ANALYSIS OF COMMON REACTIONS OF A DEVELOPED DIESEL-BIODIESEL COMBINED CHEMICAL SCHEME

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Abstract: *Diesel is a complex real fuel, which is derived from conventional petroleum sources, and is composed of hundreds of compounds. Although the composition of petroleum-based diesel fuel is highly variable, there are some trends: The carbon numbers of the components range from approximately* $C_{10} - C_{22}$*. An average carbon number is 14 or 15. These characteristics indicate the difficulty of using diesel for experimental studies associated with numerical simulations. In order to avoid these difficulties, simplified synthetic fuels, called "surrogate fuels", with shorter chain lengths and known physical chemical properties are chosen to carry combustion studies. On the other hand, biodiesel is a complex mixture of methyl esters with different chain lengths and degrees of unsaturation and can be used in pure form or may be blended with diesel without major modifications in diesel engines. However, changing the fuel in diesel engines induces changes in combustion behavior, which can impact pollutants emission. This paper presents numerical studies of diesel-biodiesel blends combustion in a 1D freely propagating gaseous premixed flame. A new chemical scheme was developed and validated by carefully combining two chemical schemes: one for diesel and one for biodiesel. This guarantees reproducing the principal features of both fuels combustion characteristics. The new combined scheme, consists of* 235 *species and* 1113 *elementary reactions. The new combined model was validated comparing laminar flame velocities of pure diesel and pure biodiesel using the combined model and the original models.*

Keywords: *Diesel, Biodiesel, Kinetic modelling, Freely propagating flames*

1. INTRODUCTION

Diesel is a complex real fuel, which is derived from conventional petroleum sources, and is composed of hundreds of compounds. Although the composition of petroleum-based diesel fuel is highly variable, there are some trends (Farrell *et al.*, 2007): The carbon numbers of the components range from approximately $C_{10} - C_{22}$. An average carbon number is 14 or 15. These characteristics indicate the difficulty of using diesel for experimental studies associated with numerical simulations. In effect, due to its complex and varying composition, the number of possible reaction pathways in a chemical reaction mechanism increases drastically. Development of models that represent all these components is prohibitive because the model would be too large for current computational resources.

In order to avoid these difficulties, simplified synthetic fuels, called "surrogate fuels", with shorter chain lengths and known physical chemical properties are chosen to carry combustion studies. The term surrogate refers to a simpler representation of a real complex fuel.

The purpose of using surrogates is to simplify the combustion mechanism by using a single fuel molecule or a blend of relatively small molecules to represent the real fuel. Numerically, the use of surrogate fuels reduces significantly the number of possible chemical reactions in the kinetic scheme, while still representing the main properties of the real fuel.

On the other hand, biodiesel is a complex mixture of methyl esters with different chain lengths and degrees of unsaturation and can be used in pure form or may be blended with diesel without major modifications in diesel engines. However, changing the fuel in diesel engines induces changes in combustion behavior, which can impact pollutants emission. Indeed, the use of biodiesel in diesel engines decreases emissions of pollutants such as carbon monoxide, unburned hydrocarbons, and particulate matter, although a slight increase in emissions of nitrogen oxides is observed in some cases (Agarwal, 2007; Zheng *et al.*, 2008). Therefore, a better understanding of diesel-biodiesel blend combustion is suitable.

Chemical kinetic mechanisms for hydrocarbon fuels have been the focus of intense research for several decades. However, there is little information on the combustion properties and pathways of common liquid fuels. This is due in part to the difficulty in representing real fuels containing hundreds of components with a wide variation in composition by surrogate fuels for the purpose of chemical kinetic modelling. Nevertheless, due to the increasing number of researchers working in the development of either kinetic modelling or experimental database of this fuel, this field has experienced a significant amount of recent developments.

This paper presents numerical studies of diesel-biodiesel blends combustion in a 1D freely propagating gaseous premixed flame. A new chemical scheme was developed and validated by carefully combining the schemes proposed by Andrae *et al.* (2011) (diesel) and Zhayou Luo *et al.* (2012) (biodiesel). This guarantees reproducing the principal features of both fuels combustion characteristics. The new combined scheme, consists of 235 species and 1113 elementary reactions. The common reactions in both chemical schemes were identified and those with different Arrhenius constants were analysed in order to obtain reasonable flame velocity results using both fuels separately.

The kinetic modelling for diesel-biodiesel blend oxidation in a 1D freely propagating gaseous premixed flame was performed using the REGATH package developed at EM2C laboratory (N. Darabiha *et al.*, 1993; Candel *et al.*, 2011; Franzelli *et al.*, 2013), that takes into account the detailed kinetic and transport phenomena (heat and mass transfer) through a numerical predictive 1D code.

The new combined model was validated comparing laminar flame velocities of pure diesel and pure biodiesel using the combined model and the original models. And then laminar flame velocities for different diesel-biodiesel blends were obtained as a function of equivalence ratio.

2. DIESEL AND BIODIESEL CHEMICAL STRUCTURE

Diesel is a complex combination of hydrocarbons produced by the distilation of crude oil. It consists of hydrocarbons having carbon numbers predominantly in the range of $C_{10} - C_{20}$. The composition varies widely, depending on the crude oils used, the refinery processes available, the overall balance of product demand, and the product specifications. The typical composition of diesel shows approximately, by volume, 66% of saturated hydrocarbons, 30% aromatics (benzene derivatives) and 4% olefins (unsaturated hydrocarbons).

Due to the complex chemical structure of diesel, simplified synthetic fuels, called surrogate fuels, with shorter chain lengths are chosen to carry numerical combustion studies.

There has been much recent progress in the area of surrogate fuels for diesel. In the last few years, experiments and modeling have been performed on higher molecular weight components of relevance to diesel fuel. Chemical kinetic models have been developed for all the n-alkanes up to 16 carbon atoms. Also, there has been experimental and modeling work on lower molecular weight surrogate components such as n-decane and n-dodecane that are most relevant to jet fuel surrogates, but are also relevant to diesel surrogates where simulation of the full boiling point range is desired (W. Pitz *et al.*, 2011).

According to a recent paper (Hernandez *et al.*, 2014), a mixture of toluene and n-heptane was the best diesel surrogate in comparison to other species surrogates. Luo *et al.* (2012) have shown that the ignition delay and smoke emission of TRF20 (80% n-heptane/20% toluene in volume) were closer to that of diesel fuel as compared with the pure n-heptane and TRF30 (70% n-heptane/30% toluene in volume) fuel.

Following these works, in this paper TRF20 (80% n-heptane/20% toluene in volume) have been chosen as a surrogate of the commercial diesel. Chemical structures of these surrogates are presented in Figure 1.

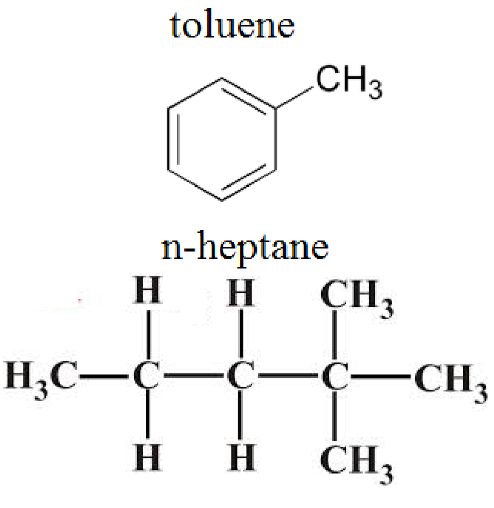

Figura 1: Diesel surrogates chemical structures

Biodiesel is a complex mixture of methyl esters with different chain lengths and degrees of unsaturation. Rapeseed-derived biodiesel (most produced around the world) concentrations and mole fractions has been estimated (D. Alviso *et al.*, 2015) and is presented in Table 1. In comparison to the literature values (Herbinet *et al.*, 2008), it corresponds to the classical composition of biodiesel obtained from this oil.

The chemical structure of each methyl ester is presented in Figure 2. The structures of these components show considerable similarities in these chemical species, each with a methyl ester attached to a large hydrocarbon chain. Main differences are:

Rapeseed biodiesel composition		
Methyl ester	CC (ppm)	Mole fraction
Palmitate	417	4.73
Stearate	157	1.78
Oleate	5660	64.21
Linoleate	1866	21.16
Linolenate	715	8.11

Tabela 1: Composition of rapeseed-derived biodiesel.

- the length of the hydrocarbon chain; 15 atoms of carbon for methyl palmitate and 17 for the other methyl esters,
- the number of double bonds in the hydrocarbon chain; no double bond for methyl palmitate and methyl stearate, one double bond for methyl oleate, two for methyl linoleate and three for methyl linolenate.

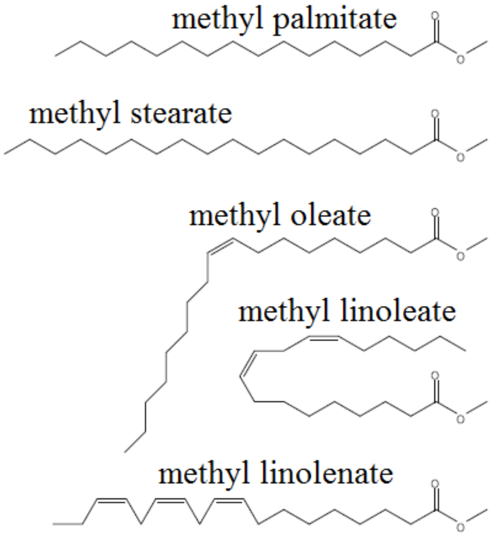

Figura 2: Structures of the main components of rapeseed-derived biodiesel

Due to the complex chemical structure of biodiesel, surrogate fuels are also chosen to carry numerical combustion studies.

In this sense, Seshadri *et al.* (2009) proposed methyl decanoate $C_{11}H_{22}O_2$ (MD) as a possible surrogate. The chemical structure of methyl decanoate MD is presented in Figure 3. More recently, Z. Luo *et al.* (2012) proposed a tri-component biodiesel surrogate, which consists of methyl decanoate $C_{11}H_{22}O_2$ (MD) , methyl 9-decenoate $C_{11}H_{20}O_2$ (MD9D) and n-heptane C_7H_{16} (chemical structures presented in Figure 3). Methyl-9-decenoate was chosen because the double bond is at the same position as the one in methyl oleate and at the same location as the first double bond in methyl linoleate and in methyl linolenate (Figure 2).

In this work, methyl decanoate $C_{11}H_{22}O_2$ (MD) and methyl 9-decenoate $C_{11}H_{20}O_2$ (MD9D) were the surrogates chosen to represent biodiesel combustion. Methyl palmitate and stearate (without double bonds) will be represented by MD. And methyl oleate, linoleate and linolenate (with one or more double bonds) will be represented by MD9D.

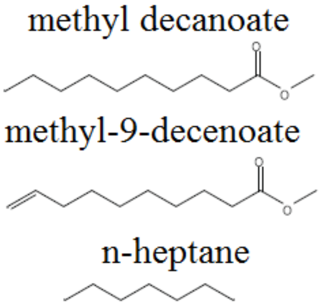

Figura 3: Biodiesel surrogates chemical structure

3. NUMERICAL APPROACH

3.1 Freely propagating premixed flames governing equations

A steady, isobaric, quasi-one-dimensional flame propagation configuration is considered. A diesel-biodiesel/air mixture is injected from one side, and a premixed flame propagates freely towards fresh gases with the corresponding laminar flame velocity. In order to get a stabilized flame (instead of a propagating flame) one must inject fresh gases with a velocity equal to the laminar flame velocity.

Gaseous phase are described by the following balance equations (mass, energy and species, respectively). It must be noted that there is one species conservation equation for each species present in the numerical model (N_{sp}).

$$\frac{\partial \rho_g v_g}{\partial z} = 0, \tag{1}$$

$$\rho_g v_g c_{p_g} \frac{\partial T_g}{\partial z} = \frac{\partial}{\partial z}\left(\lambda_g \frac{\partial T_g}{\partial z}\right) - \sum_{k=1}^{K} h_k W_k \Omega_k - \left(\sum_{k=1}^{K} \rho_g Y_k V_{kz} c_{p_{gk}}\right) \frac{\partial T_g}{\partial z}, \tag{2}$$

$$\rho_g v_g \frac{\partial Y_k}{\partial z} = -\frac{\partial}{\partial z}\left(\rho_g Y_k V_{kz}\right) + W_k \Omega_k, \qquad k = 1, ...N_{sp} \tag{3}$$

The above system of equations is completed by the ideal gas equation.

The solutions have the form: gas density $\rho_g = \rho_g(z)$, gas axial velocity $v_g = v_g(z)$, gas temperature $T_g = T_g(z)$ and species mass fractions $Y_k = Y_k(z)$, $k = 1, ...N_{sp}$ (N_{sp} is the number of species).
In these equations $c_{p_{gk}}$ and c_{p_g} are the heat capacity at local constant pressure of species k and of the mixture respectively. h_k, W_k, Ω_k are the specific enthalpy, the molar weight and the molar chemical production rate of the k^{th} species respectively, and V_{kz} is the diffusion velocity of the k^{th} species in the axial direction.

3.1.1 Numerical conditions

The kinetic modeling for diesel-biodiesel blends oxidation in the 1D freely propagating flame was performed using the 1D-PREMIXED code within the REGATH package (N. Darabiha *et al.*, 1993; Candel *et al.*, 2011; Franzelli *et al.*, 2013) with detailed thermochemical and transport properties developed at EM2C laboratory. The inputs to each simulation include a chemical kinetic reaction mechanism, a dataset of thermochemical properties and a dataset of transport properties.

Diesel-biodiesel/air freely propagating premixed flames operating condition have been chosen, considering the equivalence ratio (ϕ): from $\phi = 0.6$ to $\phi = 1.4$. The mixture stream is kept at an initial temperature of 300 K. The pressure is equal to one atmosphere.

4. KINETIC MODELING

As explained in Section 2., TRF20 (80% n-heptane/20% toluene in volume) have been chosen as a surrogate of the commercial diesel. The mechanism due to Andrae *et al.* (2011) consisting of 150 species and 759 reactions was chosen to represent diesel chemistry. This model consists of a semi-detailed description of toluene oxidation and skeletal mechanisms of iso-octane and n-heptane.

On the other hand, methyl decanoate $C_{11}H_{22}O_2$ (MD) and methyl 9-decenoate $C_{11}H_{20}O_2$ (MD9D) were the surrogates chosen to represent biodiesel combustion. The mechanism due to Zhayou Luo *et al.* (2012) with 115 species and 460 reactions for a tri-component biodiesel surrogate: methyl decanoate (MD), methyl 9-decenoate (MD9D) and n-heptane, was chosen to represent biodiesel chemistry.

In order to be able to simulate diesel-biodiesel/air flames, species representing these two fuels must be present in a single model. The kinetic model proposed here is designed from the original diesel oxidation framework of Andrae *et al.* (2011). This latter chemical scheme is used as a starting base model. Then additional species found in kinetic scheme of Zhayou Luo *et al.* (2012) (biodiesel) and the corresponding reactions are added. For the common species (which are 30), reactions present in Zhayou Luo *et al.* (2012) model and not present in Andrae *et al.* (2011) model are also added. Thus, the new combined scheme, consists of 235 species and 1113 elementary reactions. This guarantees reproducing the principal features of both fuels combustion characteristics.

In addition, the common reactions (which are 105) in both chemical schemes were identified and analysed in order to obtain reasonable flame velocity results using both fuels separately. In order to identify the sensitive reactions for the estimation of diesel and biodiesel flame velocities, 5 packages of 21 common reactions each were made, as explained below:

- Package I: reactions 1-21
- Package II: reactions 22-42
- Package III: reactions 43-63

- Package IV: reactions 64-84
- Package V: reactions 85-105

Then, 7 chemical schemes were developed as follows (considering the common reactions):

- Scheme A: Package I-V from diesel scheme
- Scheme B: Package I-V from biodiesel scheme
- Scheme C: Package I from biodiesel scheme and II-V from Diesel scheme
- Scheme D: Package II from biodiesel scheme and I, III-V from Diesel scheme
- Scheme E: Package III from biodiesel scheme and I, II, IV, V from Diesel scheme
- Scheme F: Package IV from biodiesel scheme and I-III, V from Diesel scheme
- Scheme G: Package V from biodiesel scheme and I-IV from Diesel scheme

It must be pointed out that the rest of reactions and Arrhenius constants are the same for all 7 Schemes.

Then, the flame velocities of Diesel and Biodiesel separately using schemes A to G were estimated, in order to identify the reaction package which is important for diesel and biodiesel flame velocity.

5. RESULTS AND DISCUSSION

As the laminar burning velocity provides invaluable information on the combustion properties, it was first used the kinetic scheme proposed by Andrae *et al.* (2011) and Zhayou Luo *et al.* (2012) to simulate 1-D freely propagating pure diesel and pure biodiesel (respectively) premixed flames using 1D-PREMIXED code within the REGATH package.

Pure diesel flame velocities estimated as a function of equivalence ratio are presented in Figure 4, for the original scheme of Andrae in black line and the combined model Schemes A and B (all common reactions from Andrae and Zhayou Luo, respectively) in colour lines. This figure shows that the combined scheme A reproduces fairly well the results of Andrae *et al.* (2011) for equivalence ratios higher than 1. The scheme B seems to be slightly better than scheme A for equivalence ratios lower than 1, although the difference is very small.

Pure biodiesel flame velocities estimated as a function of equivalence ratio are presented in Figure 5, for the original scheme of Zhayou Luo in black line and the combined model Schemes A and B in colour lines. This figure shows that the combined scheme A gives better results than scheme B, compared to those of the original scheme of Zhayou Luo *et al.* (2012). This is quite remarkable, as scheme B has all the common reactions from Zhayou Luo, and one would think that it would give better results than scheme A. For scheme A, in the very lean zone (from $\phi = 0.6 - 0.8$) the results are very good, then, as equivalence ratio increases, the difference between scheme A and Zhayou Luo results also increases.

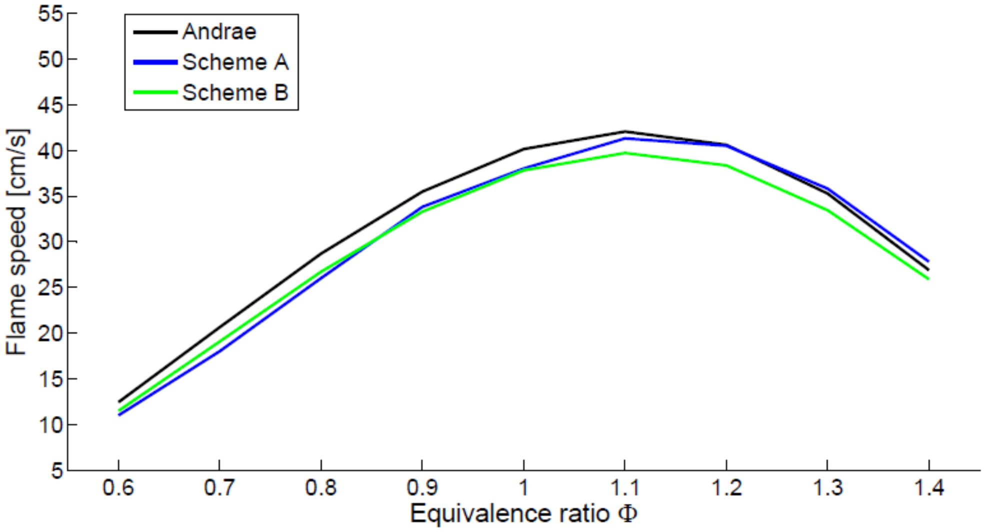

Figura 4: Diesel flame velocity as a function of equivalence ratio using the original scheme of Andrae *et al.* (2011) in black line and the combined model Schemes A and B in colour lines

Then,the combined kinetic schemes C to G were used, to simulate 1-D freely propagating flames for pure diesel and pure biodiesel.

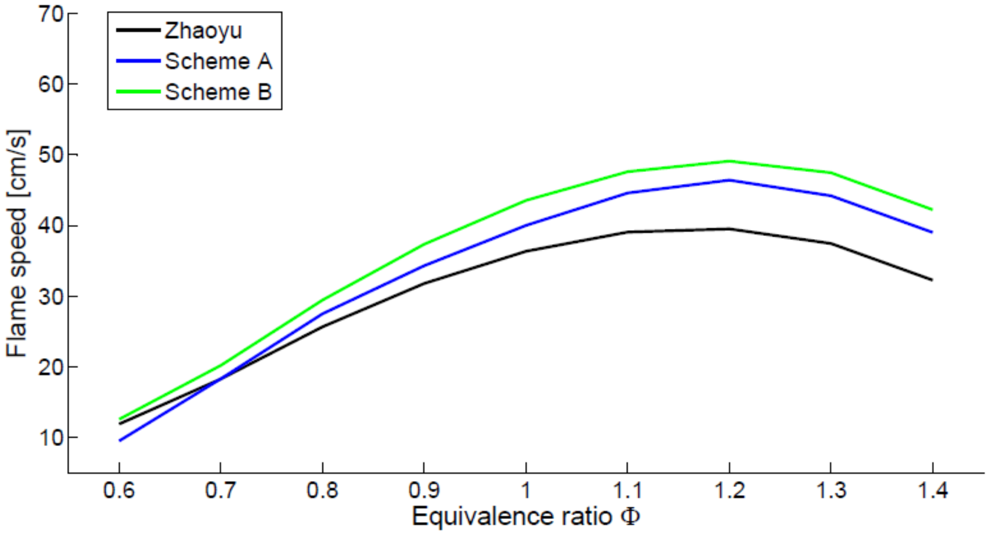

Figura 5: Biodiesel flame velocity as a function of equivalence ratio using the original scheme of Zhayou Luo *et al.* (2012) in black line and the combined model Schemes A and B in colour lines

First it was compared the flame velocities of pure diesel using the combined schemes C to G and that of Andrae *et al.* (2011). Flame velocities estimated as a function of equivalence ratio are presented in Figure 6. Results using Andrae *et al.* (2011) model are presented in black line, whereas those of the combined models C to G are presented in colour lines.

Now, in order to quantify how much close is one scheme results to the original scheme results, standard deviations of schemes A to G were estimated. The results are given in Table 5.

Tabela 2: Standard deviation of Schemes A to G compared to the original schemes of Andrae and Zhayou Luo simulating the combustion of pure diesel and pure biodiesel, respectively

	Standard deviation						
Original scheme	Scheme A	Scheme B	Scheme C	Scheme D	Scheme E	Scheme F	Scheme G
Andrae	0.55	0.63	0.29	1.43	3.49	1.25	0.54
Zhayou Luo	1.57	2.40	1.55	4.42	5.51	1.47	1.70

Figure 6 and Table 5show that for all equivalence ratios, the combined scheme C is the one that reproduces the better the results of Andrae *et al.* (2011). Scheme G also gives very good results, slightly better than Scheme A, which has all the common reactions from Andrae.

Then, there were compared the flame velocities of pure biodiesel using the combined schemes C to G and that of Zhayou Luo *et al.* (2012). Flame velocities estimated as a function of equivalence ratio are presented in Figure 7. Results using Zhayou Luo *et al.* (2012) model are presented in black line, whereas those of the combined models C to G are presented in colour lines. This figure and Table 5show that for all equivalence ratios, the combined scheme F is the one that reproduces the better the results of Zhayou Luo *et al.* (2012). Scheme C also gives very good results, slightly better than Scheme A.

Therefore, taking into account schemes C to G in the estimation of both diesel and biodiesel flame velocity results, schemes C and G are the ones that have reasonable results for both fuels. In the other hand, scheme F gives good results for biodiesel, but bad results for diesel. Consequenly, schemes C and G were combined into one scheme: Packages I and V from biodiesel scheme and II-IV from Diesel scheme. However, the results of this new combined scheme is not better than Scheme C results. In conclusion, scheme C (Package I from biodiesel scheme and II-V from Diesel scheme) is the one that gives the best results using both fuels separately.

As one can see the results of the developed combined model are much better for diesel, compared to biodiesel (see Table 5). And taking into account that in a blend, diesel is usually the majority component, this is quite convenient.

Diesel-biodiesel blends flame velocities as a function of equivalence ratio using the combined model C are presented in Figure 8. The blend correspond in volume to B20 (20% biodiesel, 80% diesel). B20 is the most common blend used around the world. Agarwal and Das 2001 found out that B20 is the optimum biodiesel blend giving maximum increase in thermal effciency, lowest brake-specific energy consumption (BSEC) and advantage in terms of lower emissions.

In Figure 8 one can see that the influence of adding biodiesel is not negligible, concerning the blend flame velocity. In fact, for equivalence ratios higher than 1, $B20$ flame velocities curve is located in average, at 20% from $B0$, and 80% from $B100$, whereas for equivalence ratios lower than 1, flame velocities of $B0$ and $B100$ are very similar, and consequently $B20$ flame velocities are very near from $B0$ and $B100$ flame velocities.

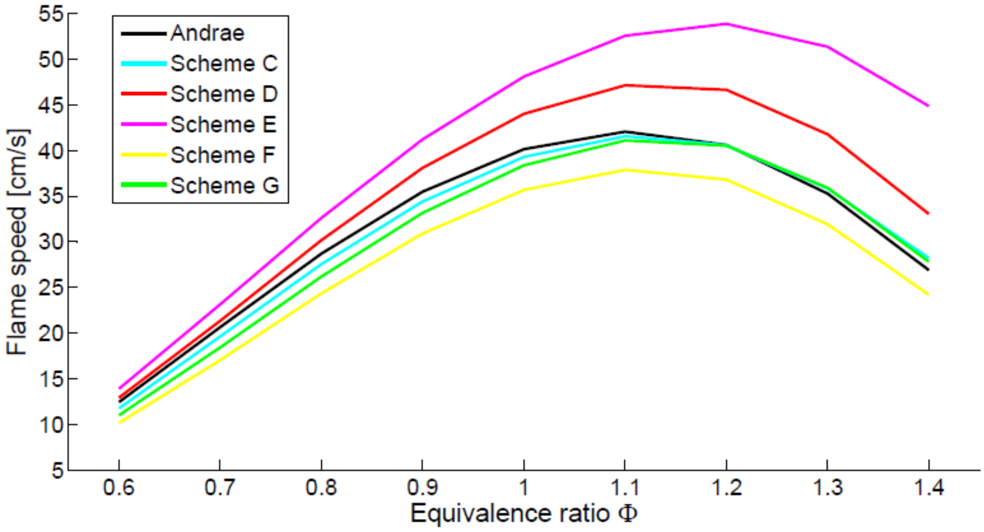

Figura 6: Diesel (Andrae) and biodiesel (Zhayou) flame velocity as a function of equivalence ratio

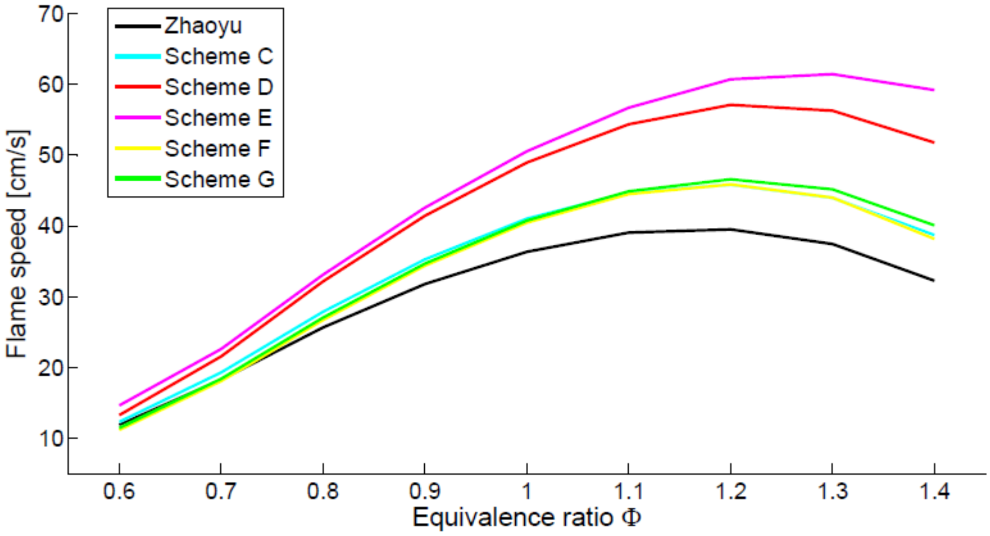

Figura 7: Diesel (Andrae) and biodiesel (Zhayou) flame velocity as a function of equivalence ratio

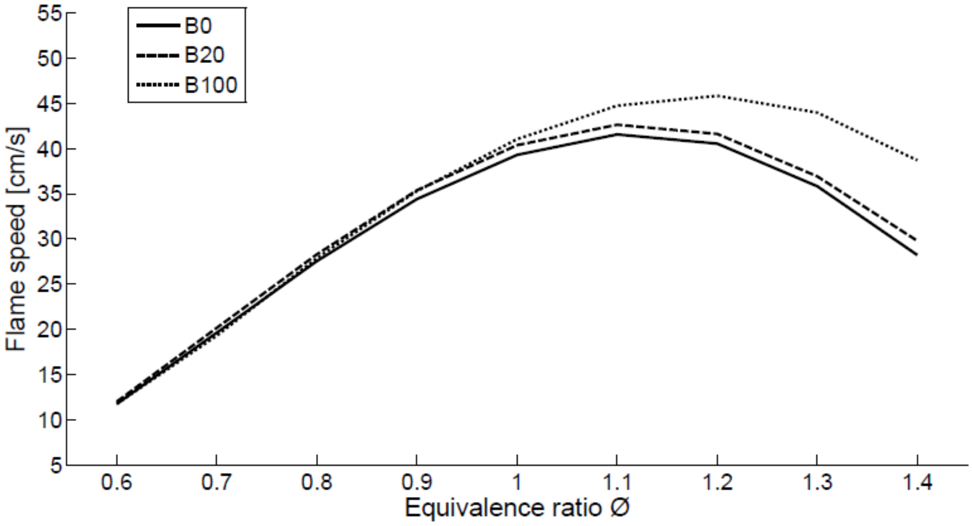

Figura 8: Diesel - biodiesel blends flame velocity as a function of equivalence ratio

6. CONCLUSIONS

This paper has presented numerical studies of diesel-biodiesel blends combustion in a 1D freely propagating gaseous premixed flame configuration.

A new chemical scheme was developed and validated by carefully combining the schemes proposed by Andrae *et al.* (2011) (diesel) and Zhayou Luo *et al.* (2012) (biodiesel). This guarantees reproducing the principal features of both fuels combustion characteristics. The new combined scheme, consists of 235 species and 1113 elementary reactions.

In addition, the common reactions (which are 105) in both chemical schemes were identified and analysed in order to obtain reasonable flame velocity results using both fuels separately. In order to identify the sensitive reactions for the estimation of diesel and biodiesel flame velocities, 5 packages of 21 common reactions each were made. Then, 7 chemical schemes called A to G were developed using these packages.

The kinetic scheme proposed by Andrae *et al.* (2011) and Zhayou Luo *et al.* (2012) were first used to simulate 1-D freely propagating pure diesel and pure biodiesel (respectively) premixed flames. Then, the flame velocities of pure diesel and pure biodiesel using the combined schemes A to G and those of Andrae *et al.* (2011) and Zhayou Luo *et al.* (2012) were compared, respectively.

The combined scheme C is the one that reproduces the better the results of Andrae, whereas scheme G also gives very good results. In the other hand, the combined scheme F is the one that reproduces the better the results of Zhayou Luo, whereas scheme C also gives good results.

Therefore, taking into account the estimation of both diesel and biodiesel flame velocity results, schemes C and G are the ones that have reasonable results for both fuels. Scheme F gives good results for biodiesel, but bad results for diesel. Consequenly, schemes C and G were combined into one new scheme. However, the results of this new combined scheme is not better than Scheme C results. In conclusion, scheme C is the one that gives the best results using both fuels separately.

The results of the developed combined model are much better for diesel, compared to biodiesel. And taking into account that in a blend, diesel is usually the majority component, this is quite convenient.

Diesel-biodiesel blend B20 (20% biodiesel, 80% diesel) flame velocities as a function of equivalence ratio using the combined model C were also presented.

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