

OXYFUEL COMBUSTION AND REACTANT PREHEATING ADAPTED TO LOW CALORIFIC GASES

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Abstract

This paper aims at pointing out the potential of the association of reactant preheating and oxyfuel combustion to enhance flame stabilisation of low calorific fuels. For this purpose, a computational study of mono-dimensional freely propagating premixed laminar flames and counter-flow diffusion flames is performed to obtain main properties of preheated oxyfuel flames of low calorific gases: thermal equilibrium, adiabatic temperature, laminar burning velocity, flame thickness and extinction strain rate. Four kinetic mechanisms are used for these calculations and show similar results. The study is focused on Blast Furnace Gas (BFG), which net calorific value is one order of magnitude lower than methane. The results show that the significant enhancement of oxyfuel flame properties induced by the preheating of BFG and oxygen can be considered as equivalent to some addition of a high calorific fuel to ensure flame stabilisation. The whole of the kinetic calculations are used to develop a tri-coaxial oxyfuel burner of BFG with a design strategy based on the calculation of a critical Damköhler number.

Introduction

The rapid growth of world energetic demand and the progressive decrease of fossil fuels availability are strong issues that faces industrial combustion sector, which has to develop new technologies turned towards alternative fuels and high thermal efficiency. For this purpose, fuels such as biogas, coke oven gas (COG), blast furnace gas (BFG), and synthetic gas derived from coal and biomass gasification, are alternative sources of energy and reliable solutions to be economically profitable and environmentally friendly compared to traditional fossil fuels. However, their substantial variations in composition during time and their low calorific value (LCV) are main drawbacks towards their use, making flame more difficult to stabilize and still significant pollution emissions. For illustration, Table 1 presents typical compositions of biogas, syngas, COG and BFG in comparison to methane as traditional fossil fuel. Stoichiometric mixture fraction Z_s and net calorific value (NCV) are also reported on Table 1. One can see that the calorific value of BFG is an order of magnitude lower than the one of methane, justifying the term 'LCV fuel' associated to these gases.

Whether oxyfuel combustion is thus an attractive solution to solve the stabilization issue of LCV flames, the additional use of a fuel with high calorific value (HCV) is still required [1]. The purpose of the present work is to investigate how the preheating of the fuel and oxygen reactants can be considered as an innovative solution to enhance the combustion of LCV fuels.

As a first step of this work, several thermochemical

calculations of major flame properties of low calorific value (LCV) fuels are performed to quantify the potential of reactant preheating and to derive guidelines for burner design.

Table 1. Compositions and main properties of typical biogas, syngas, coke oven gas (COG) and blast furnace gas (BFG) compared to methane.

concentrations (% vol.)	methane	biogas	syngas	COG	BFG
CH ₄	100	52	7	28	-
H ₂	-	-	9	62	5
CO	-	-	14	6	23
CO ₂	-	40	20	4	23
N ₂	-	8	50	-	49
NCV (kWh/Nm ³)	9.94	5.17	1.44	4.75	0.95
Z _s	0.34	0.48	0.20	0.24	0.88

1. The kinetic mechanisms for flame properties calculations

Three types of canonical flame geometries were simulated: 0D fully premixed reactor to determine thermo-kinetic equilibrium, 1D stationary planar laminar flame to determine laminar burning velocity S_L , flame thickness δ_{th} and adiabatic flame temperature T_{ad} , and stationary counter-flow diffusion flame to determine the evolution of adiabatic flame temperature with the strain rate K and its upper limit K^* before extinction. For these simulations, the COSILAB[®] package is used within four different kinetic mechanisms that are tested and compared (Table 2). Indeed, no mechanism validated for our specific

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conditions of oxyfuel combustion of LCV fuels with reactant preheating has been found in literature.

The GRI-Mech 3.0 is a detailed mechanism initially developed for methane and natural gas combustion with air, and validated in a large range of temperature, pressure and fuel equivalence ratio. [2]. It consists in 325 elementary reactions for 53 species (Table 2). Its use has been also extended to oxyfuel combustion [3-5] as well as different fuel mixtures representative of BFG [6], BFG-COG mixtures [7], syngas in air flame [8] and syngas with oxygen-enriched air [9]. The three other kinetic mechanisms also used in the present study have been especially developed for H₂-CO mixtures: 'OpenSmoke' [10], 'Sun mech' [11] and 'Davis mech' [12]. The latter has been also used for the study of oxygen-enriched-air combustion of syngas [9]. The effect of initial temperature on laminar burning velocity has been studied by Zhou *et al* [13] with the Sun mechanism and by Natajara *et al.* with GRI-Mech 3.0 and Davis mech [14].

Table 2. Numbers of species and elementary reactions involved in the four tested kinetic mechanisms.

		species	reactions
GRI-Mech 3.0	[2]	53	325
Davis Mech	[12]	14	43
OpenSMOKE	[10]	32	174
Sun Mech	[11]	16	33

For all these conditions, from the discrepancies observed between kinetic mechanism and experimental data used for validation as well as between kinetic mechanisms, no definitive conclusion on the adequacy of one or more kinetic mechanisms to simulate our specific conditions can be drawn. For this purpose, the four schemes are first compared by calculating the adiabatic temperature T_{ad} and the laminar burning velocity S_L obtained from the simulations of a freely propagating laminar BFG-O₂ oxyfuel flame of blast furnace gas in stoichiometric conditions ($\phi = 1$) for different reactant preheating temperature T_{pr} from 298 K to 850 K (Figure 1).

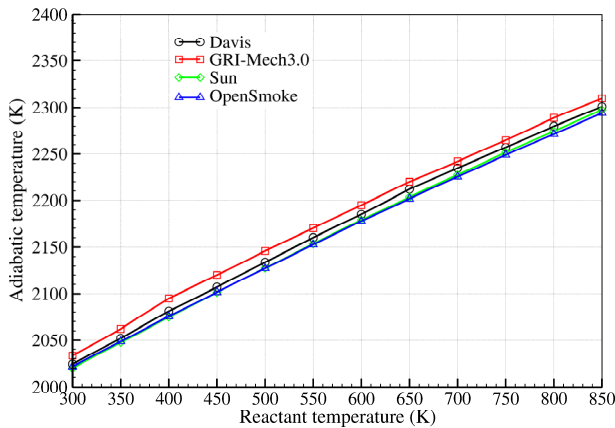


Figure 1. Evolutions of adiabatic flame temperature T_{ad} vs. T_{pr} obtained with the four tested kinetic mechanisms (BFG-O₂ flame at $\phi = 1$).

The adiabatic temperature shows almost no difference between the four kinetic mechanisms. A similar quasi-linear variation of T_{ad} with T_{pr} is observed with the four mechanisms. The maximum of difference of adiabatic temperature is 20 K. The GRI 3.0 Mech seems to have a slight over-prediction of T_{ad} compared to the other mechanisms which have almost no difference.

Figure 2 shows a more noticeable difference between laminar burning velocity S_L calculated with the four mechanisms. The discrepancy is larger for high reactant preheat temperature but the relative difference is always lower than 15%. For the four mechanisms, a similar shape of the exponential increase of S_L with T_{pr} is observed. This shows the ability of such simulations to point out the effect of initial reactant temperature on the main characteristics of oxyfuel flames of LCV fuels.

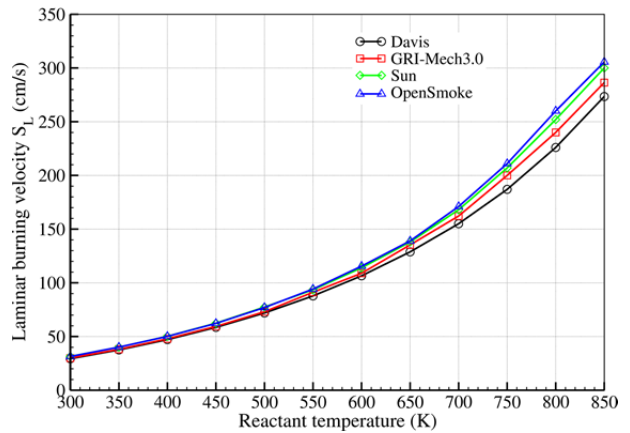


Figure 2. Evolutions of laminar burning velocity S_L vs. T_{pr} obtained with the four tested kinetic mechanisms (BFG-O₂ flame - $\phi = 1$).

Few validation comparisons are available for oxyfuel combustion, especially for these new alternative fuels. Thus the agreement between the four models can be considered correct. As the Davis mechanism has been initially developed and optimised for H₂-CO mixtures which are the only combusting components of the BFG composition considered in the present work, and already used with preheated reactants and with oxygen-enriched air combustion of LCV, this mechanism has been chosen to carry on studying the effect of the reactant preheating on oxyfuel flames of LCV fuels. Nevertheless, similar would be obtained with the other mechanisms.

2. Oxyfuel flame properties of different LCV fuels

The laminar burning velocity S_L is obtained from simulations of a mono-dimensional stationary laminar premixed flame at 298 K and atmospheric pressure. Its evolution with the equivalence ratio is presented in Figure 3 for oxyfuel flames of different LCV fuels: biogas, syngas, and BFG, which compositions are given in Table 1, and compared to a reference CH₄-air flame.

For all these cases, the S_L curve presents the traditional bell shape, but with a maximum on the lean side for the oxyfuel flames, because of the absence of nitrogen in the oxidizer.

As biogas is a 50:50 mixture of methane and CO_2 and N_2 inert gases (Table 1), the laminar burning velocity of biogas oxyfuel flame is very high. For syngas- O_2 , despite a low NCV, the range of S_L is also sufficiently large to ensure flame stabilisation when pure oxygen is used as oxidizer. These high values of S_L are not conserved with BFG- O_2 mixture: the maximum of laminar burning velocity is lower than the one of a CH_4 -air flame chosen as reference flame. This result shows that the stabilisation of a turbulent oxyfuel BFG flame would be an issue that requires to consider additional supply of heat. Thus, in the following the potential of reactant preheating from flue gas recovery on flame stabilisation of LCV oxyfuel flames is focused on the worst case of BFG- O_2 flame.

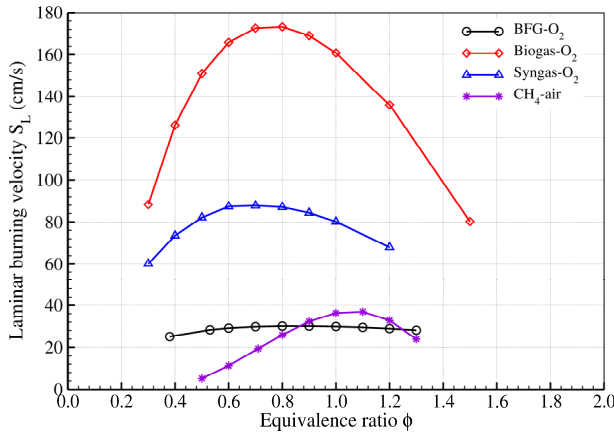


Figure 3. Evolutions of laminar burning velocity S_L with equivalence ratio for different LCV oxyfuel flames and a CH_4 -air flame (Davis mech - $T_{pr} = 298 \text{ K}$).

3. Effect of reactant preheating on oxyfuel combustion of BFG

Figure 4 shows adiabatic temperature and major species molar fractions at equilibrium versus initial preheat temperature T_{pr} for BFG- O_2 stoichiometric mixtures calculated with the chemical equilibrium code of the COSILAB package. As already mentioned, the adiabatic flame temperature increases quasi-linearly with the initial temperature, from $T_{ad} = 2020 \text{ K}$ at $T_{pr} = 298 \text{ K}$ to $T_{ad} = 2325 \text{ K}$ at $T_{pr} = 900 \text{ K}$.

Increase in initial temperature T_{pr} leads to decreases in CO_2 and H_2O molar fractions because of their thermal dissociations and then the correlated increases of CO and H_2 molar fractions. High concentration of molecular nitrogen ($[\text{N}_2] = 49 \text{ \% vol.}$) in BFG induces significant formation of NO . This shows that the reactant preheating from flue gas recovery may have a significant impact on pollutant emissions (CO_2 and NO

concentrations in flue gas) that would have to be considered in practical applications.

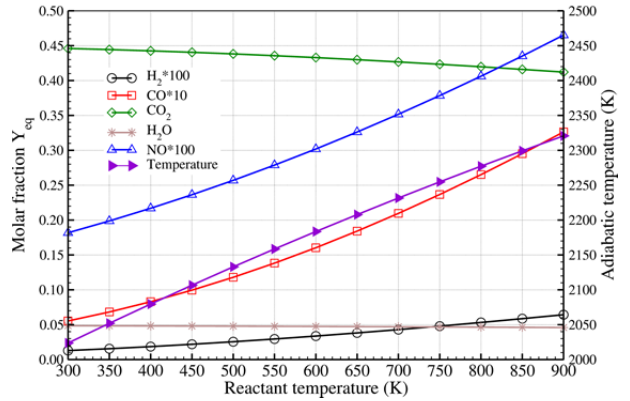


Figure 4. Adiabatic temperature and equilibrium species molar fractions vs. initial reactant temperature T_{pr} (stoichiometric BFG- O_2 mixture).

Simulations with Davis mechanism of monodimensional freely propagating premixed laminar flames of stoichiometric BFG- O_2 mixtures are performed to determine laminar burning velocity S_L and flame thickness δ_{th} , which are essential properties to control the flame stabilization in the wake of burner nozzle lip. The latter is deduced from the temperature profile across the flame front as the ratio between the temperature difference ($T_{ad} - T_{pr}$) and the maximum of temperature gradient across the flame front. Figure 5 presents the evolutions of S_L and δ_{th} versus initial reactant temperature.

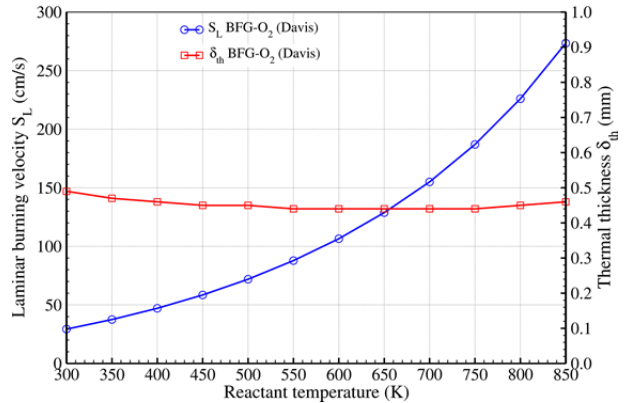


Figure 5. Evolutions of laminar burning velocity and flame thickness with initial reactant temperature T_{pr} of a stoichiometric BFG oxyfuel flame (Davis Mechanism).

The laminar burning velocity is multiplied by 9 when T_{pr} varies from 300 K to 850 K , with an exponential rise as already mentioned. High values of S_L rapidly reached augur on the potential of reactant preheating to ensure stabilization of oxyfuel flame even with very low calorific value. Without preheating, flame thickness of BFG- O_2 oxyfuel flame is high ($\delta_{th} \approx 0.5 \text{ mm}$), but has a very slight decrease when increasing T_{pr} . The narrowing of the temperature difference ($T_{ad} - T_{pr}$)

when preheating reactants is compensated by an equivalent decrease of the temperature gradient (Figure 6). Then in a turbulent BFG-O₂ flame, significant interaction between flame front and small turbulent structures can be expected with such flame thickness.

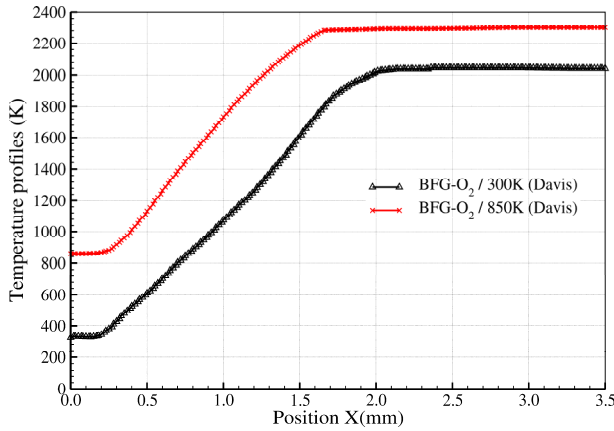


Figure 6. Temperature profiles across a laminar premixed BFG-O₂ flame front for two initial temperatures.

4. Comparison between reactant preheating and HCV fuel addition

The stabilization of an oxyfuel flame of LCV fuel can be ensured by the addition of a high calorific value (HCV) fuel such as natural gas or methane. In a previous study, we have shown that the stabilization of BFG-O₂ diffusion flame required the presence of a CH₄-O₂ pilot flame. This has led to the development of a quadri-coaxial burner studied both at 25 kW lab-scale and at 500 kW semi-industrial scale [15].

The properties of preheated BFG-O₂ flames are compared to BFG-O₂ flames enriched with methane. First, to quantify this effect and compare to BFG and oxygen preheating, laminar burning velocity have been calculated from simulations of mono-dimensional freely propagating laminar stoichiometric premixed oxyfuel flame of fuel mixtures of BFG and CH₄ in different proportions. Figure 7 presents the evolution of S_L when progressively adding methane in (BFG-CH₄) mixture at 300 K.

As far as the laminar burning velocity is concerned, the preheating of BFG and oxygen at 550 K is equivalent to a fuel mixture of 10% CH₄-90% BFG for which a large range of stable turbulent flames has been previously obtained [1]. In the largest expected reactant preheating ($T_{pr} = 800$ K), the high laminar burning velocity (close to $S_L = 200$ cm.s⁻¹) is similar to the one obtained in an oxyfuel flame of (50% CH₄-50% BFG), which has strong ability to stabilize.

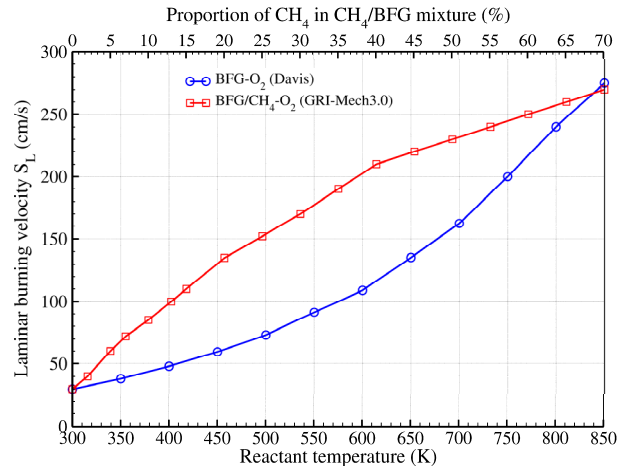


Figure 7. Comparison between the evolutions of the laminar burning velocity with reactant preheating temperature T_{pr} (lower x-axis) and with the proportion of methane in a (BFG-CH₄) fuel mixture (upper x-axis).

The comparison of reactant preheating and HCV fuel addition is also performed from simulations of mono-dimensional counterflow diffusion flames with different strain rates K , since flame extinction influences pollutant emission and heat transfer. Figure 8 presents the profiles of adiabatic temperature of such counterflow diffusion flames. The adiabatic temperature decreases when the strain rate increases up to the quenching value K^* where the flame is blown off because of too large stretch.

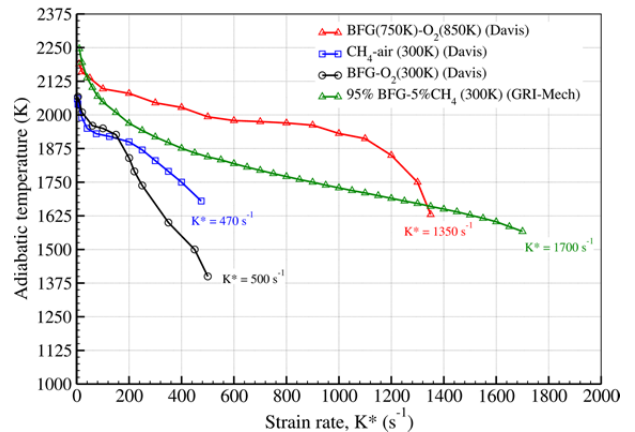


Figure 8. Adiabatic flame temperature T_{ad} vs. strain rate K of mono-dimensional counterflow diffusion flames.

For low strain rates ($K < 200$ s⁻¹), the BFG-O₂ oxyfuel flame without preheating (300 K) has similar tendency of T_{ad} with K to a CH₄-air flame considered as a reference case. From $K > 200$ s⁻¹, the adiabatic temperature of the BFG-O₂ oxyfuel flame falls down more quickly than the CH₄-air flame. Whereas, their quenching strain rate are similar, around $K^* = 500$ s⁻¹.

For the BFG-O₂ oxyfuel flame, the preheating of the fuel at 750 K and the oxygen at 850 K has two beneficial effects. As already pointed out, the adiabatic

temperature is largely increased, but also the preheating induces a significant shift of the maximum of strain rate: up to $K^* = 1350 \text{ s}^{-1}$. The latter moves closer to the quenching strain rate of an oxyfuel flame of a (5% CH_4 -95% BFG) mixture. However the preheated BFG- O_2 oxyfuel fame has higher adiabatic temperature in a large range of strain rate. Based on the results of the previous study of (BFG- CH_4) oxyfuel flames, these numerical results confirm the objective of preheating the reactants and building an experimental set-up.

5. A design strategy of oxyfuel combustion burner adapted to LCV fuels

The effect of reactant preheating on the main features of oxyfuel flames of LCV gases will be studied in a lab-scale facility equipped with electric heating systems for oxygen and fuel mixtures. A synthetic BFG mixture is provided by a dynamic mixing system of pure gases.

Based on previous experience of BFG oxyfuel combustion [1], a tri-coaxial burner configuration is chosen, with an annular BFG injection surrounded by a central (O_{2i}) oxygen injection and a peripheral (O_{2e}) oxygen injection (Figure 9). The oxygen repartition is quantified by non-dimensional parameter R_o , defined from internal $Q_{O_{2i}}$ and external $Q_{O_{2e}}$ oxygen flowrates:

$$R_o = \frac{Q_{O_{2i}}}{Q_{O_{2i}} + Q_{O_{2e}}}$$

For fixed LCV fuel composition and global equivalence ratio, the repartition of oxygen between central and external flows is used to control flame length, heat transfer along the combustion chamber and pollutant emissions in flue gas.

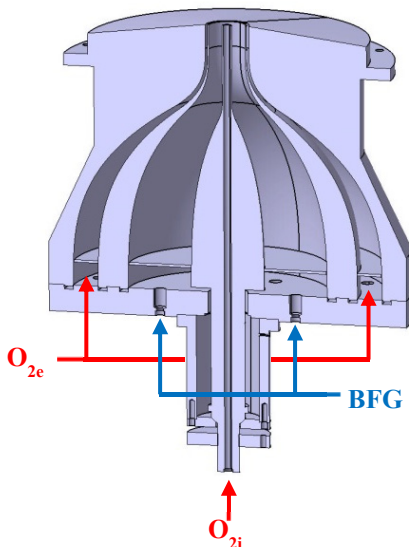


Figure 9. A diagram of the tri-coaxial O_2 /BFG/ O_2 burner.

Previous experimental study has shown that the stabilization of a BFG- O_2 turbulent diffusion flame at 300 K is controlled by a maximum value of the convection velocity $U_c = (U_{BFG} + U_{O_2})/2 = 10 \text{ m} \cdot \text{s}^{-1}$ [1]. The extinction of such BFG- O_2 turbulent diffusion flame can be characterized in terms of a critical Damköhler number representing the competition between the mixing rate within the shear layer and the low BFG- O_2 chemistry. The Damköhler number is defined as the ratio between a mixing time $\tau_t = U_c/e$, where e is the width of the burner lip [17, 18], and a chemical time τ_{ch} , obtained from the scalar dissipation rate χ_q [19]:

$$\tau_{ch} = \frac{1}{\chi_q} = \frac{D}{Z_s^2 \cdot S_L^2 \cdot (1 - Z_s)^2}$$

In the present work, the value of the critical Damköhler number at 300 K is adopted and used to design the tri-coaxial burner. First, the chemical time is determined from thermochemical calculations of the diffusivity D , the stoichiometric mixture fraction Z_s and the laminar burning velocity S_L . Then, the value of the critical Damköhler number obtained previously is used to deduce the maximum of convection velocity U_c . Finally, the inner and outer diameters of the central (O_{2i}) and co-annular injections (BFG and O_{2e}) are chosen so that the convection velocities calculated in the inner (O_{2i} -BFG) and outer (BFG- O_{2e}) mixing layers never exceed the critical velocity U_c . Figure 10 presents the evolution of the critical velocity U_c obtained from this methodology when varying reactant initial preheat temperature T_{pr} . The exponential increase in U_c points out that the reactant preheating will favors undoubtedly the stabilization of BFG- O_2 turbulent diffusion flames in the inner and outer shear layers.

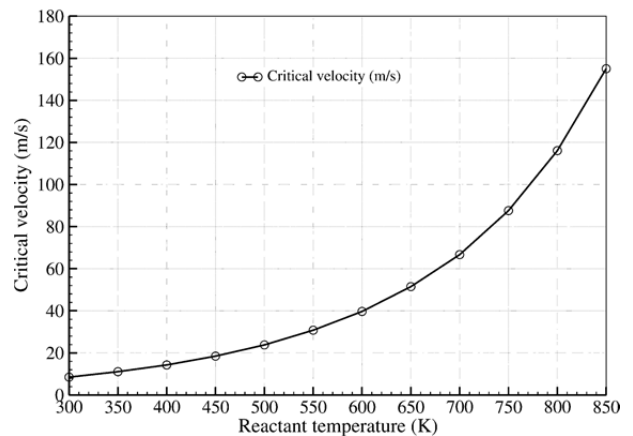


Figure 10. Evolution with the reactant temperature of the critical value of the convection velocity U_c derived from the Dahmköhler number.

The tri-coaxial burner thus designed will be tested in a lab-scale facility. The stability diagram will be built by varying oxygen and BFG preheating temperatures as well as central and peripheral oxygen repartition. The

structures of the turbulent oxyfuel BFG flames will be studied in order to derive guidelines for the design of oxyfuel burner adapted to LCV fuels.

Conclusions

In this work, the potential of the association of reactant preheating and oxyfuel combustion to enhance flame stabilisation of LCV fuels is shown from thermodynamic and kinetic calculations of flame properties. These calculations are performed from freely propagating mono-dimensional premixed laminar flames and counter-flow diffusion flames. The four kinetic mechanisms tested show similar results on the evolutions of adiabatic temperature and laminar burning velocity with the initial preheating temperature.

The study is focused on a typical composition of blast furnace gas (BFG), which main characteristics are the large proportion of CO in its composition and a net calorific value which is one order of magnitude lower than methane. Results show that the preheating of BFG and oxygen enhance significantly the adiabatic temperature, the laminar burning velocity and the extinction strain rate, whereas the flame thickness remains constant. This can be considered as equivalent to the addition of a HCV fuel to ensure flame stabilisation.

The whole of the kinetic calculations are used to develop the design of a tri-coaxial oxyfuel burner of BFG that is planned to be tested on a lab-scale facility.

Acknowledgements

This study is performed in the framework of the 'CALOHRY' project joining Air Liquide, CMI GreenLine and CORIA laboratory, with the financial support of ANR (ANR-12-SEED-0010-01).

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