

Lewis Number Effect on Explosive Transition of Stretch-Free Flat Flame

Youhi Morii¹, Akira Tsunoda^{1,2}, Kaoru Maruta¹

¹Institute of Fluid Science, Tohoku University, Sendai, Miyagi, Japan

²Graduate School of Engineering, Tohoku University, Sendai, Miyagi, Japan

1 Introduction

Understanding the deflagration to detonation transition (DDT) and the knock onset in spark-ignition engines is important but still unsolved in safety engineering and practical engine development. Prior work on DDT has been reviewed by Oran, and many studies continue to evolve from the pioneering work of Zel'dovich [1] and Gu et al. [2] with respect to understanding the mechanism of knocking.

Recently, auto-ignition assisted flames, which propagate faster than the general deflagration wave due to chemical reactions in the preheat zone, have gained attention [3–7]. Thus, understanding the relationship between ignition and deflagration has become increasingly important to understand such phenomena. Our recent theoretical and numerical study investigated this relationship [8], aiming to enhance our understanding of detonation and knocking. We identified the limits of the existence of stretch-free flat flame structures with a Lewis number over unity, referred to as the "explosive transition of deflagration" [8]. This allowed us to categorize the phenomena into three behaviors of the deflagration wave: (I) Deflagration wave with negligible preheat zone chemical reaction, (II) Deflagration wave with non-negligible preheat zone chemical reaction, and (III) Phenomena beyond the explosive transition of deflagration. A schematic diagram of the behaviors of the deflagration is shown in Fig.1. As the unburned gas temperature increases, chemical reactions in the preheat zone become non-negligible, moving from (I) to (II) and eventually to (III), where no deflagration structure exists.

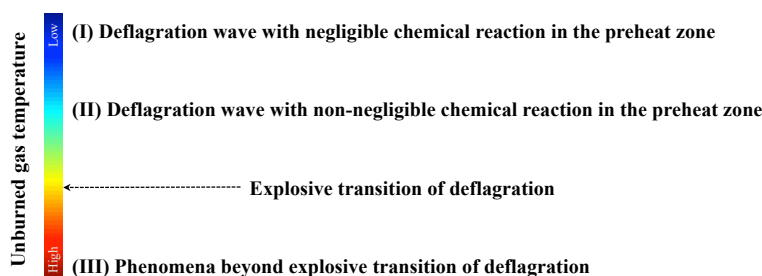


Figure 1: Behaviors of deflagration wave including explosive transition of deflagration.

This study investigates the effect of ignition on deflagration wave structure near the explosive transition, using four fuels with different Lewis numbers (hydrogen, methane, propane, and *n*-heptane), after establishing the theoretical connection between ignition and deflagration wave structure.

2 Explosive Transition of Deflagration [8]

To consider the relationship between ignition and deflagration, reaction progresses in 0d ignition and 1d deflagration were compared using a spatial-temporal transformation. The details of the theory discussed in this section can be found in [8]. A brief description is given here.

2.1 Governing equations of 0D homogeneous ignition using normalized fuel mass fraction and temperature

The governing equations for 0D homogeneous ignition are the conservation of energy and chemical species. For the constant pressure and enthalpy case, the conservation equations are given by

$$\frac{dY_k}{dt} = \frac{\dot{\omega}_k W_k}{\rho} \quad (k = 1, 2, \dots, K), \quad (1)$$

$$\frac{dT}{dt} = -\frac{\sum_{k=1}^K \dot{\omega}_k h_k W_k}{c_p \rho} \quad (2)$$

where t is time, Y_k is the mass fraction of k^{th} species, $\dot{\omega}_k$ is the chemical production rate of k^{th} species, W_k is the molecular weight of k^{th} species, ρ is the mass density, T is the temperature, K is the total number of chemical species, h_k is the enthalpy of k^{th} species, and c_p is the mean specific heat. The equation of state used for the interconversion of pressure, density, and temperature is a perfect gas given by $\rho = (p\bar{W})/(RT)$, where p is the pressure, \bar{W} is the mean molecular weight, and R is the universal gas constant. Here, the normalized temperature and the normalized fuel mass fraction are defined by $\tilde{Y}_f = (Y_{f,1} - Y_f)/(Y_{f,1} - Y_{f,0})$ and $\tilde{T} = (T - T_0)/(T_1 - T_0)$, where subscript 0 means the initial value and subscript 1 means the final value, and assuming that \tilde{T} and \tilde{Y}_f are Legendre-transformable relations. Then, the following relation holds

$$\frac{d\tilde{T}}{dt} = -\frac{d\tilde{Y}_f}{dt}. \quad (3)$$

Note that, for the Legendre transformation to be possible, \tilde{T} and \tilde{Y}_f must be convex functions, and from Eq. (3), $\tilde{T} \in [0, 1]$ and $\tilde{Y}_f \in [0, 1]$, the following conditions are required $d\tilde{T}/dt > 0$ and $d\tilde{Y}_f/dt < 0$. This constraint is always true for single-step chemical reaction models and is usually true for multi-step chemical reaction models. Note that \tilde{T} and \tilde{Y}_f are bijective, and once either \tilde{T} or \tilde{Y}_f is determined, all remaining variables, including T , ρ , and Y , are determined. The Eqs.(1)(2) can be transformed using \tilde{Y}_f and \tilde{T} as follows

$$\frac{d\tilde{Y}_f}{dt} = -\frac{\dot{\omega}_f W_f}{\rho(Y_{f,1} - Y_{f,0})} = -\frac{1}{Y_{f,1} - Y_{f,0}} \frac{dY_f}{dt}, \quad (4)$$

$$\frac{d\tilde{T}}{dt} = -\frac{\sum_{k=1}^K \dot{\omega}_k h_k W_k}{c_p \rho (T_1 - T_0)} = \frac{1}{T_1 - T_0} \frac{dT}{dt}. \quad (5)$$

2.2 Governing equations of 0D homogeneous ignition using residence time

The residence time is the total time that the fluid parcel has spent inside a control volume defined by $\tau = \int_{x_0}^x u^{-1} dx$, where u is the velocity of the fluid parcel and x is the position of the fluid parcel. Then, the total differential of the residence time can be given by $d\tau = u^{-1} dx$. The Eqs.(4) and (5) can be transformed by a spatial-temporal transformation using the residence time and the velocity of the fluid parcel as follows

$$\rho u \frac{d\tilde{Y}_f}{dx} = -\frac{\dot{\omega}_f W_f}{Y_{f,1} - Y_{f,0}} \quad (6)$$

$$\rho u \frac{d\tilde{T}}{dx} = - \frac{\sum_{k=1}^K \dot{\omega}_k h_k W_k}{c_p (T_1 - T_0)}. \quad (7)$$

Note that the Eq. (3) can be rewritten using the position of fluid parcel as follows

$$\frac{d\tilde{T}}{dx} = - \frac{d\tilde{Y}_f}{dx}. \quad (8)$$

2.3 Governing equations of 1D laminar premixed flame using \tilde{Y}_f and \tilde{T}

The conservation equations for 1D laminar premixed flame are given by

$$\rho u = \text{constant}, \quad (9)$$

$$\rho u \frac{dY_k}{dx} = \frac{d}{dx} \left(\rho D_k \frac{dY_k}{dx} \right) + \dot{\omega}_k W_k, \quad (k = 1, 2, \dots, K), \quad (10)$$

$$\rho u \frac{dT}{dx} = \frac{1}{c_p} \frac{d}{dx} \left(\lambda \frac{dT}{dx} \right) - \frac{\sum_{k=1}^K \dot{\omega}_k h_k W_k}{c_p}, \quad (11)$$

where D_k is the mixture diffusion coefficient of k^{th} species, and λ is the thermal conductivity of the mixture. The Eqs. (10) and (11) can be rewritten using \tilde{Y}_f and \tilde{T} as follows

$$\rho u \frac{d\tilde{Y}_f}{dx} = \frac{d}{dx} \left(\rho D_f \frac{d\tilde{Y}_f}{dx} \right) - \frac{\dot{\omega}_f W_f}{Y_{f,1} - Y_{f,0}}, \quad (12)$$

$$\rho u \frac{d\tilde{T}}{dx} = \frac{1}{c_p} \frac{d}{dx} \left(\lambda \frac{d\tilde{T}}{dx} \right) - \frac{\sum_{k=1}^K \dot{\omega}_k h_k W_k}{c_p (T_1 - T_0)}. \quad (13)$$

The only difference between Eqs. (6) and (7) and Eqs. (12) and (13) is the first additional term of the right-hand side in Eqs. (12) and (13). Now consider the Lewis number of the fuel defined by $Le_f = \frac{\lambda}{\rho c_p D_f}$. When $Le_f = 1$, Eqs. (6) and (7) and Eqs. (12) and (13) are equivalent. In other words, when $Le_f = 1$, the governing equations for 0D homogeneous ignition using \tilde{Y}_f and \tilde{T} are equivalent to those for 1D laminar premixed flame using \tilde{Y}_f and \tilde{T} . In the next section, simple calculations will be performed to investigate the effect of the Lewis number on the limit of the 1D laminar premixed flame.

3 Results and discussion

3.1 Numerical methods and conditions

Computations of 0D homogeneous ignition with constant pressure and enthalpy and 1D laminar premixed flame with multi-step chemical reaction models were performed using Cantera [9]. The four mixtures, hydrogen/air, methane/air, propane/air, and *n*-heptane/air, were used. The Le_f of the four mixtures for pressure of 0.1 MPa and temperature of 300 K are 0.43, 0.88, 1.66, and 2.85, respectively. The multi-step chemical reaction models are UT-JAXA model [10] for hydrogen, the Lu model [11] for methane, San Diego mechanism [12] for propane, and the reduced SIP model [13] for *n*-heptane. Note that, in order to maintain the Legendre transform even when multi-step chemical reaction models are used, the computational domain should be as short as possible because \tilde{T} and \tilde{Y}_f must be the convex functions. Numerical conditions for pressure and equivalence ratio were fixed at 0.1 MPa and 1.0, respectively. 1D laminar premixed flame simulations were performed by varying the inlet temperature

from 300 to 3000 K in 100 K increments. After confirming that the simulation failed no matter how the computational domain was decreased, the next step was to perform the simulation by varying the inlet temperature in 1K increments to determine the temperature limit at which the calculation could be performed. After performing 1D laminar premixed flame simulations, 0D homogeneous ignition simulations were performed using the initial temperatures at which the 1D laminar premixed flame could be calculated.

3.2 Lewis number effect on 1D laminar premixed flame using \tilde{Y}_f and \tilde{T}

Figure 2 shows the relation between 0D homogeneous ignition and 1D laminar premixed flames for hydrogen, methane, propane, and *n*-heptane. As discussed in section 2, for $Le_f = 1$, 0D homogeneous ignition using \tilde{Y}_f and \tilde{T} after a spatial-temporal transformation based on the residence time is equivalent to 1D laminar premixed flame using \tilde{Y}_f and \tilde{T} . For $Le_f < 1$, the profile of a 1D laminar premixed flame on \tilde{Y}_f - \tilde{T} plane is convex below the profile of $Le_f = 1$ case (i.e., 0D homogeneous ignition), while for $Le_f > 1$ the profile is convex above the profile of $Le_f = 1$ case (i.e., 0D homogeneous ignition) and can intersect the profile of $Le_f = 1$ case (i.e., 0D homogeneous ignition) in the high-temperature region of the flame (i.e., reaction zone) where the heat release is high. From now on, the discussion will be conducted in the relatively low temperature region (i.e., preheat zone) where the influence of the Lewis number is clear in Fig. 2.

In Fig. 2, at a given \tilde{T} , the consumption of \tilde{Y}_f can be considered as the progress of the reaction. Thus, the reaction progresses are in the order of $Le_f < 1$, $Le_f = 1$ (i.e., 0D homogeneous ignition), and $Le_f > 1$. Therefore, if $Le_f < 1$, there is no ignition in the preheat zone. In other words, 1D laminar premixed flame structure can exist even if the inlet temperature and pressure are very high. However, if the $Le_f > 1$, there is a possibility of ignition in the preheat zone. In other words, depending on the inlet temperature and pressure conditions, there may be conditions where 1D laminar premixed flame structure can not exist.

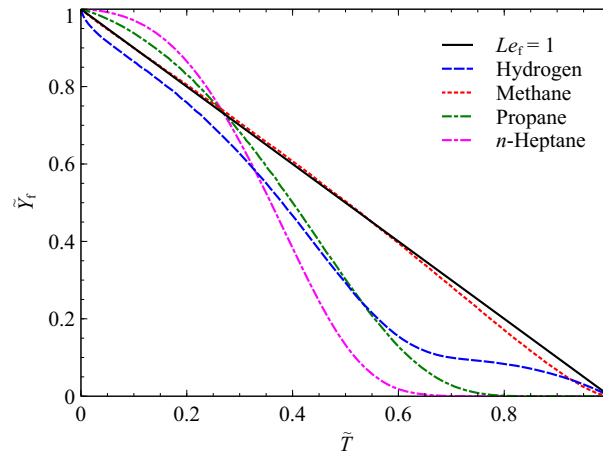


Figure 2: The relation between 0D homogeneous ignition and 1D laminar premixed flames for hydrogen, methane, propane, and *n*-heptane fuels using \tilde{Y}_f and \tilde{T} . Note that the 0D homogeneous ignition is equivalent to the 1D laminar premixed flame when $Le_f = 1$.

Figure 3 shows the results of burning velocity with varying the inlet temperature. In the case of hydrogen and methane fuels, i.e., $Le_f < 1$, the burning velocity increases with increasing inlet or initial temperature and propagating flame exist even at inlet temperature of 3000 K. In the case of propane and

n-heptane fuels, i.e., $Le_f > 1$, the burning velocity increases with increasing inlet or initial temperature, but the propagating flame could not exist over 2030 K for propane fuel and over 1270 K for *n*-heptane fuel. This is to be expected from theory. We also find that the threshold temperature depends on the Lewis number, and that the higher the Lewis number, the lower the threshold temperature.

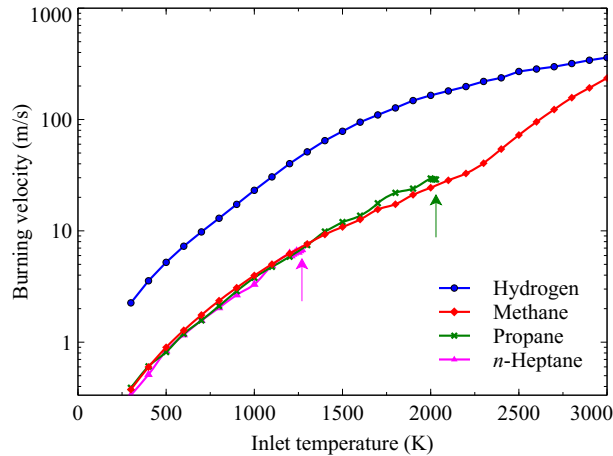


Figure 3: Burning velocity of hydrogen, methane, propane, and *n*-heptane fuels at fixed pressure of 0.1 MPa and varying inlet temperature. The green and magenta arrows refer to the temperature limits of propane and *n*-heptane fuel, respectively.

5 Conclusions

In this study, we have theoretically demonstrated the theoretical connection between \tilde{Y}_f and \tilde{T} is identical for 0D homogeneous ignition and 1D laminar premixed flames under the condition $Le_f = 1$, employing a spatial-temporal transformation. Our findings revealed that for 1D laminar premixed flames with $Le_f < 1$, ignition does not take place in the preheat zone, ensuring the existence of a flame structure. In contrast, for 1D laminar premixed flames with $Le_f > 1$, ignition may occur in the preheat zone, resulting in the absence of a flame structure when the temperature surpasses a certain threshold. We have introduced the term "explosive transition of deflagration" to describe this phenomenon where a flame structure cannot be sustained due to ignition in the preheating region. Additionally, we have established that the threshold temperature is dependent on the Lewis number, with higher Lewis numbers leading to lower threshold temperatures. It is important to mention that our study focused on stretch-free flames, while stretched flames are explored by Tsunoda et al. [14] in a separate presentation at this conference.

Acknowledgement

This work was partially supported by JSPS KAKENHI Grant Number 19KK0097.

References

- [1] Y. B. Zeldovich, "Regime classification of an exothermic reaction with nonuniform initial conditions," *Combustion and Flame*, vol. 39, pp. 211–214, 10 1980. [Online]. Available: <https://linkinghub.elsevier.com/retrieve/pii/0010218080900176>

- [2] X. J. Gu, D. R. Emerson, and D. Bradley, "Modes of reaction front propagation from hot spots," *Combustion and Flame*, vol. 133, pp. 63–74, 4 2003. [Online]. Available: <https://linkinghub.elsevier.com/retrieve/pii/S0010218002005412>
- [3] Y. Ju, C. B. Reuter, O. R. Yehia, T. I. Farouk, and S. H. Won, "Dynamics of cool flames," *Progress in Energy and Combustion Science*, vol. 75, p. 100787, 11 2019. [Online]. Available: <https://linkinghub.elsevier.com/retrieve/pii/S0360128518301539>
- [4] A. J. Susa, A. M. Ferris, D. F. Davidson, and R. K. Hanson, "Experimental observation of negative temperature dependence in iso-octane burning velocities," vol. 57. American Institute of Aeronautics and Astronautics Inc., 2019, pp. 4476–4481.
- [5] T. Zhang and Y. Ju, "Structures and propagation speeds of autoignition-assisted premixed n-heptane/air cool and warm flames at elevated temperatures and pressures," *Combustion and Flame*, vol. 211, pp. 8–17, 1 2020. [Online]. Available: <https://linkinghub.elsevier.com/retrieve/pii/S001021801930433X>
- [6] X. Gong and Z. Ren, "Flame speed scaling in autoignition-assisted freely propagating n-heptane/air flames," *Proceedings of the Combustion Institute*, vol. 38, pp. 2153–2161, 1 2021. [Online]. Available: <https://linkinghub.elsevier.com/retrieve/pii/S1540748920302455>
- [7] Y. Ju, "Understanding cool flames and warm flames," *Proceedings of the Combustion Institute*, vol. 38, pp. 83–119, 1 2021. [Online]. Available: <https://linkinghub.elsevier.com/retrieve/pii/S1540748920306805>
- [8] Y. Morii and K. Maruta, "What connects ignition and deflagration? – on explosive transition of deflagration," *arXiv*, 12 2022. [Online]. Available: <http://arxiv.org/abs/2212.01978>
- [9] D. G. Goodwin, R. L. Speth, H. K. Moffat, and B. W. Weber, "Cantera: An object-oriented software toolkit for chemical kinetics, thermodynamics, and transport processes," 2021. [Online]. Available: <https://www.cantera.org>
- [10] K. Shimizu, A. Hibi, M. Koshi, Y. Morii, and N. Tsuboi, "Updated kinetic mechanism for high-pressure hydrogen combustion," *Journal of Propulsion and Power*, vol. 27, pp. 383–395, 3 2011. [Online]. Available: <https://arc.aiaa.org/doi/10.2514/1.48553>
- [11] T. Lu and C. K. Law, "A criterion based on computational singular perturbation for the identification of quasi steady state species: A reduced mechanism for methane oxidation with no chemistry," *Combustion and Flame*, vol. 154, pp. 761–774, 9 2008.
- [12] F. Williams, "Chemical-kinetic mechanisms for combustion applications," 12 2016. [Online]. Available: <http://combustion.ucsd.edu>
- [13] Y. Sakai and A. Miyoshi, "Development of reduced chemical kinetics mechanism of gasoline surrogate fuel with oxygenated compounds (in japanese)," *Proceedings of the 29th International Combustion Engines Symposium*, 2018.
- [14] A. Tsunoda, Y. Morii, and K. Maruta, "Counterflow flame behavior at large lewis number around explosive transition of deflagration," *29th International Colloquium on the Dynamics of Explosions and Reactive Systems*, 2023.