ACCURATE MODELLING OF NO_X EMISSION IN A COKE-OVEN-BATTERY WITH VALIDATION AGAINST TEST DATA

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Abstract
In the present work, CFD simulations are used to provide an overview of a heating flue combustion chamber, a typical unit of a coke oven battery, that uses coke oven gas as fuel, with a focus on the production of NO_x, which is, along with carbon monoxide, the main source of pollution that must be minimized at the design stage and controlled during operation. In contrast to the most common approaches available in the literature, the proposed method is based on a detailed chemical reaction mechanism, with 24 species and 134 reactions, derived by reduction from the C1-C3 Polymers, and capable of accurately estimating the concentrations and distributions of radical and intermediate species, involved in NO_x formation by thermal and reaction mechanisms, and "destruction" by reburn phenomenon. The production of oxides of nitrogen pollutants is simulated by thermal and reaction mechanisms, also taking into account the reburn of NO_x. In this work, it is shown that the reburn mechanism is essential to correctly assess emission levels under the operating conditions of the heater combustion chamber. Numerical results were also validated through comparison with flue gas outlet temperature and NO_x emission at the furnace outlet.
The numerical results are shown and shed light on the peculiar characteristics of combustion in the heating stack. Finally, the content of NO_2 pollutants in the dry flues obtained from the numerical simulation is compared with the experimental data, showing almost perfect agreement. As a further result, modeling with and without reburn effect of NO is compared, showing its extreme significance.

Context and motivation
The ongoing energy transition is particularly challenging in sectors [1], such as the steel industry, where a combination of fuel flexibility, use of innovative processes, thermal efficiency, and increasingly restrictive emission limits, with a focus on nitrogen oxides (NO_x), are required. In this context, and particularly in the steel industry, advanced and validated design tools are needed to meet market needs by

optimizing processes and designing new plants, ensuring high efficiency and, at the same time, reduced emissions, under a wider range of operating conditions. In this framework, CFD (Computational Fluid Dynamics) is an extremely useful tool both for the analysis and design of individual equipment and for the optimization of the entire process.
In the specific case of coke oven batteries, the simulation of flow and combustion within the combustion chamber involves several relevant physical phenomena, which the modeling must take into account, mainly turbulence, heat exchange by conduction, convection and radiation, the chemical reaction between fuel and oxidant, and NO_x production. Therefore, validation of such complex models through comparison against experimental data coming from on-site measurements is particularly valuable for researchers and designers, and in the present study it was done by taking advantage of Paul Wurth's integrated work covering the entire process from design to commissioning and testing of the entire plant and the coke oven-battery in particular.
The key topics of this work are, therefore, the analysis of CFD fluid dynamic simulations, carried out on a combustion chamber, a typical unit of a coke oven battery, and the comparison of the results with experimental data from plant testing. The study aims at describing and providing advanced modeling validated with process data for the simulation combustion of a coke oven battery chamber which is able to accurately estimate NO_x emissions, which represent a pollutant extremely noxious both for human health and for the environment, frequently undergoing severe regulations which often require costly flues post-treatments to be met.

Problem description

In general, coke oven battery is an equipment to produce processed coke from the distillation of coal, made by a series of alternating heating flues and coking chambers, placed in rows to maximize the heat transfer rate.
The considered geometrical model consists of one “unit”, including one heating chamber, two heating wall portions and a chamotte refractory portion (Figure 1). Each individual chamber is divided into: an upward heating flue, where air and fuel are supplied and combustion occurs, and a downward heating flue, from which flue gases exit. Combustion air is divided into primary air in the base duct and secondary air at the lower and upper levels. To improve the combustion process by internal FGR (Flue Gas Recirculation) [2] two windows are symmetrically located at the lower level and act as recirculation ports (Figure 1).

Numerical modelling

CFD analyses are conducted with the Ansys Fluent-v17.2 [3]. The computational fluid and solid domains are discretized by a polyhedral mesh of about 3 million elements, obtained by a multi-block approach, generating smaller cells close to complex geometries, such as staging ports and COG inlet to correctly capture flows and combustion details.

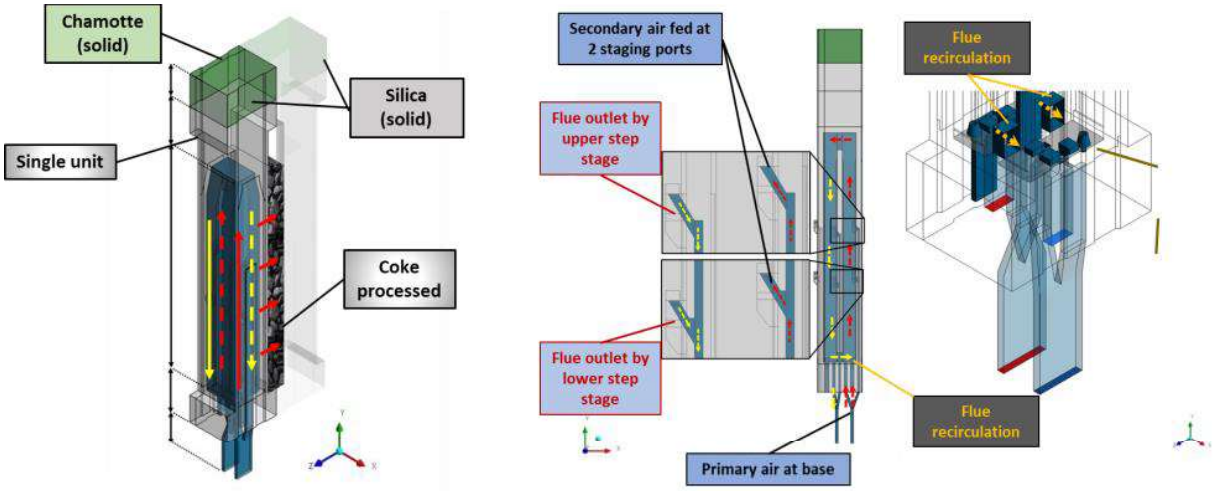

Figure 1. Heating flue chamber: isometric view (left), side view (middle) and isometric view of basement detail (right).

Numerical modeling is based on a stationary RANS (Reynolds Averaged Navier-Stokes equations) approach, in particular, the two-equation k-ε model with standard wall functions at solid boundaries are used for turbulent flow. The equation of state follows an ideal gas approach, with density depending on both temperature and absolute pressure. Combustion in coke ovens is controlled by two phenomena that occur in series: mixing of the reagents and then the chemical oxidation reactions. It is therefore the combination of fluid dynamics (mixing) and chemistry that determines the structure of the flame (extension and maximum temperature) and the level of NO_x emissions. The turbulent-chemistry interaction is modelled using the most advanced eddy dissipation concept (EDC) [4], with in situ adaptive tabulation (ISAT) model [5] to improve computational efficiency.

The need to have a direct and accurate prediction of NO_x and CO, which exploits the species and intermediates calculated in the combustion simulation, and the use of gaseous fuels derived from the steel process, such as coke oven gas (COG), has required the use of a more detailed kinetic scheme. The detailed mechanism chosen is the Polimi-23, which is a reduction of the complete C1-C3 scheme of the Politecnico di Milano for hydrocarbon combustion, with 23 species and 134 reactions. The addition of a 24th species, the intermediate CH, is necessary to "activate" the NO_x reburn mechanism, which is crucial for emission prediction, as shown in the results part.

Pollutant emissions are calculated by a post-processing method, which takes into account three mechanisms: the first is the Zeldovich mechanism, which involves the formation of thermal NO_x, and a second contribution from prompt effect. Fuel NO_x can be neglected, given the low content of nitrogen compounds in the fuel. Finally, the reburn mechanism is included, which is of paramount importance for accurate emission estimation in this case. Local turbulent fluctuations, which greatly affect NO_x production rates, are considered with a tabulated pdf approach based on temperature and species fluctuations.

To account for radiation within the gas and between the gas and the walls a Discrete Ordinates method (DO) is used, with absorption coefficients calculation based on WSGGM (Weighted Sum of Grey Gases Model) algorithm, in which a mean optical path is computed on the whole fluid domain. Finally heat conduction in solid is treated solving the Laplace equation with specific thermal conductivity for different materials.

Results

The experimental data from the plant are shown in Table 1 in the third line and are used to set up the CFD simulation and to carry out the comparison of the numerical simulation results with the experimental texts and validation of the model. In the composition used for the simulation because butane C_4H_{10} is not available among the species in the kinetic scheme used, it is replaced by the same amount (by mass) of ethane C_2H_6, as in the fourth line. The fuel flow rate for the individual chamber is estimated from the total measured fuel flow rate, while the air mass flow rate is calculated from the design value of the outlet oxygen content. The heat transfer coefficient between the wall and the coke is adjusted by matching the CFD brick temperature value to the experimental value. Finally, the concentration of nitrogen dioxide measured in the outlet flues is used as a verification and validation parameter for the model (
Table 2).

Table 1. Fuel composition, computed, and for CFD setup.

	CH_4	CO_2	CO	H_2O	H_2	C_2H_6	C_4H_{10}	N_2
Units	%	%	%	%	%	%	%	%
Exp. data	34.43	6.39	15.52	6.86	10.06	--	4.82	21.92
CFD set-up	34.43	6.39	15.52	6.86	10.06	4.82	--	21.92

Table 2. Process data by test/measurements, computed, and from CFD results

	Exp. data	CFD set-up
COG Inlet temperature	550 [°C]	550 [°C]
COG Flow rate	All chambers = 17'545 [Nm^3/h]	Single chamber = 19.56 [Nm^3/h]
AIR Inlet temperature	1200 [°C]	1200 [°C]
Outlet O_2	4.77÷5.18% [%vol. Dry basis]	Design: 4.95 [%vol. Dry basis]
NO_2	395 [mg/Nm^3] @ 5% O_2 dry out	394 [mg/Nm^3] @ 5% O_2 dry out
Outlet temperature	1274* [°C]	1274* [°C] (*tuned*)

Simulation results show a temperature field characterized by a high-temperature flame zone located at COG / air jets interface, with a peak above 1800°C, where the

highest concentrations of NO occur. A relevant concentration of hydrocarbon radical/intermediates CH_x is produced in the rich flame zone, which are involved in the reburn NO depletion mechanism (Figure 3).

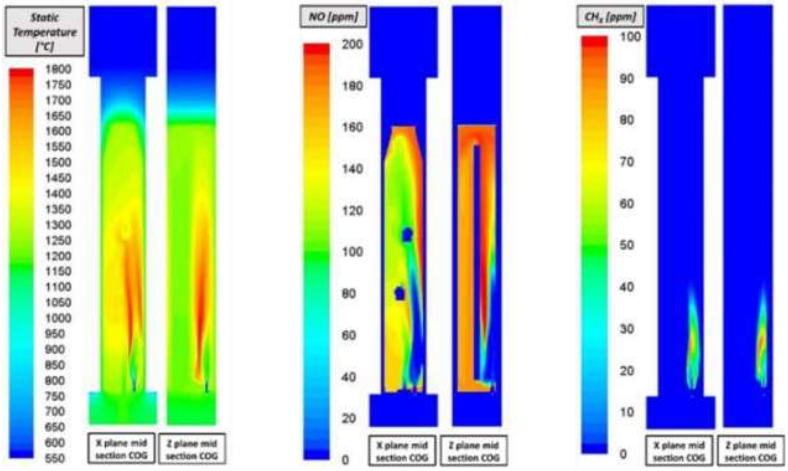

Figure 2. CFD results: contours of main fields on two planes cutting COG inlet.

As shown in Figure 3, the rates of thermal and prompt NO formation, and the rate of depletion due to reburning have the same order of magnitude, clearly explaining the relevant effect of reburn and why it cannot be neglected when simulating coke-oven-batteries. The thermal formation term is high in the fuel-air mixing zone where high gas temperature is combined with oxygen availability. On the other hand, the reburn NO depletion rate is high (in absolute value) where two conditions are met: NO and CHx intermediates are available, and temperature is above 1600 [K].

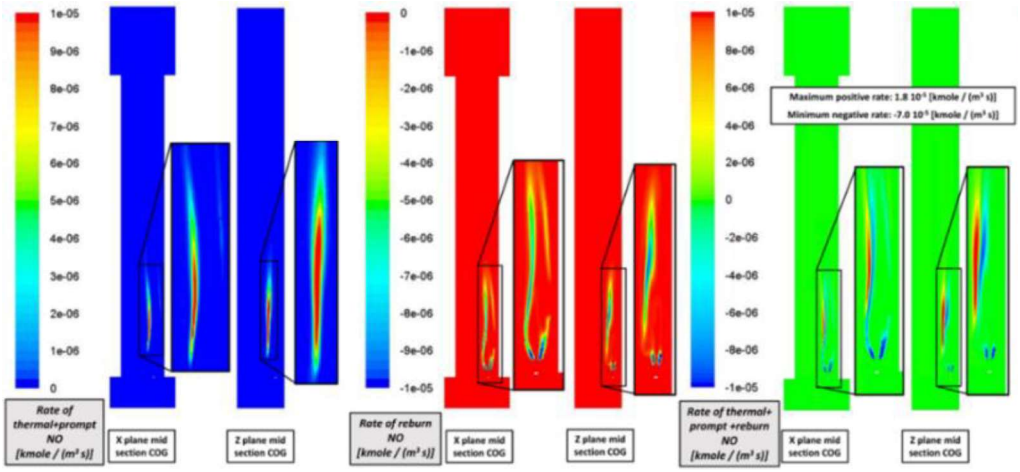

Figure 3. Rates of NO production/deplation: thermal+prompt (left), reburn (middle) mechanisms and thermal+prompt+reburn (right).

The validation of the CFD overall modelling and simulation approach is obtained comparing CFD NO_x emission estimation (394 mg/Nm^3) against the experimental data (395 mg/Nm^3), which shows almost perfect agreement (Table 1).Without the

NO reburn mechanism, a completely wrong value is obtained: 979 mg/Nm^3 versus 395 mg/Nm^3 of the tests, as shown in Figure 4.

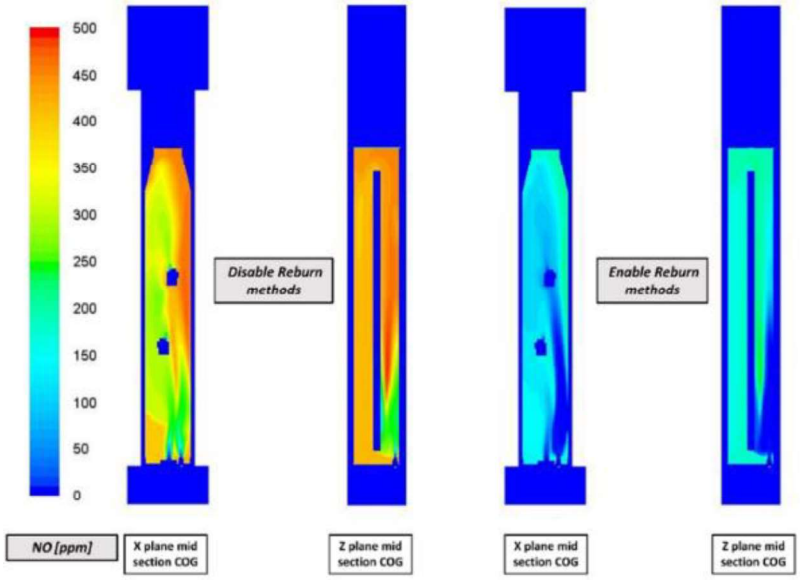

Figure 4. NO content with (right) and without (left) reburn mechanism**.**

Conclusion

In this paper, a comprehensive procedure for CFD modelling of coke oven battery combustion, with emphasis on NO_x emission calculation, is proposed to be an effective tool for design and optimization. The model is based on a detailed combustion mechanism and NO_x post-processing calculation, including thermal mechanism, thrust mechanism and rebound mechanism, based on the species fields (CH_x) calculated in the combustion simulation. The modelling approach was validated by comparing CFD results with experimental data from plant tests, demonstrating excellent agreement and the fundamental importance of the reburn reduction mechanism. The model needs no further tuning to be used for optimization of the heating combustion chamber design.

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