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Emissions of CO_2, CO, NO_x and N_2O from dried lignite combustion in oxygen-enriched O_2/CO_2 atmospheres in a circulating fluidized bed boiler

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Abstract

The paper presents the idea of oxygen-enhanced dried lignite combustion in a circulating fluidized bed (CFB) boiler as it gives a possibility of reduction of gaseous pollutants emissions. Calculations were conducted using one-dimensional model, which describes crucial processes associated with combustion of solid fuels under circulating fluidized bed conditions, e.g., hydrodynamics of bed material, fuel devolatilization, volatiles and char combustion and heat transfer. The paper is focused on CO_2, CO, NO_x and N_2O emissions from a 670 t/h CFB boiler during dried lignite combustion in O_2/CO_2 atmosphere. The dependence of the gaseous pollutants' emissions on oxygen fraction in an inlet gas as well as moisture content in lignite were studied. The oxygen fraction was increased from 21 to 30% vol., whereas moisture content was decreased from 43.5 to 10% – simulating fuel pre-drying. A case study considers solution when process is run with recirculation of CO_2. The obtained results was compared to the case where CO_2 is not recycled.

Keywords: Oxy-coal combustion; CFB; Combustion; Modeling

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Nomenclature

CO2rec, CO2	–	CO_2 concentration in the flue gas with and without recirculation, respectively, vol.%
COrec, CO	–	CO_2 concentration in the flue gas with and without recirculation, respectively, ppm
$c_{v,b}$	–	volume fraction of solids in the bottom dense zone
c_v^*	–	volume fraction of solids in the upper dilute zone
dH	–	height of each element into which the combustion chamber was divided, m
D_e	–	hydraulic diameter of combustion chamber, m
H_k	–	height of combustion chamber, m
HV	–	lover heating value
m.l.	–	moisture content in the lignite, %
$\dot{m}_s$	–	mass flux rate of sorbent, $\mathrm{kg\,s^{-1}}$
$\dot{m}_z$	–	mass flux rate of fuel, $\mathrm{kg\,s^{-1}}$
nCO2, nCO, nNOx, nN2O	–	CO_2, CO, NO_x and N_2O molar flux rate without CO_2 recirculation, $\mathrm{kmol\,s^{-1}}$
nCO2rec, nCOrec, nNOxrec, nN2Orec	–	CO_2, CO, NO_x and N_2O molar flux rate respectively, with CO_2 recirculation, $\mathrm{kmol\,s^{-1}}$
ngas	–	molar rate flux of wet flue gas, $\mathrm{kmol\,s^{-1}}$
NOxrec, NOx	–	NO_x concentration in the flue gas with and without recirculation, respectively, ppm
N2Orec, N2O	–	NO_x concentration in the flue gas with and without recirculation, respectively, ppm
O2	–	O_2 concentration in the inlet gas mixture, vol.%
Pg	–	fraction of primary gas, vol.%
Sg	–	fraction of secondary gas, vol.%
Re_k	–	Reynolds number related to the diameter of combustion chamber,
z	–	distance of cross-section area from the grid, m
z_b	–	the height of the bottom region, m
δ_{hsp}	–	thickness of wall layer, m

1 Introduction

The technology of oxy-fuel combustion in circulating fluidized bed (CFB) boilers is a representative of clean combustion technologies. The CFB is also one of the most interesting applications for the oxy-fuel combustion [1–3]. The main advantage of the oxy-fuel combustion is the increase of carbon dioxide (CO_2) concentration in flue gas. This allows the capture of CO_2 from flue gas without high, additional energy input [4]. This technology leads to the other options for coal and is competitive with noncarbon technologies including wind, solar and nuclear ones [5].

The oxy-fuel combustion process needs to recycle a certain amount of flue gas. It is important in order to keep the temperature in combustion chamber, water and steam temperatures within reasonable limits. Is also prevents slagging and fouling of the heat transfer surfaces during coal combustion [4,5]. The recycled gas is some amount of the flue gas leaving the boiler. The flue gas consists mainly of CO_2 and water vapor. When coal is burned with nearly pure oxygen, the quantity of combustion byproduct is about 25% of the amount of byproduct produced when using air [5]. Some experimental data from oxy-coal combustion in a 300 kW laboratory-scale circulating fluidized bed combustor are presented in [6]. Tests were carried out for air and oxygen-enhanced conditions with the O_2 volumetric concentration in oxidant (wet): 21, 26, and 35%. The obtained average bed temperature increased linearly with oxygen concentration in the recirculated flue gas and changed from about 700 °C for 21% O_2 to about 980 °C for 35% O_2.

The idea of near zero-emissions power generation was presented also in [7,8]. As authors underlined, the idea can be realized under oxy-fuel combustion if all of the byproducts are recirculated to the furnace and undergo reburn and re-capture within the closed-loop system [7]. The advantages resulting from the oxy-fuel combustion process application are listed in [9]. Two variants of the technical realization of the combustion process were studied, i.e., with and without recirculation of CO_2. Although the implementation process without recirculation initiated the development of the concept of combustion in oxygen-enriched atmosphere, the realization with recirculation takes a leadership role, despite the technical difficulties associated with flue gas recirculation [10–12]. A typical recirculation rate reaches 65–80%, depending on the type of coal [9].

A two-dimensional computational fluid dynamics model was used to predict the oxy-coal combustion processes in a 50 kW CFB boiler at Southeast University China [13]. The effects of combustion atmospheres on distribution of solid volume fraction, temperature and gas composition in the riser were examined including oxygen concentration in the inlet gas. The model takes into account both indirect and direct desulphurization mechanisms.

The paper presents the idea of oxygen-enhanced dried lignite combustion in a CFB boiler as it gives a possibility of reduction of gaseous pollutants emissions. It constitutes a continuation of the previous research [14]. The obtained results were compared to the case, where CO_2 is not recycled so the flue gas contains additional portion of the component.

2 Calculation procedures and main assumptions

For the purpose of this work, the model was applied that uses the experience in modeling the existing 670 t/h CFB boiler, operated in Turow Power Station in Poland [14–20]. Validation of the model was also successfully performed for the above-mentioned boiler operated under air conditions as well as for 0.1 MW_{th} oxy-fuel-CFB test rig both operated with air and oxy-fuel atmosphere [15].

The calculations presented have been made for 670 t/h CFB boiler. The dimensions of cross-section area of the combustion chamber in its lower part, just above the grid, are 21.2 m×5.2 m. Then, the cross-section area increases to reach dimensions of 21.2 m×9.9 m at a height of 6.7 m above the grid level. The total height of the combustion chamber is 48 m. Two outlet windows, each of dimensions of 7.8 m×3.4 m, were situated in this way that the axis of cross-section area were located at a level of 38 m. All calculations are done for 100% load. The boiler is fired with the lignite. The properties and size distribution of the fuel are given in Tabs. 1 and 2, respectively. The limestone is also fed to the combustion chamber in parallel with coal. The properties and size distribution of sorbent are shown in Tabs. 3 and 4.

Table 1: Properties of lignite (as received state).

Lower heating value, MJ/kg	10.603
Average solid concentration, kg/m^3	1.265
Moisture, %	43.5
Ash, %	16.4
Volatile meter, %	19.9
Carbon, %	28.0
Hydrogen, %	2.85
Sulphur, %	0.33
Nitrogen, %	0.40
Oxygen$^{by\ diff.}$, %	8.52

The primary gas is supplied to the boiler through the gas distributor, in the bottom part of the boiler whilst the secondary gas – onto two levels: 0.75 and 1.25 m above the grid. To retain the regime of the circulating fluidized bed with the variable oxygen content in a supplied gas mixtures, there was the need to modify the overall dimensions of combustion chamber. It allows to maintain a constant gas velocity in the whole volume of combustion chamber at a level of

Table 2: Particles size distribution of lignite (simplified to three ranges).

Mean diameter, mm	0.20	0.58	2.06
Mass fraction, –	0.32	0.35	0.33

Table 3: Properties of limestone.

$CaCO_3$, %	$MgCO_3$, %	Inert, %	Moisture, %
92.0	2.5	4.5	1.0

Table 4: Particles size distribution of sorbent (simplified to three ranges).

Mean diameter, mm	0.20	0.58	2.06
Mass fraction, –	0.99	0.05	0.05

5.4 m/s. In the presented model, only one of the three characteristic chamber dimensions, i.e., the chamber depth, was subjected to changes that ranged from 9.9 m for oxygen content in inlet gas 21% to 7.0 m for O_2 content equal 30%. It corresponded to the reduction of the chamber volume by about 30% (from 8457 to 5920 m^3).

Because of the fact that following tests described in the paper correspond to different moisture content in lignite, the fuel feed rate increases with the increase in moisture content, to keep the constant heat flux to the boiler. Table 5 shows the properties and fuel feed rates corresponding to different moisture content in lignite.

Applied model consists of several submodels describing hydrodynamics of the bed material, chemical reactions (e.g., fuel devolatilization, volatiles and char combustion, where the char is considered as the porous solid residue) and heat transfer in the combustion chamber. Model is based on solids and gas balances and energy equations. The distribution of solids concentration in the lean zone is determined using the following expresion [16]:

$$c_v = (c_{v,b} - c_v^*) \exp\left[-a\,(z - z_b)\right] + c_v^* \,. \tag{1}$$

Table 5: Properties of the lignite as the function of moisture content (as received state).

Moisture, %	Carbon (c), %	Lower heating value, kJ/kg	Feed rate, kg/s
43.5	28.0	10991	63.0
30.0	34.7	14215	48.7
20.0	39.6	16603	41.7
10.0	44.6	18990	36.5

The Werdermann's empirical correlation has been used to obtain the thickness of the wall layer for a given height of cross-section area in th combustion chamber [21]

$$\delta_{hsp}(z) = \left[1.1\mathrm{Re}_k^{-0.33} \left(\frac{H_k}{D_e} \right)^{0.68} \left(\frac{H_k - z}{H_k} \right)^{0.92} \right] D_e \, . \tag{2}$$

The reaction space in the combustion chamber is divided into 96 elements, each of a height of $dH = 0.5$ m. The model covers homogeneous and heterogeneous reactions describing: char oxidation, formation and destruction of NO and N_2O, which are based on experiences given, among others, in [22–25]. The reaction rates are determined using Arrhenius-type equation, whereas the kinetic parameters for reactions used in the model are reported in [15–17]. A detailed description of the model, i.e., the structure, hydrodynamics and heat transfer equations as well as the system of chemical reactions can be found elsewhere [15–17].

The calculations have been done for the following conditions and main assumptions:

- oxygen concentration in O_2/CO_2 atmosphere from the range of 21–30%;
- moisture content in fuel of 43, 30, 20, and 10%;
- constant heat flux to the boiler;
- primary to secondary gas ratio: Pg/Sg = 60/40;
- constant density of burning fuel particles;
- the existence of two zones in the combustion chamber, i.e., a dense zone in the lower part of the combustion chamber and a dilute zone in the upper part, above the secondary gas feed points;
- particles pass from the lower dense zone to the upper dilute zone and are replenished with particles coming from the return system;
- fuel particles in the dense zone are distributed uniformly;
- the fuel drying and volatiles release is completed in the dense zone.

The method covered by the model belongs to the clean combustion technologies and constitute a supplement of the methods described by Dors [26].

3 Results and discussion

Calculations were performed for the case, when process runs with recirculation of CO_2 flux (Fig. 1a). The recycled CO_2 is the part of CO_2 contained in the flue gas. As the results the fallowing parameters have been obtained: (1) the fluxes of gaseous pollutants and their concentrations in dry flue gas, (2) flux of wet flue gas. The obtained results were compared to the case without CO_2 was not being recycled (Fig. 1b).

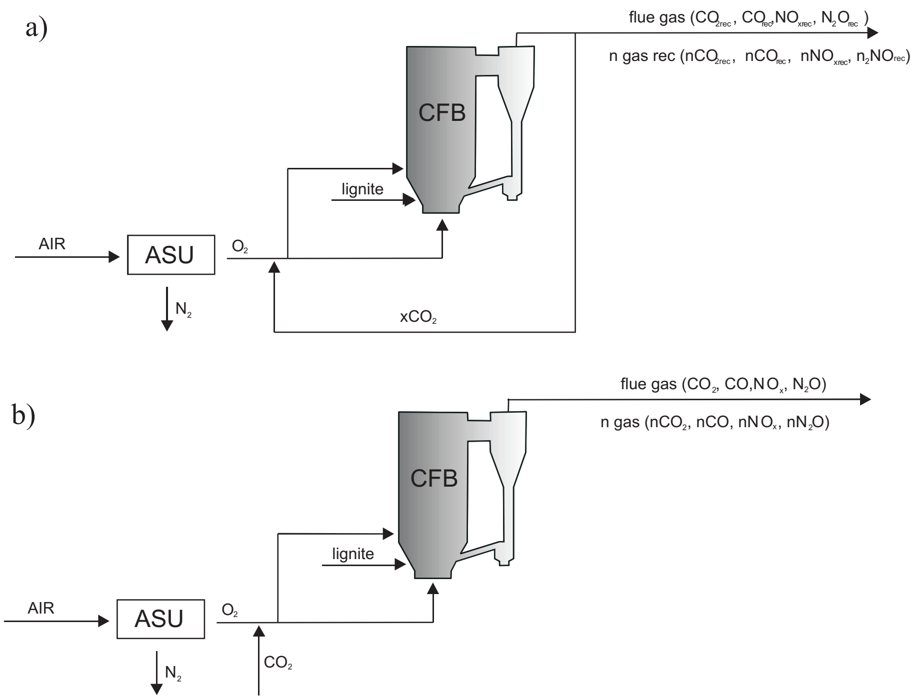

Figure 1: The two different implementations of the process: a) with the CO_2 recirculation, b) without the CO_2 recirculation: ASU - air separation unit, CFB – circulating fluidized bed.

The results of calculations are shown in Figs. 2–6. As it is shown the increase of the oxygen concentration leads to a decrease of CO_2 concentration. The decrease in CO_2 concentration for O_2/CO_2 environment comes from the excess of O_2. The decline is of about 80% with 21% of oxygen concentration in the inlet gas and about 73–78%, depends on the moisture level in the lignite, for oxygen-enriched conditions, with 30% of O_2 concentration in the mixture of O_2/CO_2 (Fig. 2).

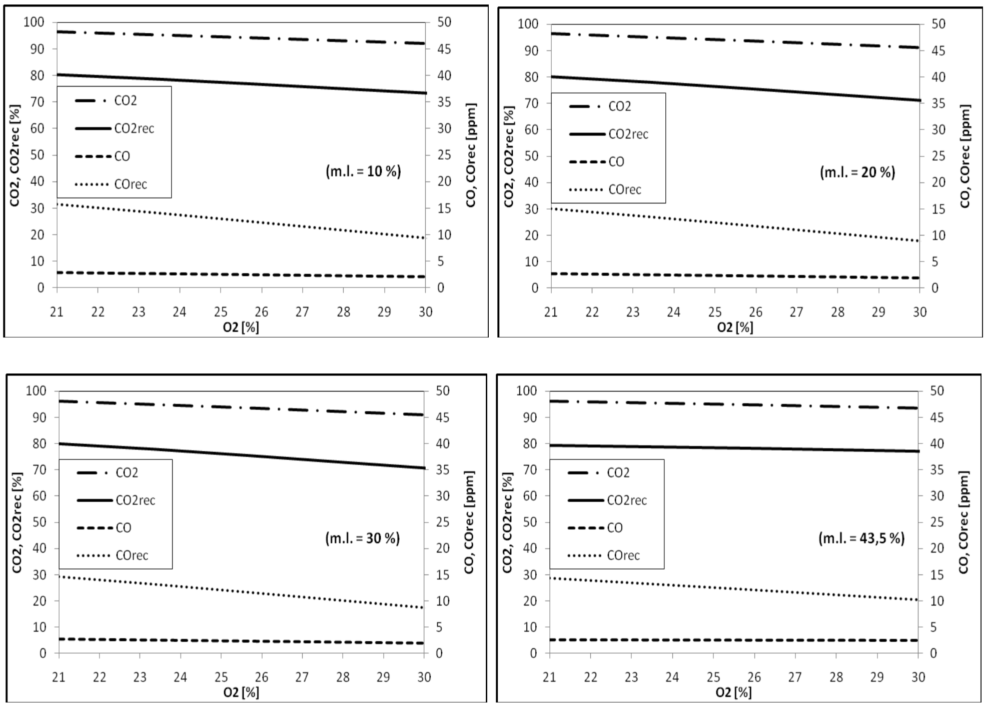

Figure 2: CO and CO_2 concentration in dry flue gas as a function of the oxygen content in inlet gas for different moisture levels in the lignite.

The CO_2 concentration is lower than that one without CO_2 recirculation. The decrease is of about 16% with 21% O_2 and 16–19% with 30% of oxygen concentration in the inlet gas. It is due to recycling back a portion of CO_2 flux in the implementation of the process with the CO_2 recirculation. The CO concentration in flue gas is higher than the one without recirculation. It is due to the fact, that the recirculation of CO_2 flux, condenses other flue gas components. The carbon monoxide (CO) in turn is conducive to nitrogen oxides (NO_x) reduction. The CO concentration changes from about 14–16 ppm with 21% O_2 to about 9–10 ppm with 30% O_2 concentration in the inlet gas. Due to recirculation a portion of CO_2 flux, the NO_x and N_2O concentration in flue gas is higher than the one without CO_2 recirculation (Fig. 3). The explanation is the same as above, related to CO concentration.

Similar to the previous case, when CO_2 is not recirculated, the reduction of NO_x and N_2O concentration occurs as the moisture content in lignite decreases and O_2 concentration in inlet gas increases, however in this case the slope of the decrease lines is steeper than the one in the case without CO_2 recirculation (Fig. 3).

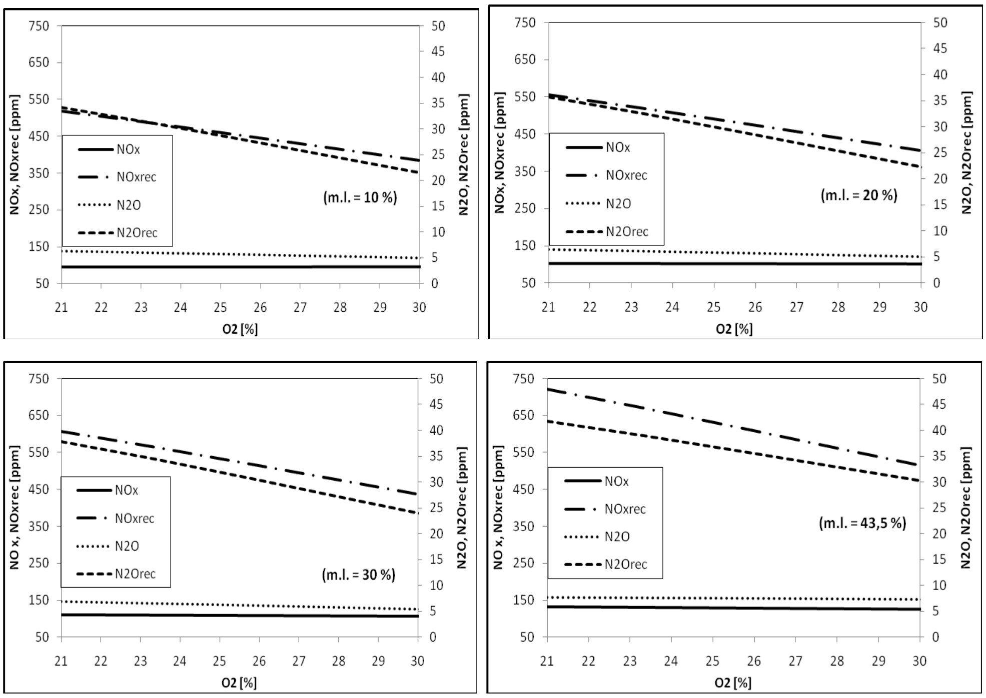

Figure 3: NO_x and N_2O concentration in dry flue gas as a function of the oxygen content in inlet gas for different moisture levels in the lignite.

When the moisture level in lignite declines from 43.5 to 10% the NO_x emission changes from 721 to 519 ppm, with 21% O_2 and from 532 to 394 ppm, with 30% O_2, whilst the N_2O emission changes from 42 to 34 ppm, with 21% O_2, and from 32 to 23 ppm with 30% O_2 in the mixture O_2/CO_2. The CO_2 flux is generally independent of the moisture level of lignite and oxygen concentration in inlet gas (Fig. 4). The CO, NO_x and N_2O flux is the same as in the previous case, without CO_2 recirculation. It is obvious because, only a portion of CO_2 flux is subjected to recycle (Figs. 4 and 5). The wet flue gas flux is independent of the oxygen content in inlet gas, but the flux of wet flue gas declines of about 38% as the moisture content in fuel decreases from 43.5 to 10% (Fig. 6).

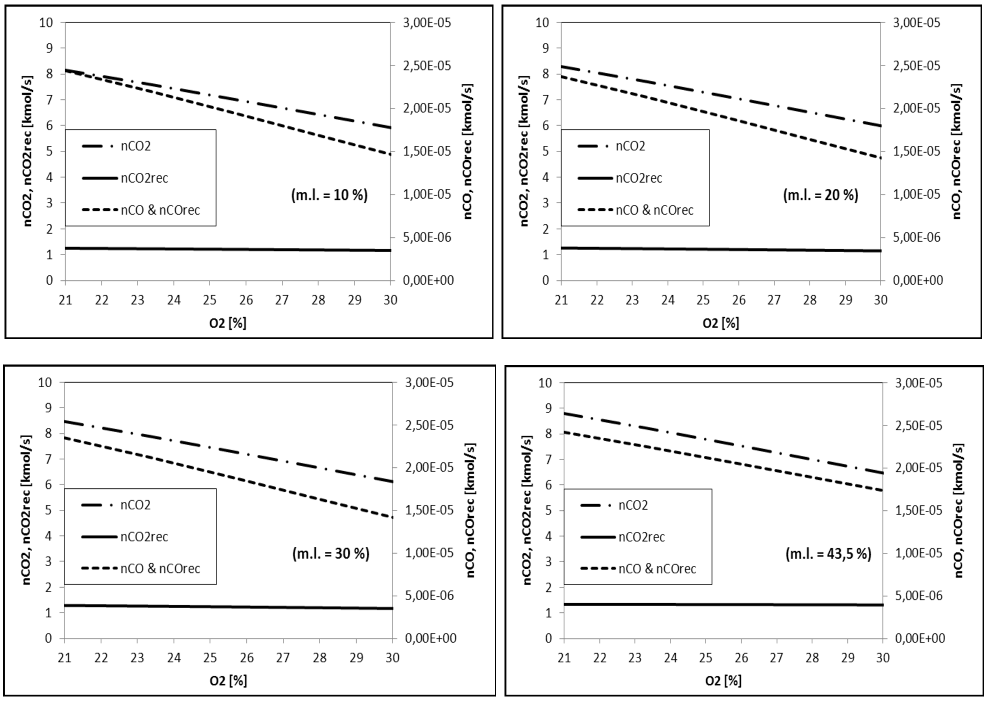

Figure 4: CO and CO_2 flux in flue gas as a function of the oxygen content in inlet gas for different moisture levels in the lignite.

The case, in which the CO_2 is not recycled has been also considered. The obtained results show that, in O_2/CO_2 conditions without CO_2 recirculation, the CO_2 concentration is generally independent of the moisture content in solid fuel (Fig.2). The increase of the oxygen concentration leads to the decrease of CO_2 concentration from 96 with 21% of oxygen concentration to about 92–93% during the oxy-combustion with 30% of oxygen concentration, which results from the excess of O_2 in flue gas. The CO concentration is of about 2-3 ppm and is almost independent of both: the moisture level and the oxygen concentration in inlet gas.

It could be attributed to two countercurrent mechanisms via the $CO/O_2/H_2O/$ char reactions with the respect of H_2O:

$$H_2O + \text{char} \rightarrow H_2 + CO \,, \tag{3}$$

$$CO + \frac{1}{2}O_2 \rightarrow CO_2 \,. \tag{4}$$

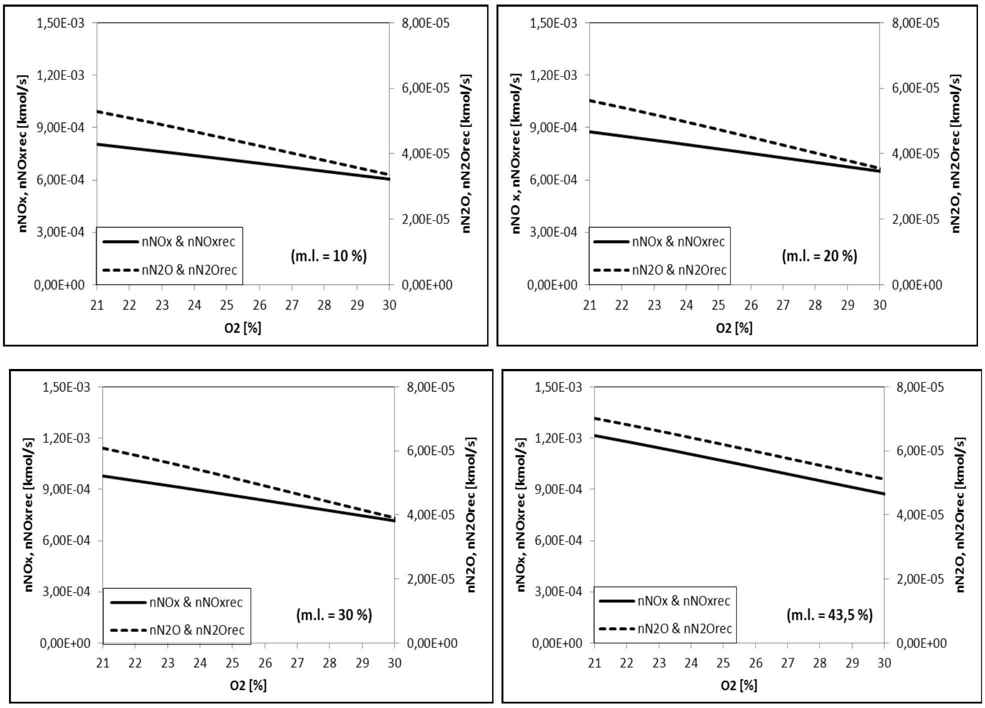

Figure 5: NO_x and N_2O flux in flue gas as a function of the oxygen content in inlet gas for different moisture levels in the lignite.

High concentration of H_2O promotes the CO formation, Eq. (3). This effect is reduced by countercurrent mechanism related to reaction (4). Higher H_2O concentrations accelerate CO reduction via Eq. (4), it was underlined and explained in [13].

A slight reduction of NO_x and N_2O concentrations occurs as the moisture content in lignite decreases (Fig. 3). When the moisture level in lignite declines from 43.5 to 10% the NO_x reduction changes from about 28% with 21% O_2 to about 24% with 30% O_2. For 21% of O_2 in inlet gas the NO_x concentration declines from 132 ppm with 43.5% of moisture content in lignite to about 95 ppm with the moisture level of 10%. For oxygen-enhanced conditions, when O_2 concentration in the mixture O_2/CO_2 equals 30%, the NO_x concentration in flue gas changes from 125 ppm with 43.5% of moisture content in lignite to about 95 ppm with the moisture level of 10%. When the moisture level in lignite declines from 43.5% to 10% the N_2O reduction changes from 25% with 21% O_2 to about 38% with 30% O_2 in inlet gas. For 21% O_2 concentration in the mixture O_2/CO_2 the NO_2 con-

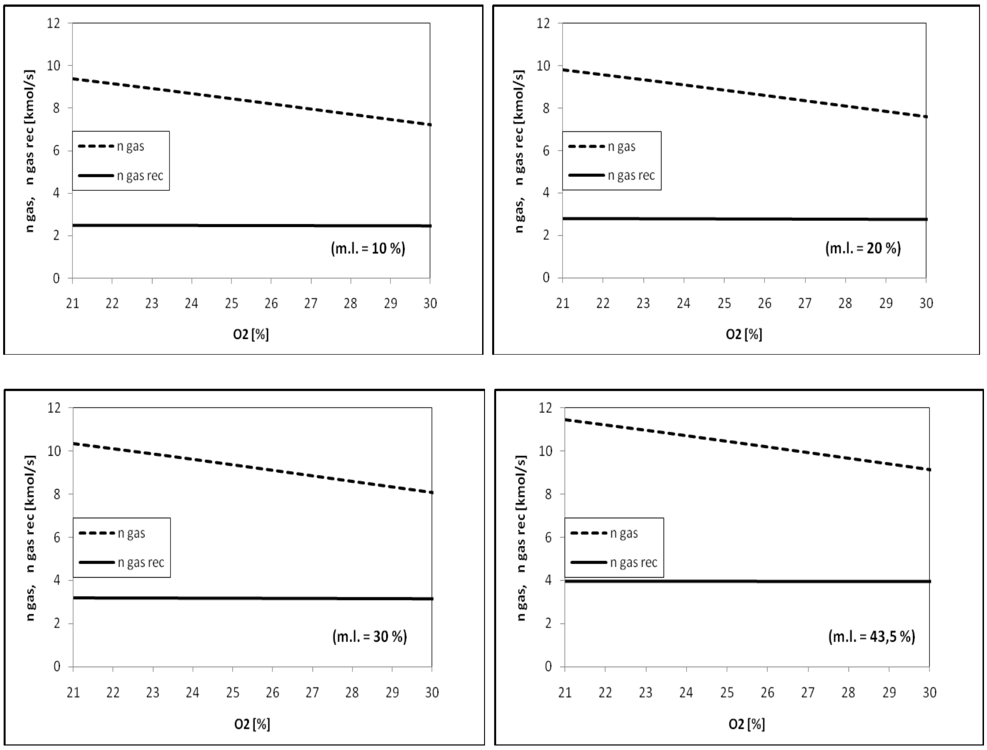

Figure 6: The flux of the wet flue gas as a function of the oxygen content in inlet gas for different moisture levels in the lignite.

centration declines from about 8 ppm with 43.5% of moisture content in lignite to about 6 ppm with the moisture level of 10%. For 30% O_2 concentration in inlet gas mixture the N_2O concentration in flue gas changes from 8 ppm with 43.5% of moisture content in lignite to about 5 ppm with the moisture level of 10%.

The CO_2 and CO fluxes are generally independent of the moisture level of solid fuel in examined range of O_2 concentration. A decline of CO_2 and CO fluxes is noticed as the oxygen concentration in inlet gas increases as the effect of higher oxygen volume fraction of the gas mixture supplied to the combustion chamber. The CO_2 flux declines of about 26% and CO flux of about 33% as oxygen concentration in the mixture O_2/CO_2increases from 21 to 30% (Fig. 4).

Figure 5 shows NO_x and N_2O fluxes versus O_2 content in the inlet gas for different moisture level. The decline of NO_x and N_2O fluxes occurs as a result of decreasing moisture level of fuel. As the oxygen concentration in inlet gas increases a decline of NO_x and N_2O fluxes are also noticed. The declines of NO_x

emission may be attributed to the reduction of NO to molecular nitrogen through the NO/CO/char reactions including the Boudouard's reaction [17]. The decline of N_2O flux could also be explained in a similar way as it was presented in [17], where it is suggested that the decrease in N_2O emission may be attributed to the difference between the rates of its formation and destruction in higher temperatures. When the temperature increases the rate of N_2O reduction exceeds the rate of its formation, and hence, the net result is a decrease in N_2O emissions [17]. As moisture level decreases from 43.5 to 10%, the NO_x reduction changes from 33 with 21% O_2 in inlet gas to about 31% with 30% O_2 in the mixture O_2/CO_2. For N_2O the reduction is of about 24% with 21% O_2 and 32% with 30% O_2 in inlet gas. The increase of O_2 concentration in the mixture O_2/CO_2 leads to the NO_x reduction. The NO_x decline of is of about 24% while for N_2O the reduction changes from 32% with the moisture content of 10% to about 24% with the moisture level of 43.5%.

The moisture content of fuel and the oxygen concentration in the inlet gas largely influence the overall wet flue gas flux (Fig. 6). A flux of wet flue gas declines of about 18–20% as the moisture content in fuel decreases from 43.5 to 10 %. It is due to the lower feed rate, when the moisture content decreases. The increase of oxygen content in the mixture of O_2/CO_2 from 21 to 30% reduces the wet flue gas flux by about 19–21%.

4 Conclusions

The results of the calculations conducted using the model of dried-lignite combustion in oxygen-enriched atmosphere of O_2/CO_2 in 670 t/h CFB boiler has been presented. The influence of the moisture content in lignite and of the oxygen content in inlet gas on emission of CO_2, CO, NO_x, N_2O was studied.

The main conclusions are as follows:

- combustion of dried-lignite in oxygen-enhanced leads to the reduction of gaseous pollutants emission,
- a decline of the CO flux is noticed as the oxygen concentration in inlet gas increases,
- higher O_2 concentration in inlet gas leads to higher NO_x and N_2O reduction,
- a reduction of NO_x and N_2O concentration occurs as the moisture content in lignite decreases,
- the performed model of the solid fuels combustion and gaseous pollutants emission is a useful tool to simulate processes that occur in CFB boilers.

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