

Influence of high ethane content on natural gas ignition

Influencia del alto contenido de etano en la ignición del gas natural

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Como citar: H. Yepes y A. Cardona, "Influence of high ethane content on natural gas ignition", *Revista Ingenio*, vol. 16, n°1, pp.36-42, 2019, doi: <https://doi.org/10.22463/2011642x.2384>.

Received date: 01 de agosto de 2018

Approval date: 16 de noviembre de 2018

ABSTRACT

Keywords:

Ethane, Ignition delay time, Ignition regime, Natural gas.

The effect of ethane on combustion and natural gas autoignition was studied in the present paper. Two fuel mixture of natural gas with high ethane content were considered, 75% CH₄ – 25% C₂H₆ (mixture 1), and 50% CH₄ – 50% C₂H₆ (mixture 2). Natural gas combustion incidence was analyzed through the calculation of energy properties and the ignition delay time numerical calculations along with an ignition mode analysis. Specifically, the strong ignition limit was calculated to determine the effect of ethane on natural gas autoignition. According to the results, ignition delay time decreases for both mixtures in comparison with pure methane. The strong ignition limit shifts to lower temperatures when ethane is present in natural gas chemical composition.

RESUMEN:

Palabras claves:

Etano, Tiempo de retraso a la ignición, Régimen de Ignición, Gas Natural.

En el presente artículo se estudió el efecto del etano sobre la combustión y la autoignición del gas natural. Se evaluaron dos mezclas con alto contenido de etano, 75% CH₄ – 25% C₂H₆ (mezcla 1) y 50% CH₄ – 50% C₂H₆ (mezcla 2). La incidencia en la combustión del gas natural fue analizada mediante el cálculo de las propiedades energéticas del combustible y los cálculos numéricos del tiempo de retraso a la ignición junto con el análisis del modo de ignición. Específicamente el límite de ignición fuerte fue calculado para determinar el efecto del etano en la autoignición del gas natural. De acuerdo con los resultados encontrados, el tiempo de retraso a la ignición disminuye para ambas mezclas estudiadas en comparación del metano puro. El límite de ignición fuerte se mueve hacia temperaturas menores cuando hay etano en la composición química del gas natural.

1. Introduction

The augmentation of natural gas consumption between 2008 and 2016 was 16.36%, exceeding widely the oil and coal growth that was 9.87% and 5.77%, respectively. The different issues of alternative energies in some countries also contributes to this behavior [1-2].

QAlthough in many cases, the composition of natural gas is principally CH₄, the extraction from the different wells produces that the chemical composition varies considerably and the amount of higher hydrocarbons like C₂H₆ and C₃H₈ takes important values. The volume fraction of these fuels in the natural gas composition can varies between 0.5% - 13.3% and 0 - 23.7%, respectively [3]. In other cases, the variability is less, but still relevant to the amount of C₂H₆ varies from 4.8% to 14.33% in volume, and C₃H₈ achieves values of 6.24% [4]. By this variability, in most cases, the liquefied natural gas typically contains a higher content of C₂H₆ than traditional natural gas. In this way, higher hydrocarbons' content can reach 50% of the composition in some cases, being C₂H₆, the principal

component of these fuels [5-6].

According to the expose above, it is evident that C₂H₆ is one of the principal components of both conventional and unconventional natural gas. Therefore, it can be that most affect the combustion process.

The variations in the chemical composition of natural gas can impact the generation of emissions along with the performance in combustion devices due to the changes in the fuel properties and the chemical kinetics [7-8]. In this sense, the ignition delay is one of the fundamental properties in the performance of internal combustion engines and gas turbines; it is used to predict and avoid undesirable phenomena like knocking in engines and pre-ignition in turbines. It can also be used as a security factor in thermal device design [9-10]. Due to the importance of these properties, some studies about the effect of natural gas's chemical composition have been carried out in the last years [11-12]. However, the amount of C₂H₆ in natural gas is below 20% (in vol.) in these studies.

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Considering the aspects mentioned above, this paper addresses natural gas ignition with high C₂H₆ content in its chemical composition. A numerical study of the changes in ignition delay with the C₂H₆ volume fraction was carried out. Finally, the effect of the amount of C₂H₆ on the ignition type was established by means of the determination of the strong ignition limit at different conditions of pressure and temperature. No study was found in the literature that had covered such a thorough range of conditions, data type, mixture composition, and detailed kinetics modeling for methane/ethane blends, as presented in this paper.

2. Methane/Ethane Blends

The selected fuel mixture for natural gas established from the composition reported in the literature [13-14]. Table 1 shows the chemical composition of representative mixtures on a volume basis. M0 and M3 correspond to pure fuels, CH₄ and C₂H₆, respectively.

The calculations were carried out over a range from lean to rich conditions for all mixtures; the equivalence ratio was varied from 0.8 to 1.4 and was conducted at atmospheric conditions at 1550 m.a.s.l, namely a pressure of 0.84 atm and a temperature of 295 K for the case of adiabatic flame temperature (T_{ad}).

Table 1. Fuel mixture compositions.

Component (% by Volume)		M0	M1	M2	M3
Methane	CH ₄	100	75	50	0
Ethane	C ₂ H ₆	0	25	50	100

The ignition delay time (τ) and the ignition mode analysis were performed for all mixtures for stoichiometric conditions.

3. Numerical Methodology

Detailed chemical kinetic simulations were performed based on the combustion mechanisms of hydrocarbons. The numerical models used to calculate the energy properties, T_{ad} and the ignition delay time, were subroutines of EQUIL and SENKIN Software Chemkin 19.0 [15-16]. To improve the ignition delay time calculation, the maximum time step was fixed at the total simulation time divided by 100 and the time interval for saving data divided by 400.

3.1 Detailed Reaction Mechanism

The detailed kinetic reaction mechanisms were selected from the results exposed in other studies in predicting SL for methane and ethane separately. Two mechanisms were selected to perform the calculations, San Diego [17] mechanism with 235 reactions and 50 species and

GRI-Mech 3.0 [18] mechanism with 325 reactions and 53 species.

These mechanisms were selected to carry out the numerical calculations of ignition delay times due to the good results found in other studies. Furthermore, Diamantis et al. [19] conducted a kinetic study of the reactions supporting or opposing explosive modes for a homogenous mixture of methane/air using the GRI-Mech 3.0 mechanism [18]. Finally, Hu et al. [20] obtained that San Diego Mech [17] can give good agreement with experimental data for ignition delay times of ethane.

3.2 Strong Ignition Limit

In order to determine the effect of C₂H₆ on natural gas ignition mode, the strong ignition limit was determined for various conditions of pressure (1 – 42 atm) and temperature (850 – 1500K), using the Sankaran criteria in the form defined by Mansfield and Wooldridge [21]. The criteria allow distinguishing between strong ignition and mixed or weak ignition through comparing the thermal sensibility ($d\tau/dT$) and the inverse of the product between laminar burning velocity (SL) and the temperature gradient (dT/dx) using equation (1).

$$\frac{d\tau}{dT} < \left(\frac{dT}{dx} S_L \right)^{-1} \quad (1)$$

According to the criteria, if the inequality is correct for a specific condition of temperature and pressure, a strong ignition occurs; conversely, when the inequality is false, the mixed or weak ignition occurs.

To determine the thermal sensibility, two expressions were used, proposed by Zeng et al. [22] for the CH₄ (equation (2)) and proposed by Kuppa et al. [23] for CH₄/C₂H₆ mixtures, see equation (3).

$$\frac{d\tau}{dT} = \left| \tau \left(\frac{-169690}{RT^2} \right) \right| \quad (2)$$

$$\frac{d\tau}{dT} = \left| -\tau \cdot d(HC) \left(\frac{T_0}{T^2} \right) \right| \quad (3)$$

In both equations T is the temperature, whereas in equation (2), R is the gas constant and in equation (3) d is a regression constant, and HC is the C and H ratio in the fuel mixture, defined by Kuppa et al. as[23]:

$$HC = \frac{4X_{CH_4} + 6X_{C_2H_6} + 8X_{C_3H_8}}{X_{CH_4} + 2X_{C_2H_6} + 83} \quad (4)$$

Where X is the volume fraction of the component in the natural gas fuel mixture. The values of τ in equations (2) and (3) were calculated using Chemkin 19.0, as was mentioned above. According to the results of Mansfield and Wooldridge [21], the value of the thermal gradient was established in 5 K/mm. SL values were calculated by the mean of two methodologies, for pressures between 1 to 10 atm and temperatures from 850 to 1000 K, the premix routine was used according to the procedure described by Burbano et al. [24], and a correlation was used for the highest pressures and temperatures. The correlation form for all fuel is described in equation (5)

$$S_L = S_{L0} \left(\frac{T_U}{T_0} \right)^\alpha \left(\frac{P_U}{P_0} \right)^\beta \quad (5)$$

Where T_U and P_U are the temperature and pressure at unburned conditions, T_0 and P_0 are the temperature and pressure at reference conditions, and α y β are constants. The values for the constants α and β for CH_4 are 2.5 and -0.26, respectively [25-26]. In the case of CH_4/C_2H_6 mixtures, the methodology to calculate the constants proposed by Kuppa et al. [27] was applied. Reference conditions of 300 K and 1 atm were used for S_{L0} for all the studied fuels

4. Results and Discussion

4.1 Fuel Properties

Table 2 shows some combustion properties calculated in volumetric terms, for the methane, ethane, and methane-ethane blends. Among the fuel properties considered when qualifying a fuel for a combustion system are the stoichiometric air volume (V_a), the lower (LHV) and higher (HHV) heating values, and the Wobbe index (W_o). For the mixtures, W_o was calculated using equation (6).

$$W_o = \frac{\sum X_i HHV_i}{\sqrt{\sum X_i d_i}} \quad (6)$$

Where X_i is the volume fraction and d_i is the specific density of the i component. The addition of ethane to methane in 25% and 50% volumetric proportions increased the LHV by 20% and 40%, respectively. It increased the HHV by 19% and 38%, respectively, due to ethane's higher heat of reaction.

Despite the higher specific density of ethane, this fuel's addition increased the W_o due to the higher HHV. Similarly, when ethane was added to methane in 25% and 50% volumetric proportions, the Wobbe index

increased 8% and 15%, respectively.

Table 2. Energy volumetric properties.

Property	CH_4	M1	M2	C_2H_6
$V_a \left(\frac{m_{st}^3 \text{ air}}{m_{st}^3 \text{ fuel}} \right)$	9.52	11.31	13.09	16.66
LHV $\left(\frac{kWh}{m_{(st)}^3} \right)$	9.43	11.31	13.19	16.95
HHV $\left(\frac{kWh}{m_{(st)}^3} \right)$	10.49	12.50	14.52	18.55
$W_o \left(\frac{kWh}{m_{(st)}^3} \right)$	14.09	15.22	16.27	18.21

On the other hand, the T_{ad} for different equivalence ratios of M2, at ambient temperature and pressure, is shown in Figure 1. The adiabatic flame for all mixtures was simulated using GRI-Mech 3.0 and San Diego mechanisms. As can be seen in Figure 1, T_{ad} predictions by two mechanisms are very similar.

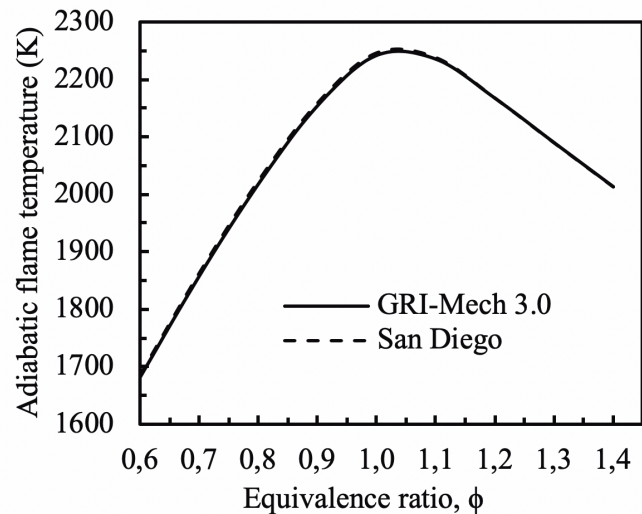


Figure 1. T_{ad} at $P_i = 0.84$ atm and $T_i = 295$ K.

The GRI-Mech 3.0 and San Diego mechanisms' differences do not exceed 1% for all equivalence ratios. The behavior is the same for all the gases of this study; for visualization reasons, the results of M2 are only shown.

The effect of ethane addition on the T_{ad} can be seen in Figure 2. At $\phi = 1$, the T_{ad} was 2218.69 K for methane, 2232.15 K for M1, 2241.16 K for M2, and 2253.28 K for ethane. Ethane had a higher temperature with respect to the other gases.

In general, the results show that the T_{ads} of the mixtures and pure gases were very comparable.

However, Figure 2 shows that the maximum T_{ad} for ethane was near $\phi = 1.1$, and the maximum T_{ad} for methane was near to $\phi = 1$. Simultaneously, mixtures 1 and 2 behaved more similarly to ethane, exhibiting the ethane's high reactivity with respect to methane. Higher T_{ad} values increase the burning velocity because the mixtures increased reactivity; this effect is analyzed in the following sections.

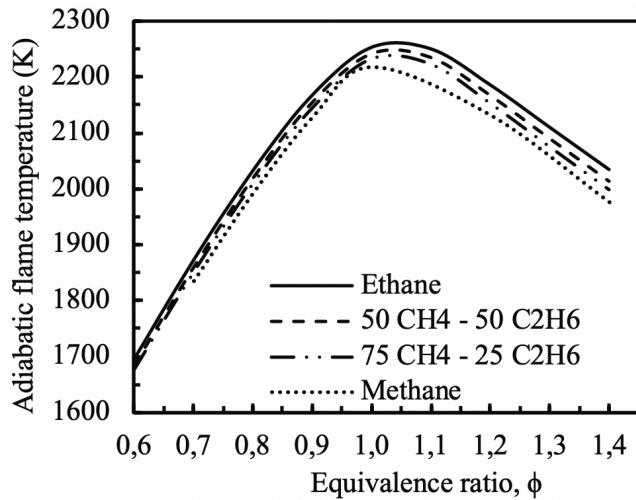


Figure 2. T_{ad} at $P_i = 0.84$ atm and $T_i = 295$ K

4.2 Ignition delay time

A database of ignition delay times was obtained for the studied mixtures, methane and ethane $\phi = 1.0$. The ignition delay time was estimated using the Chemkin 19.0 and GRI-Mech 3.0 mechanism; the results are shown in Figure 3 and Figure 4. The results showed that the ignition delay time is higher for the ethane than the two blends. An important result is observed when the ignition delay times for pressures of 1 atm and 5 atm are compared; in general, the ignition delay time at 5 atm has lower value respect to the one at 1 atm.

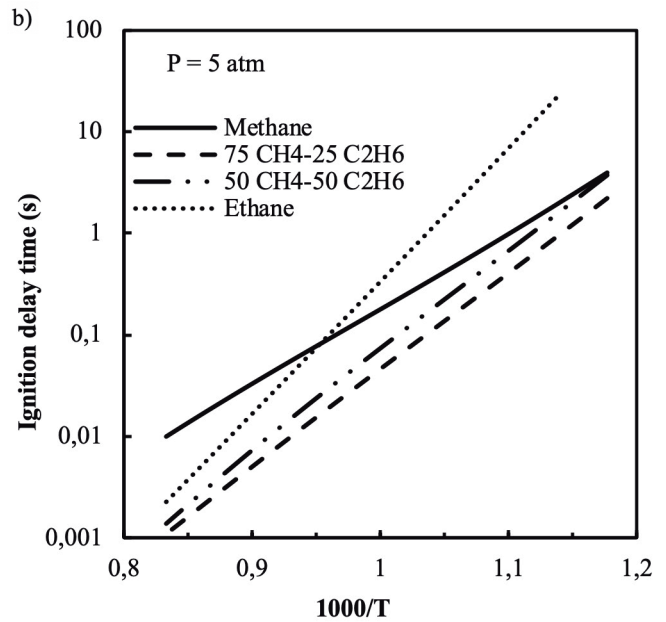
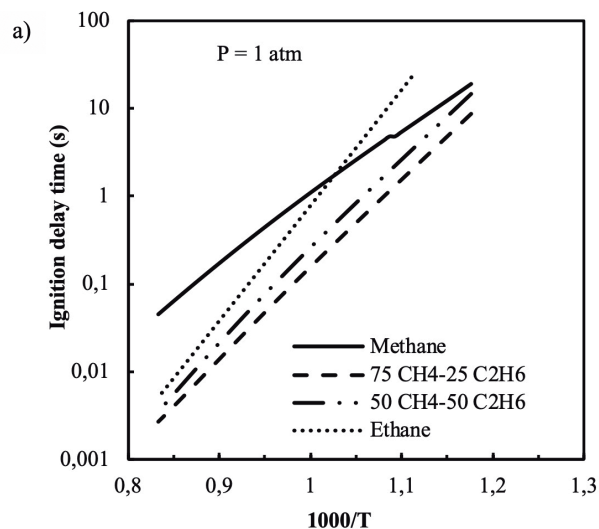
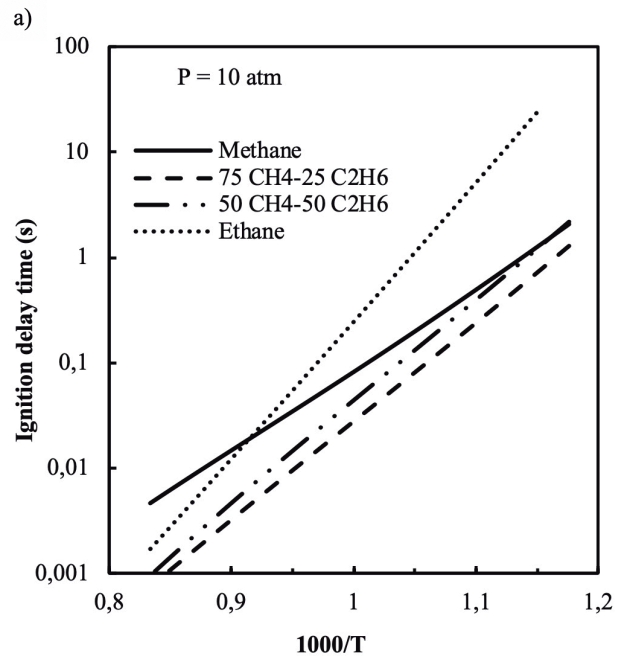


Figure 3. Ignition delay time all mixtures at $T_i = 295$ K, and $\phi = 1.0$. a) $P_i = 1$ atm, b) $P_i = 5$ atm.



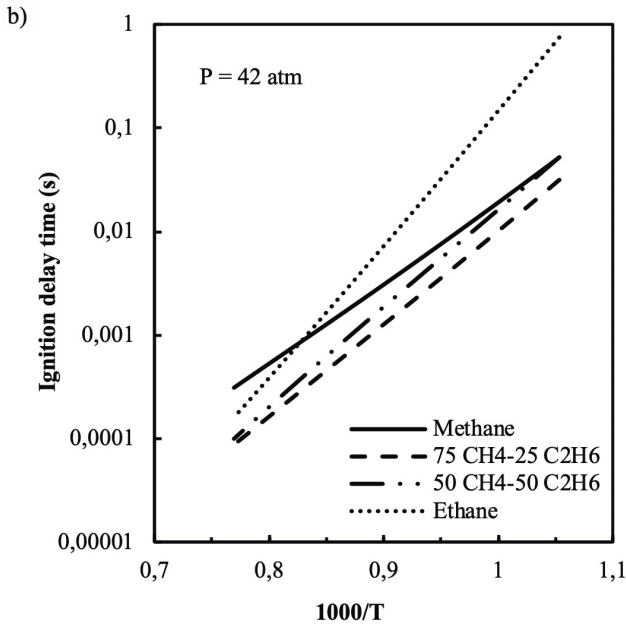


Figure 4. Ignition delay time all mixtures at $T_i = 295$ K, and $\phi = 1.0$. a) $P_i = 10$ atm, b) $P_i = 42$ atm.

As is expected, the ignition delay time decreases when C_2H_6 is added to natural gas composition due to the higher LHV of the mixture compared with pure CH_4 . The energy released during pre-ignition reactions increases with the amount of C_2H_6 in the mixture. It promotes the formation of radicals that accelerated the reaction, therefore the ignition of the fuel mixture.

However, the ignition delay time of pure C_2H_6 is higher than the mixtures, which suggest that the decrease in this parameter is not only by the thermal effect and the chemical kinetic have an important role in the behavior, especially at lower temperatures. The detailed kinetic analysis is beyond the scope of this work and is considered for future study.

4.3 Strong ignition limit

In order to establish the effect of the C_2H_6 content in the natural gas on the auto-ignition modes under different pressure and temperature conditions, the strong ignition limit was calculated. This limit separates the ignition modes in two groups, the strong ignition where behavior is considered spatially uniform as a detonation wave, and mixed or weak ignition where the ignition is an inhomogeneous process with localized reaction sites and a deflagration process takes place [21].

The methodology used to calculate the strong ignition limit was validated with the experimental data of Huang et al. [28] for the case of pure CH_4 . Figure

5 shows the experimental results (symbols) and the strong ignition limit (dashed line) calculated in the present work.

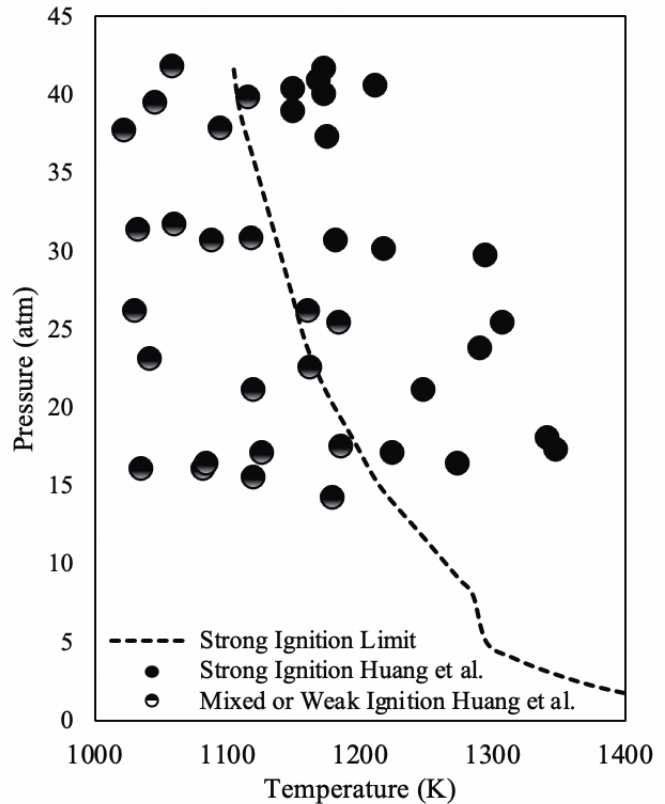


Figure 5. Ignition regimes for CH_4 . Experimental data are results from Huang et al. [28]

As can be seen, the calculated strong ignition limit is according to the experimental data and delimits in an adequate form of the division between the types of ignition. The adequate delimitation of the ignition regimes made by the calculated strong ignition limit, confirms the suitability of the adopted methodology applied to the fuels with C_2H_6 . Figure 6 shows the strong ignition limit for the CH_4 and mixtures 1 and 2. As can be expected, the strong ignition limit moves to lower temperature values as the pressure increases for all the studied fuels. This behavior is according to what is reported in the literature for other fuel mixtures [21-28]. As it was shown by Burrell et al. [29], SL decreases when pressure increases, and for high values, the reduction is more critical; this drop in SL generates that the local flame front of a hot spot travels more slowly to the unburned mixture, allowing that the unburned mixture starts the auto-ignition process.

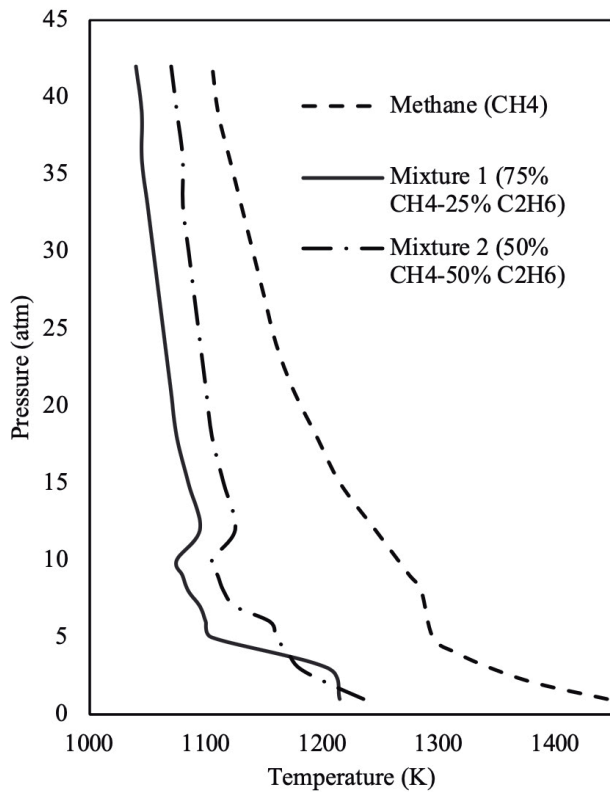


Figure 6. Strong ignition limit for all the fuel mixtures.

According to the results shown in Figure 6, the addition of C₂H₆ in the fuel mixture generates a shift in the strong ignition limit to lower temperatures; therefore, the strong ignition zone is broader in comparison with CH₄. This fact is associated with the increase in the Wobbe index when C₂H₆ is added; the energy per volume unit increases, which allows a higher energy release in the ignition process. The highest amount of released energy produces lower ignition delay times for the mixtures, as can be seen in the previous section, allowing a more homogeneous ignition.

However, it does not exist a monotonous behavior with the increase of C₂H₆ volume fraction in the fuel, since for M1 the strong ignition limit is achieved at lower temperatures with respect to M2, despite in the former the C₂H₆ volume fraction is half of the one for M2. This trend can be associated with two aspects. The first is due to C₂H₆ is a more complex molecule, and therefore when the concentration is higher, it takes more time to start with the ignition. The results of the ignition delay time in the previous section confirm that fact. The second aspect is related to the higher SL values when the C₂H₆ volume fraction is higher, which allows that deflagration takes place and can achieve the unburned mixture before it ignites, allowing an inhomogeneous ignition.

For relatively low pressures (less of 5 atm), the strong ignition limit of the mixtures tends to change, and the limit for M2 takes place at a lower temperature compared to M1.

5. Conclusion

Analysis of ethane content on combustion properties and auto-ignition of natural gas was performed by determining ignition delay and strong ignition limit for methane, ethane and two mixtures of natural gas. Numerical calculations of some properties were also performed with different reaction mechanisms. Analysis of the obtained results leads to the following conclusions:

- The higher content of C₂H₆ in the chemical composition of natural gas increases the Wobbe index due to the higher LHV of this fuel.
- Natural gas with high C₂H₆ content has a lower ignition delay time in comparison with pure CH₄. Therefore, it would be necessary to modify combustion devices when this type of fuel is used to reduce the damage probability.
- The presence of C₂H₆ in the chemical composition of natural gas has an important role in the types of ignition of the fuel mixture. The strong ignition limit shifts to lower temperatures when C₂H₆ is present due to the higher energy density in comparison to CH₄.

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