

Laminar burning velocities of primary reference fuels and simple alcohols

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ABSTRACT – Laminar burning velocities for methanol, ethanol, and binary and quaternary mixtures of these with iso-octane and n-heptane, have been determined using the heat flux method on a flat flame adiabatic burner. Measurements were done for an equivalence ratio range between 0.7 and 1.5 and for a range of temperatures between 298 K and 358 K at atmospheric pressure. The present study expands the available data on laminar burning velocities of alcohol-hydrocarbon blends and validates simple mixing rules for predicting the laminar burning velocity for a wider range of fuel blends of hydrocarbons with methanol and/or ethanol. It is shown that simple mixing rules that consider the energy fraction of the blend's components are accurate enough to predict the experimentally determined laminar burning velocity of the mixtures.

INTRODUCTION

The use of fossil fuels on a massive scale has led to an intolerable stress on the local (air quality) and global (greenhouse effect) environment. An important and increasing share in the energy consumption goes to the transportation sector. Alcohols, methanol and ethanol in particular, are promising alternatives for fossil fuel replacement. These liquid fuels are largely compatible with the current fuel and distribution infrastructure and are easily stored in a vehicle. They can be used in the current vehicle internal combustion engines with little or no changes. Methanol and ethanol are also miscible with gasoline which enables a soft start to an alternative transport energy economy and in contrast to other alternatives, they have the well-known potential to increase the engine performance and efficiency over what is achievable with gasoline [1, 2]. Much of the recent research efforts evaluating the potential of alcohols have focused on ethanol [3]. However, methanol is more versatile from a production point of view, its properties are even more beneficial for the power output and efficiency of internal combustion engines compared to ethanol and the addition of methanol to ethanol-gasoline blends can also have an economic advantage [4, 5].

To fully exploit the characteristics of alcohols for increased efficiency and performance, engine cycle simulation models, which have become indispensable tools in the design of advanced engines, should be adapted to the combustion characteristics of these alcohol fuels. A fundamental property of a fuel and a key parameter in simulation tools for the characterization of the combustion behavior is the laminar burning velocity (u_l). Accurate and fast calculation of the laminar burning velocity is needed in simulation tools. Sileghem et al. [6] investigated if mixing rules could be used to determine the laminar burning velocity of fuel blends from the burning velocity of the fuel components with sufficient accuracy and without being computationally too

demanding. In that work, different mixing rules for the prediction of the laminar burning velocity of ethanol-hydrocarbon mixtures were tested and compared. Three different mixing rules gave results in good agreement with the experimental determinations of the laminar burning velocities for mixtures. The mixing rules have been explained by Sileghem et al. [6], but are repeated here for the clarity of the present paper:

Mixing rule based on energy fraction of the fuels' components [7].

$$u_{l,blend}(\Phi) = \sum_{i=1}^n \alpha_i \cdot u_{l,i}(\Phi) \quad (1)$$

In this expression α_i is the energy fraction of fuel component i. The energy fraction can be calculated as follows:

$$\alpha_i = \frac{\Delta cH_i^\circ \cdot x_i}{\sum_{i=1}^n \Delta cH_i^\circ \cdot x_i} \quad (2)$$

cH_i° is the heat of combustion and x_i is the mole fraction of the fuel component i.

Mixing rule based on Le Chatelier's rule [6].

$$u_{l,blend}(\Phi) = \frac{1}{\sum_{i=1}^n \frac{\alpha_i}{u_{l,i}(\Phi)}} \quad (3)$$

where α_i is the energy fraction of the fuel component (Eq. (2)).

Le Chatelier [8] first proposed a similar empirical mixing rule for predicting the flammability limit of lean fuel-air mixtures:

$$LFL_{blend} = \frac{1}{\sum_{i=1}^n \frac{x_i}{LFL_i}} \quad (4)$$

where x_i is the mole fraction of the i^{th} component and LFL_i is the lower or lean flammable limit of the i^{th} component in volume percent. Di Sarli and Di Benedetto [9] used Le Chatelier's rule to predict the laminar burning velocity of hydrogen-methane blends. They used the mole fraction instead of the energy fraction. These two approaches have been compared by Sileghem et al. [6] and it was found that the energy fraction gave better results for the laminar burning velocity of ethanol-hydrocarbon mixtures.

Mixing rule developed by Hirasawa et al. [10].

Hirasawa et al. [10] developed an empirical mixing rule, which depends on a mole fraction weighted average of the burning velocities and flame temperatures.

The expression for laminar burning velocity becomes:

$$u_{l,\text{blend}} = \prod_{i=1}^n u_{l,i}^{\beta_i} \quad (5)$$

with

$$\beta_i = \frac{x_i n_i T_{f,i}}{n_{\text{blend}} T_{f,\text{blend}}} \quad (6)$$

x_i is the mole fraction and n is the total amount of moles of the combustion products and diluents, and T_f represents the adiabatic flame temperature. 'i' refers always to the i^{th} fuel component and 'blend' refers to the fully blended fuel. More detailed information can be found in [6, 10, 11].

These three mixing rules gave similar predictions for ethanol-hydrocarbon blends [6] indicating that the flame temperature is the dominant factor for the laminar burning velocity, as was found by Hirasawa et al.[10]. Sileghem et al. [6] concluded that the mixing rule by Le Chatelier, based on energy fraction, was the most useful to represent the data of their study since it is relatively

simple and give a prediction with satisfying accuracy. The data consisted of experimental laminar burning velocities of ethanol/n-heptane and ethanol/n-heptane/iso-octane mixtures at 1 atmosphere and 298K and 338K and modeling data of an ethanol/n-heptane blend and blends of ethanol and a toluene reference fuel at higher pressure and temperature. However, due to uncertainty limits and the limited data set used in the study of Sileghem et al. [6], further validation is needed. In this study, the laminar burning velocities of blends of methanol, ethanol, iso-octane and n-heptane at atmospheric pressure have been investigated. The laminar burning velocities for the pure fuels have been determined previously [12, 13], but the data for methanol and ethanol were extended within the present work. For further discussion on the comparability of the laminar burning velocities of the pure fuels with previous determinations we refer to Sileghem et al. [14] in the case of ethanol, iso-octane and n-heptane.

EXPERIMENTAL SETUP

The measurements were performed using the heat flux method on a perforated plate burner. The experimental setup for the adiabatic flame stabilization is shown in Figure 1. This method has been proposed by de Goey et al. [15] and was further developed by van Maaren and de Goey [16]. The present experimental rig is also used in the study of Sileghem et al. [14] to measure the laminar burning velocity of iso-octane, n-heptane, toluene and gasoline. Important features of the method are, therefore, only shortly outlined in the following.

The heat flux burner has two major parts: a burner head with a heating jacket supplied with thermostatic water to keep the temperature of the burner plate constant at 368K and a plenum chamber with a separate temperature control system. This control system enables to set a temperature of the fresh gas mixture from 298 to 358 K. A burner plate of 2 mm thickness

perforated with small holes (0.5 mm in diameter) is attached to the burner outlet. A theoretical analysis of the heat flux method has been given by de Goey and van Maaren [15, 16]. A mixing panel shown in Figure 1 was used to provide a controlled flow of the vaporized fuel and air at the required equivalence ratio. The key part of this mixing panel is the Bronkhorst High-Tech CORI-FLOW MassFlow Controller (MFC) connected to the Controlled Evaporator and Mixer (CEM, Bronkhorst High-Tech). The liquid fuel flow from the fuel reservoir, pressurized by nitrogen, is metered by the CORI-FLOW MFC and fed to the CEM. Part of the air flow, controlled by the gas MFC-1, is used as a carrier gas to facilitate vaporization in the CEM at temperatures up to 473 K. Another part of the air flow controlled by the gas MFC-2 is varied to provide the required mixture composition and is added downstream. For the measurements reported in this study, the tube connecting the CEM with the burner was a heated tube to avoid condensation of the fuel on its way to the plenum chamber.

ERROR ASSESSMENT

Detailed analysis of these uncertainties was performed earlier [14, 17, 18]. The overall accuracy of the measurements presented in this work is commonly better than ± 1 cm/s. Only for rich mixtures above the equivalence ratio of 1.3, the error could be slightly larger (maximum 1.3 cm/s). The laminar burning velocities measured by van Lipzig et al. [12] and Vancoillie et al. [13] using the same heat flux setup as in the present work are systematically higher than the present measurements by a few cm/s. The difference compared to the present work is larger than the stated experimental error. One of the reasons for the difference could be that in the present work the gas mixture was transported from the evaporator to the burner through a heated tube while in the previous experiments an insulated but unheated tube was used. A comparison of the

measurements with and without a heated tube strongly indicated that some condensation did occur without it. Because condensation is more likely to happen at higher equivalence ratios, higher laminar burning velocities for rich mixtures can possibly be a result of condensation and thus leaner gas mixtures than expected. Additionally, attention has been paid to the stability of the CORI-FLOW performance, resulting in more stable and reliable results for the laminar burning velocities, especially at lower velocities. As explained in [14], the previous measurements were probably also affected by the non-symmetry effect that was manifested in significant over-evaluation of the burning velocities obtained in those studies [12, 13].

RESULTS AND DISCUSSION

Methanol and ethanol

First, the laminar burning velocities of the pure fuels (methanol, ethanol, iso-octane and n-heptane) were measured. Results for iso-octane and n-heptane are published elsewhere [14]. In this section, the focus will be on the laminar burning velocity of methanol and ethanol. For both fuels, the laminar burning velocity has been measured at 298K, 318K, 328K, 338K and 358K. Due to limitations in the experimental setup, measurements on methanol performed by Vancoillie et al. [13] were limited to equivalence ratios below 1.1 and equal to 1.5, however, in the present work the whole range of equivalence ratios from 0.7 to 1.5 was reached. Figure 2 compares the measurements for methanol at 298K, 318K and 338K with the data found in the literature. As described before, the results of Vancoillie et al. [13], done on a similar setup, are slightly higher than the measurements done in this study. Further, it is clear that there are still large deviations with the other data in the literature. At 298K, deviations with the measurements of Gibbs and Calcote (G&C) [19] are large but it has been shown that the measurement methods

of Gibbs and Calcote produced unreliable results [20, 21]. For the measurements of Metghalchi and Keck (M&K) [22], the agreement is good for lean mixtures. Metghalchi and Keck did not take flame stretch and instabilities into consideration in their closed vessel studies. Failing to perform stretch corrections for the spherical flames can lead to over- or underestimation of the true laminar burning velocity, depending upon the sign of the Markstein number. At 318K, the laminar burning velocities of Egolfopoulos et al. [23] are higher than the present measurements for equivalence ratios from 1 to 1.3. Egolfopoulos et al. did take flame stretch effects into account but because of the typically small strain rate in their flames (about 100 s^{-1}), they used a linear extrapolation to zero stretch. This linear extrapolation has recently been reported to lead to overestimations of the burning velocity by 5-10% [23, 24]. At 338K, measurements were compared to those of Veloo et al. [25] at 343K. Veloo et al. recently repeated the measurements by Egolfopoulos et al. [23] on methanol-air flames under a restricted set of conditions. They used the counterflow twin-flame burner in combination with a particle image velocimetry method. The unstretched laminar burning velocity was derived using a nonlinear extrapolation approach based on direct numerical simulations of the experiments. This led to an improved accuracy of the measured burning velocity compared to the linear extrapolation employed by Egolfopoulos et al. [23]. From Figure 2 it is clear that the effect of the equivalence ratio on the laminar burning velocity agrees well with the present measurements, although the measurements were done at a different temperature.

In Figure 3, the measurements for ethanol at 298K and 358K are compared with the data found in the literature. For ethanol, the scatter between the different measurements is already less than for methanol. The measurements of Konnov et al. [26], van Lipzig et al. [12] and Dirrenberger et

al. [27] were all done with the same measurement method, i.e. the heat flux method. The measurements done by Dirrenberger et al. compare very well at 358K, especially at lean and rich mixtures, but are slightly higher at 298K. The measurements of Konnov et al. [26] report lower laminar burning velocities for leaner mixtures of ethanol. For rich mixtures, the agreement is better but the burning velocities are slightly higher. Van Lipzig et al. [12] stated that the differences between his measurements and the measurements of Konnov et al. [26] were due to erroneous readings from the edge thermocouple that affected temperature measurements but when we compare the present measurements with the measurements of Konnov et al., there seems also to be a shift to richer mixture both for 298K and 358K. A possible cause for this shift could be fuel condensation. Special attention was paid to prevent fuel condensation during this study by installing a heated tube between the evaporator and the burner. Note also that the correspondence with the recent data sets of Bradley et al. [28] is reasonable although these authors gathered their data with completely different measurement method.

Another way to compare different measurements from the literature is to look at the effect of the unburned mixture temperature on the burning velocity. The most widely used correlation describing the effect of initial temperature on the burning velocity:

$$u_l = u_{l0} \left(\frac{T_u}{T_{u0}} \right)^\alpha \quad (7)$$

where u_l is the laminar burning velocity and T_u is the unburned mixture temperature, was used in this or equivalent form since the 1950s, e.g. in [29]. The subscript “0” refers to the values at reference conditions (usually 298 K and 1 bar). α represents the power exponent of the temperature dependence. In Figure 4, both the present data and the data of Vancoillie et al. [13] are plotted as a function of the temperature on a log-log scale. The slope of the measurements

represents the power exponent of the temperature dependence. As can be seen, there is less deviation from the linear behavior for the present measurements on methanol. It is clear that the changes to the setup (heated tube, stability of the CORI-FLOW) result in more stable and reliable results. In Figure 5, the power exponents α , derived from the present measurements on methanol, are compared with the power exponents found by Vancoillie et al. [13]. Around stoichiometry, there is still a good agreement. In Figure 5, the power exponents derived from modeling results using the mechanism by Li et al. [30] are also shown. This mechanism also produces a minimum for slightly rich mixtures and it agrees very well with the measurements of Vancoillie et al. [13], especially for lean mixtures. For the present measurements, the agreement is good around equivalence ratios of 1-1.3 but the deviations are larger for lean mixtures. More details about the modeling can be found in [13].

In Figure 6, the laminar burning velocities of ethanol are shown as a function of the temperature on log-log scales. The difference in temperature dependence between the present measurements and the measurements of Dirrenberger et al. [27] and Konnov et al. [26] corresponds also to the difference in deviations between the laminar burning velocities at different temperatures. In Figure 7, the power exponents, derived from the present measurements, the data of Dirrenberger et al. [27] and the data of Konnov et al. [26], are compared. The shape of the curve for power exponents for the present measurement is consistent with the previous measurements on the same setup [13, 14], the minimum is around the equivalence ratio of peak burning velocity. This is also reflected in the measurements of Konnov et al. [26], while the shape of the power exponents derived from the measurements of Dirrenberger et al. [27] is very different. The power exponent of Dirrenberger et al. [27] was derived from measurements at only three different

temperatures which is less accurate and could be the cause of the different shape. In Figure 7, calculated power exponents using the Konnov mechanism [31] were added. The power exponents agree well with the present experimental data especially in rich mixtures. The power exponents derived from the data of Konnov et al. [26] indicate the trend to level off with increasing equivalence ratio. This effect was also predicted in methane-air flames [32]. This cannot be seen in the present measurements and it seems that the experimental trend follows the simulations by the Konnov mechanism; therefore, further measurements of the laminar burning velocity of rich mixtures are desirable.

Mixtures of methanol, ethanol, iso-octane and n-heptane

To further validate the mixing rules tested by Sileghem et al. [6], measurements have been done on different fuel blends: ethanol/iso-octane (25% v/v liquid, 50% v/v and 75% v/v ethanol), methanol/iso-octane (75% v/v methanol) and a mixture of methanol, ethanol, iso-octane and n-heptane (25% v/v each). For the binary mixtures, the measurements have been limited to alcohol/iso-octane mixtures because the difference in laminar burning velocity between iso-octane and the light alcohols is bigger compared to n-heptane. The measurements on methanol/iso-octane could not be extended to lower methanol blends because of phase separation. Methanol/iso-octane mixtures with 75% v/v methanol were found to be the practical lower limit.

In Figure 8, the measurements for all ethanol/iso-octane blends are shown at 338K and in Figure 9, a comparison of the different mixing rules, described in the introduction, is made for ethanol/iso-octane with 50% v/v ethanol. As expected, the laminar burning velocities of the fuel blends are in between the laminar burning velocities of the pure fuels. In Figure 9, the best

prediction is made by the Le Chatelier rule based on energy fraction, especially for richer mixtures, in agreement with what was shown by Sileghem et al. [6]. Mixing rules based on mole or mass fraction were also evaluated by Sileghem et al. [6] and it was concluded that they gave less accurate predictions. An example of this applied to the present study can be seen in Figure 10, where the laminar burning velocity is plotted for different equivalence ratios as a function of either energy fraction of ethanol or mole fraction of ethanol. From the Figure 10 we can conclude that predicting the laminar burning velocity with a linear interpolation based on the mole fraction is not a good option. Predicting the laminar burning velocity of ethanol-hydrocarbon blends based on the energy fraction is a better option with only a slight overprediction which can also be seen in Figure 9.

In Figure 11, the three different mixing rules are tested for methanol/iso-octane. Here, the energy fraction mixing rule gives the best prediction and the Hirasawa et al. mixing rule and Le Chatelier mixing rule underpredict the measurements. The laminar burning velocity of methanol/iso-octane is again in between the laminar burning velocity of iso-octane and methanol.

In this study, a quaternary mixture with both ethanol and methanol was measured because complex mixtures with more than one alcohol component could play an important role in the future [4]. The burning velocities of the pure components and the mixtures are shown in Figure 12 at 338K. In Figure 13, the laminar burning velocity of the mixture is shown together with the predictions of the three mixing rules and with the predictions of a linear interpolation based on the mole fraction. Again, it is clear that the mole fraction is not the right choice to calculate the

laminar burning velocity of fuel blends out of the laminar burning velocities of the pure components. As a result, a mole fraction mixing rule should not be used in engine simulation models. The three mixing rules give similar results with the best agreement for the energy fraction mixing rule. Based on these results and the fact that there are also small errors in the laminar burning velocity of the pure fuels, we can conclude that these three mixing rules give accurate predictions for laminar burning velocities of blends with methanol and ethanol. These mixing rules can be invaluable as a way to calculate this property in a computationally efficient way in simulation programs.

CONCLUSION

Measurements of the laminar burning velocity have been done for methanol, ethanol, iso-octane, n-heptane and binary and quaternary mixtures of these fuels for a wide range of temperatures at atmospheric pressure using the heat flux method on a perforated plate burner. First, the temperature dependence of the laminar burning velocity as a function of the equivalence ratio has been shown for methanol and ethanol. This temperature dependence can be used to validate reaction mechanisms. Both fuels have the temperature dependence with a minimum around the equivalence ratio of peak burning velocity. Second, different mixing rules for the prediction of the laminar burning velocity of fuel blends were validated. The energy fraction mixing rule, the mixing rule developed by Hirasawa et al.[10] and the Le Chatelier's rule based on energy fraction gave very good results, both for mixtures with methanol and ethanol. Le Chatelier's rule gave slightly better results for the ethanol blends while the energy fraction mixing rule gave better results for methanol blends. Mixing rules are not expected to be linear in the fuel blend composition, proven by the poor predictions of the mole fraction mixing rule. Keeping in mind

the uncertainty limits of both the laminar burning velocity of the mixture and the laminar burning velocity of the pure fuels, we can conclude that the three proposed mixing rules can be used in simulation models to calculate the laminar burning velocity with acceptable accuracy. However, further experimental validation is needed at higher temperatures and pressures.

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FIGURE CAPTIONS

Preference for color: on the Web only.

Figure 1: Figure 1: Experimental heat flux setup for liquid fuels.

- Figure 2: Figure 2: Laminar burning velocities of methanol at different temperatures.
- Figure 3: Figure 3: Laminar burning velocities of ethanol at different temperatures.
- Figure 4: Figure 4: Log-log plot of laminar burning velocities of methanol-air flames at atmospheric pressure and different initial temperatures. Squares, present experiments; Triangles, Vancoillie [13].
- Figure 5: Figure 5: Comparison between the power exponent α of methanol from measurements and the power exponent α derived from the mechanism of Li et al. [30].
- Figure 6: Figure 6: Log-log plot of laminar burning velocities of ethanol-air flames at atmospheric pressure and different initial temperatures. Squares, present experiments; Triangles, Konnov [26]; Crosses, Dirrenberger [27].
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- Figure 9: Figure 8: The laminar burning velocity ethanol/iso-octane (50% v/v ethanol) at 338K together with the predictions of the different mixing rules.
- Figure 10: Figure 9: The laminar burning velocity of iso-octane, ethanol and ethanol/iso-octane blends (25% v/v, 50% v/v and 75% v/v ethanol) at 338K as a function of either energy fraction of ethanol or mole fraction of ethanol.

Figure 11: Figure 10: The laminar burning velocity methanol/iso-octane (75% v/v methanol) at 338K together with the predictions of the different mixing rules.

Figure 12: Figure 11: The laminar burning velocity of iso-octane, n-heptane, methanol, ethanol and their quaternary mixture (25% v/v each) at 338K.

Figure 13: Figure 12: The laminar burning velocity methanol/ethanol/iso-octane/n-heptane mixture (25% v/v each) at 338K together with the predictions of the different mixing rules.

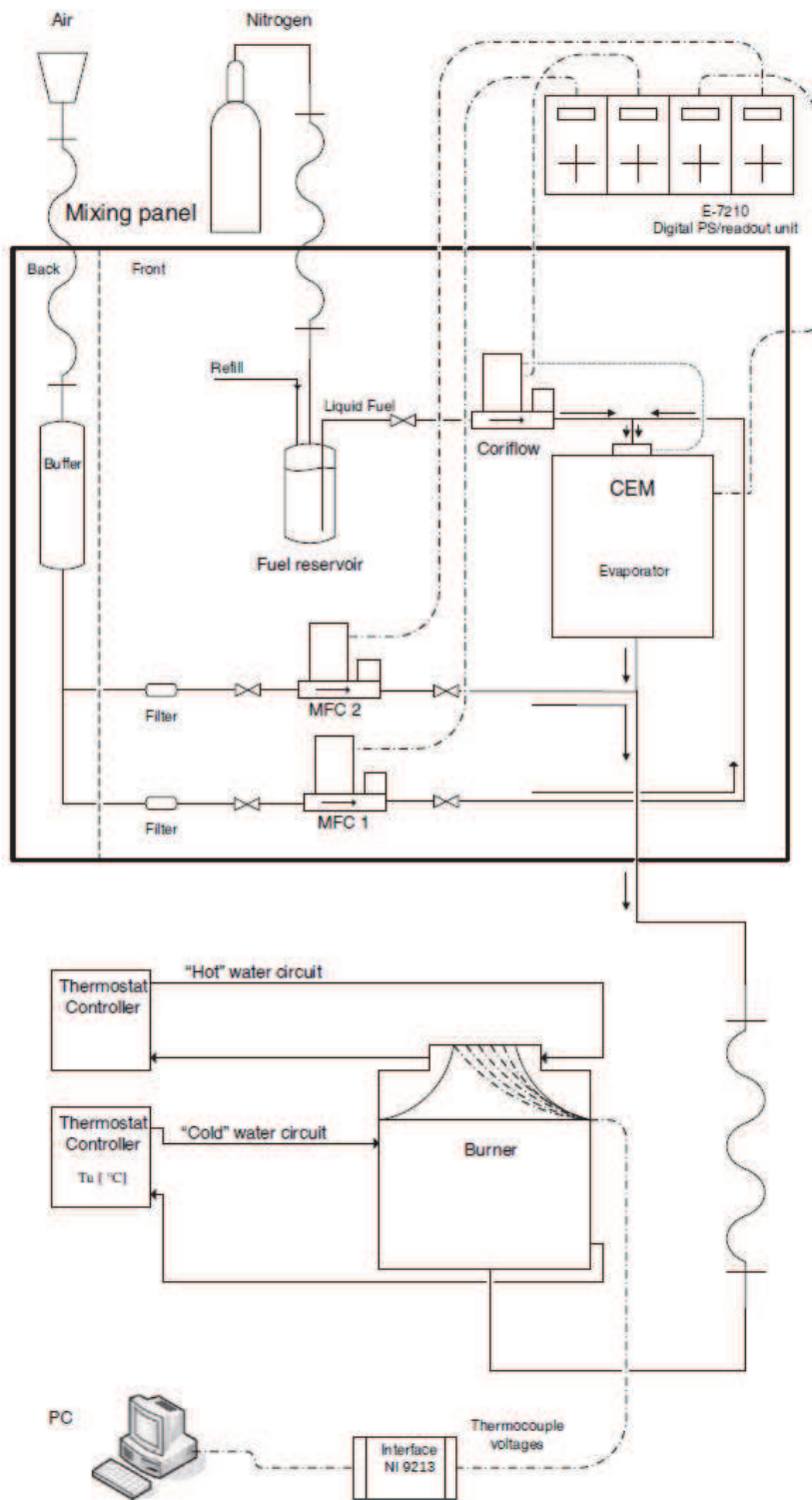
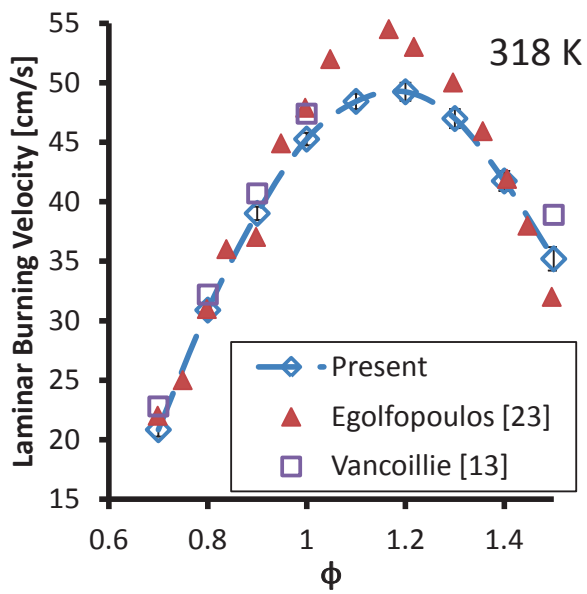
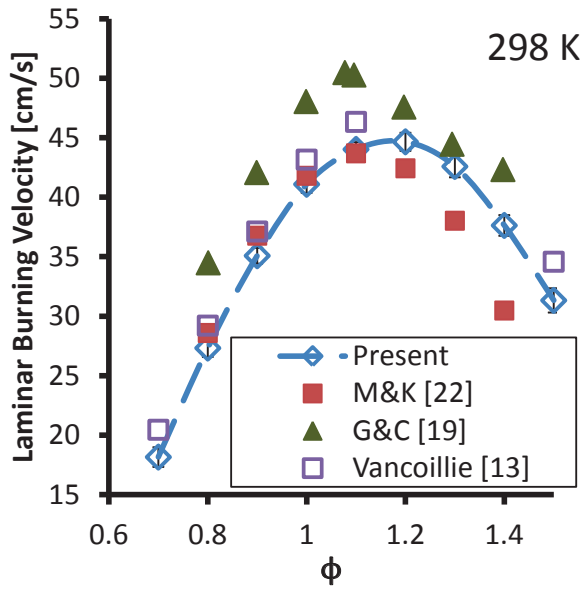


Figure 1: Experimental heat flux setup for liquid fuels.



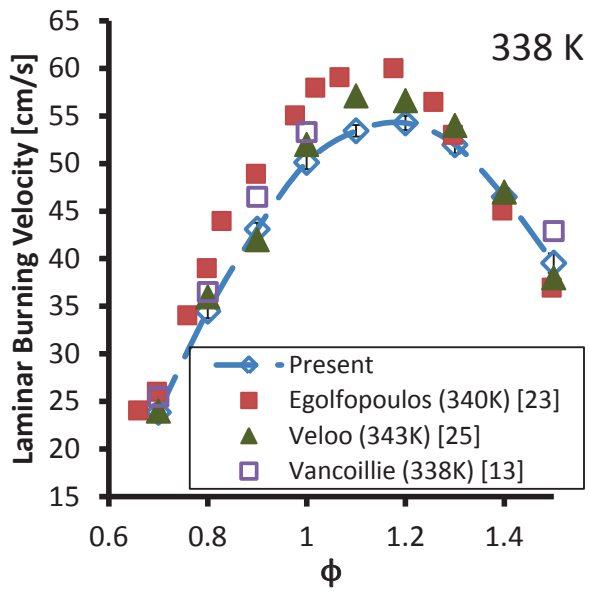


Figure 2: Laminar burning velocities of methanol at different temperatures.

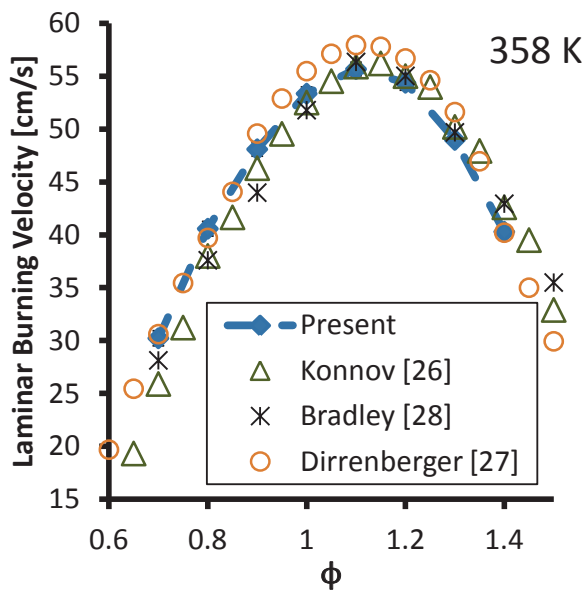
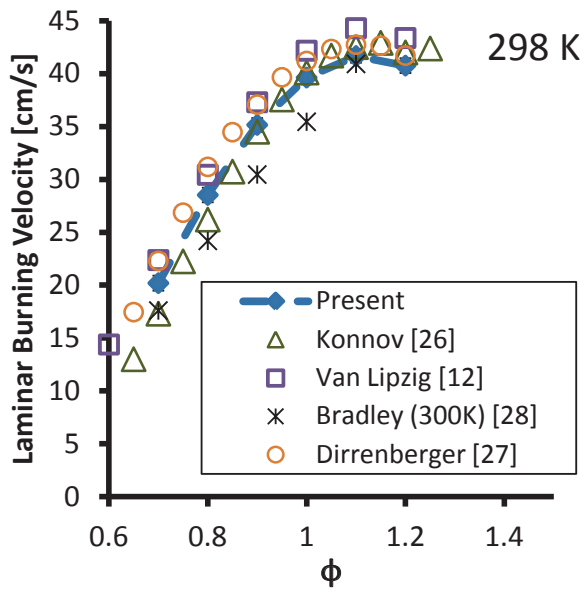


Figure 3: Laminar burning velocities of ethanol at different temperatures.

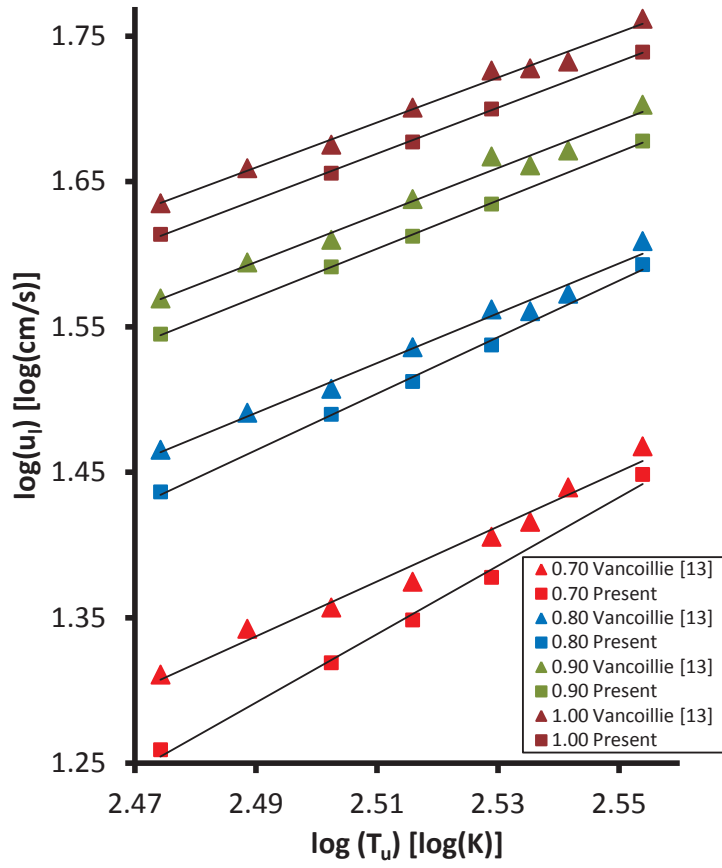


Figure 4: Log-log plot of laminar burning velocities of methanol-air flames at atmospheric pressure and different initial temperatures. Squares, present experiments; Triangles, Vancoillie [13].

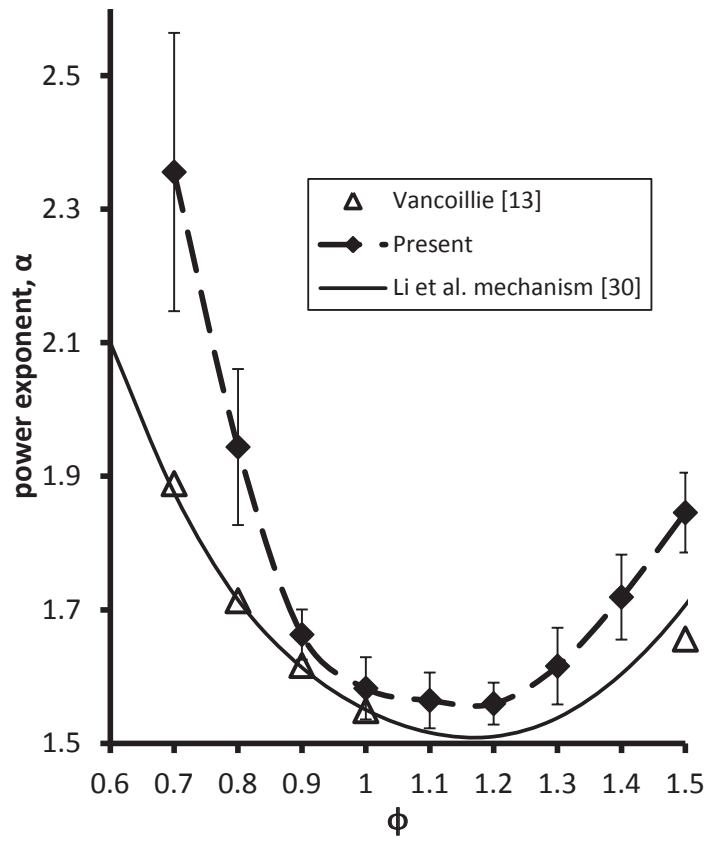


Figure 5: Comparison between the power exponent α of methanol from measurements and the power exponent α derived from the mechanism of Li et al. [30].

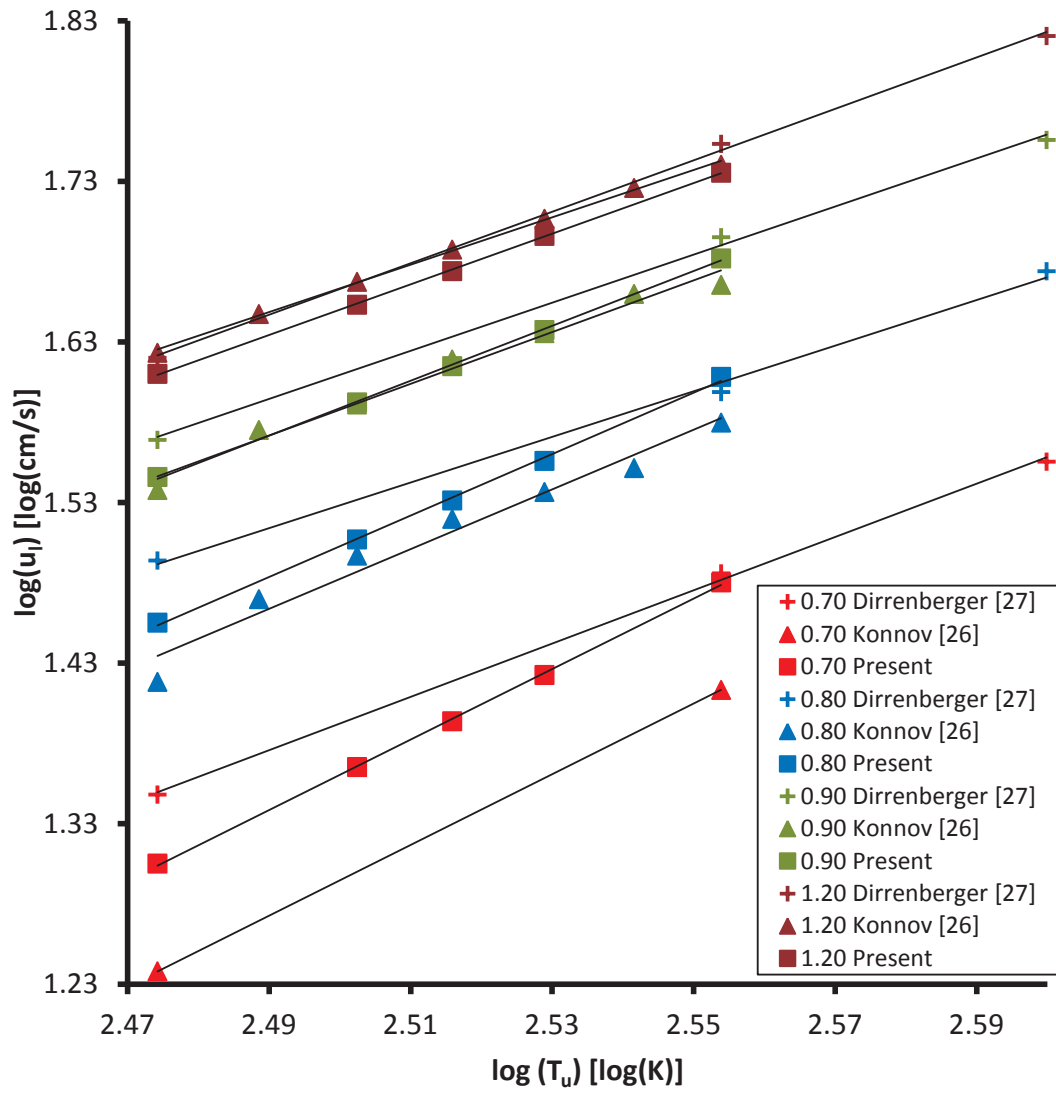


Figure 6: Log-log plot of laminar burning velocities of ethanol-air flames at atmospheric pressure and different initial temperatures. Squares, present experiments; Triangles, Konnov [26]; Crosses, Dirrenberger [27].

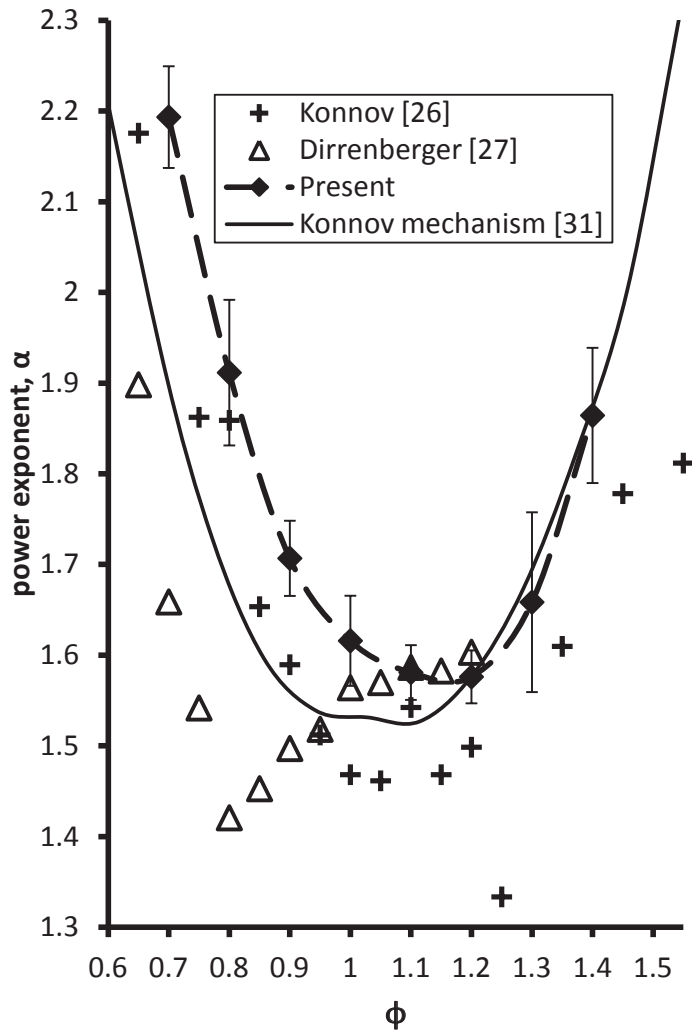


Figure 7: Comparison between the power exponent α of ethanol from measurements and the power exponent α derived from the Konnov mechanism [31].

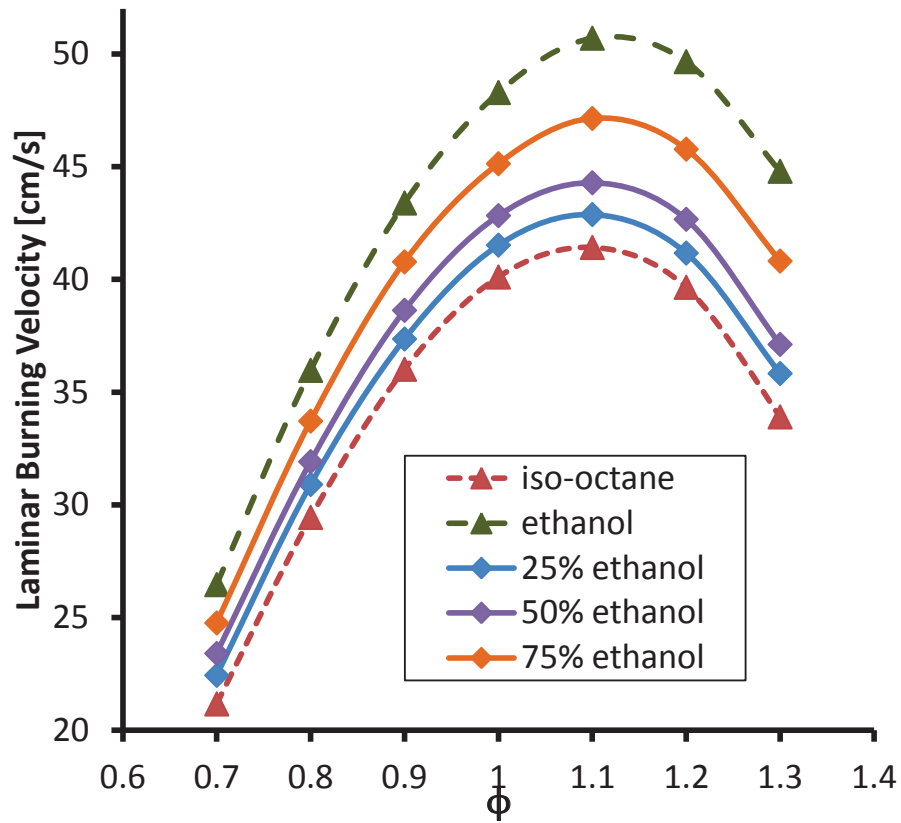


Figure 8: The laminar burning velocity of iso-octane, ethanol and ethanol/iso-octane (25% v/v, 50% v/v and 75% v/v ethanol) mixtures at 338K.

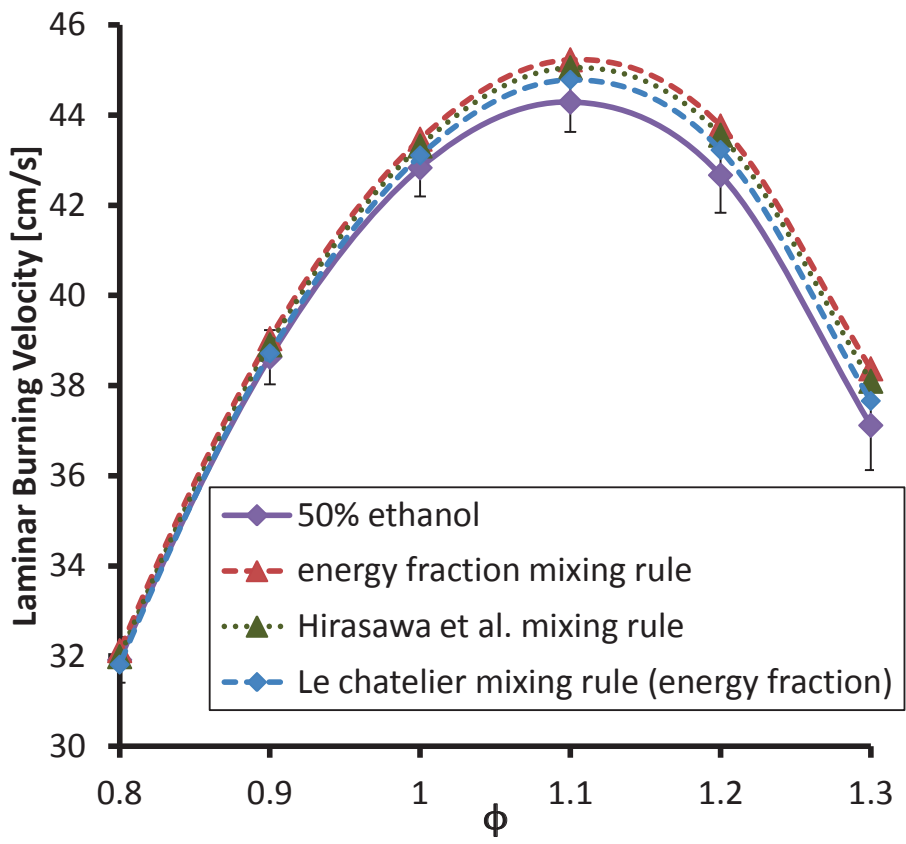


Figure 9: The laminar burning velocity ethanol/iso-octane (50% v/v ethanol) at 338K together with the predictions of the different mixing rules.

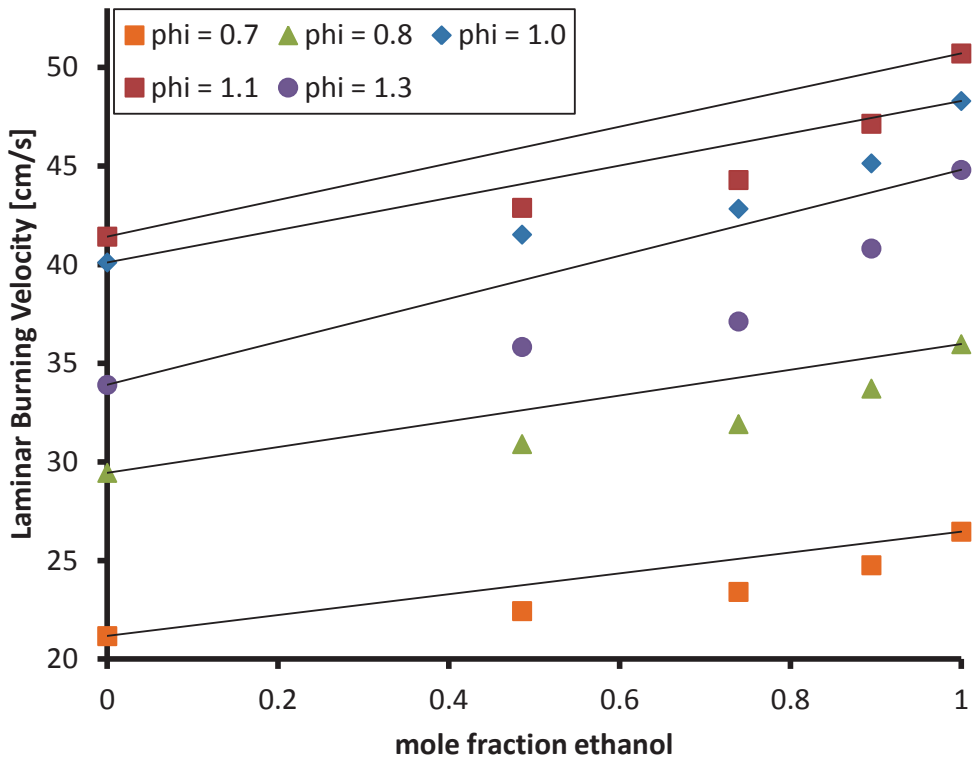
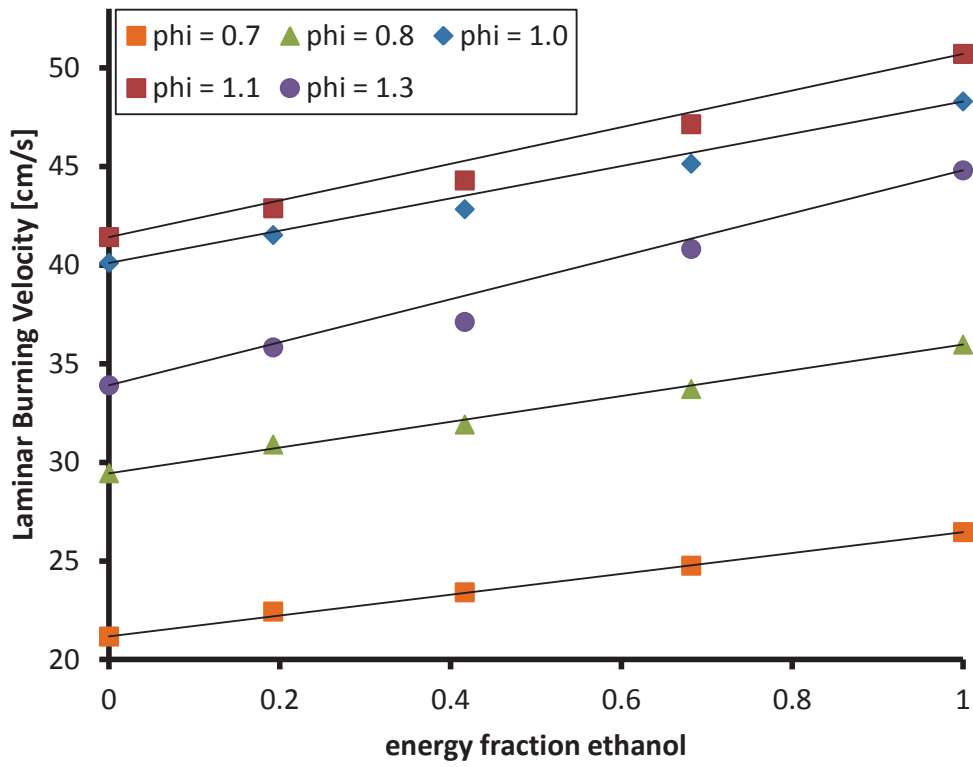


Figure 10: The laminar burning velocity of iso-octane, ethanol and ethanol/iso-octane blends (25% v/v, 50% v/v and 75% v/v ethanol) at 338K as a function of either energy fraction of ethanol or mole fraction of ethanol.

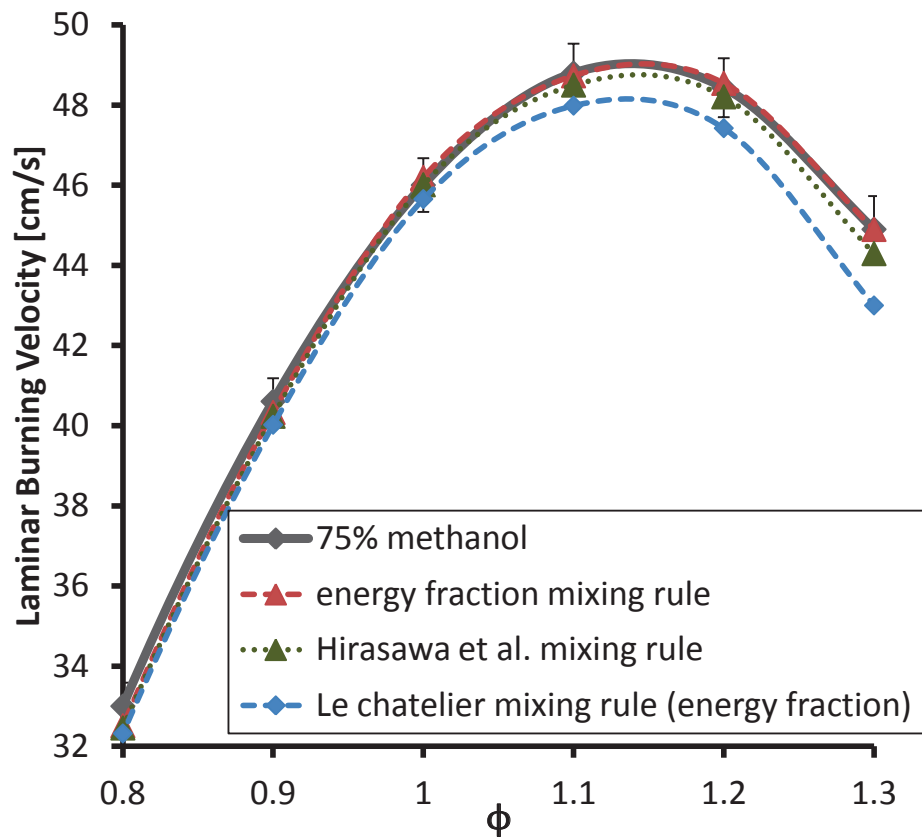


Figure 11: The laminar burning velocity methanol/iso-octane (75% v/v methanol) at 338K together with the predictions of the different mixing rules.

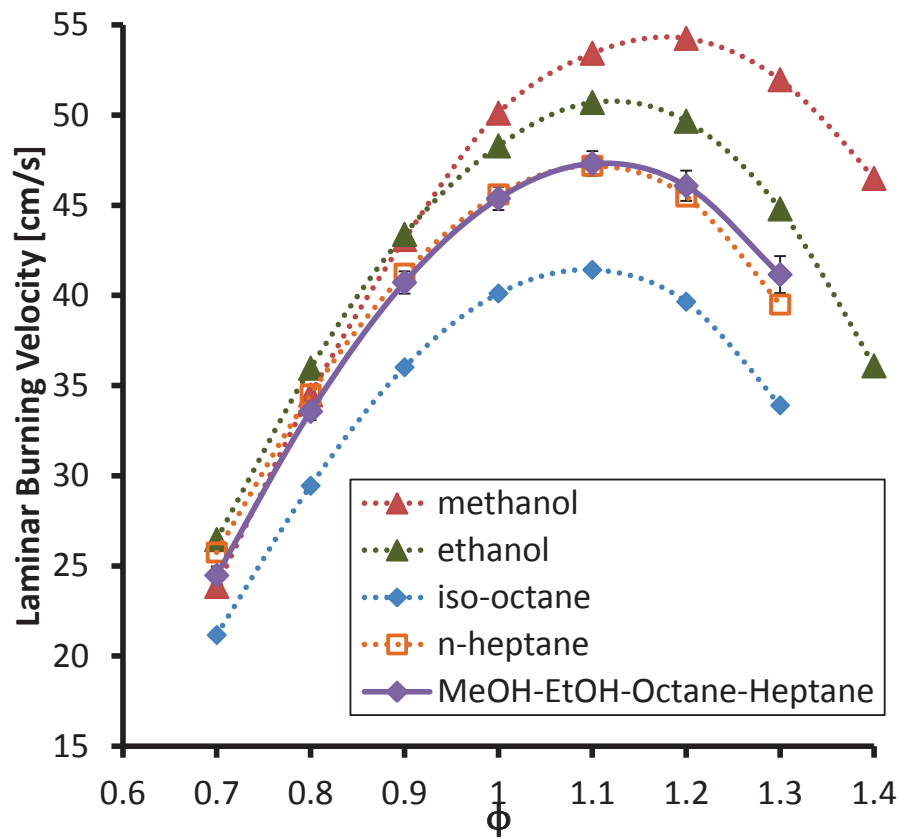


Figure 12: The laminar burning velocity of iso-octane, n-heptane, methanol, ethanol and their quaternary mixture (25% v/v each) at 338K.

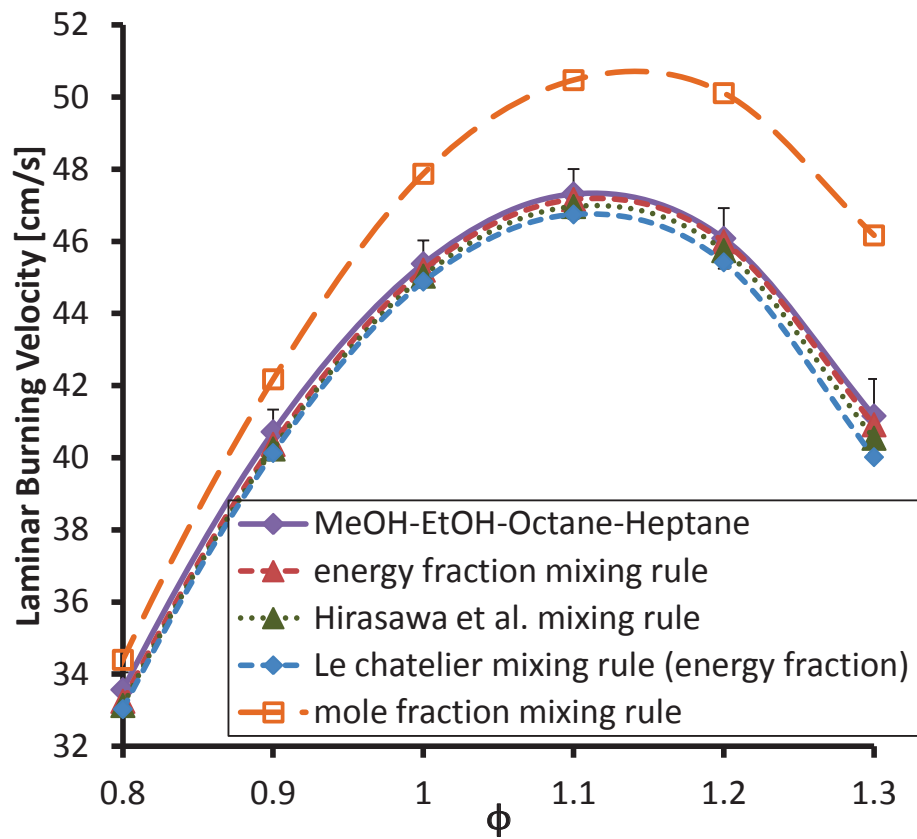


Figure 13: The laminar burning velocity methanol/ethanol/iso-octane/n-heptane mixture (25% v/v each) at 338K together with the predictions of the different mixing rules.

Supplementary Material

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