

Particle Thermometry in Radiatively Ignited Explosive Mixtures

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1 Introduction

Fine powders can induce ignition of a surrounding explosive charge when heated rapidly by radiation. In vapour cloud explosions, particles likely to be present in the unburnt gas mixture can be ignited upon radiation from the main flame front [1–3]. Similarly, light emanating from a faulty optical fibre can heat fine dusts or single particles leading to accidental ignitions [4–7]. Furthermore, Atkinson and Cusco [8] have suggested that the forward thermal radiation emitted from the main flame during a vapour cloud explosion may potentially influence the corresponding flame propagation. However, in practical incidents, these radiatively induced ignition kernels have to be formed within reasonably short timescales and well in front of the advancing flame in order to have a notable effect. Recently, Beyrau et al. [9] have shown that laser irradiated powders can indeed ignite an adjacent explosive atmosphere using a near infra-red (NIR) laser source. An experimental study was carried out which examined fine powders with very different characteristics (type, size, morphology, etc.) and the corresponding ignition times established. Ignition times $\simeq 100$ ms were obtained at an irradiance < 600 kW/m² using a commercially available carbon black powder (acetylene black) coated on a glass substrate. Although the actual level of thermal radiation emitted during a real incident remains conjectural, findings suggest that radiation induced ignition in typical vapour cloud explosions can be significant.

The objective of the current study is to investigate further the relationship between absorption-emission properties of different powders by the determination of particle temperatures under different irradiance using emission spectroscopy (ES) [10]. Visually black materials used by Beyrau et al. [9] like carbon black and graphite feature a relatively well defined emissivity, hence, particle thermometry is used to obtain useful information on this novel ignition mechanism.

2 Experimental Configuration

The experimental facility used for these experiments is shown in Fig. 1. A stainless steel flame tube with a 90 x 60 mm rectangular cross-section and 240 mm length was used filled with stoichiometric butane-air mixtures. The desired equivalence ratio was achieved using the method of partial pressures measured using a pressure transducer. Mixture homogeneity was achieved after circulating the mixture for 70 flow-through times in a closed loop. For a butane-air mixture of unit stoichiometry, the error in the equivalence ratio was less than ± 2 %. The open end of the tube was temporarily sealed using a thin aluminium foil that provided sufficient sealing for mixture preparation. Upon ignition, the pressure

rise, caused by the propagating flame, bursts the foil avoiding a significant pressure increase inside the flame tube. Candidate particulates were placed at the centre of the flame tube and illuminated by a NIR continuous wave laser diode (808 nm, 50 W). The spatial distribution of laser power was determined using a CCD camera, a maximum of 1.6 MW/m^2 over an area of approximately 32 mm^2 could be delivered. These boundary conditions were used throughout the current study unless otherwise stated and are consistent with Beyrau et al. [9]. Ignition times were obtained from the camera images recorded at a rate of 208 fps corresponding to an uncertainty of $\pm 5 \text{ ms}$. This value includes the particle and gas heating times – until the surrounding fuel-air mixture ignites and is hence different from a conventional induction time.

Time-resolved spectroscopic measurements were performed using an Ocean Optics Red Tide 650 spectrometer (350 - 1000 nm) fitted with notch filter (808 nm) to suppress the scattered laser light. The spectrometer collected the emitted radiation from the illuminated powder at a rate of 80 fps using an integration time of 3 ms. The highest rate of temperature rise obtained was of the order of 100 K per exposure (corresponding to 3 ms). Hence, we believe that the integration time used was sufficiently short. The spectrometer spectral response was corrected using a calibrated light source. Particle temperatures were obtained using ES, a non-intrusive optical diagnostic technique which has been successfully used for measuring soot temperatures in flames. The technique is based on the analysis of the spectrally resolved emitted radiation. Temperatures were obtained by fitting of the theoretical spectra according to Planck's law of radiation to the experimentally determined one over a spectral range of 400 - 780 nm. More details on the method can be found elsewhere [11, 12]. For the current investigation the emission coefficient for all carbon based powders was assumed to be $\epsilon = 1$, this is a good approximation for visually black matter. In the next section, temperatures are presented with the average value obtained from three measurements at each condition.

3 Results & Discussion

Temperature histories of two different carbon blacks, acetylene black and Printex XE2-B, obtained in air using an irradiance of 1.6 MW/m^2 can be seen in Fig. 2a. The vertical dashed lines represent the average times to ignition ($\tau_{ign, Ace} = 20 \text{ ms}$ and $\tau_{ign, XE2B} = 1.1 \text{ s}$) obtained by Beyrau et al. [9] in a stoichiometric butane-air mixture. Moreover, these were the extremes measured with different powders, coated on a glass substrate, using the same irradiance. Emission spectroscopy can provide useful information in terms of equilibrium temperature and rate of temperature rise with both powders. However, there is a lack of data points during the initial heating stages. This poses a limitation for the case of acetylene black since the ignition of the surrounding charge takes place within this period. Reasons behind this deficiency are discussed later in the text. Scanning Electron Microscope (SEM)

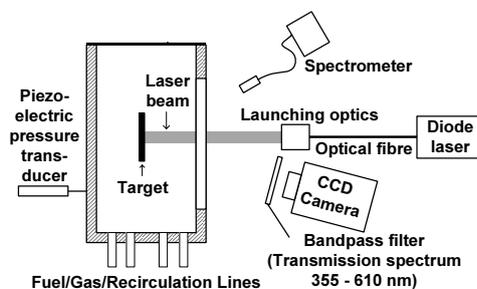


Figure 1: Schematic of the experimental configuration.

images have previously shown that Printex XE2-B is characterised by a higher level of primary particle fusion; this minimises the overall surface area and increases the mass and thermal capacity [9]. This results to a lower rate of temperature rise compared to acetylene black while the longer ignition times may also relate to the lower equilibrium temperatures achieved. Temperature histories obtained with natural and synthetic graphite powders can be seen in Fig. 2b along with their corresponding average ignition times in a stoichiometric butane-air mixture ($\tau_{ign, GS} = 65$ ms and $\tau_{ign, GN} = 130$ ms). Both graphite powders show a lower rate of temperature rise and lower equilibrium temperature than acetylene black. Ignition times obtained with the synthetic graphite in butane-air were approximately half of those obtained with the natural equivalent. The difference in equilibrium temperatures may relate to the absorptivity of the individual powder.

Higher irradiance yields higher equilibrium temperatures as well as rate of temperature rise; both directly relate to the time to ignition of an adjacent charge. The effect is shown in Fig. 3a where acetylene black temperatures measured in air are presented. Interestingly, results obtained even with the lowest flux are well above the high temperature ignition temperatures promoted by chain branching. Equilibrium temperatures reached in air at various irradiance for acetylene black are compared with data from Adler et al. [4] obtained with soot coated fibrous targets heated by light of a comparable wavelength (1064 nm). Temperature dependence on irradiance shows a similar trend for the two materials as it can be obtained from Fig. 3b. However, it is also obvious that the current ES technique over-estimates temperatures since results are higher than the theoretical $I = \varepsilon\sigma(T^4 - T_o^4)$. We believe this is due to the assumption of the emission coefficient $\varepsilon = 1$ used. Thus an improved determination of the individual powder emissivity would most certainly enhance the accuracy of the experiment. However, we also believe that the temperature is only slightly over-predicted and that has a minor effect in the following discussion.

Ignition temperatures for various carbon based powders were obtained in a surrounding stoichiometric butane-air mixture under identical irradiance. Ignition temperature, in this case, refers to the particle temperature obtained using ES just before the appearance of visible flame in the camera images. For acetylene black, $\tau_{ign, Ace} = 20$ ms, the experiment is limited by the signal strength prior to ignition thus reliable measurements could only be obtained after ignition. Nevertheless, it can be reported that for this carbon black which features the shortest ignition time, ignition temperature appears to be below 1720 K. The lack of signal strength prior to ignition is a combination of the spectrometer's sensitivity and the low ignition temperature. From Wien's law, the wavelength peak of the spectral irradiance of a black body at 1720 K is 1.68 μm and also most of the irradiance is emitted in the NIR part of the spectrum. Thus, the current visible spectrometer is unable to collect sufficient light. Longer integration times could be selected to increase the amount of photons collected, however, this would comprise the time resolution. As the soot temperatures rise both the peak wavelength and emitted irradiance shifts towards the visible part of the spectrum enhancing the accuracy of the experiment.

In contrast, with Printex XE2-B there is sufficient signal before and after ignition – until the aluminium foil bursts, enabling reliable temperature measurements. A typical temperature history is shown in Fig. 4b, for this specific experiment $\tau_{ign, XE2B} = 940$ ms. Printex XE2-B featured an ignition temperature of 1971 K which appears to be, in the best case, approximately 250 K higher than acetylene black. This shows that the particle temperature is not the sole ignition criterion. The corresponding camera images of mixture ignition by Printex XE2-B on coverslip can be seen in Fig. 4a. Spectra obtained before and after ignition are presented in Fig. 5a and 5b respectively. Interestingly, sodium lines could be observed in the volume of burned gas. These were also observed for other powders, albeit much less pronounced. The colour of the flame suggests strong presence of sodium. Ignition temperatures around 1826 K were obtained with Printex 80, $\tau_{ign, P80} = 85$ ms, 1760 K with natural graphite, $\tau_{ign, GN} = 130$ ms while with the synthetic graphite, $\tau_{ign, GS} = 65$ ms, ignition temperature appeared to be below 1710 K since the signal strength was limited before ignition. Finally, previous SEM analysis [9] has indicated that the morphological features of acetylene black closely resamples car exhaust

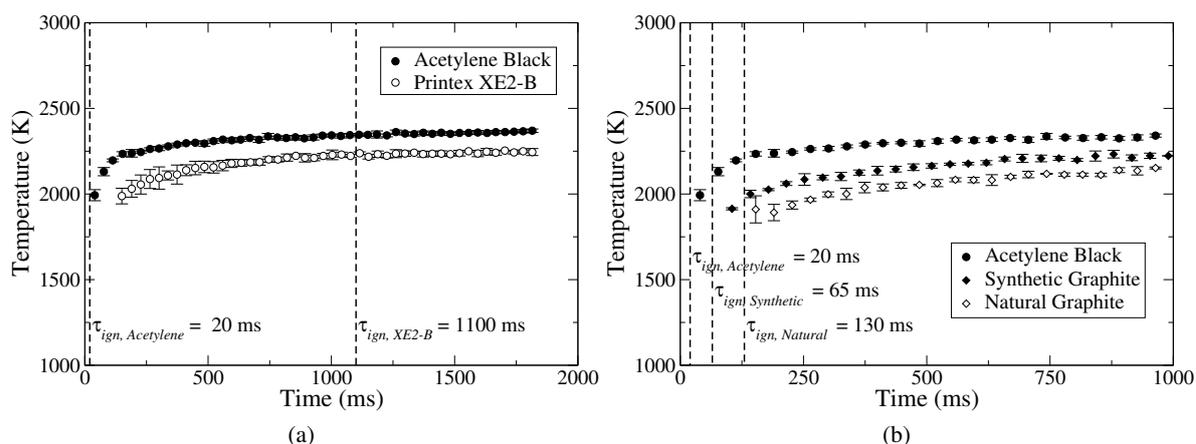


Figure 2: Temperature histories of: (a) acetylene black and Printex XE2-B and (b) natural and synthetic graphite on a microscope coverslip in air, irradiance = 1.6 MW/m^2 . Ignition times (τ_{ign}) correspond to the times obtained in a stoichiometric butane-air mixture using the irradiance.

related particles. Results presented in the current study revealed that this type of particles show the most favourable ignition characteristics with low ignition temperatures, in addition to the shortest ignition times.

4 Conclusion

Emission spectroscopy, a method for temperature measurements in sooting flames, has been used to determine the temperature of irradiated powders under radiation levels relevant to radiation induced ignition in flammable atmospheres. Graphite powders showed both a lower rate of temperature rise and equilibrium temperature than acetylene black when irradiated in air. The temperature dependence on radiation flux showed a similar trend with soot coated fibrous targets [4], albeit the current ES technique overestimates temperatures. A reliable validation method is required to allow the correction of this offset. Ignition temperatures were also obtained in a stoichiometric butane-air mixture. The temperature appears to be not the only ignition criterion indicated by the different values measured with powders that feature similar times to ignition. For powders featuring the shortest ignition times, reliable measurement could not be obtained due to limited signal strength prior to ignition. Car exhaust related particles show the most favourable ignition characteristics with low ignition temperatures. Further particle temperature spectroscopy will be carried out in flammable atmospheres with a NIR spectrometer. This may enable the measurement of ignition temperatures where the signal strength is not sufficient with the current visible spectrometer.

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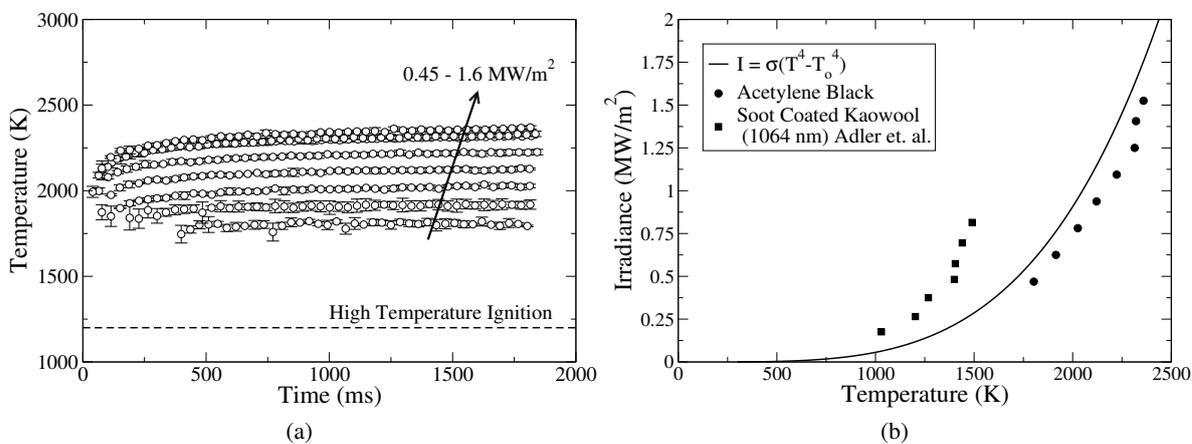


Figure 3: (a) Temperature histories and (b) equilibrium temperatures for various irradiance using acetylene black on a coverslip in air.

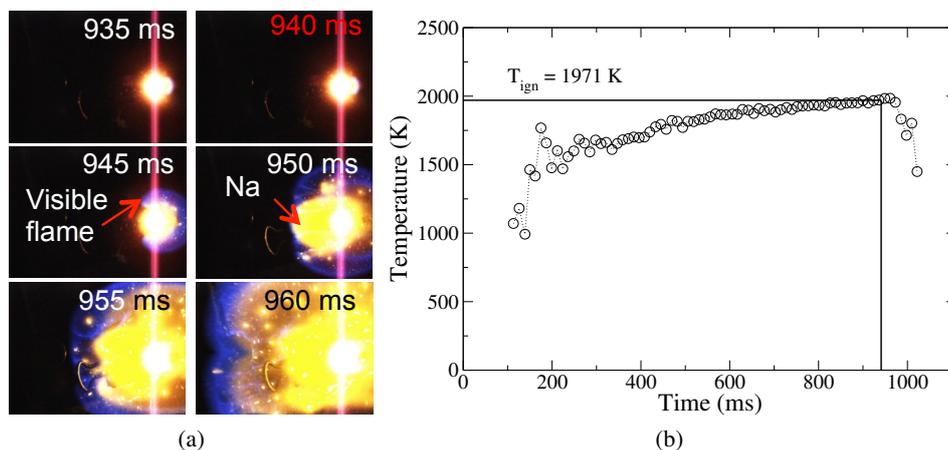


Figure 4: (a) Images of stoichiometric butane-air mixture ignition by Printex XE2-B on a coverslip, irradiance = 1.6 MW/m². For this specific experiment $\tau_{ign} = 940$ ms. The colour of flame suggests strong presence of sodium. (b) Temperature history, note that temperature measurement could be obtained before and after ignition - until the aluminium foil bursts.

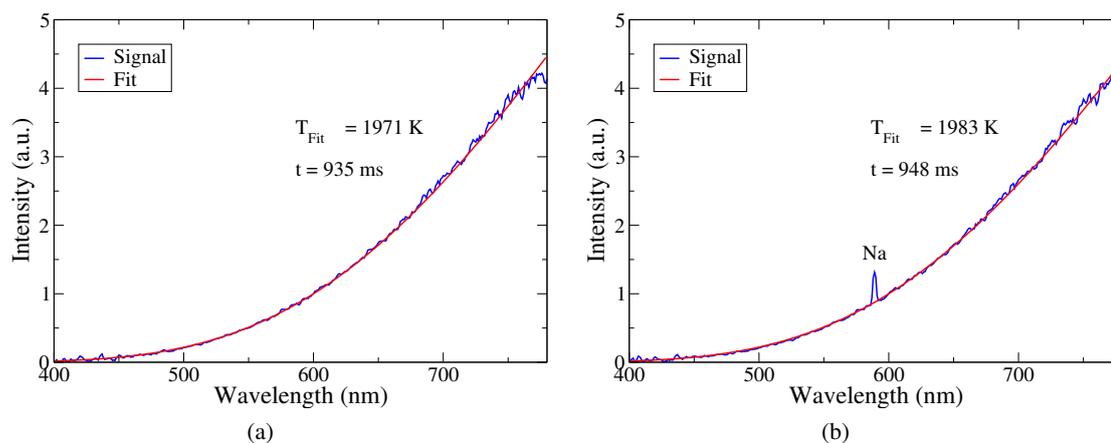


Figure 5: Emitted spectra measured using Printex XE2-B on a coverslip in a stoichiometric butane-air mixture, irradiance = 1.6 MW/m^2 . (a) Ignition temperature = 1971 K (obtained shortly prior to ignition); (b) spectra emitted shortly after ignition, a sodium line can be observed in the volume of burned gas.

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