Detonation onset chemiluminescence: an experimental analysis to choose light filters

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1 Introduction

Advancements in light-emitting diodes (LEDs) technology have made them useful for high-speed schlieren applications. They can emit short and uniform bright light pulses without the high cost associated with lasers. To have good image quality, it is necessary to have a high-power LED with narrow emission bandwidth to reduce the chromatic aberrations [1]. Since most of the LEDs are moderately narrowband emitters with an approximately Gaussian spectral shape, it is common practice to use bandpass filters to block longer and shorter wavelengths [2].

By selecting a filter whose transmission spectrum matches the light emitting one, it is possible to improve the contrast of the images and to control the changes that may occur over time in ambient lighting conditions. However, there are some applications where the spontaneous chemiluminescence inherent to the respective process can emit strong light in a large range of wavelengths. This could be a problem during flow visualization using the schlieren technique since the chemiluminescence light may have comparable intensity or even stronger than the LED light, obtaining very bright saturated images with poor contrast. This problem is worsens if the chemiluminescence spectrum matches with the LED spectrum because the filter allows light from both sources to pass through. In this case, the criterion for choosing adequate filters is not so straightforward because if the transmission wavelength range is selected as wide as the LED emission, a large amount of light from the chemiluminescence will pass through the filter. Surprisingly this topic has received very little attention in the literature.

In the present work, a criterion to choose bandpass filters for visualizing detonation onset using schlieren visualization is proposed. To do so, the detonation onset spectra of flames accelerating in tubes and transiting to detonation in $CH_4 + 2O_2$ and $2H_2 + O_2$ mixtures are measured and analyzed. Furthermore, as a preliminary test of the validity of the criterion proposed, schlieren images are compared using different commercial filters and a blue LED.

2 Experimental methodology

Figure 1 shows a schematic of the experimental set-up used. To obtain the light intensity during the detonation process, the detonation is generated in a polycarbonate 10 mm x 10 mm squared channel of 1 m length. For more information about the channel, consult the work made by Ballossier et al.

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[3]. A USB2000+ Ocean Optics spectrometer was used to measure the light intensity. With this is possible to measure the light emission over the range of wavelengths of 200 - 1100 nm, i.e., from ultraviolet to infrared, with an optical resolution of 0.1 nm. The detector correspond to a 2048-element linear silicon CCD array. The spectrometer was connected to a 1000 μm diameter premium optical fiber with a collimating lens used to collect the light in long distances. The lens was placed 60 cm away from the channel, pointing toward the detonation onset distance ($\bar{x}_{\text{DDT}}$) of each mixture. Two reactive mixtures were tested: CH_4 + $2O_2$ ($\bar{x}_{\text{DDT}} = 440$ mm) and $2H_2$ + O_2 ($\bar{x}_{\text{DDT}} = 424$ mm). $\bar{x}_{\text{DDT}}$ was obtained in previous experiments using the schlieren visualization technique. The integration time of the spectrometer was fixed at 2 ms during all the tests. With this integration time it is possible to collect light during the whole flame acceleration process until the detonation onset.

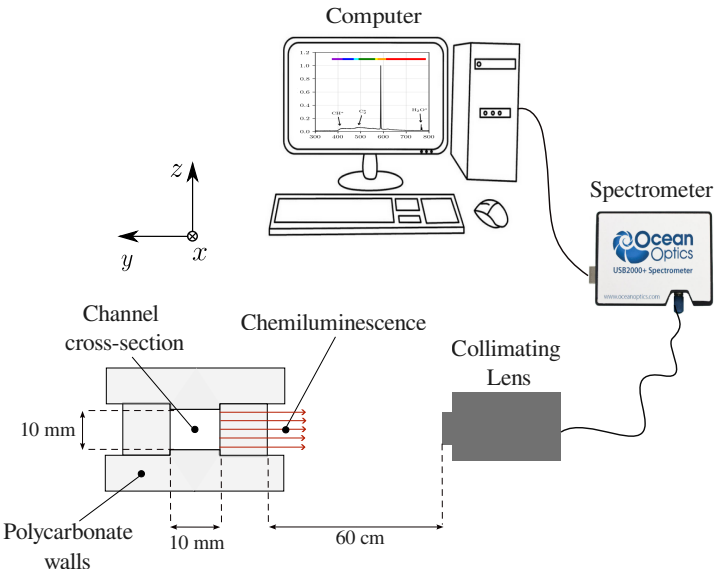

Figure 1: Experimental setup used to measure the light intensity during the detonation process.

3 Results

3.1 Light emission in CH_4 + $2O_2$ and $2H_2$ + O_2 combustion

Figure 2 shows the normalized light emission intensity ($I(\lambda)$) spectrum for (a) CH_4 + $2O_2$ and (b) $2H_2$ + O_2 detonation. The light intensity was normalized with the maximum value for each test. Three replicates were carried out for each condition, showing high reproducibility. For the CH_4 + $2O_2$ mixture, four characteristic peaks are present at 420 nm, 484 nm, 588 nm, and 766 nm. After a literature survey about chemiluminescence for hydrocarbons and hydrogen flames, it is possible to differentiate the following molecules for the wave lengths λ corresponding to: CH^* ($\lambda = 420$ nm), C_2^* ($\lambda = 484$ nm), H_2O^* ($\lambda = 766$ nm) and OH^* ($\lambda = 310$ nm) [4, 5]. The signal of the OH^* was not captured in the present study because the polycarbonate walls block the ultraviolet light. Note that the peak obtained at 588 nm was not reported in the cited papers, and exhibits the largest emission intensity obtained in the current work. Regarding the $2H_2$ + O_2 mixture, two peaks are present at 588 nm and 766 nm; an additional emission line should be present at $\lambda = 310$ nm but is blocked by our experimental setup. The molecules responsible of these peaks are as follows: H_2O^* ($\lambda = 766$ nm) and OH^* ($\lambda = 310$ nm) [6, 7]. Again, the peak obtained at 588 nm was not reported in these works.

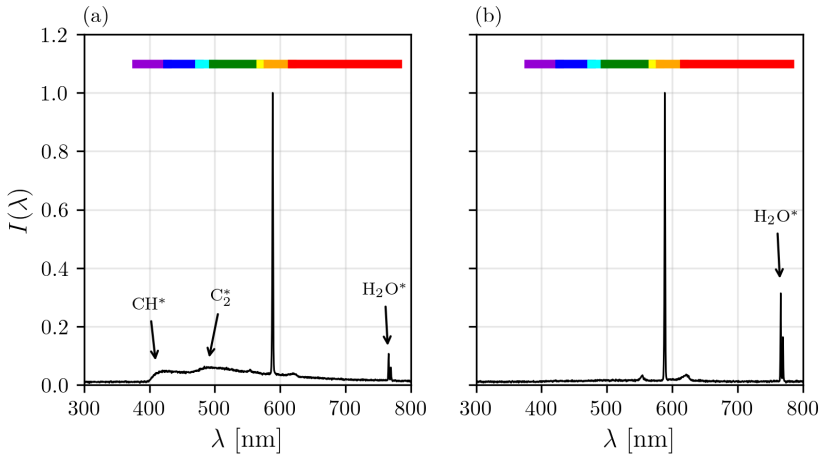

Figure 2: Detonation onset spectra for (a) CH_4 + $2O_2$ and (b) $2H_2$ + O_2 mixtures.

Regarding the origin of the signal centered at 588 nm, it seems there is not a conclusive explanation in the literature. Only two papers report it. The work by Pan et al. [8] presents the spectrum of a diffusion H_2/air flame. A pronounced peak centered at 589.3 nm was obtained. They speculate that this may come from the combustion of sodium in the air since sodium has spectral lines of 589 nm and 589.6 nm. This explanation is plausible for diffusion flames operating in places near the sea, where a high amount of sodium could be found in the air. However, in the present study, the detonation was performed in a closed channel in which, after applying vacuum (with pressures below 10 mbar), was filled with mixtures composed of high purity O_2 (99.9997%), H_2 (99.95%) and CH_4 (99.95%). It is thus unlikely that sodium is responsible for this peak.

In the work by Goyal et al. [9] the light spectrum of propane (C_3H_8)/air flames was recorded under high-pressure conditions (up to 30 bar). The authors measured three excited radicals at 590 nm, 625 nm, and 645 nm but their presence had not been detected nor studied in previous works. Again, 590 nm is the spectral line with the highest intensity. Based on the intensity and wavelengths of these signals, they suggested that it may be due to the emission of C_2^* or water vapor via the H + OH $\rightarrow$ H_2O + $h\nu$ reaction. Since in the present work the peak at 588 nm appears with very high intensity for both H_2 and CH_4 detonation, the last hypothesis is more likely since C_2^* is not produced during the combustion of H_2. Furthermore, the authors showed that these radicals appear after the ignition delay time of the mixture defined based on an arbitrary pressure rise or OH^* threshold. Note that in [9] the experiments were carried out in a closed chamber and the reactive mixture consisted of research plus grade ultra-high purity argon (99.9999 %), nitrogen (99.999 %) and oxygen (99.994 %), and research grade propane (99.99 %). It is thus unlikely that sodium or another airborne sand during their tests be present. Based on these results, it is concluded that H_2-O_2 and CH_4-O_2 detonations are mainly orange in color and it is probably due to the H_2O emission at high pressure.

3.2 Criteria for filters selection

Figure 3 (a) shows the light emission during the detonation of the mixture CH_4 + $2O_2$. In addition, the filters' transmission is presented. The filters properties are shown in Table 1. The acronyms CWL and FWHM are, respectively, the Center Wavelength which represents the wavelength (λ) value where the filter transmission is centered, and the Full Width at Half-Maximum which represents the wavelength's width at half the maximum amplitude of the filter's transmission. The LED spectrum is also shown in

Figure 3 (a) to determine the portion of the LED's intensity that is blocked by the filters.

Table 1: Filters used during the experiments.

Filter	CWL [nm]	FWHM [nm]	Trans. [%]	Brand	Reference
F1	470	155	> 95	ThorLabs	FESH0550
F2	460	25	> 90	Semrock	FF01-460/14-25

Since the spectrometer measures $I(\lambda)$ over a wide λ interval, it is important to compute the light intensity in a specific wavelength range for the LED and the detonation onset's emission. This is carried out using the expression for the total intensity shown in Equation 1 and setting the interval of integration to $[a, b] = [\lambda_1, \lambda_2]$ which corresponds to the area under the $I(\lambda)$ curve in the respective range. To have the best image contrast, the filter should allow to pass through as much light as possible coming from the LED but as little light as possible coming from the detonation spontaneous chemical emission, i.e., chemiluminescence. The latter requirement translates into maximizing the LED's light to detonation chemiluminescence ratio, $I_{\text{led}}/I_{\text{deto}}$, in the transmission wavelength range of the filters. This can be estimated via Equation 1. Since the analysis is based on the area under the curve of the light emission spectra, it is possible to create an ideal filter based on a qualitative approach. Ideally, this filter should have the same CWL of the LED spectrum, and with the methodology proposed the FWHM that maximizes both the $I_{\text{led}}/I_{\text{deto}}$ and $\%\ I_{\text{total,led}}$ can be estimated. However, to avoid having to order a custom filter we constrained our selection to commercially available filters for which the CWL and FWHM change simultaneously.

$$I_{\text{total}} = \int_{a=-\infty}^{b=+\infty} I(\lambda) d\lambda; \quad \lambda_{1,2} = CWL \pm \frac{FWHM}{2} \tag{1}$$

Figure 3 (b) shows $I_{\text{led}}/I_{\text{deto}}$ for filters F1, F2 and without filter; the percentage of the total LED intensity $\%\ I_{\text{total, led}}$ that passes through the filter is also reported. The latter is of interest since dark images are expected for low $\%\ I_{\text{total,led}}$. The $I_{\text{led}}/I_{\text{deto}}$ ratio is 0.27 and 1.07 for F1 and F2 filters, respectively, and 0.12 without filter. $\%\ I_{\text{total,led}}$ is $98\ \%$ for F1, $59\ \%$ for F2, and $100\ \%$ without filter, as expected. It means that for the filter F2 higher contrast but lower luminosity is expected in the images since the $I_{\text{led}}/I_{\text{deto}}$ is bigger but $\%\ I_{\text{total,led}}$ is lower compared with filter F1.

Figure 4 shows schlieren images of the detonation onset (DO) process for the CH_4 + $2O_2$ mixture without filters and using both filters. The camera exposure time (110 ns) and framing rate (5 MFPS) were kept constant to better isolate improvements coming from the use of the filters themselves. In Figure 4 the images reported at time $t = t_{ref}$ correspond to the moment when the DO occurs for each test. The DO is visualized as a white spot due to the high luminosity which saturates the camera. Note that different amounts of DO points occurred for each test, i.e, $1, 2$, and 3 for the Figure 4 (a), (b) and (c), respectively. For the last one, the third point is not visualized at $t = t_{ref}$ but the Blast Wave (BW) is visible after. At $t = t_{ref}$ the Precursor Wave (PW) is visualized for all the tests since the chimiluminescence is not so high at this stage. However, for filter F2 less bright images are obtained due to the lower $\%I_{total,led}$ (see Figure 3 (b)). Nevertheless, when comparing the images obtained $3.0\ \mu$s after DO for each test, much more contrast for the recorded with the filter F2 is obtained, being possible to observe the BW/burned gas interaction. In contrast, the images obtained with the filter F1 and without the filter are very bright and do not allow to observe of any detail, due to the low $I_{\text{led}}/I_{\text{deto}}$ relation, as presented in Figure 3 (b). Note that the λ range of the filter F1 fits well with the range for the blue LED (see Figure 3 (a)), meeting the condition that is normally required for the band-pass filter´s selection. However, due to the low $I_{\text{led}}/I_{\text{deto}}$ ratio very bright images are obtained due to the

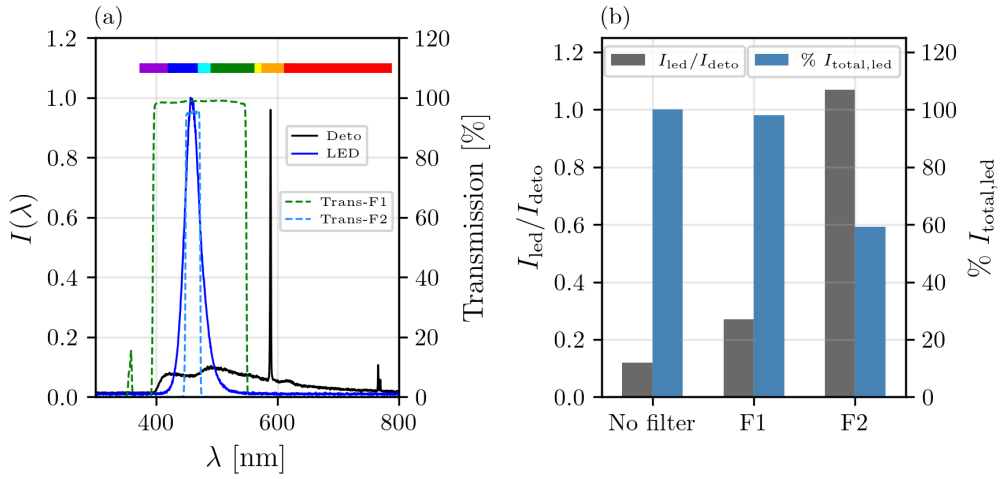

Figure 3: (a) LED and detonation light emission spectrum of CH_4 + $2O_2$. (b) I_{led}/I_{deto} and % $I_{total,\,led}$ for filters F1, F2, and without filter.

high chemiluminescence that passes through the filter. Our preliminary results suggest that the criterion developed here for choosing filters that maximize the I_{led}/I_{deto} ratio together with the % $I_{total,led}$ is one key to improve the image quality in a process with high spontaneous chemiluminescence occurring at similar wavelengths of the light source used for the flow visualization technique; schlieren in our case.

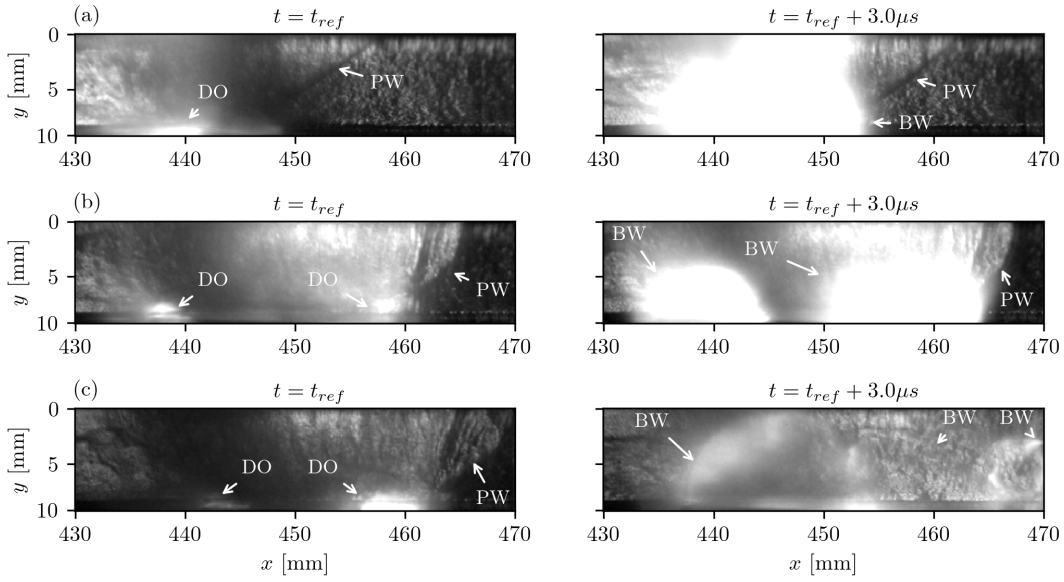

Figure 4: Schlieren images during the detonation onset in CH_4 + $2O_2$ mixture (a) without filter, (b) with filter F1, and (c) with filter F2. The acronym stands for DO: Detonation Onset; PW: Precursor Wave; BW: Blast Wave.

4 Conclusions

The light emission spectrum of detonation onset in a narrow channel is measured and analyzed for CH_4 + $2O_2$ and $2H_2$ + O_2 mixtures. Comparing it with the emission spectrum of the light source (blue

LED) used for schlieren visualization it was possible to assess the criterion proposed for bandpass filter selection. The main learnings from this study are:

- There is a pronounced emission peak at $\lambda = 588$ nm in the spectrum measured for both mixtures tested that has not received much attention in the literature. It is likely due to the light emission from reaction H + OH $\rightarrow$ H_2O + $h\nu$.
- Maximizing the $I_{\text{led}}/I_{\text{deto}}$ ratio and the $\%I_{\text{total,led}}$ value resulted in a significant improvement in the quality of the images obtained using schlieren visualization.
- The methodology proposed in this work allows for the optimization of the filter selection by playing with the filter's parameters. This is important since it is possible to select the best filter for the respective application without testing it under experimental conditions.

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