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Maria Clara de Jesus Vieira, Luis Fernando Figueira da Silva. Flame flashback critical Damköhler number for CO<sub>2</sub> diluted CH<sub>4</sub> and C<sub>3</sub>H<sub>8</sub> mixtures with air. 2021. hal-03457136v1

**HAL Id: hal-03457136**

**<https://hal.science/hal-03457136v1>**

Preprint submitted on 30 Nov 2021 (v1), last revised 28 Jun 2022 (v4)

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# Flame flashback critical Damköhler number for CO<sub>2</sub> diluted CH<sub>4</sub> and C<sub>3</sub>H<sub>8</sub> mixtures with air

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## Abstract

The premixed laminar flame flashback phenomenon in tubes has been known and studied for several decades. However, the effect of the CO<sub>2</sub> dilution has not been addressed, which is relevant for assessing the safety of oil-producing facilities. Furthermore, even if numerical studies have underscored the important role played by the mixture Lewis number ( $Le$ ), these lack an experimental validation. For this reason, specific studies of premixed flame flashback in laminar flows have been undertaken. The objective is to assess the flame flashback critical conditions in mixtures of hydrocarbons – methane or propane diluted by various amounts of CO<sub>2</sub> – with air. This is effected by determining the critical Damköhler number, which is computed using both experimental and numerical data. The former leads to the flashback critical velocity gradient, whereas the latter to a characteristic chemical time scale. The effect different flame thickness definitions on the Damköhler number ( $Da$ ) is examined, evidencing than an order of magnitude discrepancy may arise depending on the definition choice. For methane/air mixtures the critical  $Da$  increases slightly with the equivalence ratio, whereas a decrease of nearly two orders of magnitude has been obtained for propane/air mixtures. The original results show that CO<sub>2</sub> dilution increases  $Da$  only when the percentage of dilution on fuel is larger than 25 % and 50 %, for

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methane and propane, respectively, situations which correspond to flame extinction after flashback. The propane/air/CO<sub>2</sub> mixture results exhibit a  $Da = Le^{5.6}$  dependency which closely follows the trend computed previously, whereas the methane/air/CO<sub>2</sub> results evidence the thermal boundary condition at the tube wall influence.

*Keywords:* Boundary layer flashback, Flashback propensity, Laminar, Premixed flames

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## 1. Introduction

The flame flashback phenomenon in laminar flows has been known and studied for several decades [1]. A significant amount of results is available, which demonstrate the influences exerted by fuel, dilution and tube diameter choices [2], for instance. However, a recent review of the state of the art [2] indicates that significant knowledge gaps remain, in particular with regard to the combustible mixture Lewis number influence, and the effect of fuel dilution by CO<sub>2</sub>. Indeed, concerning the latter, CO<sub>2</sub> is used to displace oil in mature reservoirs, and is naturally found in some geological formations, which leads to fuel gas produced with dilutions that may reach 75 %. Therefore, these knowledge gaps prevent the use of existing results for the prediction of flame flashback, in particular under the CO<sub>2</sub>-abundant gas production conditions of the Brazilian pre-salt offshore platforms. For this reason, specific experimental studies of CO<sub>2</sub>-diluted hydrocarbon/air premixed flames flashback in laminar flows are undertaken here.

Classically [1], the flame flashback through the boundary layer is assumed to occur when the velocity of flow,  $u(y)$ , is smaller than the burning velocity,  $S_L$ , at some point near the wall, as seen in Fig. 1. The position at which the burning velocity velocity is equal to the flow velocity is defined as the penetration distance,  $\delta_p$ . The flame is supposed to be extinguished at a certain distance from the wall, called the quenching distance ( $\delta_q$ ). When the flow velocity at  $y = \delta_p$  is equal to the laminar flame speed the critical condition for flame flashback

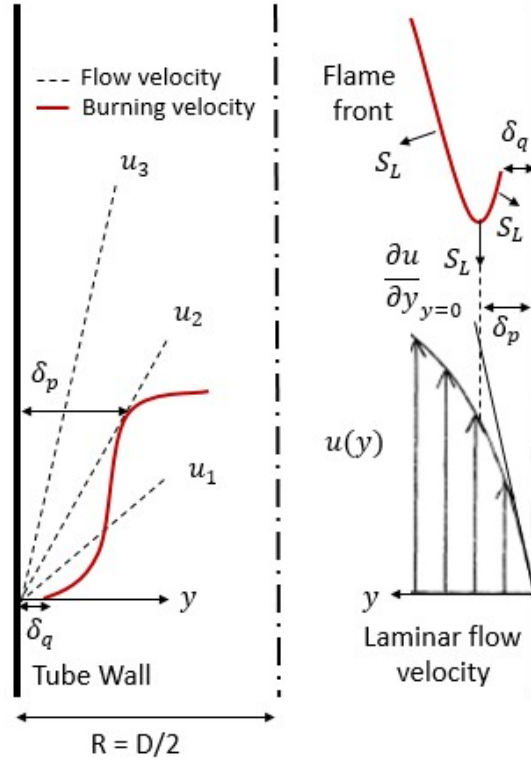


Figure 1: Schematic model of the interaction between flame and boundary layer that leads to the concept of the critical velocity gradient. Velocity profiles of the laminar flow and flame front under the flashback condition.

arises. This model further assumes that no flame/fluid interaction occurs and, therefore, that there are no disturbances in the laminar flow velocity upstream  
 25 to the flame.

During the past decades, the critical conditions leading to laminar boundary layer flashback in tubes have been subject of experimental and numerical studies. Pioneering experimental studies [3] have identified the role of the flow velocity gradient at the wall on the flashback onset. The critical value of this  
 30 gradient has been measured, for instance, as a function of the fuel type, mixture equivalence ratio and oxygen content, and summaries of these results are avail-

able [1, 2]. Numerical studies of this phenomenon have extended the critical velocity gradient concept by means either of a Damköhler or a Karlovitz number [4, 5, 6, 7]. The computed critical Damköhler number has been shown to  
 35 be an increasing function of the Lewis number, and to decrease when the tube wall boundary condition changes from isothermal to adiabatic. To the best of our knowledge, a systematic experimental investigation of the Lewis number influence on the flashback propensity has not been performed, however. More recently, flame stretch effects were accounted for and shown to decrease the critical  
 40 bulk velocity for flashback to occur for near-stoichiometry H<sub>2</sub>-air flames [8]. However the stretch influence on flashback was found to be less pronounced for methane/air flames.

In order to further define the laminar boundary layer flashback controlling parameters, Fig. 1 shows a schematic model of this process, where a stabilised  
 45 flame front can be seen inside a tube near the wall. On the right side of this figure, the velocity of the flow,  $u(y)$ , is represented by a parabolic profile. On the left side of Fig. 1, which considers the stabilised flame front immediately upstream the edge of the tube, represents the three conditions conditions for flame/flow interaction. The horizontal axis,  $y$ , represents the distance from the  
 50 tube wall, and the dashed lines 1, 2 and 3 symbolise different flow velocities. The solid curve represents the velocity of the laminar flame front near the wall. Note that, in this model, the laminar flame front velocity is zero within the quenching region, i.e.,  $0 < y < \delta_q$ . When the flow velocity is greater than  $S_L$ , the flame leaves the tube and stabilises on the outside, a situation which is  
 55 represented by line 3. Line 2 indicates the critical condition for observing flame flashback, when the flow velocity is equal to the laminar flame front speed at the flame penetration distance,  $y = \delta_p$ . If the flow velocity is reduced (line 1), the flame propagates into the tube. Given this picture, the velocity gradient used to characterise the flashback in the critical situation may then be defined

60 as

$$g_c = \left( \frac{du}{dy} \right)_{y=\delta_p} = \frac{S_L}{\delta_p}. \quad (1)$$

In the classic premixed flame flashback model [1, 2], the flame penetration distance within the boundary layer,  $\delta_p$ , that controls the flashback phenomenon, is considered proportional to the laminar flame thickness, i.e.,  $\delta_p = K\delta_L$ , where  $K$  is assumed to be constant. This model further supposes that a developed  
65 laminar flow exists within the tube, which enables the critical condition for flashback to occur through the boundary layer to be expressed by equating the flow's and the modelled expressions of the critical velocity gradient, i.e.,  $(du/dy)_{y=\delta_p} = 8\bar{U}/D = S_L/(K\delta_L)$ , where  $D$  is the tube diameter and  $\bar{U}$  is the bulk flow velocity, both which stem from measurements here.

70 One should note that  $K$  may be interpreted as a Karlovitz number or, conversely, as the inverse of a Damköhler number [4, 5]. The critical Damköhler number for flashback to occur may then written as

$$Da = \frac{S_L}{g_c\delta_L} = \frac{DS_L}{8\bar{U}\delta_L}. \quad (2)$$

An extra usual assumption concerns the definition of the laminar flame thickness, which is taken to be proportional to the mixture thermal diffusion coefficient,  $\delta_L = \alpha/S_L$ , thus leading to:  
75

$$Da = \frac{S_L^2}{g_c\alpha} = \frac{DS_L^2}{8\bar{U}\alpha}. \quad (3)$$

However, such a flame thickness definition is known to under-predict the computed thermal thickness of premixed flames [9]. Equations (2) and (3) suggest that, if the Damköhler number critical value is constant, the flashback propensity in terms of the critical velocity gradient should be reduced when  $S_L$  decreases, which is the case for hydrocarbon-air CO<sub>2</sub> diluted mixtures of interest  
80 to the present work.

The goal of this work is thus to determine the influence of the mixture composition and fuel dilution by CO<sub>2</sub> on the flame flashback limit, which is represented by a Damköhler number. This is effected by combining original experimental data of the critical flashback conditions, and by performing detailed  
85 chemical kinetics computations to determine the flame propagation velocity and thickness. More specifically, the question of whether  $Da$  may be assumed to be

constant is addressed, as is the influence of the mixture effective Lewis number. Furthermore, the extent to which different flame thickness definitions may  
90 change the critical Damköhler number values is examined also.

This paper is organised as follows: the experimental and computational methodologies used to determine the Damköhler and effective Lewis numbers for CO<sub>2</sub>-diluted methane and propane mixtures with air are first presented. Then  
95 is assessed the influence on the Damköhler number of the flame thickness definition, of the mixture equivalence ratio and fuel CO<sub>2</sub> dilution, and of the effective Lewis number. Finally, the main conclusions of this work are presented.

## 2. Methodology

### 2.1. Experimental methodology

To study the boundary layer flashback phenomenon in laminar flows an  
100 experimental bench was set up, which is schematically shown in Fig. 2. Its dimensions were chosen based on data found in the literature [2], which indicated that determining the flashback limits throughout the flammability range of methane and propane mixtures with air should require tubes with different diameters and a wide range of flow meters. Accordingly, this bench consists of  
105 interchangeable  $D = 17, 6, 5,$  and  $4$  mm inner diameter quartz tubes, which are vertically mounted, the larger diameters being used to determine the flashback around stoichiometry, and the smaller for either fuel lean or fuel rich mixtures. These tube diameters are known with a 0.6% accuracy. Note that for tubes with diameters smaller than 2 mm, extinction, instead of flashback, is expected  
110 to occur for undiluted mixtures [1]. The tubes have a length  $L$  of 1 m, that ensures a fully developed laminar flow for all the Reynolds numbers considered, i.e.,  $L/D > 0.05 \text{ Re}$ .

As shown in Fig. 2, Alicat Scientific flow controllers are used to measure the reactants flow rates prior to mixing. These MC-series controllers, with ranges of  
115 0.5, 5, 50 and 500 SCCM, 5 and 20 SLPM, have a standard accuracy calibration of  $\pm 0.6\%$  of reading or  $\pm 0.1\%$  of full scale, whichever is greater. Throughout

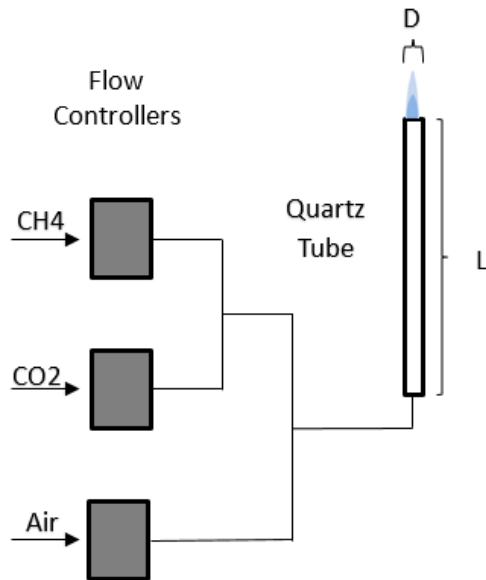


Figure 2: Experimental apparatus scheme, showing the fuel ( $\text{CH}_4$  or propane), air, and  $\text{CO}_2$  mass flow controllers, mixing tubes, vertical quartz tube, and flame position prior to flashback.

the experiments performed these flow controllers were used in different gas lines, so as to cover fuel/air mixtures from the lean to the rich flammability limit, and  $\text{CO}_2$  dilution in the fuel stream up to 75 %. Mixing is effected in a 6 mm  
 120 outer diameter tube which length is at least twice that required for obtaining a fully developed flow. The fuels studied here were methane and commercial propane, which constituents are  $\text{C}_3\text{H}_8/\text{CH}(\text{CH}_3)_2(\text{C}_2\text{H}_5)/\text{C}_2\text{H}_6/1\text{-C}_4\text{H}_8/\text{C}_3\text{H}_6$ , and molar basis composition is (89.1, 4.16, 3.12, 2.94, 0.68) %.

Each experiment begins with the flame stabilised at the quartz tube rim for  
 125 a chosen  $\text{CO}_2$  dilution in the fuel stream. The air flow rate is then increased or decreased, so that the flame flashback arises either from the fuel rich or the fuel lean equivalence ratio, respectively. The steps with which the air flow rate is varied was adapted in order to achieve an adequate, repeatable, resolution of the flame flashback. Indeed, for each flow-meter, these steps are always smaller than



130 1 % of the corresponding full scale. The time interval between those flow rate  
 steps was also larger for smaller flow rates, so that several flow residence times  
 within the tube were allowed for before changing the flow rate. For a range of  
 equivalence ratios at the vicinity of stoichiometry the flame may stabilise in a  
 configuration in which a partial entrance in the tube is observed [1]. In this  
 135 work flashback is assumed to occur only when the flame visually propagates  
 upstream in the tube, and such a partial entrance was not annotated. The  
 set of experiments enables to determine the critical velocity gradient ( $g_c =$   
 $32 \dot{V}/(\pi D^3) = 8\bar{U}/D$ ), where  $\dot{V}$  and  $\bar{U}$  are the measured mixture volume flow  
 rate and the corresponding mean flow velocity when flashback occurs, which are  
 140 functions of the equivalence ratio,  $\phi$ , and fuel (methane or propane) dilution by  
 CO<sub>2</sub>. Note that all experiments have been conducted under ambient conditions,  
 i.e., 1 atm and 25 C, and that the tube wall temperature was not measured.  
 The shortcomings associated to this will be addressed in section 3.3.

## 2.2. Numerical methodology

145 In order to determine  $Da$  via Eq. (2),  $S_L$ , the laminar flame front speed at  
 which the flat, isobaric, adiabatic flame freely propagates must be provided. To  
 this end, numerical calculations were performed using the Ansys CHEMKIN  
 software, version 2020 R1. The chemical kinetic mechanisms adopted were: (1)  
 the GRI-Mech 3.0 mechanism [10] for the methane/air/CO<sub>2</sub> mixtures, and (2)  
 150 for propane/air/CO<sub>2</sub> mixtures the San Diego Mech [11]. These mechanisms  
 were chosen to represent the chemical kinetics of the studied mixtures based on  
 their predictive ability demonstrated in previous review works [12, 13]. For the  
 sake of conciseness the computed flame velocities are not reported here.

The classical flame flashback analysis defines the flame thickness,  $\delta_L$ , as the  
 155 ratio of the fresh gases mixture thermal diffusion coefficient and the flame speed,

$$\delta_{L,T} = \frac{\alpha}{S_L}, \quad (4)$$

thus leading to Eq. (3). In this work the computed flame thickness was also de-  
 termined using a correction to  $\delta_{L,T}$  that accounts for variable thermal properties

across the flame [14],

$$\delta_{L,B} = 2 \frac{\alpha}{S_L} \left( \frac{T_b}{T_u} \right)^{0.7}, \quad (5)$$

and the temperature derivative maximum [15], i.e.,

$$\delta_{L,max} = \frac{T_b - T_u}{\max(dT/dx)}, \quad (6)$$

160 where,  $T_b$  and  $T_u$  are the temperatures of the burned gases at equilibrium and of fresh mixture, respectively, and  $dT/dx$  was obtained from the premixed flame computations. When performing such flame computations care was taken to check for domain and mesh independence of the results for all studied fuel/dilutant/air mixtures.

165 The Damköhler number is determined here using both experimental and computed data as  $Da = DS_L/(8\bar{U}\delta_L) = \pi D^3 S_L/(32\dot{V}\delta_L)$ . Therefore, the maximum experimental errors associated to  $Da$  are 2.4 % and 4.0 % for methane/air and propane/air respectively, whereas those corresponding to  $\phi$  are 1.1 % and 1.8 %. These errors stem from those associated to the tube diameter and reactants flow rates measurements.

In order to investigate the mixture composition influence on  $Da$ , two different representations are adopted here, i.e., (1) the equivalence ratio of the fresh gases, and (2) their effective Lewis number. The effective Lewis number is determined using a formulation [16] that relies on the Zel'dovich number to continuously vary between the fuel lean and fuel rich limits,

$$Le = Le_1 + H(Le_2 - Le_1), \quad (7)$$

where  $Le_2$  and  $Le_1$  are the fuel-rich and oxidiser-rich Lewis numbers, and

$$H = n \frac{G(m, n-1, A)}{G(m, n, A)}, \quad G(m, n, A) = \int_0^\infty \xi^m (\xi + A)^n \exp(-\xi) d\xi, \quad (8)$$

$m$  and  $n$  are the orders of reaction with respect to the deficient and abundant reactants, respectively, the normalised final concentration of the abundant reactant is either  $A = \beta(\phi-1)/Le_2$  or  $A = \beta(\phi-1)/Le_1$ , and  $\beta = E_a(T_b - T_u)/(RT_b^2)$  is the Zel'dovich number. For the sake of simplicity it is assumed that  $m = n =$

1; note that different reaction orders would lead to different slopes of the  $Le(\phi)$  curve [Eq. (7)].

The Zel'dovich number is determined using a recently proposed methodology, which uses an extended Arrhenius expression that is optimally fitted to the  
 185 computed premixed flame heat release rate [17]:

$$\dot{q}(\theta) \propto \{1 - \exp[\beta_1(1 - 1/\theta)]\}^{\beta_2} \exp\left\{\frac{\beta(\theta - 1)}{(1 + \gamma\theta)/(1 + \gamma)}\right\}, \quad (9)$$

where  $\theta = (T - T_u)/(T_b - T_u)$  is the temperature based reaction progress variable, and  $\gamma = (T_b - T_u)/T_u$  represents the thermal expansion across the flame. In this heat release rate expression,  $\beta_1$  and  $\beta_2$  control, respectively, the initial and the final  $\dot{q}$  decrease towards equilibrium as  $\theta \rightarrow 1$ . For the sake of com-  
 190 pleteness, the Zel'dovich number correlations resulting from such a procedure that are used in Eq. (8) read:

$$\text{methane/air : } \beta = 10.4 \phi^4 - 49.6 \phi^3 + 98.7 \phi^2 - 90.7 \phi + 33.4, \quad (10)$$

$$\text{propane/air : } \beta = -21.7 \phi^4 + 106 \phi^3 - 164 \phi^2 + 94.2 \phi - 12.3, \quad (11)$$

for which  $R^2 > 0.98$ .

### 3. Results and Discussion

#### 3.1. Flame thickness definition influence

195 In this section is examined the influence of three different flame thickness definitions on the flame flashback critical Damköhler number, Eq. (2). More specifically, the considered flame thickness definitions are given by Eqs. (4), (5) and (6). Figure 3 shows the evolution with the mixture equivalence ratio of the computed flame thickness, as well as the corresponding critical Damköhler  
 200 numbers, for methane/air and propane/air mixtures. For the sake of simplicity undiluted fuel mixtures results are given and discussed in this section only.

In Figs. 3a and 3b it can clearly be seen that the three flame thickness definitions lead to minimum values at equivalence ratios slightly larger than

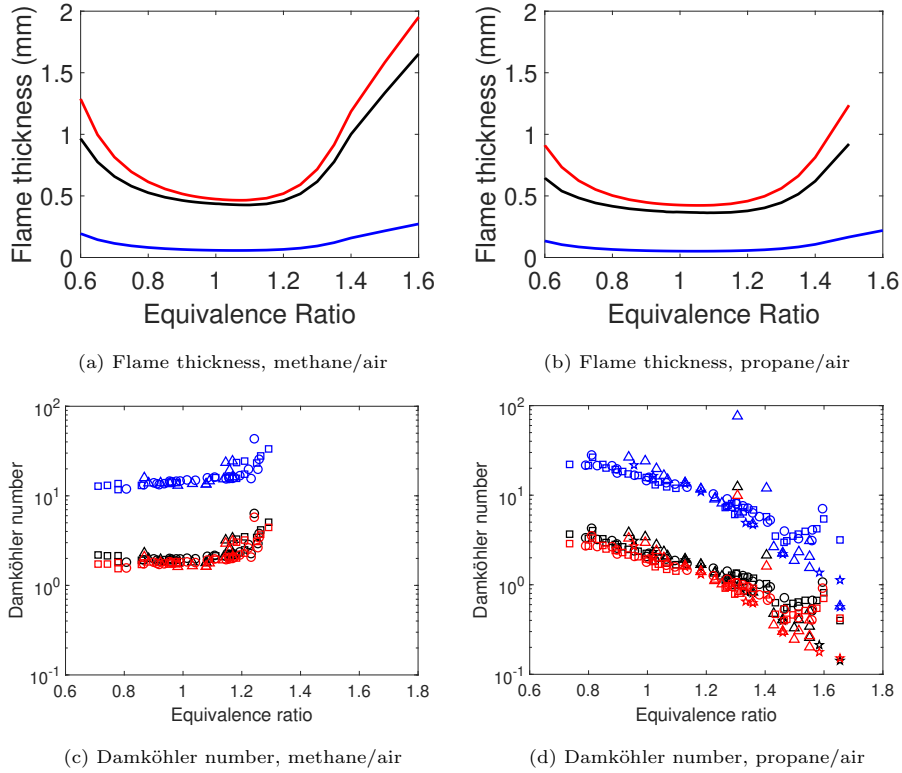


Figure 3: Flame thickness and Damköhler number as a function of the equivalence ratio for undiluted fuel/air mixtures,  $\delta_{L,T}$  : blue,  $\delta_{L,B}$  : red,  $\delta_{L,max}$  : black. The symbols correspond to different tube diameters, i.e.,  $D = 4$  ( $\Delta$ ),  $5$  ( $\star$ ),  $6$  ( $\circ$ ) and  $17$  ( $\square$ ) mm.

stoichiometry, which is related to the well-known rich shift of maximum  $S_L$  [18].

205

Indeed, regardless of the flame thickness definition chosen, both methane and propane mixtures with air exhibit a thickness increase for leaner and richer mixtures. For instance, the leanest methane/air flame thermal thickness is 3.3 times that of a stoichiometric flame, whereas this ratio is 2.2 when the maximum temperature gradient thickness is considered. Concerning lean propane/air flames, these ratios are somewhat smaller, i.e., 2.5 and 1.8, respectively. Rich fuel/air mixtures exhibit similar trends, albeit with larger absolute values for this thickness ratio. Furthermore, at stoichiometry, the flame thickness values

210

computed via Eqs. (4), (5) and (6), i.e.,  $\delta_{L,T}$ ,  $\delta_{L,B}$ , and  $\delta_{L,max}$ , are respectively  
 215 0.058, 0.47, and 0.43 mm for methane/air and 0.051, 0.42, and 0.36 mm for  
 propane air mixtures. This reasonable agreement between  $\delta_{L,max}$  and  $\delta_{L,B}$  is  
 verified throughout the considered equivalence ratio range, which confirms that  
 the latter is a good approximation to the former. Furthermore, Figs. 3a and 3b  
 indicate that the choice of flame thickness definition should exert a significant  
 220 impact on the Damköhler number computed via Eq. (2), since  $\delta_{L,T} \ll \delta_{L,max}$ .

Indeed, Figs. 3c and 3d clearly show that the critical Damköhler number  
 computed using the flame thermal thickness is an order of magnitude larger  
 than that resulting from the two other formulations. Nevertheless, the overall  
 behaviour with the mixture equivalence ratio is independent of the flame thick-  
 225 ness definition. For instance, examining first the computed  $Da$  values using  
 $\delta_{L,max}$  as flame thickness definition, for methane mixtures, Fig. 3c shows that  
 the Damköhler number is practically independent of the equivalence ratio for  
 $0.7 \leq \phi \leq 1.1$ , that is,  $Da \approx 2.5$ , when  $\delta_{L,max}$  is chosen. However, for  $\phi > 1.1$ ,  
 an increase in the Damköhler number with equivalence ratio is observed. Us-  
 230 ing the same thickness definition for undiluted propane mixtures, as seen in  
 Fig. 3d, the Damköhler number decreases steadily from  $\approx 4$  to  $\approx 0.2$  with  $\phi$ ,  
 to  $0.7 \leq \phi \leq 1.5$ . These results confirm early numerical results [4], which in-  
 dicated that the constant  $Da$  assumption should not be used to describe flame  
 flashback. However, methane/air and propane/air mixtures exhibit increasing  
 235 and decreasing  $Da$  trends with  $\phi$ , respectively, which discussion is deferred to  
 section 3.3

### 3.2. Equivalence ratio and CO<sub>2</sub> dilution influence

The results of the critical Damköhler number for mixtures of methane/air/-  
 CO<sub>2</sub> and propane/air/CO<sub>2</sub> are presented in Fig. 4, which depicts the  $Da$  values  
 240 as a function of the equivalence ratio, for several tube diameters. Since the three  
 flame thickness definitions examined in section 3.1 yielded analogous trends,  
 the one adopted here is  $\delta_{L,max}$  only. This figure shows that, for moderate CO<sub>2</sub>  
 dilutions, the overall trends of  $Da$  with  $\phi$  are identical to those seen in Fig. 3

for undiluted mixtures, i.e., for methane/air mixtures a moderate  $Da$  increase  
245 with  $\phi$  is seen, whereas propane/air mixtures exhibit a corresponding decrease  
that spans over more than an order of magnitude. Since the adopted model uses  
the assumption that  $Da = \delta_L/\delta_p$ , this suggests that flame penetration distance  
is relatively smaller in leaner propane/air mixtures, when compared to richer  
ones.

250 Concerning now the higher  $\text{CO}_2$  dilutions, as seen in Fig. 4a, methane mix-  
tures exhibit a  $Da$  increase as  $\text{CO}_2$  is added beyond 30 %. For example, in  
the  $D = 17$  mm tube, for stoichiometric methane/air mixtures, the Damköhler  
number is 1.7 and for methane/air/30% $\text{CO}_2$  mixtures it is 2.8. As may be  
seen in Fig. 4b propane mixtures have a similar behaviour with respect to  
255  $\text{CO}_2$  dilution. Indeed, a stoichiometric propane/air mixture characterised by  
a Damköhler number of the order of 1.7, whereas a stoichiometric mixture of  
propane/air/75 %  $\text{CO}_2$  is of the order of 4.6. It should be stressed that, in the  
particular case of those highest fuel dilutions, and for the smallest tube diame-  
ters, the flame flashback is followed by an nearly immediate extinction, i.e., the  
260 flame does not sustain stable propagation throughout the tube length. In this  
sense, these particular conditions do not strictly represent the flame flashback  
phenomenon. Thus, if those flame extinction results are discarded, Fig. 4 indi-  
cates that the critical  $Da$  value is independent of the tube diameter regardless  
the  $\text{CO}_2$  dilution considered.

### 265 3.3. Lewis number and $\text{CO}_2$ dilution influence

In order to address the relation between the Lewis number and the critical  
flashback conditions, Fig. 5 depicts the effective Lewis number for the different  
combustible mixtures studied. This figure shows that the effective Lewis number  
behaviour as a function of equivalence ratio is a classical one, tending to the  
270 value of the limiting reactant for very lean or very rich mixtures. The increase  
of  $\text{CO}_2$  dilution in the fuel leads to a decrease of the effective Lewis number  
for methane mixtures and for rich propane mixtures, which is related to the  
 $\text{CO}_2$  smaller thermal diffusivity. However, dilution by  $\text{CO}_2$  exerts a nearly

negligible influence on the effective Lewis number of lean propane mixtures,  
 275 which is associated to nearly identical molecular weights of  $C_3H_8$  and  $CO_2$ .

Figure 6 depicts the critical Damköler number as a function of the effective  
 Lewis number for the different mixtures studied. The present results are shown  
 as symbols, whereas the lines represent those of previous numerical studies [6].  
 Again, each symbol represents a tube diameter, whereas the colours stand for  
 280 different fuel dilutions by  $CO_2$ . It is important to stress that the solid and  
 dashed lines represent limiting cases of isothermal (300 K) and adiabatic bound-  
 ary conditions at the tube wall, respectively, and that the Zel'dovich number  
 used in the depicted computational results is  $\beta = 10$  [6]. These computational  
 results clearly show that the flame flashback  $Da$  increases with the Lewis num-  
 285 ber and decreases when the tube wall approaches an adiabatic condition. This  
 former trend was also observed in the present study for propane/air flames, as  
 may be verified in Fig. 6b, which closely follow the adiabatic boundary tendency,  
 with the notable exception of those flashback cases that lead to flame extinction  
 (solid symbols). Indeed, within the range of the current measured data, the  
 290 following correlation holds:

$$Da \propto Le^\ell, \quad \text{with } \ell = 5.6, \quad (12)$$

which may be compared with a previously determined value of  $\ell = 1.68$  [7] for  
 turbulent hydrogen/air flames.

The observed methane/air mixtures behaviour could seem, at first, to con-  
 tradict that of the propane/air mixtures. However, for these mixtures, which  
 295 are characterised by a rather small Lewis number variation, the effect domi-  
 nating the critical  $Da$  seems to be that of thermal boundary condition at the  
 tube wall. Indeed, when using this  $(Le, Da)$  representation, Fig. 6a results in-  
 dicates that smaller tube diameters tend to lead to larger  $Da$  values, which was  
 not readily apparent in Fig. 4a. For these smaller tubes, the ratio between the  
 300 quenched mixture region and flame surface areas could be expected to increase,  
 thus influencing the flashback conditions. This results underscore the need to  
 include, in the  $Da$  model, a Péclet number, which could be modelled as [19]

$Pe = \delta_q/\delta_L = 1/\varphi - 1$ , where the quenching distance,  $\delta_q$  is schematically shown in Fig. 1, and  $\varphi$  is the ratio between the measured flame heat transfer to the wall and the flame heat release. The present experimental apparatus does not enable a direct measurement of the former, and thus of  $\varphi$ . Therefore the development of a more general correlation, i.e,  $Da(Le, Pe)$  is out of reach of the present endeavour. Another interesting avenue for such a development could be the use of artificial neural networks to avoid systematic measurements of the tube tip thermal boundary conditions [20].

#### 4. Conclusion

The critical Damköhler number for flame flashback to occur in  $\text{CH}_4$  and  $\text{C}_3\text{H}_8$  mixtures was determined as a function of the fuel, the equivalence ratio and  $\text{CO}_2$  dilution. To this end, measured flashback critical velocity gradient data, which provides a fluid time scale, were combined with detailed chemical kinetics simulation results of laminar premixed flames. These computations enabled to define the flame time scale using three different flame thickness definitions, i.e., (i) the classical thermal thickness, (ii) that based on the maximum temperature gradient, and (iii) one that corrects for temperature dependence of the thermal diffusion coefficient in (i) and thus approximates (ii). These length scales led to critical Damköhler numbers that exhibit a similar trend with equivalence ratio, but which value is nearly an order of magnitude larger when (i) is used.

Methane/air/ $\text{CO}_2$  flames exhibited a slightly increasing Damköhler number with equivalence ratio. On the other hand, propane/air/ $\text{CO}_2$  flashback is characterised by almost a two order of magnitude Damköhler number decrease as the equivalence ratio goes from 0.7 to 1.6. For both mixtures,  $\text{CO}_2$  dilution results in a Damköhler number increase only when flashback leads to flame extinction within the tube.

The Lewis number influence on the critical Damköhler number of propane mixtures was found to follow existing numerical simulation results trends, leading to a  $Da \propto Le^{5.6}$  dependency. This good agreement between the present results



and those numerical simulations suggest that the critical Damköhler number defined using the temperature gradient flame thickness is more suitable than the one that uses the thermal thickness. On the contrary, methane mixtures results  
335 showed a lesser sensitivity to Lewis number variations – which are rather small – but a seemingly larger one to tube diameter. Indeed, smaller tube diameters led to larger critical Damköhler number values, approaching those of the constant tube temperature simulations, which could be related to the thermal boundary conditions at the wall, that were not characterised in this study. Such a result  
340 indicates that the quenching region near the flame wall should be accounted for in a more general Damköhler number model, which could use a Péclet number, for instance.

## 5. Acknowledgments

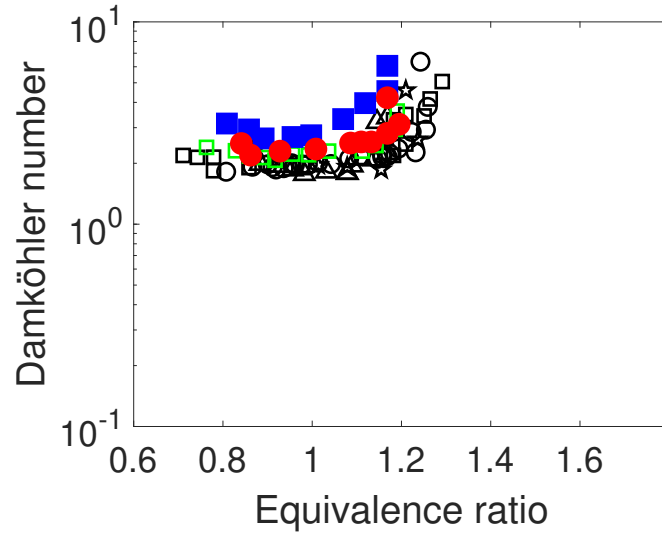
This work was performed while L.F. Figueira da Silva was on leave from  
345 the Institut Pprime (UPR 3346 CNRS, France). This work was supported by Petrobras contract no. 5900.0111688.19.9 under the technical monitoring of Dr. P. R. Pagot. Support was also received from Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq, Brazil) under the Research Grant No. 304444/2018-9. This study was financed in part by the Coordenação  
350 de Aperfeiçoamento de Pessoal de Nível Superior - Brasil (CAPES) - Finance Code 001.

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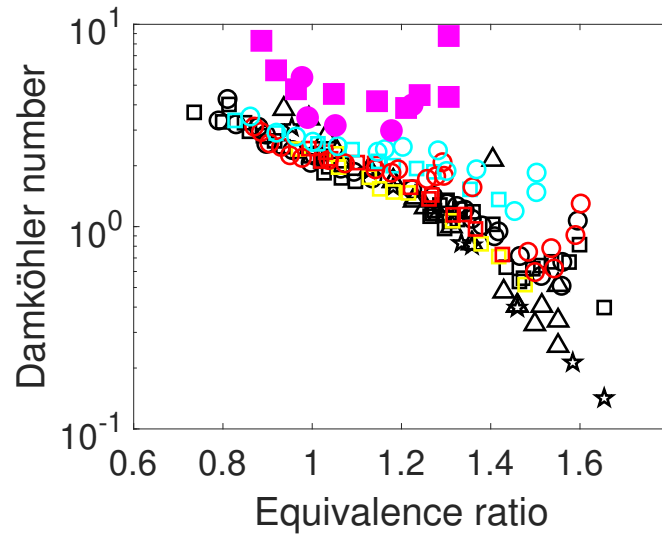
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(a) Methane/air



(b) Propane/air

Figure 4: Damköhler number as a function of the equivalence ratio for mixtures in tubes of  $D = 4$  ( $\Delta$ ),  $5$  ( $\star$ ),  $6$  ( $\circ$ ) and  $17$  ( $\square$ ) mm. The symbols colours are organised such as: black - undiluted, green - 10 %, yellow - 15 %, red - 25 %, blue - 30 %, cyan - 50 % and magenta - 75 %. The filled symbols represent the highest dilution percentage.

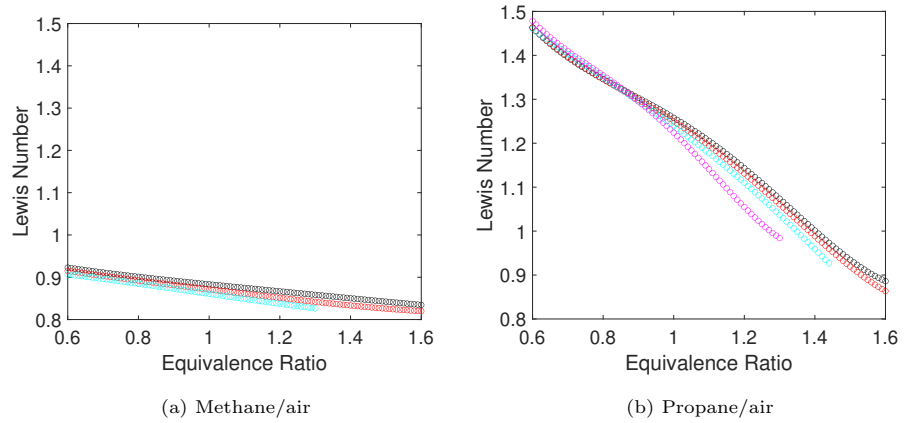
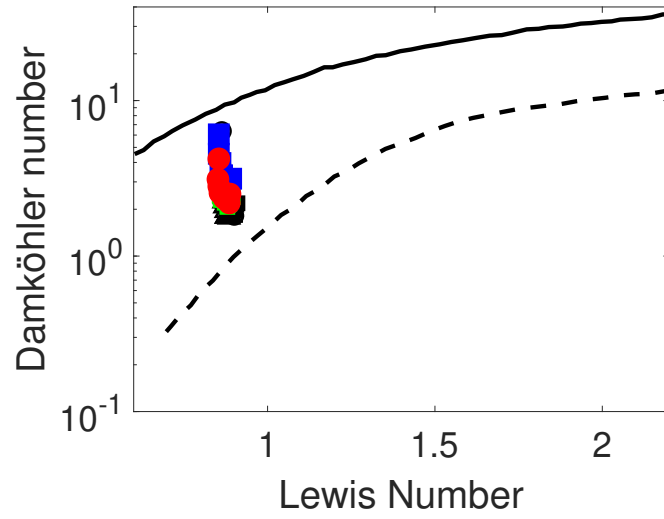
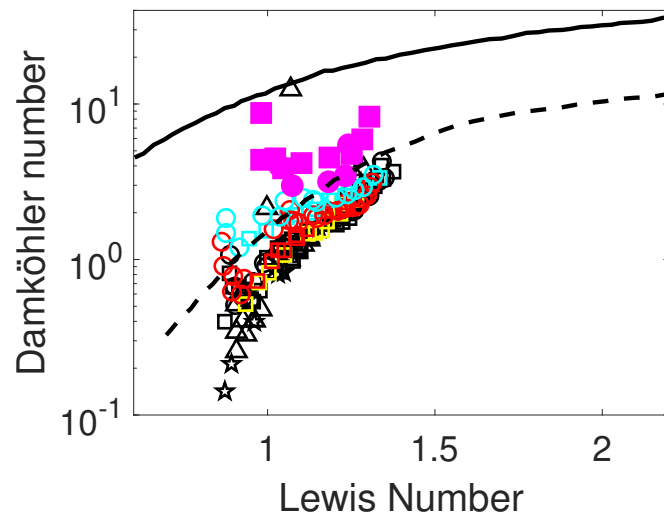


Figure 5: Computed effective Lewis number as a function of the mixture equivalence ratio for different CO<sub>2</sub> dilutions in the studied fuels. The symbols colours are organised such as: black - undiluted, red - 25 %, cyan - 50 % and magenta - 75 %.



(a) Methane/air



(b) Propane/air

Figure 6: Damköhler number as a function of the effective Lewis number for mixtures in tubes of  $D = 4$  ( $\Delta$ ),  $5$  ( $\star$ ),  $6$  ( $\circ$ ) and  $17$  ( $\square$ ) mm. The symbols colors are organized such as: black - undiluted, green - 10 %, yellow - 15 %, red - 25 %, blue - 30 %, cyan - 50 % and magenta - 75 %. The filled symbols represent the highest dilution percentages. The lines are the previous computed results [6]: dashed - adiabatic, solid - isothermal (300 K) tube wall.