

Challenges of the Induction Zone Length Measurements by Laser-Induced Fluorescence of Nitric Oxide in Various Conditions

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1 Introduction

Over the last 20 years, more and more laser diagnostics are employed to characterize the propagation of detonation waves in canonical configurations [1] or detonation engines [2]. Recently, Rojas Chavez et al. [3] conducted induction zone length (IZL) measurements in stoichiometric hydrogen-air detonation, at 18 kPa and 293 K initial conditions, by applying the laser-induced fluorescence of nitric oxide (NO-LIF) technique. The NO-LIF technique is enabled by seeding the mixture with 2000 ppm of NO and the selection of a laser excitation wavelength, at 225.120 nm, for which the LIF signal is closely linked to the NO number density evolution along the detonation wave. Thus, the NO-LIF signal evolution is strongly correlated to the thermodynamic changes across the detonation, enabling a correlation between the LIF signal decay and the end of the induction zone (see [3] for details). This study aims to numerically determine the possibility to perform IZL measurements by NO-LIF in other conditions. It relies on Zeldovich von Neumann Döring (ZND) simulations, coupled with our in-house spectroscopic code, called KAT-LIF, to simulate the NO-LIF profiles, as performed for H₂-air in [3, 4]. The study has two objectives: (i) to assess the challenges associated to the IZL measurements by NO-LIF in four different detonable mixtures: Mixture A: H₂-O₂-Ar (2H₂-O₂-3.76Ar), Mixture B: H₂-NO₂/N₂O₄, Mixture C: CH₄-air, and Mixture D: CH₃NO₂-O₂ at $T = 293$ K and $P = 20$ kPa initial conditions; (ii) to assess the challenges associated to the IZL measurements at higher pressure than 20 kPa for Mixture A: H₂-O₂-Ar.

2 Numerical methods and procedures

Conditions of interest This study is divided into two parts. Firstly, four different stoichiometric mixtures are investigated at $T = 293$ K and $P = 20$ kPa initial conditions: Mixture A (H₂-O₂-Ar), Mixture B (H₂-NO₂/N₂O₄), Mixture C (CH₄-air), and Mixture D (CH₃-NO₂). Mixture A aims to assess the effect of the argon dilution on the IZL measurement method, as well as to validate the technique on more regular mixtures (i.e., lower stability parameter χ) than H₂-air. Mixtures B, C, and D are selected due to their irregular nature (i.e., higher stability parameters χ) than both H₂-air and Mixture A. Also, those mixtures have the propensity to present double cellular structures under different conditions [5–7], as well as presenting strong thermal NO production in the reaction zone (Mixture B and D, mainly) [5, 6, 8]. Mixtures C and D are selected to assess the validity of the IZL determination method in hydrocarbon detonations. Secondly, the IZL determination method is assessed at higher initial pressures, 50 and 100 kPa, with Mixture A only. The compositions of Mixtures B and D are determined according to previous studies [7, 9].

Steady detonation and spectroscopic modeling The ZND simulations are conducted with ZNDkin, as previously employed in [3, 4, 10]. The following reaction models Mével et al. [11], Joubert et al. [6], Mével et al. [12], and Chaumeix et al. [13] are employed for Mixture A, B, C, and D, respectively.

The NO-LIF profiles are simulated with our in-house spectroscopic code, KAT-LIF, already presented and validated in [3, 4, 10]. Only a brief presentation is recalled here. KAT-LIF code is a spectroscopic code enabling the simulation of the NO-LIF profiles, in the linear regime of fluorescence, along a given beam path, and according to a limited number of user-specified inputs, such as thermodynamic parameters (e.g., temperature and pressure), species composition, and laser parameters (e.g., laser excitation wavelength, laser bandwidth, or laser fluence). For frontal orientation of the laser, as defined in [14], the ZND outputs can be directly employed in KAT-LIF to simulate the LIF profiles in detonation waves. All the KAT-LIF simulations consider the same laser parameters and optical paths as in [3].

Induction zone length determination method Figure 1a recalls the temperature (T), pressure (P), thermicity ($\dot{\sigma}$), and the normalized NO-LIF (LIF) profiles, obtained for a H_2 -air detonation at 18 kPa and 293 K and a laser excitation wavelength at 225.120 nm presented in [3]. From the NO-LIF profile obtained in Figure 1a, the IZL is determined by the distance L_{mod} which corresponds to the distance between the LIF signal rise (i.e., defined as the maximum LIF gradient near $X=0$) and 84% of the LIF signal decay (i.e., a drop of LIF signal from 100% to 41%, considering 30% as the minimum LIF signal). Note that the 84% LIF signal decay was determined in [3] and validated on a large range of H_2 -air conditions: $\phi = 0.5 - 2$ and $P = 10 - 100$ kPa. This study aims at assessing the validity and robustness of the definition proposed in [3] to other mixtures and conditions. Figure 1b presents a typical numerical result obtained for the H_2 - O_2 -Ar mixture, where both the thermicity and the normalized NO-LIF evolution are presented as a function of the distance from the shock. The NO-LIF profiles are normalized by the absolute counts obtained in H_2 -air conditions in Figure 1a. The agreement between L_{mod} and Δ_i will be discussed for each mixture, as well as the normalized LIF value for each mixture.

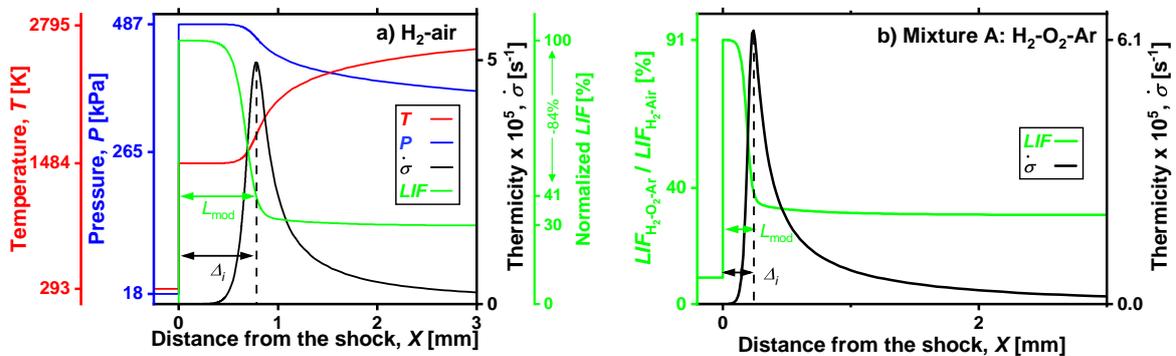


Figure 1: The induction zone length determination method is presented in a), reproduced from [3]. Typical numerical results from this study are represented in b), where the thermicity and the normalized NO-LIF profiles across the detonation are depicted. Negative X values correspond to the fresh gas.

3 Results and discussions

Effect of NO on the detonable mixtures Prior performing NO-LIF simulations, the negligible effect of the 2000 ppm of NO must be confirmed. Figure 2 summarizes ZND results, which aim at evidencing the effect of 2000 ppm of NO on the four mixtures of interest. The symbols depict the numerical results for the neat mixtures (i.e., without NO) and the solid lines represent the numerical results for the seeded mixture. For mixtures A (H_2 - O_2 -Ar), B (H_2 - NO_2 / N_2O_4), and D (CH_3NO_2 - O_2), a negligible

effect of the NO addition is observed on both the Δ_i ($\pm 0.3\%$) and the evolution of the temperature, pressure, and thermicity profiles. The results obtained with mixtures A, B, and D are consistent with our initial findings [3] and the experimental mean cell width results obtained in the same conditions for H_2 -air ($2\text{H}_2\text{-O}_2\text{-}3.76\text{N}_2$) and $\text{H}_2\text{-O}_2\text{-Ar}$ ($2\text{H}_2\text{-O}_2\text{-}3.76\text{Ar}$) mixtures [15]. For mixture C (CH_4 -air), a significant increase (+34%) of the Δ_i is observed, as well as a delayed beginning of the reaction zone. The numerical results obtained for mixture C (CH_4 -air) tend to indicate that a lower NO addition should be employed for enabling the NO-LIF diagnostic in CH_4 -air detonation. Nevertheless, it must be reminded that none of the models available in the literature were validated for NO sensitization in detonation conditions, as discussed for cell size predictions in [15]. Thus, the observed effect of NO could be either due to the validity range of the model itself or due to the effect of NO on the characteristic length scales. The origin of this effect cannot be stated for now and require new experimental data, which are out of the scope of the present study. Note that, a negligible effect of NO is obtained for some other hydrocarbons (e.g., C_3H_8 -air). The IZL obtained with 2000 ppm of NO is used as a reference for the next section in order to assess the robustness of the technique rather than the effect of NO addition.

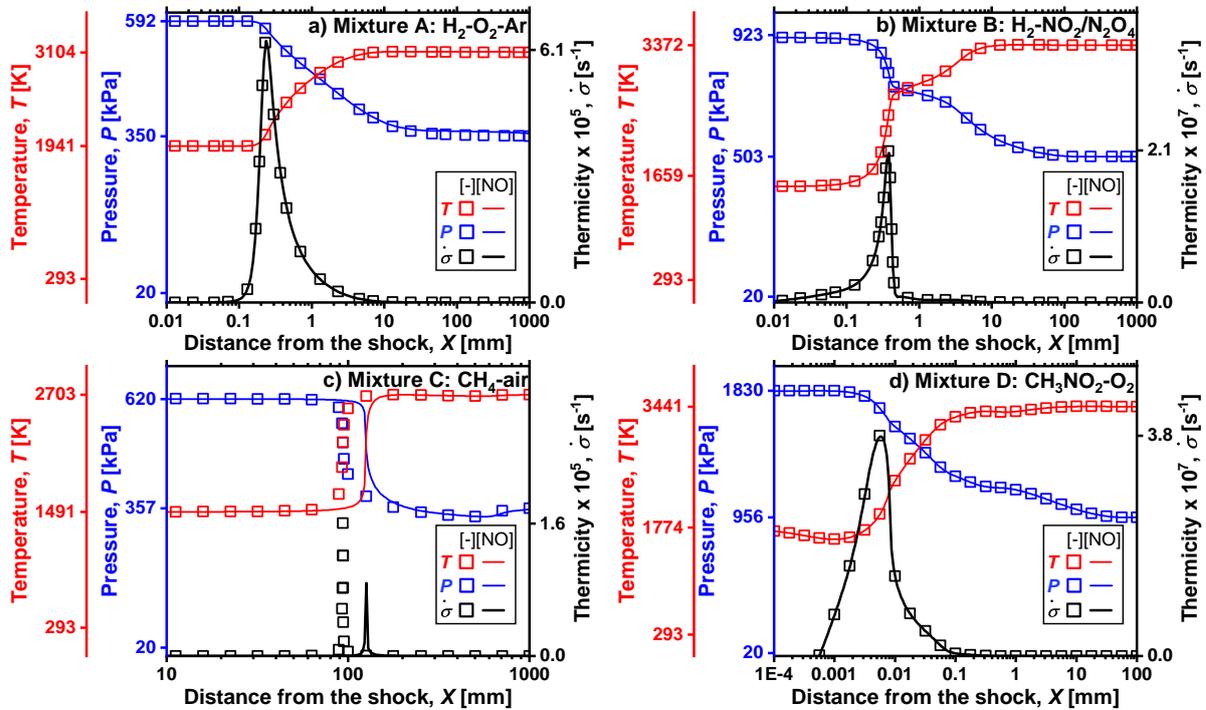


Figure 2: Effect of NO addition on different mixtures at $P = 20$ kPa and $T = 293$ K. Empty symbols and solid lines correspond to conditions without NO and with 2000 ppm of NO, respectively.

Effect of the mixture on the induction zone length measurements Figure 3 presents both the thermicity and the normalized NO-LIF profiles obtained for each mixture. Results can be divided into two categories. First, the IZL measurements are achievable for mixtures A and C with a satisfactory level of signal in the von Neumann state, with less than 30% signal decreased, compared to [3], and a satisfactory accuracy at predicting the IZL (within $\pm 0.3\%$), similar to the values reported in [3]. Surprisingly, a non-monotonic LIF signal decay is observed in Fig.3.c for $0 < X < 130$ mm. This non-monotonic decay can be accounted for the laser energy absorption (from 0 to 100 mm), the thermal NO production from 100 to 120 mm, and the sharp LIF signal decay (near 125 mm) in the reaction zone. Second, the IZL determination method (i.e., -84% of the LIF signal decay) cannot be employed on Mixtures B and D, due to the different evolution of the NO-LIF profile. In fact, the NO-LIF signal is maximized near

the thermicity peak instead of the von Neumann state (i.e., right after the shock). On both mixtures, the changes in the NO-LIF profile evolution are due to a strong thermal NO in the reaction zone, which induces a significantly stronger NO-LIF signal in the reaction zone than in the von Neumann state (near 100 folds). In addition, the detection of both the shock and the thermicity peak does not seem achievable with the current excitation strategies (i.e., 225.120 nm) for Mixture B and D due to the restricted dynamic range of the intensified cameras. Alternative excitation strategies will be discussed in the future to mitigate this limitation.

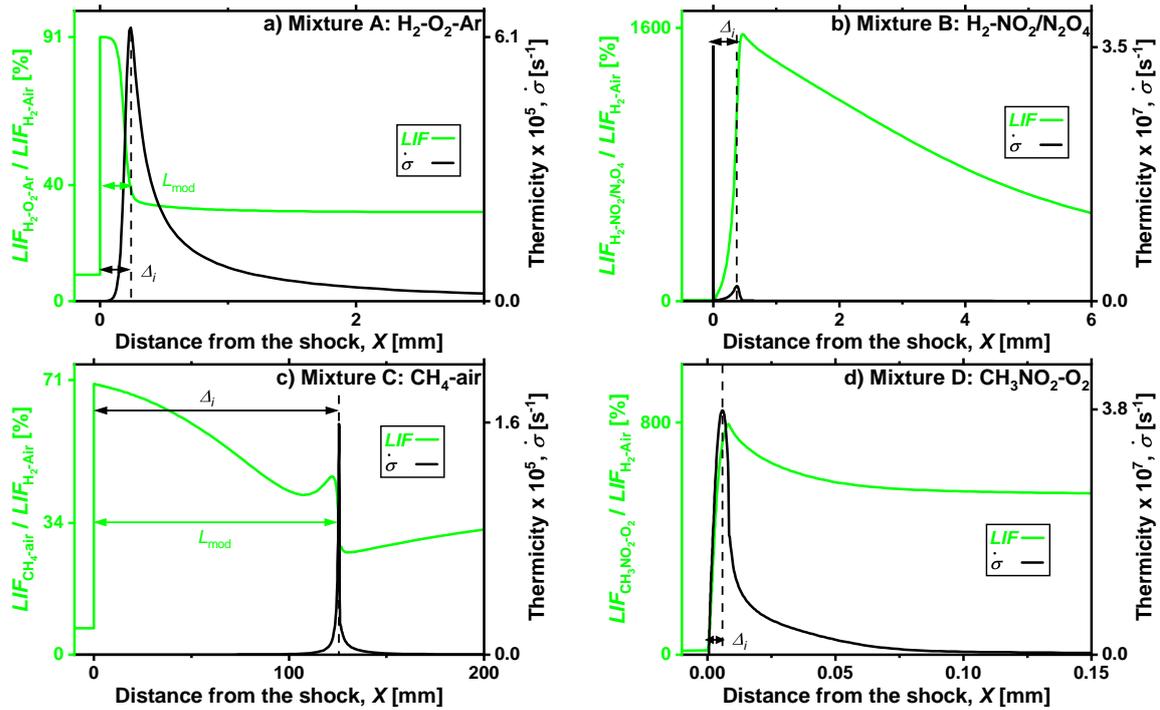


Figure 3: Normalized NO-LIF profile, in green, and thermicity profiles, in black, obtained for each mixture of interest from KAT-LIF and ZNDkin, respectively. Conditions are $\phi = 1$, $P = 20$ kPa, $T = 293$ K. Simulations are normalized by the absolute NO-LIF counts obtained for H_2 -air in [3]. In b), the first thermicity peak is due to high thermicity plateau after the shock, already reported in [6], and is not correlated with the IZL.

Effect of the pressure on IZL measurements Figures 4a and 4b present the simulation results obtained for H_2 - O_2 -Ar mixtures at 50 and 100 kPa, respectively. In both conditions, the IZLs are satisfactorily predicted within 4 and 1% errors for 50 and 100 kPa, respectively. This discrepancy is slightly larger than the one reported at low pressure for H_2 - O_2 -Ar mixture but is in close agreement with the results reported in [3] for H_2 -air. The LIF signal maximum is 2 to 4 times lower than H_2 -air condition at low pressure. The present results indicate that IZL measurements are theoretically possible but less favorable than at low pressure. Experimentally, those measurements may become challenging for high-pressure conditions ($P \geq 50$ kPa), with the present laser excitation wavelength (225.120 nm), due to the lower NO-LIF signal than H_2 -air, the required spatial resolution, and the strong OH^* chemiluminescence. These challenges could be mitigated by employing appropriate filters to suppress most of the OH^* chemiluminescence and by accounting for the transition line pressure shift (i.e., larger than 10 pm at 100 kPa). Thus, alternative laser excitation wavelengths must be determined for different pressure ranges to maximize the NO-LIF signal. Determining these wavelengths and their validity range will be performed in a future study.

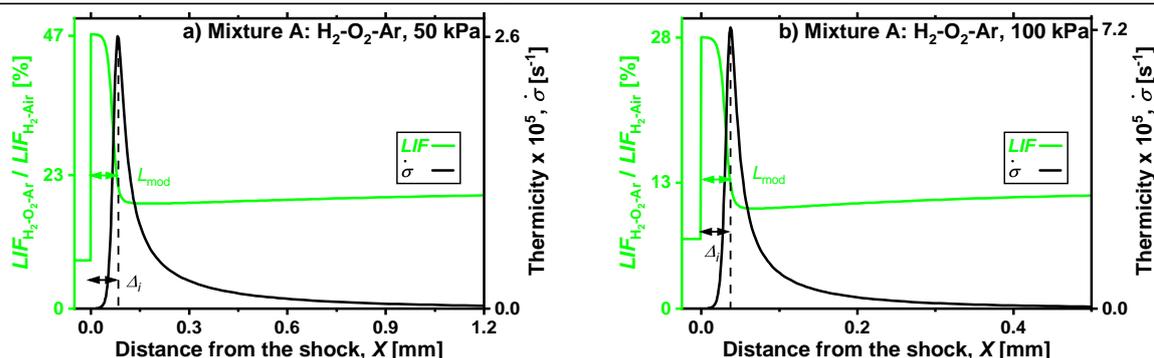


Figure 4: Simulation results obtained for stoichiometric $\text{H}_2\text{-O}_2\text{-Ar}$ at 50 and 100 kPa in a) and b), respectively.

4 Conclusions

This study numerically investigated the possibility to perform induction zone length measurements (IZL) by NO-LIF in various mixture and pressure conditions. The main conclusions of the study are:

- Based on ZND simulations, all the investigated mixtures revealed a negligible effect of the NO addition on the detonation properties, except $\text{CH}_4\text{-air}$. This may indicate that a lower seeding level ($X_{\text{NO}} < 2000$ ppm) may be required for some specific mixtures to enable the IZL measurements by NO-LIF. Experimental investigations, such as [15], are required to confirm this trend, as none of the reaction models was validated for NO sensitization in detonation conditions.
- IZL measurements can be achieved in $\text{H}_2\text{-air}$, $\text{H}_2\text{-O}_2\text{-Ar}$, or $\text{CH}_4\text{-air}$ detonations with a satisfactory accuracy ($< 1\%$) and a satisfactory NO-LIF signal, at 20 kPa initial pressure.
- The IZL measurements cannot be conducted for $\text{H}_2\text{-NO}_2/\text{N}_2\text{O}_4$ and $\text{CH}_3\text{NO}_2\text{-O}_2$ mixtures due to a strong NO production in the reaction zone. This indicates that alternative IZL determination methods (e.g., LIF signal correlation or laser excitation wavelength) must be identified for these two mixtures or, potentially, for any mixture presenting similar strong NO production (i.e., mixtures with nitrogen-containing fuel or NO_x as oxidizer).
- The IZL measurements are determined at 50 and 100 kPa for $\text{H}_2\text{-O}_2\text{-Ar}$ mixture within a 4- and 1%-error, respectively. Note that the NO-LIF signal is 2 to 4 times lower than in $\text{H}_2\text{-air}$, which may limit the application of the diagnostic to high-pressure conditions. Part of this signal loss could be mitigated by accounting for the transition line pressure shift at high pressure.

Thus, it appears that the IZL measurement technique can be generalized to any simple hydrogen or hydrocarbon detonations (i.e., without NO_x as an oxidizer or nitrogen-containing fuel) over a large range of pressure, as long as a low enough NO seeding level is employed to not perturb the detonation properties. Future works will focus on experimentally confirming the effect of the NO addition on the cell size, as started in [15], for different initial pressures, N_2 and Ar dilutions, dilution levels, and fuels in order to confirm the applicability of IZL determination to any detonable mixture. In addition, alternative excitation strategies will be determined in order to mitigate part of the current challenges and limitations of the IZL determination methods (i.e., signal intensity and mixture with strong thermal NO).

Acknowledgments

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