank, using simple mass and energy balance; level II: core-annulus, 1.5-D with broad consideration of combustion and other related processes; level III: 3-D models based on Navier-Stokes Equation with detailed consideration of chemical kinetics and individual physical processes. Similar classification was also discussed by Gungor and Eskin (2008). Miller and Bowman (1989) described the nitrogen chemistry taking into account complex reactions network. The authors provided 234 reactions in Appendix A and 73 reactions in Appendix B.
Leckner and Lyngfelt (2002) developed a two-dimensional coal combustion model in a CFB boiler. The model was successfully validated against data from a pilot-scale 50 kW CFB combustor as well as an industrial – scale 160 MW CFB boiler.
A review of macroscopic (semi-empirical) models for fluid dynamics of circulating fluidized bed boilers was given by Pallares and Johnsson (2006). The authors underlined, that as the nature of the industrial processes is often non-linear and extremely complex, models usually include some empirical parameters to provide necessary data in the cases where up-to-date modeling is unsatisfactory. It happens e.g. to adjust parameters of the model, which could not be determined immediately, especially for different operating conditions.
This situation also refers to nitrogen chemistry, NO_x and N_2O emissions from chemical looping combustion (CLC) units. The emissions of NO_x and N_2O are the result of complex of competing formation and destruction mechanisms (Mondal et al., 2012). Among them are: volatile content of fuel, bed temperature, air excess, gas residence time in the furnace and the presence of limestone in the combustion chamber (Czakiert et al., 2012; Diego et al., 1996; Knobig et al., 1998; Krzywanski and Nowak, 2017; Leckner et al., 2004; Liukkonen, 2011; Scheffknecht et al., 2011; Tan et al., 2012; Toftegaard et al., 2010; Zhao et al., 2012).
The NO_x concentration increases whilst N_2O concentration decreases with the increase of bed temperature. Both NO_x and N_2O concentrations increase with the increase in excess air. Limestone addition leads to the increase in NO_x and decrease in N_2O concentrations (Diego et al, 1996; Zhao et al., 2012). These results are consistent with the opinion that low rank coals can yield more NO_x than higher rank ones (Hayhurst and

Lawrence, 1996). The residence time of gas in the bottom zone of the combustion chamber leads to the NO_x decomposition more dominant mechanism over the NO_x formation and the atmosphere with limited oxygen concentration leads to conversion of volatiles-N to N_2 instead of NO_x (Gungor and Eskin, 2008).

There are also some empirical correlations and rules, which are applied to predict gaseous pollutants emissions by manufactures of industrial fluidized bed boilers. These formulas take into account basic dependencies between emissions and the operating parameters and are quantified based on data obtained from operation of both pilot- and large-scale units.

Such approach was adopted in this paper to develop a model of NO_x and N_2O emissions form the complex geometry Fluidized-Bed Chemical-Looping-Combustion of Solid-Fuels (FB-CLC-SF) equipment. An empirical model of NO_x ($NO + N_2O$) and N_2O emissions has been developed to study gaseous pollutants emissions from a complex geometry FB-CLC-SF unit. Simulation results agree well with experimental data not only quantitatively. The proposed methodology allows achieving also qualitative agreement of calculated data with experimental results.

MATERIALS AND METHODS

The presented idea of solid fuel combustion combines two technologies: Chemical Looping Combustion (CLC) and In-situ Gasification Chemical Looping Combustion (iG-CLC). As the result an innovative Hybrid Chemical Looping Combustion came into existence with these two complementary technologies.

The studies, calculations and model validation were performed on the 5-7 kW_{th} hot Chemical-Looping-Combustion of Solid-Fuels facility, given in Fig. 1.

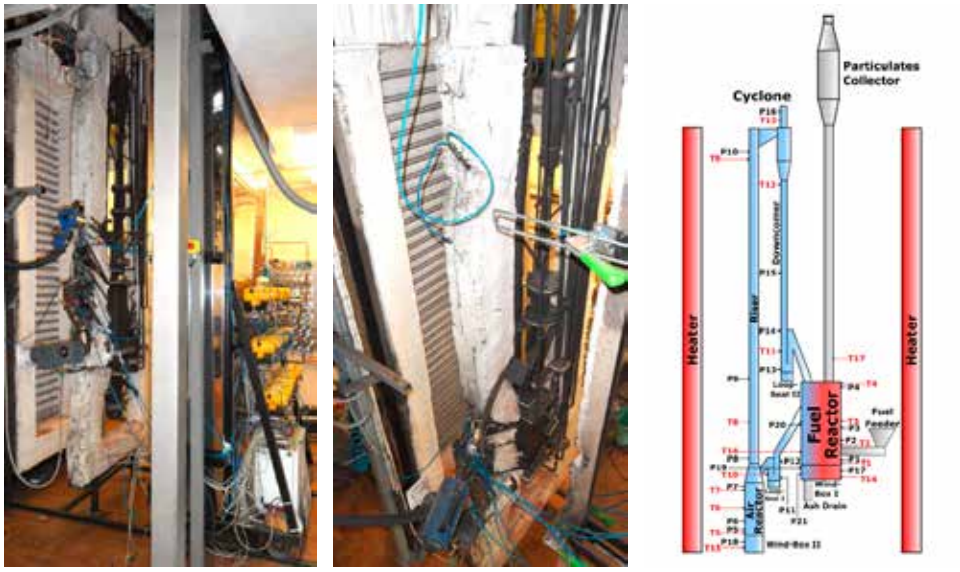

Fig. 1. The CLC unit.

The geometry of the fuel reactor is rather complex. The equipment consists of two main parts: air reactor (AR) and fuel reactor (FR). The FR consists of several chambers, connected each-others, but two of them (Chambers I and II) are the largest of all cells in FR. The cross section of the AR is circular, whereas the one of FR is rectangular. The geometry of the equipment is described in Tables 1 and 2. The fluidization regime which exists in the fuel reactor corresponds to bubbling fluidized bed, whereas AR operates under circulating fluidized bed conditions.

The study was conducted for three hot tests (test 1 – 3), at which the unit operated stably and smoothly. The main difference between tests is the atmosphere at which the combustion occurred. The tests 1 and 2 were carried out in air, and gas mixture composed of 21% O_2 and 79 % CO_2, respectively.

Table 1: The main dimensions of the air reactor and cyclone.

The diameter of the air reactor, [m]	0.098
The total height of the riser and air reactor, [m]	2.55
The diameter of the riser, [m]	0.04
The efficiency of the cyclone, [%]	99.0
The inner diameter of the cyclone, [m]	0.066
The height of the cylindrical portion of the cyclone, [m]	0.190
The height of the conical part of the cyclone, [m]	0.130
The diameter of the downcomer, [m]	0.028

Table 2: The main dimensions of the fuel reactor (Chambers I and II).

The inner equivalent diameter of combustion chamber, [m]	0.074
The height of combustion chamber, [m]	0.500

During test 3 CO_2 was fed into the FR as a fluidizing gas. The ilmenite with the Sauter mean diameter of particles of 161 μm, true density of 3879 kg/m^3 and the sphericity of 0.65 was also used during the Test 3 (Fig. 2).

Fig. 2. Microscopic photograph of ilmenite - research material used in the study

Since ilmenite is mainly composed of $FeTiO_3$ ($FeO{\cdot}TiO_2$), the iron oxide is the active phase which behaves as oxygen-carrier (Adanes et al., 2012).

The sand with the Sauter mean diameter of particles of 166 μm, true density of 3200 kg/m^3 and the sphericity of 0.7 were used during the investigations (Fig. 3).

Fig. 3. Microscopic photograph of sand - research material used during the investigations.

The Sobieski bituminous Polish coal was fed into the unit with CaO content in ash 6.81 %. The properties of the fuel are shown in Table 3.

Table 3: Properties of coal (as received).

LHV	Proximate analysis/wt. %				Ultimate analysis/wt. %				
[MJ kg^{-1}]	W	VM	A	FC$^{\text{by diff.}}$	C	H	S	N	O$_2$$^{\text{by diff.}}$
23.429	13.3	29.49	8.2	49.01	61.9	3.66	1.39	0.99	10.56

The main operating parameters for all tests, including gas mass flow rate, bed pressures and temperatures as well as the total mass of solids, residence time and excess air are given in Table 4.

Table 4: Main operational conditions of the FB-CLC-SF unit.

No of test / gas to FR	**1**/air	**2**/21% O_2 / 70% CO_2	**3**/CO_2 + Ilmenite
Gas mas flow rate in AR, [$^{x}3600^{-1}$ kg s^{-1}]	4.21	4.21	4.21
Total gas mas flow rate in FR, (chambers I and II) [$*3600^{-1}$ kg s^{-1}]	3.49	5.41	5.73
Fuel mas flow rate m_f, [kg s^{-1}]	$1 \cdot 10^{-4}$	$1 \cdot 10^{-4}$	$4 \cdot 10^{-5}$
Bed temperature T, [K]	1147	1135	1117
Absolute pressure below the gas distributor in AR, [Pa]	105 087	105 873	104 149
Gas residence time in FR τ, s	1.24	1.33	1.38
Excess air in FR λ, -	1.13	1.75	0.592
Absolute pressure below the gas distributor in FR, [Pa]	105 083	105 654	105 360
Total mass of solids in the AR, [kg]	1.90	2.79	1.42
Total mass of solids in the FR, [kg]	2.56	2.40	2.36

For the purpose of this work an empirical model of NO_x and N_2O has been developed to study gaseous pollutants emissions from complex geometry Fluidized-Bed Chemical-Looping-Combustion of Solid-Fuels equipment.

The procedure involves two steps. During the first step the NO_{xbase} and N_2O_{base} emissions are determined at model base conditions. The second stage constitutes in corrections of the previously obtained emissions due to deviations of the experimental conditions from the model base conditions. Such approach, often applied by commercial manufactures of industrial-scale boilers is a non-iterative and easy to use procedure to predict gaseous pollutants emissions.

The validation of the developed models has been successfully performed using data from a hot fluidized bed CLC equipment. The maximum relative errors between experimental and calculated results do not exceed 10 %. The model allows to study the influence of the operating parameters on the NO_x and N_2O levels emissions from the considered of complex geometry, FB-CLC-SF unit.

RESULTS AND DISCUSION

The procedure combines experience in modelling of gaseous pollutant emissions in fluidized beds (Krzywanski et al., 2010a, 2010b, 2011, 2013, 2014, 2016, 2017). Some data presented in the paper do not indicate references, since some of the results used in the paper are confidential.

For the purpose of this work the empirical model was developed allowing to estimate the NO_x (i.e. NO+NO_2) and N_2O emissions from solid fuels combustion in the considered FB-CLC-SF facility.

The NO_x emissions

The nitrogen oxides emissions depend mainly on the volatile content o fuel, bed temperature, lime content in the ash and excess air (Zhao et al., 2012, Gungor and Eskin, 2008). Since the volatile content of the fuel is one of the most important factor the NO_{xbase} emission level can be quantified as the function of VM/LHV (1):

$$\mathrm{NO}_{\mathrm{xbase}} = 127239\left(\frac{\mathrm{VM}}{100 \cdot \mathrm{LHV}}\right)^2 + 2349\left(\frac{\mathrm{VM}}{100 \cdot \mathrm{LHV}}\right) + 66.015 \quad (1)$$

The obtained data correspond to NO_x emissions in dry flue gas (normalized to 6% O_2 basis), bed temperature 1144 K and excess air 1.2. For the considered VM content and LHV of fuel the NO_{xbase} concentration in dry flue gas is equal 115.7 ppm.
Such NO_{xbase} emission level was used in further calculations during the second stage. Since the bed temperature during all tests are different than the temperature at the above given base conditions and was changing in the range of 1117-1147 K (see Table 3) the additional temperature correction factors k_T should be taken into account:

$$k_T = 0.0000359\,T^2 - 0.0803\,T + 45.909 \quad (2)$$

It is also well known that lime content influences the NO_x emissions since the CaO has catalytic effect on the NO formation (Diego 1996; Duan et al., 2011; Krzywanski and Nowak, 2017). Therefore the lime correction factor k_{Ca} should be also predicted in the present consideration. Taking into account the experimental results the k_{Ca} factor can be described as follows:

$$k_{Ca} = 0.0079 \cdot M^3 - 0.0044 \cdot M^2 + 0.0901 \cdot M + 0.9861 \quad (3)$$

where:

$$M = \frac{CaO}{m_f \cdot LHV}, \quad [g \cdot MJ^{-1}].$$

The excess air is another factor which greatly influences the NO_x emissions and should be also considered in the model. The excess air correction factor k_λ can be determined by the formula:

$$k_\lambda = -3.2997 \cdot \lambda^2 + 10.471 \cdot \lambda - 4.6187 \quad (4)$$

Finally the fallowing Eq. (5) can be applied to determine the total NO_x emission level from the FB-CLC-SF unit in dry flue gas:

$$NO_x = NO_{xbase} \cdot k_T \cdot k_{Ca} \cdot k_\lambda \quad (5)$$

For such performed model we managed to predict the NO_x emissions in good accuracy. The comparison between measured and predicted by the developed empirical NO_x emissions model is given in Table 5.

Table 5: The NO_x emission (normalized to 6% O_2 basis) for the considered CLC tests.

No of test/gas to FR	NO_x^d [ppm]	NO_x^p [ppm]	Err [%]
1/air	393	355	9.7
2/21% O_2 / 70% CO_2	380	417	-9.7
3/CO_2 + Ilmenite	56	52	7.1

The N_2O emissions

The main parameters influencing the nitrous oxide emission level are: the fuel rank, bed temperature, oxygen excess and gas residence time (Czakiert, 2013; Diego et. al., 1996; Feng et al., 1996; Leckner et al. 2004).
For the purpose of this work the oxygen content of the coal is applied as a parameter defining its rank (Basu 2013; Jenkins et al., 1998). The N_2O emission level can be quantified in accordance with the following function:

$$N_2O_{base} = 0.0008 \cdot (O_2)^2 - 0.1202 \cdot O_2 + 6.1249 \quad (5)$$

The obtained data correspond to N_2O emissions in dry flue gas (normalized to 6% O_2 basis), bed temperature 1144 K and gas residence time 5s. For the considered O_2 content of fuel (expressed in dry, ash free conditions) the N_2O_{base} concentration in wet flue gas is equal to 4.9 ppm.
Such N_2O_{base} emission level was used for further calculations in the second stage. The following equation was applied to determine the influence of the excess air factor k_λ :

$$k_\lambda = -6.2754 \cdot \lambda^2 + 19.202 \cdot \lambda - 8.7855 \quad (6)$$

Finally the N_2O emission level from the FB-CLC-SF unit can be predicted by the Eq. (7):

$$N_2O = 2.34 \cdot N_2O_{base} \cdot k_\lambda \exp(-k\tau) \quad (7)$$

where:

$$k = k_0 \exp\left(-\frac{E}{RT}\right),$$

For such performed empirical model of N_2O emissions we managed to predict the N_2O emissions level in good accuracy. The comparison between measured and predicted N_2O emissions for the considered tests are given in Table 6.

Table 6: The N_2O emission (normalized to 6% O_2 basis) for each of the considered tests.

No of test/gas to FR	N_2O^d [ppm]	N_2O^p [ppm]	Err [%]
1/air	54	49	9.3
2/21% O_2 / 70% CO_2	53	58	-9.4
3/CO_2 + Ilmenite	5	4.6	8.0

The maximum relative error between experiment and simulation result does not exceeded 10 %.

NOx and N2O emissions for the real conditions

Such developed model allows to determine the influence of operating conditions on NO_x and N_2O emissions. As an example the effect of excess air is given in Fig. 4.

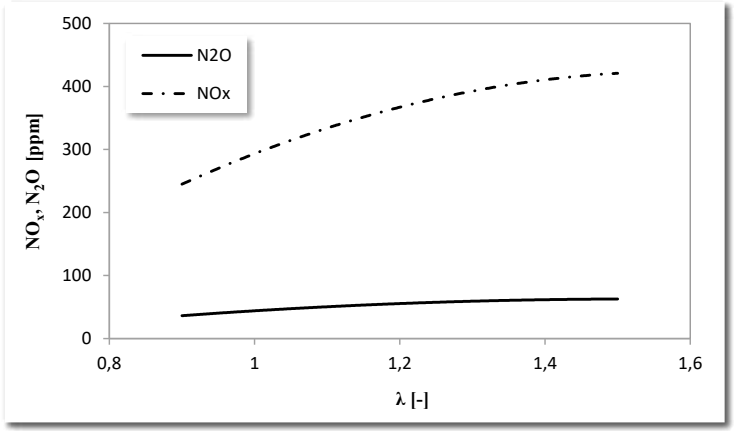

Fig. 4. Effect of excess air on NOx and N2O emissions, T=1123 K., τ = 1.3 s.

Both NO_x and N_2O emission increases with the increase in excess air. Similar behavior was reported by Diego et al. (1996).

CONCLUSION

An empirical model of NO_x ($NO + N_2O$) and N_2O emissions has been developed to study gaseous pollutants emissions from a complex geometry CLC unit. First simulations of NO_x and N_2O emissions from solid fuel combustion in CLC conditions with ilmenite, in O_2/CO_2 gas mixture and in air are shown in the paper.

The validity of the model was successfully performed on a 5-7 kW_{th} hot FB-CLC-SF equipment. The maximum relative error between experiment and simulation result does not exceed 10%. This confirms that the applied modeling approaches constitute an effective method for modeling of CLC processes.

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NOTATION

A ash content of the fuel, %

C carbon content, %

CaO lime mass flow rate, kg s^{-1}

E activation energy, E = 250 kJ mol^{-1}

F.C. fixed carbon content of the fuel, %

H hydrogen content of the fuel, %

k_{Ca} lime correction factor, -

k_0 pre-exponential factor, $k_0 = 2.733 \times 10^{10}$ s^1

k_λ correction factor for oxygen excess, -

k_T temperature correction factor, -

LHV the lover heating value, MJ kg^{-1}

m_f fuel mass flow rate, kg s^{-1}

N nitrogen content, %

NO_x concentration of NO_x ($NO + NO_2$), ppm

O_2 oxygen content of the fuel, %

R gas constant, R = 8.314 J mol^{-1} K^{-1}

S sulfur content of the fuel, %

T temperature, K

τ gas residence time, s

V.M. volatile matter content of the fuel, %

W moisture content of the fuel, %

Superscripts

d desired (measured)

p predicted (calculated)

Acronyms

CLC Chemical Looping Combustion

Err the relative error, %

FB-CLC-SF Chemical Looping Combustion of Solid-Fuels

iG-CLC In-situ Gasification Chemical Looping Combustion

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