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Numerical study of natural gas reforming by non-catalytic partial oxidation based on the Virtuhcon Benchmark

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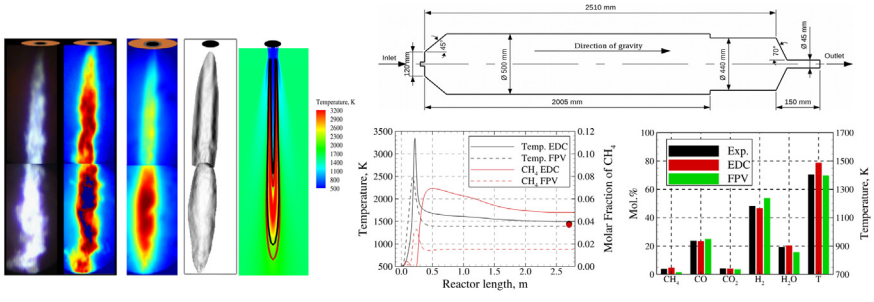
HIGHLIGHTS

- Numerical study of non-catalytic natural gas reforming under HT/HP conditions.
- Validation of the numerical results against semi-industrial test plant experiments.
- Comparison of EDC and FPV approaches for turbulence-chemistry interaction modeling.
- The models applicability was discussed in terms of flame and reactor characteristics.

GRAPHICAL ABSTRACT



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ABSTRACT

The non-catalytic reforming of natural gas to syngas was studied numerically. The numerical simulations focused on the Virtuhcon Benchmark, which is a set of experimental data based on the semi-industrial scale test plant HP POX (high-pressure partial oxidation). The experimental data comprises reactor characteristics such as product gas composition and wall temperatures across the reactor for temperatures between 1473 and 1673 K and pressures between 50 and 70 bar(g), and optically estimated flame characteristics such as flame length and width. For turbulence-chemistry interactions, the widely used Eddy Dissipation Concept model and an advanced Flamelet/Progress-variable-based approach developed for POX processes were applied. Contrary to standard Flamelet approaches, the advanced model can describe correctly both the reaction zone and the comparatively slow chemical processes in the almost homogeneous post-flame zone. Based on the experimental data, the applicability of the different numerical models will be discussed carefully. In contrast to several literature work, the model evaluation is based not only on global reactor characteristics, but also on optical flame analyses from inside of the semi-industrial test plant, which allows to evaluate the capability of the numerical model to predict local reactive flow effects inside industrial HP/HT processes. The results reveal that both approaches allow a reliable prediction of the syngas composition, flame length, and flame width. With respect to the outlet temperature, the Eddy Dissipation Concept tends to overpredict the resulting temperature, or, from a different point of view, to underpredict the progress of the endothermic reforming conversion processes.

1. Introduction

Extensive resources and high production profitability make natural gas one of the most promising sources of hydrocarbons. Due to

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the low costs for natural gas as a chemical feedstock, gas chemistry is becoming one of the fastest-growing sectors of modern fuel and energy complexes [1]. High-pressure partial oxidation (HP POX) is one promising strategy for syngas production without the need for catalysts. Due to the rapidly changing political and economical conditions, the classical, time- and cost-intensive development of new or improved technology from lab to pilot and finally to demo scale is no longer feasible. For that reason, numerical modeling has become more and more attractive to investigate the physical processes in HP/HT systems, to adjust existing systems to different length scales and feedstock, and to develop new technologies. The process of natural gas partial oxidation plays an important role in Gas-to-Liquid (GTL) technology, which is widely used in the chemical industry and has been developed since the 1970s. One of the largest scales of POX facilities is applied in Shell GTL technology [2]. POX is also the key stage in the Shell Middle Distillate Synthesis Process (SMDS) described in [3].

The numerical modeling can be subdivided into different approaches. Kinetic models can easily provide basic reactor characteristics with little computational effort, see e.g. [4–11]. CFD models, on the other hand, allow a more detailed analysis of the fluid flow, species and temperature distribution, and the impact of processes such as radiation or turbulent mixing on the overall processes. One of the most challenging parts of this approach is coupling the turbulent flow with the chemical reactions. Compared to combustion processes, different reaction regimes occur in POX reactors with large variations in length and time scales [12,13]. On the one hand, the fast oxidation reactions, often using pure O_2 as an oxidizer, occur near the burner and are mainly governed by the diffusion of non-premixed reactants. On the other hand, in the region downstream of the flame the slow reforming reactions take place in a premixed regime in nearly homogeneous mixtures. This zone is characterized by larger time and length scales than the oxidation zone.

The quality of the numerical results depends strongly on the validity and accuracy of the underlying models. Especially for the CFD modeling of reactive flows, the turbulence-chemistry interaction is a dominant mechanism and has to be modeled carefully. In the literature, the Eddy Dissipation Concept (EDC) has been widely used for modeling partial oxidation reactors with detailed kinetic mechanisms [14–17]. EDC is a robust and widely used model, but computationally very demanding, especially in combination with the detailed mechanisms required to describe the slow reforming reactions. Rehm et al. [14] examined the influence of the EDC model parameters on the accuracy of gasification modeling, showing that EDC cannot accurately describe endothermic reforming reactions without the model parameters being adjusted. These results were confirmed by the DNS performed by Caudal et al. [13], which showed the tendency of the EDC model to underestimate the reaction rates in the reforming zones if standard parameters are applied.

To reduce the computational effort required to simulate the turbulence-chemistry interaction, reduction techniques are usually applied [18]. The chemistry is separated from the solution of the turbulent flow and modeled using simplified reduced approaches, such as equilibrium, homogeneous reactors, 1D diffusion and premixed flames. The results of the reduced chemistry models are usually calculated in advance and stored in look-up tables. The tables are commonly parametrized using scalar variables, representing the main chemistry parameters, such as the mixture fraction and the progress variable, etc. These look-up tables are coupled to the CFD solutions through the same controlling variables, which are solved together with the turbulent flow. Finally, the influence of the unresolved fluctuations is accounted for by integrating with a probability density function (PDF) of the main controlling variables; this approach is widely used in combustion simulations [19]. The PDFs are generally modeled using presumed functions parametrized by the mean/filtered values and their variances. The presumed PDF approach, coupled with chemical equilibrium was used by [16,17]. Although this approach is computationally efficient due to the tabulated chemistry, it is not able to describe the slow reforming reactions in partial oxidation reactors. Similarly, the steady laminar diffusion flamelet (SLDF) approach used by Vegendla et al. [20] also relaxes to equilibrium too fast because of the low strain rates in the reforming zone.

The tabulation of reactive structures under POX conditions is quite challenging. Especially the large (compared to combustion) fuel-to-air ratios must be adequately taken into account when generating the tables. As shown in Vascellari et al. [21], both the reaction and the post-flame zone can be modeled accurately using a newly developed Flamelet/Progress variable approach (FPV) that is based on premixed 1D flames and coupled with ANSYS® Fluent®. Using non-premixed reactive structures (which is the standard approach in the FPV context) would allow only to describe the flame zone. The comparatively slow chemical processes in the almost homogeneous post-flame zone would not be correctly accounted for.

The complexity of all the models applied in a CFD calculation necessitates an intensive validation against experiment data, which should reflect conditions in the real industrial process. Industrial-scale HP-POX plants are operated at high pressures and temperatures (more than 1700 K and 60 bar(g)), which limits the access to the reaction chamber for measurements. For that reason, in situ process data, e.g. optical measurements, are generally not available for HP-POX plants. In the literature, experimental results are mostly limited to lab-scale equipment, e.g. see [22–26]. In the work by [27] a tabulated chemistry model is applied that focuses on the soot formation in ultra-rich natural gas combustion processes. The current work compares numerical results to experimental values and offers suggestions for soot formation modeling. However, in the present study, the soot formation is not taken into account due to its low concentration, and, therefore, the model assumes that soot formation is negligible and has a correspondingly negligible effect on the overall process. In addition, Xu et al. [17] provided the outlet composition for an industrial-scale HP POX plant, and the corresponding reactor geometry and input streams. Due to the limited access to local process data, most works in the literature have focused on validating the model against global reactor performance data, e.g. the syngas composition at the outlet.

Recently, Richter et al. [28] published experimental data gained from the semi-industrial scale test plant HP POX, operated at TU Bergakademie Freiberg. The set of experiments, known as the Virtuhcon Benchmark, comprises detailed information about the burner and reactor geometry, temperature measurements along the reactor wall, and species analysis at the outlet of the reactor. Additionally, optical in situ measurements of the flame were carried out, providing details about the flame shape, e.g. the flame length and width. These data provide a comprehensive set of validation data for the CFD modeling of partial oxidation processes, comprising local flow phenomena and global reactor performance data on an industrially relevant scale.

In this work, the non-catalytic reforming of natural gas is studied numerically, with a special focus on the Virtuhcon Benchmark. This means that, reference values for the model setup and its validation are taken directly from [28]. Special attention was paid to the comparison of two different modeling approaches for the turbulence-chemistry interaction, namely the Eddy Dissipation Concept and a Flamelet/Progress Variable approach. The results from the two approaches are carefully analyzed and compared with experimental data in terms of computational time, flame characteristics, outlet conditions, temperature and species