MODELING OF CHAR COMBUSTION IN CO_2/O_2 AND N_2/O_2 ATMOSPHERES

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ABSTRACT

The combustion of coal particles suspended in a gas stream is a complex process including devolatilization, gas phase oxidation and solid char combustion. The impact on these processes by oxyfuel atmospheres, which exhibit a very high fraction of carbon dioxide, has been investigated by the Institute of Combustion and Power Plant Technology (IFK) at the University of Stuttgart. The experiments on the combustion of chars of Lusatian lignite in N_2/O_2 and CO_2/O_2 atmospheres are used for comparison with CFD-Simulations by Hitachi Power Europe (HPE). The comparison shows that by extension of the existing char combustion model with the Boudouard reaction the oxygen consumption in the test furnace can be calculated for most cases quite well.

1. INTRODUCTION

Oxyfuel combustion is one of the promising technologies to enable carbon dioxide capture and sequestration (CCS) for new and existing coal-fired power plants to reduce overall global carbon dioxide emissions required for stabilizing atmospheric carbon dioxide concentration and global warming. The combustion of coal in recirculated flue gas enriched with pure oxygen leads to atmospheres with very high contents of carbon dioxide and water and almost no nitrogen. This will lead to different gas properties like higher density, higher specific heat capacity and a much higher radiative gas emission. Experimental investigations and numerical simulations have to be done, to get a better understanding of these changes on the combustion processes. This knowledge may prove to be important for future works, for example for a proper retrofit of existing coal-fired power plants to oxyfuel-combustion.

The effect of the CO_2-rich environment on the combustion processes has been investigated experimentally by the Institute of Combustion and Power Plant Technology (IFK) at the University of Stuttgart [1,2]. The results of the measurements are used for validation of CFD-simulations by Hitachi Power Europe (HPE). Since most of the models used to simulate combustion processes in typical coal burners were developed for atmospheric conditions, examinations are necessary whether these models are valid also for oxyfuel atmospheres or if adjustments have to be made or even new models have to be implemented in the CFD code.

The combustion of coal particles suspended in a gas stream is a complex process including devolatilization, gas phase oxidation and solid char combustion. These processes have been investigated separately in different reactors by IFK, which allows a comparison with the corresponding numerical model in the CFD code. The ob-

Table 1. Proximate and ultimate analyses of coal [1,2].

	Moisture [%, ar]	Ash [%, ar]	Volatiles [%, daf]	Fixed C [%, daf]	C	H	N	S	O	LHV [kJ/kg]
Lausitz Coal	10.20	4.79	55.09	44.56	65.96	5.00	0.77	0.36	26.58	21455
Char from N_2-atm.	0.90	11.40	16.99	83.01	86.55	1.60	0.89	0.65	10.32	28741
Char from CO_2-atm.	1.10	14.20	17.23	82.76	87.25	1.63	1.00	0.61	9.51	30376

Table 2. Experimental conditions for char combustion in O_2/N_2 and O_2/CO_2 atmospheres [1,2].

	O_2 fraction at inlets [vol-%]	Overall stoichiometry [-]	Carrier flow rate [Nm^3/h]	Primary flow rate [Nm^3/h]	Secondary mass flow [Nm^3/h]	Coal mass flow [g/h]	O_2 excess [vol-%]
Char from N_2-atm.	5, 8, 15	2.2, 1.52, 1.22	1.50	2.55	5.95	149, 347, 808	2.74
Char from CO_2-atm.	5, 8, 15	2.2, 1.52, 1.22	1.50	2.55	5.95	152, 354, 824	2.74

jective of this work is to check, whether the available char combustion models are suitable both for air and oxyfuel environments with same model constants.

2. EXPERIMENT

The experiments are made at the Institute of Combustion and Power Plant Technology at the University of Stuttgart. Since the objective of this work is to examine the influence of different atmospheres on char combustion, first of all the volatile fraction of the coal should be minimized to reduce the influence of the devolatilization model on the results. IFK uses an entrained flow reactor to accomplish the coal devolatilization step during coal combustion under different atmospheres. For this work a lignite from the Lausitz area in Germany is prepared in the reactor as well in pure N_2- as in pure CO_2-atmosphere. The coal has been analyzed before and after devolatilization in the reactor, results are listed in Table 1. For convenience purpose the resulting materials will be denoted as chars, although there is still some volatile matter left.

Combustion experiments are performed in a cylindrical once-through 20 kW furnace (Figure 1) [1,2]. Its dimensions are 2500 mm in length and 200 mm in diameter. The ceramic walls are heated electrically so that a constant wall temperature of 1300°C is provided. The gas flow is divided into carrier, primary and secondary stream and injected without swirl through the annular jet burner at the top of the furnace. The outer diameter of the burner is 34 mm. Atmospheric and oxyfuel environments are established by gas streams consisting of oxygen volume fractions of 5, 8 or 15 vol.-% (dry) and the rest being either nitrogen or carbon dioxide, respectively. The char produced in pure CO_2 will be used for combustion experiments in CO_2 environments whereas the char from devolatilization in pure N_2 will be used for combustion experiments in N_2 environments. The conditions for the six conducted combustion experiments are listed in Table 2.

Gas samples are collected via an oil-cooled sampling probe, which is inserted at the center of the furnace exit and can be moved axially up to the

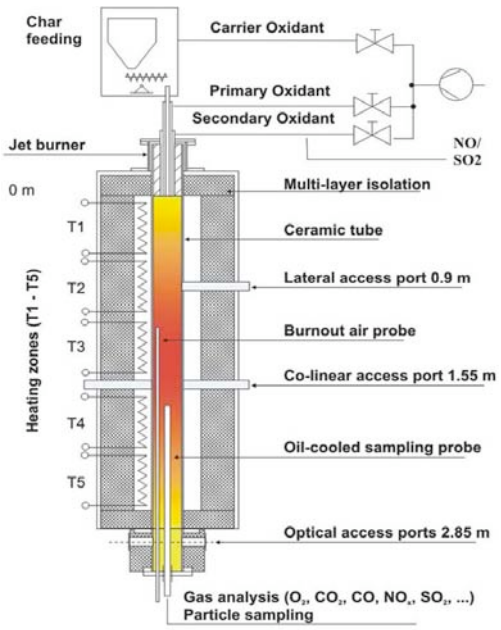

Figure 1. Schematic diagram of the once-through 20 kW furnace [1].

burner exit. The probe is cooled to 180 °C to ensure quenching of the flue gases. A volume flow of 1.5 Nm^3/h is generated by a pump, to transport the combustion gases to the analyzer. The maximum outer diameter of the cone shaped sampling probe is 63 mm.

3. CFD-SIMULATION

The cylindrical geometry of the jet burner and the furnace is discretized with two-dimensional axisymmetric meshes with quadrilateral cells. Since the diameter of the sampling probe is relatively large, it represents a significant flow obstacle in the furnace. To identify its influence on the results, simulations with and without sampling probe are made. Five different meshes were used, one without probe and each of the remaining four with a different position for the probe. The number of cells in the meshes varies between 160000 and 220000.

The steady state CFD-simulations are made with the uncoupled solver by Fluent. The fluid is assumed to be incompressible and involving friction. The species involved are volatiles, carbon monoxide, carbon dioxide, water vapour, oxygen and nitrogen. In addition to the eulerian gas phase a lagrangian particle phase is accounted for with the discrete phase model. Turbulence is calculated with the standard k-ε-model. The heat radiation is calculated with the discrete ordinates methods including gas and particle emissions. Whereas the particle emission is assumed to be constant, the gas emission is calculated using the weighted sum of grey gases model [3]. The homogenous reaction rates of the volatiles with oxygen to carbon monoxide and oxidation of the latter to carbon dioxide are modeled as the minimum of the kinetic reaction rates and the turbulent mixing rates calculated with the eddy dissipation model. Simulations with two different char combustion models are made two check their suitability for oxyfuel combustion. The first one is Fluent´s intrinsic model based on Smith´s model [4], which assumes that the char reaction rate includes the effects of both diffusion of the oxidizing species to the particle through the gas phase and the chemical reaction on the particles internal surface. The model is designed for char reaction with one oxidant and is approved for atmospheric combustion. The simulations made with the intrinsic model regard the following heterogeneous reaction:

$$\text{char} + 0.5\,O_2 \rightarrow CO \qquad (1)$$

In oxyfuel combustion however, because of the high concentration of carbon dioxide the Boudouard reaction becomes important:

$$\text{char} + CO_2 \rightarrow 2\,CO \qquad (2)$$

Regarding more than one reactant for the coal requires switching to the surface combustion model. This model follows the relationships for calculating char particle burning rates presented by Smith [4] and also considers the bulk diffusion of the reactants, particle properties like surface area and mass fraction of the surface species in the particle. Furthermore a kinetic rate of Arrhenius form is included. The Arrhenius constants chosen for this work are taken from Hobbs [5].

4. RESULTS

4.1. Impact of the sampling probe

The CFD-simulations show, that the impact of the sampling probe on the species fractions at the measuring point is not negligible. Figure 2 shows the species volume fractions along the flame axis versus the residence time. While the volume fraction of the oxygen varies only marginal, the simulations with sampling probes result in considerably lower CO volume fractions which are closer to the experimental results. Figure 3 (a) shows the axial velocities of the fluid near the burner and the sampling probe. The probe disturbs the free flow of the gas streams and from the turbulent kinetic energy k in Figure 3 (b) can be seen, that around the probe turbulence is present. Except from the region near the burner inlets, the highest values for k are near the probe. This leads to a higher turbulent mixing rate of the species at the probe and thus to higher homogeneous reaction rates. Because of the reaction stoichiometry of the CO oxidation, the rise of the CO volume fraction due to the higher turbulent reaction rates becomes more measurable than the drop of the O_2 volume fraction. Because of the apparent influence of the sampling probe on the results, it is included in all following simulations. The probes are positioned at the same axial coordinates for all six different environments. Because of the slightly different flow velocities due to different gas densities, the residence time at the probe positions vary between the cases.

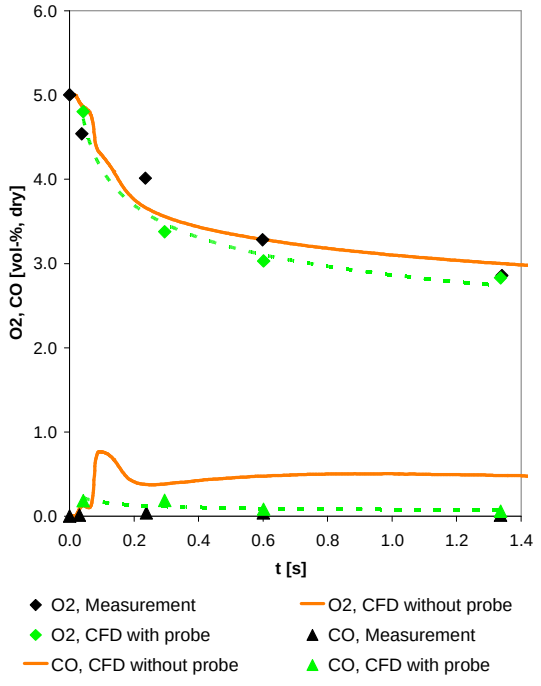

Figure 2. Measured and calculated species volume fractions for the case of 95 % N_2 and 5 % O_2. Comparison of CFD-simulations made with and without sampling probe.

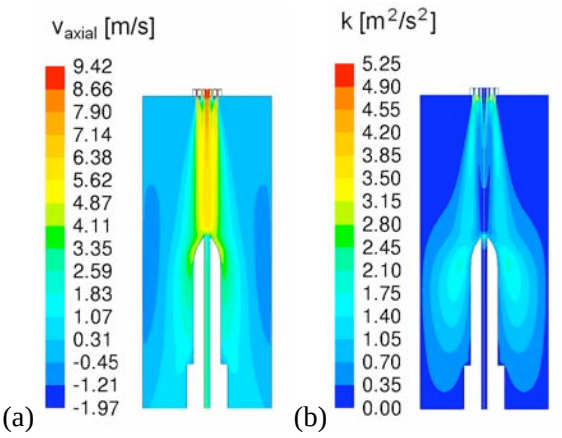

Figure 3. Calculated axial velocities (a) and turbulent kinetic energy (b) for the case of 95 % N_2 and 5 % O_2 and a probe position Δx = 0.22 m away from the burner.

4.2. Impact of the combustion model

The comparison of the numerical results with two different combustion models is made for the environment with the highest carbon dioxide volume fraction of 95 % and 5 % oxygen. Figure 4 shows that inclusion of the Boudouard reaction leads to a slightly higher predicted CO volume fraction. Since the CO volume fraction is very low at the flame axis, both models calculate a large relative but a very small absolute deviation. The bigger difference between both models appears in the calculated oxygen volume fractions. This is a direct consequence of the char reaction rates. The highest reaction rates of approx. $r_{char,ox.}$ = 350 mol m^{-3} s^{-1} are achieved by the char

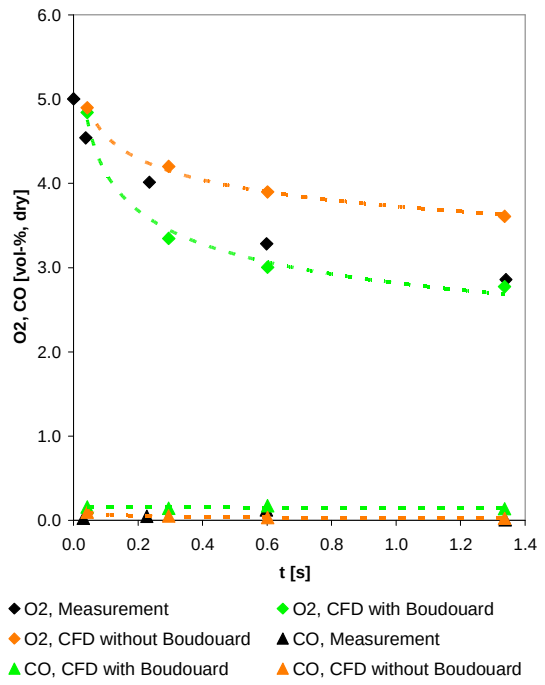

Figure 4. Measured and calculated species volume fractions for the case of 95 % CO_2 and 5 % O_2. Comparison of CFD-simulations made with and without Boudouard reaction.

oxidation when using the surface combustion model. The maximum rate of the Boudouard reaction is with $r_{char,Boud.}$ = 180 mol m^{-3} s^{-1} nearly half as big. Both reactions together result in a maximum burnout ratio of $r_{burnout,Boud.}$ = 250 g m^{-3} s^{-1} that is five times bigger than the maximum burnout ratio of $r_{burnout,intr..}$ = 50 g m^{-3} s^{-1} in the simulation with the intrinsic combustion model without Boudouard reaction. The higher reaction rates predict an oxygen consumption near the burner which is a little bit to fast in this environment. However the oxygen volume fraction is predicted better more downstream when using this char reaction model. On the other hand the simulation only regarding the char oxidation predicts the oxygen consumption only near the burner roughly correct. With increasing distance to the burner the difference between measurement and simulation is getting bigger. The calculated oxygen volume fractions then become too high. All in all the results from the CFD-simulations including the Boudouard reaction are rated as more accurate, so this model is used to simulate char combustion in the six different examined atmospheres.

4.3. Impact of the environment

The measured and calculated volume fractions of oxygen and carbon monoxide for all six examined atmospheres are shown in Figure 5. The measurements reveal a faster oxygen consumption up to a residence time of $t \approx 0.5$ s in environments with high nitrogen fractions than in those with high carbon dioxide fractions. While the difference in oxygen consumption is relatively small for low oxygen fractions, it becomes bigger as the oxygen fraction at the inlet increases. On the other hand, the maximum carbon monoxide fraction is observed in CO_2-rich atmospheres. However the maximum is reached at a later residence time than in N_2-rich atmospheres. With increasing oxygen fraction the measured CO values rise, which suggests that carbon monoxide production by the char oxidation is promoted.

The results of the CFD-simulations show the same tendencies in oxygen consumption as the experiments, but they are less pronounced. Nevertheless the agreement between experiment and simulation at least for the atmospheres with oxygen fractions of 5 and 8 vol-% is quite well. The biggest deviation from the experiment appears in the atmosphere of 85 vol-% nitrogen and 15 vol-% oxygen. It has to be pointed out, that the dotted lines in Figure 5 are only fit-curves through the values from CFD-simulations to clarify the tendencies of oxygen consumption and carbon monoxide formation. Certainly comparisons can only be made at the examined sampling probe positions. More simulations with probe positions in the region with high gradients in oxygen consumption would probably lead to a better agreement between experiment and simulation than indicated by the fit-curves. But the already calculated value for t = 0.29 s indicates, that there still will be some deviation left. In the atmosphere with 85 vol-% carbon dioxide and 15

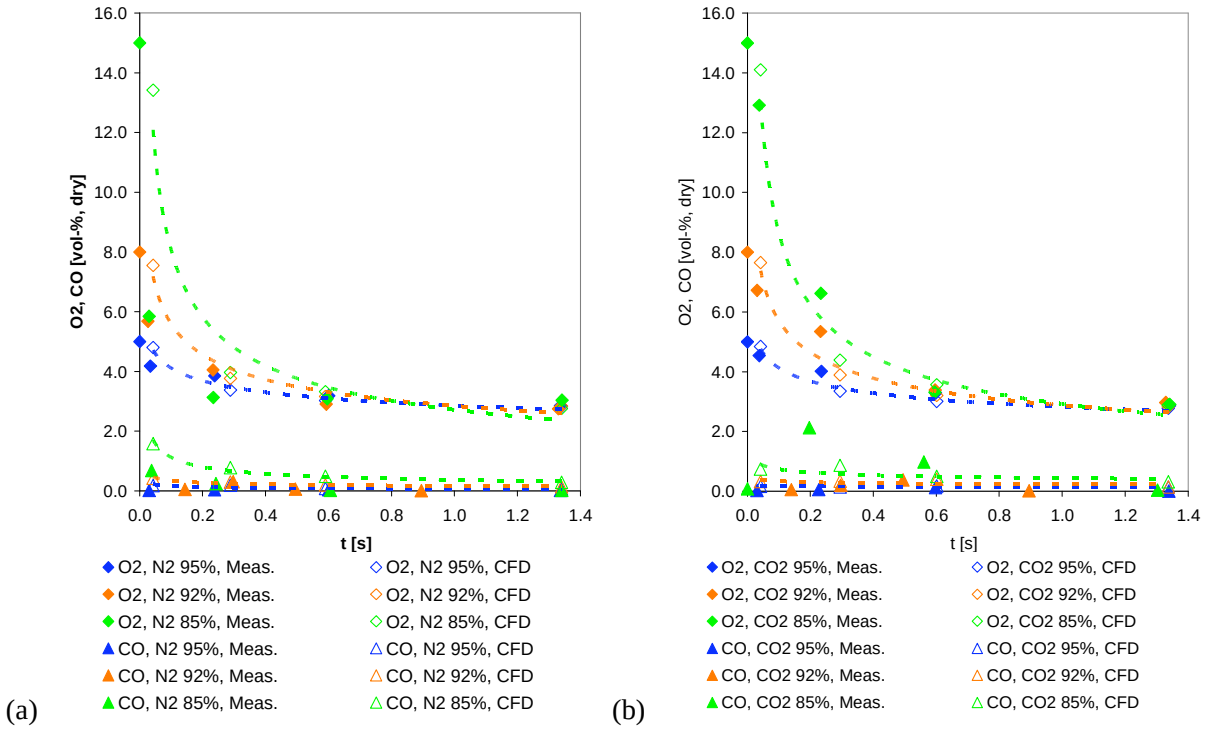

Figure 5. Measured and calculated species volume fractions for six different atmospheres: (a) Values in N_2-environments; (b) Values in CO_2-environments. The dashed lines are fit-curves through the values from the CFD-simulations.

vol-% oxygen the agreement between experiment and simulation is like in the atmospheres with lower oxygen fractions much better.

Since the carbon monoxide volume fractions are mostly below 0.4 vol-% (dry) the rating of the calculated values is somewhat difficult. The deviation is relative high even for the atmospheres with oxygen fractions of 5 and 8 vol-%, respectively. On the other hand one may consider, that the absolute deviation is in fact very small. For both simulations with higher oxygen fractions of 15 vol-% there is a bigger difference between prediction and measurement of carbon monoxide fractions. Especially the considerably higher CO volume fraction in the atmosphere of 85 vol-% carbon dioxide and 15 vol-% oxygen is not yet predicted.

5. CONCLUSIONS AND OUTLOOK

The comparison between experiments and CFD calculations shows, that exact boundary conditions and geometries have to be established for the simulations. In the case of this work the relative large sampling probe has a big impact on the results, so that its integration in the numerical mesh is required.

The CFD-simulations without the Boudouard reaction do not predict the species volume fractions as good as the simulations with this reaction. The inclusion of the Boudouard reaction leads to a quite well agreement with most of the experiments also in oxyfuel atmospheres. The correct modeling of the char combustion processes in oxyfuel environments will be important for future works on this topic.

Bigger differences between experiment and simulation arise in the atmospheres with the highest oxygen volume fractions. Further investigations will show, whether the prediction especially of the carbon monoxide fraction can be improved. One possibility for improvement is the adjustment of the radiation model. In this

work the gas emissivity is calculated with the weighted sum of grey gases model using the coefficients defined by Smith [4] for air-blown combustion with a high concentration of non-radiating nitrogen. HPE´s newly adopted polynomial coefficients for CO_2-rich atmospheres [6] will be used in further simulations to allow for a more accurate modeling of the heat radiation in such environments. This might result in different flame temperatures which will again affect the temperature depending reaction rates.
Another possibility to improve the predictions by the CFD-simulation is the extension of the combustion mechanism by regarding more reactions like the heterogeneous watergas reaction or the hydro gasification and the subsequent homogeneous reactions.

LITERATURE

[1] Al-Makhadmeh, L., Maier, J., Scheffknecht, G., Proceedings of the 34th International Conference on Clean Coal & Fuel Systems, 2009, Clearwater, Florida, USA, 112-123
[2] Al-Makhadmeh, L., Coal Pyrolysis and Char Combustion under Oxy-Fuel Conditions, Thesis, University of Stuttgart, 2009
[3] Smith, T. F., Shen, Z. F., Friedman. J. N., J. Heat Transfer 104 (1982), 602-608
[4] Smith , I.W., 19th Symp. (Int.) Comb. (1982), The Combustion Institute, Pittsburgh, 1045-1065
[5] Hobbs, M.L., Radulovic, P.T., Smoot, L.D., AIChE 38 (1992) 5, 681-702
[6] Rehfeldt, S., Kuhr, C., Ehmann, M., Bergins, C., Tigges, K.-D., Wu, S., Proceedings of the 35th International Conference on Clean Coal & Fuel Systems, 2010, Clearwater, Florida, USA, accepted