Chemical Engineering Science 134 (2015) 694–707

Contents lists available at ScienceDirect

Chemical Engineering Science

journal homepage: www.elsevier.com/locate/ces

# Flamelet/progress variable modeling of partial oxidation systems: From laboratory flames to pilot-scale reactors

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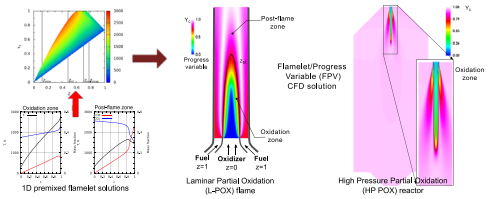
## HIGHLIGHTS

- Flamelet Progress Variable approach is used for modeling partial oxidation.
- Chemical solution is pre-computed with premixed and diffusion 1D flamelets.
- Premixed flamelets accurately describe oxidation and reforming zones.
- FPV results are compared with a DNS of a laminar POX.
- FPV accurately estimates the outlet composition for a high pressure POX.

## GRAPHICAL ABSTRACT




## ARTICLE INFO

*Article history:*
Received 31 March 2015
Received in revised form
20 May 2015
Accepted 23 May 2015
Available online 3 June 2015

*Keywords:*
Partial oxidation
Natural gas
CFD modeling
Flamelet progress variable

## ABSTRACT

Partial oxidation (POX) reactors are characterized by different reaction regimes: the near burner zone is mainly governed by the mixing of the initially non-premixed reactants and by the fast oxidation zone, while the post-flame zone is mainly governed by the slow reforming reactions under almost fully premixed conditions. Advanced turbulence–chemistry interaction models are required to describe different regimes in POX reactors. This work investigates the use of the flamelet/progress variable (FPV) approach in laminar and turbulent POX reactors. The FPV approach allows different times and length scales to be captured, using a progress variable to describe the advancement of the reactions in reacting mixtures. The solution of the chemical reactions is generally separated from the solution of the flow and mixing by pre-calculating solutions of one-dimensional canonical flames. The solution of the chemistry is stored in look-up tables and linked to the solution of the flow using the mixture fraction and the progress variable scalars. In this work, different methods for generating the flamelet look-up tables are analyzed for a laboratory reference laminar POX flame. The results are compared to the direct numerical simulation with the goal of identifying the most suitable flame structure for tabulation in POX systems. These results are then used to build a FPV table. Finally, a pilot-scale high-pressure POX reactor was investigated and the numerical results are validated using the measured composition of the syngas at the exit of the reactor.



*Abbreviation:* $Y$, mass fraction; $Y_c$, progress variable; $Z$, mixture fraction; DLR, Deutsches Zentrum für Luft- und Raumfahrt; EDC, Eddy dissipation concept; FGM, flamelet generated manifold; FLUT, flamelet look-up table; FPI, flamelet prolongated ILDM; FPV, flamelet/progress variable; SLDF, steady laminar diffusion flamelet; HP POX, high pressure partial oxidation; ILDM, intrinsic low dimensional manifold; L POX, laminar partial oxidation; PDF, probability density function; POX, partial oxidation; ULF, universal laminar flame

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http://dx.doi.org/10.1016/j.ces.2015.05.042
0009-2509/

## 1. Introduction

Partial oxidation (POX) is a process in which a solid, liquid or gaseous hydrocarbon feedstock is converted into a synthesis gas (syngas), primarily composed of hydrogen and carbon monoxide in a global fuel-rich mixture. The syngas obtained from the POX processes generally represents an intermediate step from hydrocarbons to bulk chemicals (methanol, DME, ammonia, acetic acid, oxo-alcohols, isocyanates) and fuel synthesis, e.g. Fischer–Tropsch (Christensen and Primdahl, 1994).

Numerical models have largely been used to predict the performance of partial oxidation reactors, especially for large-scale reactors, which are usually not equipped with instrumentation for analyzing the process and validating the models. Partial oxidation processes have been modeled by many authors using kinetic models (Zhu et al., 2001; Yu et al., 2006; Olivieri and Vegliò, 2008; Zamaniyan et al., 2008; Zahedi-Nezhad et al., 2009; Brüggemann et al., 2010; Zhou et al., 2010) and CFD (Rehm et al., 2009; Guo et al., 2012; Vegendla et al., 2011; Xu et al., 2014), respectively. Recently, Richter et al. (2014) presented the results of an experimental campaign in a pilot-scale high-pressure partial oxidation reactor with the goal of providing a test-case for validating numerical solutions and modeling approaches.

With the increasing availability of high-performance computing resources for scientific modeling on both laboratory and pilot-plant scales, numerical simulations have become a well-established tool for understanding such complex reactive flows, with the goal of developing models for process optimization. CFD simulations are now an important part of the design process for advanced reactors, since they help to gain detailed local and global knowledge of the process.

The coupling of the turbulent flow with the chemical reactions is one of the most challenging problems when modeling partial oxidation processes (Caudal et al., 2015). In fact, different reaction regimes with strongly varying length and time scales occur in POX reactors (Wu et al., 2010; Caudal et al., 2013; Prüfert et al., 2014), potentially requiring different modeling strategies. On 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 the 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 and this zone is characterized by larger time- and length-scales than the oxidation zone (Vegendla et al., 2011; Hunger et al., 2013).

The Eddy Dissipation Concept (EDC) (Gran and Magnussen, 1996) has been often used for modeling POX reactors (Rehm et al., 2009; Guo et al., 2012; Xu et al., 2014) with detailed mechanisms. EDC is computationally very demanding, especially in combination with the large kinetic mechanisms required to describe the slow reforming process. In addition, Rehm et al. (2009) demonstrated that EDC cannot accurately describe the endothermic reforming reaction, and the agreement between measurement and simulation results can only be improved if the model parameters are adjusted. These findings have been confirmed by Caudal et al. (2015), who performed a turbulent DNS of the reforming zone, showing that EDC generally underestimates the reaction rates in the reforming zone with respect to the averaged results of a DNS.

To reduce the computational effort for simulating the turbulence–chemistry interaction, reduction techniques are usually applied. 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 pre-calculated and stored in look-up tables (LUTs). The tables are commonly parametrized using scalar variables, representing the main chemistry parameters, such as the mixture fraction and the progress variable. Look-up tables are coupled to the CFD solutions through the same input variables, which are solved together with the turbulent flow. Finally, the influence of the unresolved turbulence fluctuations is accounted for by integrating with a probability density function (PDF) of the main input variables; this approach is extensively used in combustion simulations (Veynante and Vervisch, 2002). The PDFs are generally modeled using presumed functions parametrized by the mean/filtered values and their variances. The presumed PDF approach, coupled with precomputed chemical equilibrium, was previously used to simulate POX reactors (Guo et al., 2012; Xu et al., 2014). However, this approach is not able to describe the slow reforming reactions in partial oxidation reactors, since the slow reforming reactions cannot be modeled by assuming chemical equilibrium. Similarly, the steady laminar diffusion flamelet (SLDF) approach (Peters, 1986) used by Vegendla et al. (2011) also relaxes to equilibrium too fast due to the low strain rates in the reforming zone.

On the other hand, the flamelet/progress variable (FPV) (Pierce and Moin, 2004) model was applied successfully to capture slower processes such as $NO_x$ formation and flame-radiation coupling (Ihme et al., 2005; Ihme and Pitsch, 2008). Another popular approach is the flamelet generated manifolds (FGM) (Oijen and Goey, 2000), which is very similar to FPV, using initially premixed flames to build the look-up tables (Oijen and Goey, 2000). More recently, diffusion flames were also used within the FGM context for table generation (Vreman et al., 2008).

This work investigates the use of the FPV approach for modeling partial oxidation processes, with a special emphasis on the modeling of the flame structure. At first, a laboratory laminar partial oxidation flame (L POX) (Stelzner et al., 2012, 2013) is simulated using the FPV approach. Two flamelet look-up table (FLUT) generation strategies are investigated based on the solutions of 1D laminar diffusion and premixed flamelets, respectively. The results of the FPV simulations are analyzed and compared to the laminar DNS solution of the L POX flame in order to determine the physically most suitable method for generating the chemical database. Then, a 5 MW pilot-scale high-pressure partial oxidation reactor (HP POX), already studied by Rehm et al. (2009) and Vegendla et al. (2011), is modeled using the FPV approach.

The remainder of the paper is structured as follows: Section 2 presents the FPV approach and the methods used for computing the flamelet solution and storing them in look-up tables. In Section 3, the L POX flame is investigated, presenting at first the comparison between the chemical databases generated using premixed and diffusion one-dimensional laminar flames, and then the results obtained from the FPV approach are compared to the direct numerical simulation of the laminar reacting flow. In Section 4, the HP POX reactor is investigated, presenting the analysis of the premixed look-up table and the results of the CFD simulations. Finally, Section 5 summarizes the findings of the present work.

## 2. CFD modeling of partial oxidation flames

The laminar and turbulent reacting flows in POX reactors are simulated using the 2D axisymmetric version of the coupled pressure–velocity solver of the commercial CFD code Ansys-Fluent (ANSYS, 2014). Convective fluxes in all transport equations are discretized with a second-order upwind scheme and the pressure gradients are discretized with a second-order accurate scheme. Radiation is modeled with the P-1 model (Cheng, 1964). The radiating properties of the gas were modeled assuming a gray-band model, based on the Weighted Sum of Gray Gases (WSGG)