

# Experimental and numerical analysis of the laminar burning velocity of oxygen-enriched ethylene mixtures

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## Abstract

Ethylene is largely adopted in the chemical industry. It is a key intermediate in combustion modelling, as well. Most of the processes involving this species take place in an oxidative environment. Nonetheless, the reactivity of mixtures containing ethylene, oxygen, and nitrogen are poorly investigated, especially at non-standard conditions. This work is intended to fill the abovementioned gaps utilizing either experimental or numerical analyses. In particular, the heat flux burner was adopted to characterize mixtures of ethylene/oxygen/nitrogen at low temperatures. The collected measurements were compared against numerical estimations resulting from detailed kinetic mechanisms, for the sake of the quantification of the estimation quality. Then, the flammability limits were estimated through the limiting laminar burning velocity theory. Estimated compositions were tested experimentally and compared with flammability limits from the current literature.

## Introduction

Light alkenes are important substances in material production as well as key intermediate compounds in the combustion of commercial fuels [1]. Among them, ethylene holds the largest share in global market production [2], being adopted as an intermediate compound for several bulk chemicals [3]. Indeed, the utilization of ethylene for the production of ethylene oxide via direct oxidation has continued to grow in the last years [4], posing additional interests in the development of kinetic models - including the low-temperature chemistry - suitable for the process optimization. Besides, the combustion properties of ethylene have been intensively investigated because of their relevance in larger fuel combustion mechanisms [5], including the formation of soot precursors [6]. The majority of the investigations unravelling the chemistry of ethylene flames have been carried out on ethylene/air [7], [8], mixtures containing additional fuels (primarily methane) [9], [10] or diluents (primarily nitrogen) [11], [12]. On the other hand, despite its relevance, experimental data on oxygen-enriched and pure oxygen flames are less abundant, especially at low temperatures. Indeed, in a recent literature review on the subject [13], measurements of ethylene/oxygen/argon in shock tubes at a temperature above 800 K have been reported, exclusively. Hence, the collection of additional data on ethylene/oxygen flames at

low temperatures may have several beneficial effects and help to get a fundamental understanding of hydrocarbons chemistry. Taking into account the conditions of interest, the evaluation of kinetic models suitable for oxidative conditions can be performed by comparing numerical predictions against experimental results of the laminar burning velocity ( $S_u$ ) [14]–[16]. Moreover, the evaluation of this parameter is also of interest in the view of the characterization of explosions scenarios [17] and flammability limits [18].

A closed vessel, counter-flow and heat flux burner can be utilized to collect experimental data [19]. The first utilizes stainless steel, vessel, commonly spherical, where the gaseous mixtures are fed and ignited, temperature and pressure are monitored to evaluate the  $S_u$  [20]. The counter-flow method consists of two opposed jets where premixed fuel and oxidant are fed [21]. Quite obviously, the main drawbacks of this technique regard the production of stretched flames, which may be relevant for highly reactive substances [22]. Besides, the heat flux burner (HFB) consists of a flat plate where the flame is stabilized by a cooling/heating system [23]. It should be noted that steady-state conditions and unstretched flames are achieved in the last case, only, promoting the utilization of the HFB.

Experimental data representing in a single physic-chemical parameter the overall reactivity, such as the  $S_u$ , are essential for the evaluation of the estimation accuracy of detailed kinetic mechanisms [24]. For these reasons, this work presents experimental data collected by using the heat flux burner and compared with detailed kinetic mechanism estimations of  $S_u$  and flammability limits.

## 2 Methodology

This study presents either experimental or numerical analyses on the reactivity of ethylene in an oxidative environment at low temperature, thus the methodological section was divided into two subsections accordingly.

### 2.1. Experimental procedure

The laminar burning velocity data for ethylene/oxygen/air mixtures at the initial temperature of 298 K and pressure of 1 atm were collected by using the heat flux burner. Previous studies have already reported in detail the experimental system, the procedures for the measurement of the  $S_u$  and the estimation of the uncertainties adopted in this work [25][26]. Hence, only a summary is given in the following. The  $S_u$  data were collected by varying the feeding velocity for a given composition, measuring the radial, parabolic temperature distribution on the burner plate, and interpolating by the resulting second-order coefficient. under the heat flux method developed by de Goey et al. (1993) [27]. The compliance with the one-dimensional assumption was monitored by measuring the difference between the inlet and outlet temperature of the cooling fluid. The initial compositions investigated in this work were expressed in terms of oxygen index (E, Equation 1) and equivalence ratio ( $\phi$ , Equation 2). The former is included within the range 0.21 – 1, whereas the latter laid within the range 0.5 – 2.0, at first, then it was conveniently varied to reach the conditions where unstable flames were observed to evaluate the flammability limits.

$$E = \frac{x_{ox}}{x_{ox} + x_{dil}}$$

$$\phi = \frac{x_{fu}/x_{ox}}{(x_{fu}/x_{ox})_{st}}$$

where  $x$  stands for the mole fraction in the initial mixtures, the subscripts *ox*, *dil*, *fu* and *st* represent the oxygen, diluent (i.e., nitrogen), fuel (i.e., ethylene), and stoichiometric conditions, respectively.

More specifically, the flammability limits were individuated by feeding decreasing (for the case of lower flammability limits, LFLs) or increasing (for the case of upper flammability limits, UFLs) amount of

fuel and keeping constant E. The absolute variation in fuel composition ( $\Delta x$ ) was kept constant and equal to 0.1 %v/v. An ignition source was provided after a given time to guarantee a stable flow rate and composition. In case of absence of ignition, the procedure was repeated three times to reduce the effect of uncontrolled unknowns and guarantee experimental reproducibility.

## 2.2. Numerical procedure

The  $S_u$  was estimated under the same initial compositions utilized during the experiments by using the detailed kinetic mechanism developed at the University of Bologna (KiBo) [37], because of its proven accuracy in the estimation of ethylene chemistry at low temperature [12][38]. Mono-dimensional, adiabatic, and steady-state flow was assumed to disregard chemical to physical phenomena, following the common practices for the evaluation of kinetic of reactive systems [39]. The grid refinement parameters were selected as optimal values in terms of computational costs and result robustness, as resulting from a grid convergency analysis reported elsewhere [40]. The flammability limits were determined by implementing the limiting laminar burning velocity ( $S_{u,lim}$ ) theory and an iterative procedure described elsewhere in detail [28]. Briefly, different compositions were tested numerically until the following condition was reached:

$$S_u \in S_{u,lim} \pm 0.01 \cdot S_{u,lim}$$

## 3 Results and discussion

The laminar burning velocity (Figure 1) and flammability limits (Figure 2) were experimentally and numerically determined by using the described procedure for ethylene/oxygen/nitrogen mixtures at initial conditions of 298 K and 1 atm were reported. Additional data retrieved from the current literature were added for the sake of comparison. More specifically, experimental data obtained through closed vessels [29][30], digital particle velocimetry [31], counterflow flame [32][33], heat flux burner [10], and predictions resulting from a thermodynamic-based approach [34] were included in this stage.

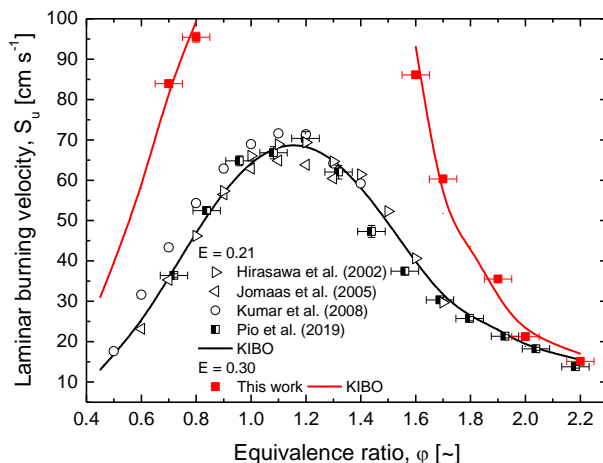


Figure 1. Laminar burning velocity experimentally and numerically obtained in this work at atmospheric conditions. Data from the literature have been added for the sake of comparison.

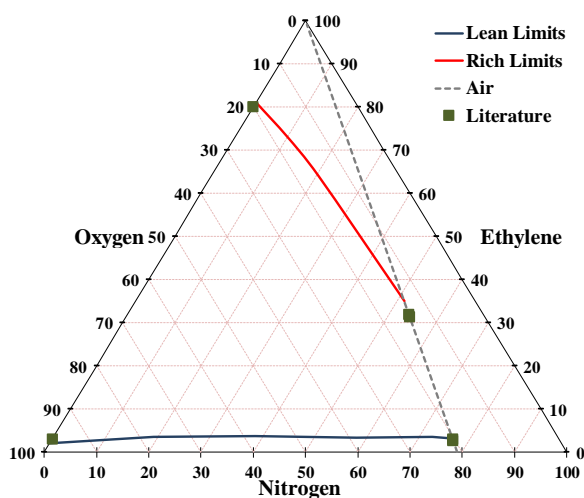


Figure 2. Ternary plot of ethylene/oxygen/nitrogen mixture as obtained by using the detailed kinetic mechanism KiBo at 298 K and 1 atm. Symbols represent experimental [29][30] and numerical [34] data from the literature.

Results demonstrate that the numerical approach can satisfactorily reproduce the trend observed for the experimental data in the case of oxygen-enriched air, as well. Besides, the typical bell-shaped curve concerning the equivalence ratio shows a maximum within 1.1-1.2, regardless of the oxygen content in the oxidant. The faster reactivity at near-stoichiometric conditions results in an increased slope of this curve at lean composition, implying a dramatic variation in the overall reactivity at the near lean limit. Hence, once the flammability limits are compared, oxygen content has an almost negligible impact on the lower flammability limit. Conversely, the lower flammability limit is strongly affected by the composition of the adopted oxidant, due to the asymptotic behaviour reported in Figure 1.

## 4 Conclusions

The present work reports an experimental and numerical investigation on the reactivity of ethylene in presence of oxygen-enriched air in terms of laminar burning velocity and flammability limits. The very good agreement between measurements collected in this work and those available in the literature guarantee the validity of the proposed procedure for the calculation of the flammability limits by using the heat flux burner. Indeed, the first stage of the presented comparison among experimental and numerical data can be intended as per mechanism validation, being the HFB a standardized apparatus for the measurement of  $S_u$  at the investigated conditions. Conversely, the second step of this analysis combines well-established theory with innovative experimental procedures.

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