Evaluation of Global Reaction Mechanisms for CFD Modelling of Oxy-Fuel Combustion

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Keywords: Oxy-fuel; Combustion; CFD; Global reactions; Propane

1. Introduction

In modelling of large-scale combustion systems and complex flames using CFD it is common to apply a simplified reaction mechanism (global mechanism/scheme) to represent the combustion chemistry and thereby reduce the computational effort compared to a complete reaction mechanism. Although these mechanisms only include a small number of reactants and reactions and greatly simplify the chemistry their prediction of major in-flame species can be of essential use in industrial scale CFD modelling work. Global schemes are, however, limited to specific conditions in order to be applicable. Typically, this means air-fired conditions for which most schemes have been developed. According to our knowledge three global fuel oxidation schemes have been derived for oxy-fuel conditions: Leiser et al. (2007) (denoted: 3-step), Andersen et al. (2009) (4-step), and Frassoldati et al. (2009) (6-step). All these are based on the well-known global scheme of Jones and Lindstedt (1988) (JL) derived for air-fired conditions. The present work evaluates the description of the combustion chemistry by these four mechanisms against a detailed reaction mechanism validated for oxy-fuel conditions. Furthermore, the predictive capability of the mechanisms is tested by inclusion in a basic CFD simulation and comparing the simulation results with measurements in the Chalmers 100 kW test facility.

2. Methodology

The combustion chemistry of the four global mechanisms is evaluated against a detailed reaction mechanism based on the work by Mendiara and Glarborg (2009) and Frassoldati et al. (2003) in a plug-flow reactor (PFR) model. To evaluate the mechanisms under realistic flame-like conditions the temperature profile and the oxygen concentration profile (controlled by the mixing between fuel and oxidizer) was predefined in the plug-flow reactor. The profiles were achieved by tracking a single mass-less particle and its trajectory starting at the fuel inlet and finishing at the outlet of the furnace. The trajectories are obtained from 3D CFD simulations of experiments in the Chalmers 100 kW unit, previously reported by Andersson (2007). More details on the methodology used are given in Hjärtstam et al. (2012). The CFD calculations are performed with ANSYS FLUENT, applying the k-ε model to account for turbulence and the eddy dissipation concept to describe the interaction between turbulence and...
chemistry. The chemistry of the global reaction mechanism is evaluated by their individually generated concentration profiles of O₂, C₃H₈, H₂ and CO and important reaction rates. The CO profile is a good indicator of combustion, as it is an important intermediate in all mechanisms investigated.

3. Results

Figure 1 gives profiles of measured and predicted (by CFD) CO concentration along the centerline of the test unit (Fig. 1a). In Fig. 1b the plug-flow calculations for the investigated reaction mechanisms are presented. The global mechanisms have in many cases a different behavior in the plug flow environment compared to the CFD model where the differences may be reduced by the descriptions of the heat transfer process and the turbulence-chemistry interaction. Yet, the CFD calculations performed in the present work clearly points out that the choice of global mechanism is critical to the results. Both the JL mechanism and the 6-step mechanism have sharp peaks in the concentration of CO along the centerline in the CFD simulations, whereas the 3-step and the 4-step mechanisms predict a clearly different behavior of the formation and oxidation of CO. The 3-step mechanism predicts considerably lower peak concentrations than the other models. Compared with the JL scheme, the oxy-fuel mechanisms predict a longer CO rich zone along the centerline.

In the plug-flow calculations it is seen that the break-down of the fuel is quite similar in the oxy-fuel derived mechanisms, but in comparison to the detailed mechanism all of them predict slower fuel decomposition. The JL mechanism, on the other hand, predicts a break-down of the fuel that is initially faster than the detailed mechanism, even though the time for complete fuel oxidation is similar for both mechanisms. The faster fuel break-down of the JL mechanism is seen in the predicted concentration of CO (Fig. 1b), where the peak is reached sooner with the JL-mechanism than with the detailed mechanism. The principle shape of the CO profile of the detailed scheme is captured by the JL mechanism and the 6-step mechanism, even though the latter strongly underestimates the peak concentration of CO. The JL and the 3-step mechanisms yield peak concentrations of CO in line with the detailed mechanism, and also predict a burnout of CO in agreement with the detailed mechanism. All mechanisms predict a complete burnout of CO after ~0.4 seconds. After 0.4 seconds O₂ is still being fed to the reactor, which results in an increased O₂ concentration.

To understand how the mechanisms describe the progress of combustion, the rates of the CO forming reactions of the global mechanisms are compared with those of the detailed scheme during flame propagation. Reaction rates for fuel-CO and CO₂-CO reactions are presented in Fig. 2. In the detailed mechanism, CO is mainly formed from oxidation of fuel-radicals during fuel oxidation. Furthermore, decomposition of CO₂ through the reaction CO₂+H=CO+OH also contributes to the formation of CO. The global mechanisms do not include fuel-radicals and CO is formed directly from the fuel through irreversible reactions with oxygen and/or water. CO may also in this case form by decomposition of CO₂ through reversible reactions. In all cases, the fuel oxidation by water is surprisingly slow compared to the other fuel-CO reactions.

![Graph showing CO concentration along the centerline and over time](image1.jpg)

Figure 1. a) Measured and modeled concentration of CO along the centerline in the Chalmers 100 kW OF 27 flame. Results of the CFD calculations using the global reaction mechanisms. b) Calculated concentration of CO as a function of time. Results from the 1D PFR calculations of the Chalmers OF 27 flame, using an O₂ injection and temperature profile based on the CFD analysis.
In general, none of the global mechanisms have the same timing in the CO peak as the detailed mechanism. The global mechanisms yield a fuel-related formation of CO that starts earlier and lasts longer than in the detailed scheme. The early start is expected since there are several fuel-related reactions in the detailed scheme that must be completed before CO is formed, whereas CO is formed directly from the fuel in the global mechanisms. The JL mechanism predicts a fast and strong CO formation, while the 3-step and 4-step mechanisms predict lower reaction rates which spans over a longer time. The 6-step mechanism has a delayed CO formation compared to the other global mechanism, a result more in line with the detailed scheme. The reaction rate peak is, however, substantially lower and with a much longer time span than the detailed mechanism.

Most interesting is the relation between the fuel oxidation reaction and the CO-CO$_2$ reaction. A flame develops in such a way that the hydrocarbons are oxidized before CO, leading to a CO peak which is oxidized when all hydrocarbons have been oxidized. This behavior is described by the detailed scheme where the CO-CO$_2$ reaction switches direction once the oxidation of hydrocarbons is completed. During the oxidation of hydrocarbons, CO has actually been formed from CO$_2$, which is a phenomenon that is not so pronounced during air fired conditions when the in-flame concentration of CO$_2$ is much lower. The only global mechanism that captures this behavior is the JL mechanism, although it clearly overestimates the reduction compared to the detailed mechanism. The 3-, 4-, and 6-step mechanism predicts a completely different behavior with CO being oxidized to a large extent in the presence of C$_3$H$_8$. This is not expected as the C$_3$H$_8$ should primarily consume the available O$_2$. Andersen et al. (2009) partly developed their 4-step mechanism in order to lower the CO concentration compared to the JL scheme, an effect that clearly can be seen in Fig. 1b, but compared to the detailed mechanism there are large differences.

4. Conclusions

The present work evaluates global reaction mechanisms which have been developed for implementation in CFD simulations of oxy-fuel combustion. The JL mechanism is the only one of the investigated schemes that captures a reaction history of CO that is in line with the detailed mechanism, with an initial formation of CO from the fuel followed by an oxidation of CO to CO$_2$. The global oxy-fuel mechanisms all predict simultaneous oxidation of fuel and CO, a behavior in contradiction with the detailed scheme. Hence, simply adjusting the reaction parameters to compensate for quantitative errors (such as over-predicted CO concentration or combustion temperature) may lead to errors in the reaction dynamics. The CO$_2$/CO reactions of the global oxy-fuel mechanisms evaluated in the present investigation have a qualitatively incorrect behavior, which in turn reduces the generic nature of the mechanism. The present paper shows that there is a significant improvement potential for global oxy-fuel reaction mechanisms and, thus, a need to develop a refined global reaction mechanism.

![Figure 2](image_url)

Figure 2. Results from the 1D PFR calculations of the Chalmers OF 27 flame, using an O$_2$ injection and temperature profile based on the CFD analysis. Reaction rates of fuel-CO reactions (black solid line) and CO$_2$-CO reactions (red dotted line) as function of time for the mechanisms. Positive values for the CO$_2$-CO reaction means that CO is formed, negative values that CO is oxidized.