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# On the effect of preferential diffusion in autoignition of $CH_4/H_2$ flames. Part 1: Development of a flamelet-based model

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ABSTRACT

This study reports on the development of a flamelet-based reduction method for autoignition of hydrogen enriched methane-based fuels. The main focus is on the inclusion of preferential diffusion effects in the Flamelet Generated Manifolds (FGM) technique for autoigniting flames. Such a development of the FGM methodology is inevitable since investigations with detailed chemistry indicate that preferential diffusion strongly affects autoignition of these mixtures. First, a novel flamelet configuration based on Igniting Mixing Layer (IML) flamelets is proposed to accommodate preferential diffusion in a flamelet database. At the next stage, transport equations for controlling variables are derived with additional terms to account for preferential diffusion effects. The extended FGM model has been evaluated by comparing its predictions with those of detailed chemistry in both laminar and turbulent situations. In laminar situations, it is revealed that the model is able to predict accurately autoignition time scales of one-dimensional hydrogen enriched flames. The turbulent situations are studied by performing Direct Numerical Simulations (DNS) of a two-dimensional unsteady mixing layer. In this configuration, the proposed model yields a precise prediction of autoignition time scales as well. The model has also been assessed using the widely used Igniting Counter-Flow (ICF) flamelets instead of IML flamelets which leads to less accurate predictions especially at high hydrogen contents. The predictive power of the proposed model combined with simplicity of its implementation introduces an attractive reduced model for the computation of turbulent flames.



## 1. Introduction

Autoignition of a fuel mixture by a hot oxidizer plays an important role in several new combustion concepts and technologies, e.g. HiTAC, Flox and MILD combustion. These concepts, to which we will refer as MILD combustion, have been introduced as promising technologies due to an increased thermal efficiency and decreased pollutant formation [1,2]. In spite of the enormous potential of the MILD combustion regime, it has been mainly limited to lab-scale burners due to stabilization issues in practical burners. The stabilization mechanism of MILD combustion is often governed by autoignition of a fuel jet in a hot and diluted environment. This mechanism is highly sensitive to variations in the fuel and oxidizer composition and operating conditions. In the experiments by Dally et al., [3] for instance, hydrogen was added to the fuel in order to improve the stabilization. Numerical modeling of MILD combustion of hydrogen rich fuel mixtures poses a significant technical and research challenges due to the complexity of autoignition under large preferential diffusion effects.

MILD combustion can be realized in many configurations depending on preheating and dilution of fuel and/or oxidizer streams. A few examples of this can be found in previous works of de Joannon et al. such as in Hot-Fuel-Diluted-Fuel [4], Hot-Oxidant-Diluted-Fuel [5] and Hot Oxidant Diluted Oxidant [6]. A number of experimental studies has been performed to study the Jet-in-Hot Coflow (JHC) burner as a model system for MILD combustion [3,7–9]. In most of these experiments, a turbulent lifted flame is observed in a hot environment of oxidizer diluted with burned gas. Recently, hydrogen enriched natural gas flames in JHC burners have been investigated systematically in the experiments of Arteaga et al. [10] on the Delft Jet-in-Hot Coflow (DJHC) burner. They observed that the addition of $H_2$ has a large influence on the lift-off height and stabilization mechanism of these lifted flames. The addition of only 5% $H_2$ resulted in a more than 50% reduction of the lift-off height. This effect cannot be explained by the change in autoignition delay of homogeneous mixtures due to $H_2$ addition.

Recent direct numerical simulations of autoigniting mixing layers of $CH_4/H_2$ mixtures with detailed chemistry and transport models by van Oijen [11] have shed some light to this issue. It was shown that the presence of hydrogen enhances the role of molecular diffusion due to the fact that $H_2$ diffuses out of the fuel mixture into the hot

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**Table 1**
Temperature and molar composition of the fuel stream for the different cases. The oxidizer stream has the same composition for all cases: $T = 1437$ K, $X_{O_2} = 0.0485$, $X_{H_2O} = 0.1452$, $X_{CO_2} = 0.0727$, $X_{N_2} = 0.7336$. $\zeta_{st}$ is the stoichiometric mixture fraction.

| Case | $T$(K) | $X_{H_2}$ | $X_{CH_4}$ | $X_{C_2H_6}$ | $X_{N_2}$ | $\zeta_{st}$ |
|---|---|---|---|---|---|---|
| $D00H_2$ | 448 | 0.00 | 0.813 | 0.037 | 0.15 | 0.0178 |
| $D05H_2$ | 448 | 0.05 | 0.763 | 0.037 | 0.15 | 0.0179 |
| $D10H_2$ | 448 | 0.10 | 0.713 | 0.037 | 0.15 | 0.0180 |
| $D25H_2$ | 448 | 0.25 | 0.563 | 0.037 | 0.15 | 0.0183 |

oxidizer due to its significant preferential diffusion. This leads to a much faster ignition process governed by hydrogen chemistry. However, there is still little knowledge available about the complex role of preferential diffusion in large scale reacting flows.

Numerical modeling of $H_2$ enriched MILD combustion in the large scale flows, as in the DJHC burner, requires reduced models for turbulence and chemistry. These models should be able to predict adequately complex autoignition events under large preferential diffusion effects. Successful reduction techniques to accommodate preferential diffusion are mainly based on flamelets [12], such as FGM (Flamelet Generated Manifolds) [13], FPV (Flamelet Progress Variable) [14] and REDIM (Reaction–Diffusion Manifolds) [15]. Inclusion of preferential diffusion in FGM has been studied by van Oijen et al. [13] and later on by de Swart et al. [16] in the context of premixed flames. It has been shown that two controlling variables are needed to account for local variations in equivalence ratio and mass burning rate. Preferential diffusion in the context of non-premixed flames has been taken into account by Pitsch et al. [17,18]. In their work, a set of flamelet equations was derived with extra terms to account for non-unity Lewis number effects. This model has been used in some studies of turbulent non-premixed flames, for example in [19]. Flamelet-based models were also used to study autoignition in turbulent jet flames (e.g., [20], but the effect of preferential diffusion on ignition was not investigated. To the authors' knowledge, there is no literature about the incorporation of preferential diffusion effects in a flamelet-based technique for autoigniting non-premixed flames.

In part 1 of this two-part paper, the focus is on the extension of the FGM technique to account for preferential diffusion effects in autoignition of $CH_4/H_2$ mixtures. In Section 2, a new type of flamelet called Igniting Mixing Layer (IML) flamelet is introduced with the relevant governing equations. The IML flamelets are analyzed and compared with the commonly used Igniting Counter-Flow diffusion flamelets (ICF flamelets) in terms of preferential diffusion effects in Section 3. In Section 4, the tabulation of the IML flamelets is discussed. In Section 5, an appropriate set of transport equations for the controlling variables is derived to account for non-unity Lewis number effects. The performance of the proposed FGM model is evaluated and validated by comparison with predictions of detailed chemistry first in laminar one-dimensional configuration in Section 6 and afterwards, in turbulent two-dimenstional situations in Section 7. Finally, conclusions are drawn. Part 2 as a separate paper consists of application of the proposed FGM model based on IML flamelets in LES of the Delft JHC burner. The main purpose of part 2 is to evaluate influence of preferential diffusion effects on the lift-off height and stabilization mechanism of lifted flames.

## 2. IML flamelets

In this paper, we aim to develop a flamelet model that can predict the effect of preferential diffusion on autoignition of methane-hydrogen mixtures at the conditions in JHC experiments. In total, four cases are studied containing 0 to 25 percent of $H_2$ by volume, which are summarized in Table 1. These cases correspond to the mean boundary conditions of the DJHC burner experiments [10]. In these

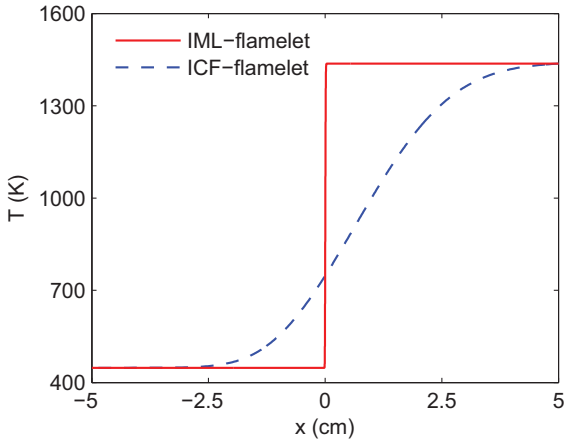


**Fig. 1.** Comparison of the initial temperature profile ($t = 0$ s) between an ICF-flamelet and an IML-flamelet.

experiments, the fuel and oxidizer streams are initially separated. Once the fuel is injected into the hot oxidizer stream, mixing starts, which is then followed by autoignition. Igniting Mixing Layer flamelets (IML flamelets) are introduced here to model this process of mixing and autoignition such as in the DJHC burner. IML flamelets are basically similar to the commonly used one-dimensional Igniting Counter-Flow diffusion flamelets (ICF flamelets) with a notable distinction in the initial condition and inflow momentum. In ICF flamelets, it is a common practice [19–22] to generate an initial condition by assuming a steady-state molecular mixing field between the fuel and oxidizer stream with frozen chemistry ($\dot{\omega} = 0$) as it is shown in Fig. 1. This situation implies that a steady-state mixing field is reached before any chemical reaction takes place. This assumption is mainly valid if the time scale of mixing is much shorter than the chemical time scales. However, such an assumption might lead to unrealistic predictions if molecular diffusion terms are comparable in size to the chemical source terms (for example in $H_2$-enriched methane mixtures). In this case, molecular diffusion has a large influence on autoignition time scales. This situation is extensively discussed in Section 3.

In IML flamelets, in contrast to ICF flamelets, fuel and oxidizer streams are initially unmixed as it is shown in Fig. 1. Such an unmixed profile is adopted here to include preferential diffusion effects in the pre-ignition stage. In this situation, the initial thermo-chemical properties have a step-function profile in physical space. Their values are equal to the fuel boundary on one side of the domain ($x < 0$ in Fig. 1) and equal to the oxidizer boundary at the other side ($x > 0$ in Fig. 1). Due to the steep gradient of mixture fraction at the interface, the scalar dissipation rate $\chi = 2D(\partial Z/\partial x)^2$ is very large at this point. During the molecular mixing process, the scalar dissipation rate decreases and chemical reactions may start at any time during the mixing process. In IML flamelets, the gradient of mixture fraction is not enforced by an inflow momentum (i.e. an applied strain). However, it is governed purely by molecular diffusion. In the absence of an applied strain, the species mass fractions and temperature approach chemical equilibrium for infinite time.

The configuration of IML flamelets resembles practical non-premixed systems in which mixing of the fuel and the oxidizer initiates after their injection from the nozzle exit at very large scalar dissipation rates. In these systems, chemical equilibrium can be reached at a sufficiently large distance from the burner where scalar dissipation rates approach zero.

The mathematical formulation of IML flamelets is described by the following set of one-dimensional transport equations:

$$\frac{\partial \rho}{\partial t} + \frac{\partial \rho u}{\partial x} = 0 \tag{1}$$