# Visualisation and Characterisation of Flame Radical Emissions through Intensified Spectroscopic Imaging

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**Abstract**. Combustion flames contain strong emissions from excited radical species produced by the combustion process. The monitoring and characterisation of such emissions is important for an in-depth understanding of fuel energy conversion and pollutant formation processes. In this paper, an approach combining emission spectroscopy with intensified digital imaging techniques is proposed for visualising and quantifying the radiative characteristics of free radicals of combustion flames. Recent advances in CCD technology, especially in EM image intensification, have made it possible to obtain high resolution emission images of isolated spectral emissions from particular flame radicals. These can be used to study emission intensity and distribution, with the aim of correlating combustion emission products with flame spectral emission patterns.

## 1. Introduction

Air pollution from fossil fuel fired power plant emissions is a long-standing environmental concern, and technologies that can increase combustion efficiency and reduce harmful emission are continually being sought. Information about combustion emissions is usually obtained through flue gas analysis, but there can be a significant delay between the combustion process at a particular time and the acquisition of the flue gas analysis arising from that combustion. In addition, the uncertainty of the fuel being burnt means that flue gas analysis is an unreliable way of optimising combustion parameters, as in general it will not be indicative of the immediate burner conditions. One strategy to determine combustion emissions is to correlate them with the presence and distribution of certain chemical species in the combustion flame. It has been established that certain intermediary radical species in the flame such as $OH^*$, $CH^*$, $C_2^*$ and $CN^*$ (when there is nitrogen content in the fuel) are involved in the chain reaction mechanisms, and play important roles in the formation of combustion emissions such as NOx and unburnt particulates [1]. There are several techniques to identify these chemical species in the flame, the most common being emission spectroscopy and laser based techniques (such as laser induced fluorescence) [2-4]. In conventional emission spectroscopy, the emission data are collect by detecting the light from a particular region of the flame through a monochromator in order to give an overall value for the magnitude of light, or using an array of light sensitive diodes to capture the light from different parts of the flame, giving a limited amount of spatial resolution [5]. Laser based techniques require an external excitation source which may be intrusive to the flame, and are generally used in laboratories, being unsuitable for practical combustion systems due to their complex system set-up and high capital cost. Attempts have also been made to use the chemiluminescence of radicals, especially $OH^*$ and $CH^*$, to infer combustion characteristics

including localised heat release [5, 6], equivalence ratio [7], and NOx emissions from hydrocarbon fuels [6].

Although there have been studies suggesting correlations between flame radical emissions and various combustion parameters, a comprehensive description of the chemical processes governing the formation of these radicals is not yet fully understood. Difficulties in obtaining simultaneous high resolution images of the emission from various combustion radicals of interest have limited the scope of the investigation. This paper presents the visualisation and characterisation of flame radicals using emission spectroscopic and imaging techniques. A combination of optical splitting and filtering, image intensification and digital signal processing techniques offers a promising solution for measuring the emission profiles of flame radicals (OH*, CN*, CH*, and $C_2$*) simultaneously and continuously. Technical issues, including currently available image intensification techniques, are addressed. Preliminary results obtained on a laboratory-scale gaseous burner are presented and discussed.

## 2. Methodology

### 2.1. Spectral characteristics of a flame

The combustion of a fossil fuel is a complex chemical/thermal reaction. The characteristics of the flame can be identified by the nature and degree of the spectral emissions from fuel molecules/particles participating in the exothermic reactions. The emission spectra given off by the flame contain two different elements, i.e., emission lines from the species participating in the chemical processes (discrete or band spectra) and the underlying blackbody curve (continuous spectra). The continuous spectra in the flame are generally observed in the luminous region where radiative energy emitted by soot and other solid particles is distributed in a common manner over a broad spectral range, and complies with the Planck radiation theory. The discontinuous spectra are attributed to isolated atoms or free radicals (intermediate species) in the flame. When the fuel reacts with oxygen at high temperature, a number of free radicals are produced in excited electronic states, and are characterised by strong spectral bands. The spectral bands of hydrocarbon flame emissions in the 250-600nm spectral region are dominated by OH* (308nm), CN* (387nm), CH* (432nm) and $C_2$* (514nm) [8] which are the interests of this investigation.

### 2.2. Technical approach

Figure 1 is the block diagram of the technical approach for the monitoring and quantifying the radiative characteristics of the flame free radicals. A major challenge in the design and implementation of the system lies in the simultaneous acquisition of multiple images of flame radicals using a single camera. The problem can be solved through the use of a dedicated optical splitting unit which divides the light of the flame into four identical beams, each filtered by a band-pass optical filter for the spectral range of the chosen radical. The radiative signals of the radicals are then collected concurrently by a CCD camera and transmitted to a computer system. Digital image and signal processing algorithms are developed to 'fingerprint' the radicals and to quantify the emission characteristics of the radicals based on their characteristic parameters. In addition, as the flame temperature and its distribution is one of the most important factors relating to the emission formation, the flame temperature is measured simultaneously using images captured by a RGB camera from an additional optical pass based on multi-colour pyrometry [9]. The quantitative relationships between the emission characteristics of the radicals and the flue gas emissions (e.g., NOx and unburn particulates) can then be achieved by analysing the signatures of the radical emissions with reference to verified historical data of flames, fuels and air supplies.
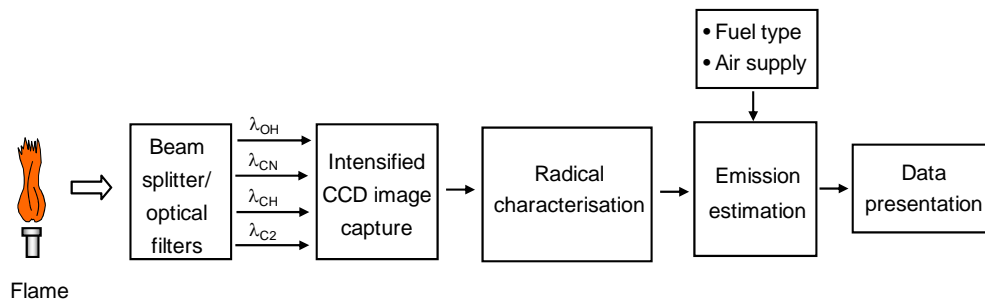
**Figure 1**. Block diagram of the proposed imaging system.

It has been understood that the radiative intensities of the radicals in the flame are relatively weak. Furthermore, the optical splitting and filtering arrangement considerably attenuates the intensity of spectral emissions from the radicals. Some form of image intensification must, therefore, be employed to amplify the signals. Technological improvements have made it increasingly feasible to use CCD technology in order to capture the low intensity of the spectroscopic emissions. There are two main types of intensification available.

*a) Photomultiplier tube image intensifiers*
The photomultiplier tube is the most established method of image intensification. In this type of system, a high voltage in the photomultiplier tube causes incoming photons to create high energy electrons that in turn strike a phosphor screen. This results in more light than from the original source photons, and the intensified signal is then captured by a CCD array. These imaging systems, often referred as ICCDs, can be very sensitive, but the resolution of the final image is often lower than in a non-intensified image, and noise levels can be high, often causing large snowy flecks in the image due to spurious electrons striking the phosphor screen.

*b) Electron multiplication image intensifiers*
Electron multiplication (EM) has recently become available for CCD cameras. In this system (referred as EMCCD), the charge built up in each pixel due to the incoming light is multiplied by using a higher than normal voltage as it is shifted it along the CCD array towards the output register. Extra electrons are generated, resulting in a higher total charge produced from the pixel. They are able to produce a high resolution image, with light sensitivity approaching that of photomultiplier tube intensification.

## 3. Preliminary Results
Choosing a suitable image intensification technique is crucial for the successful development of the proposed imaging system, and thus experiments were undertaken to examine the suitability of commercially available ICCD and EMCCD cameras. A portable propane/butane burner was used to generate a premixed flame. Radiative images of the flame at wavelengths corresponding to OH* (308nm), CH* (387nm), CN* (432nm) and $C_2$* (514nm) were obtained using interference filters with 10nm FWHM (Full-Width Half-Maximum). Both ICCD and EMCCD cameras were used to capture the images. The ICCD camera was equipped with a single stage MCP (Micro-Channel Plate) intensifier and a 12-bit CCD sensor. Figure 2 shows images captured by the ICCD camera with an exposure time of 2ms using maximum gain. In all cases, the strongest emission is observed at the area close to the nozzle outlet. Noise from the intensifier is, however, evident as large flecks in the image, as is characteristic of this type of intensification, and in some cases it is difficult to differentiate between radical emission and noise. Figure 3 shows an example of the image quality available from the EMCCD camera (16-bit) with a 500μs exposure time. Although some fine grain is evident, slightly greater detail of the emission zone is seen. This may be a benefit of the self-cooled design of the EMCCD camera, where the number of the hot/dark pixels is minimised by keeping the sensor at -75C to reduce the the dark current. Also, EMCCD sensors are less likely to be damaged by exposure to

excess light (except strong laser) --all that will happen is that the sensor will saturate. This is particularly useful for a highly fluctuating light source such as a combustion flame.
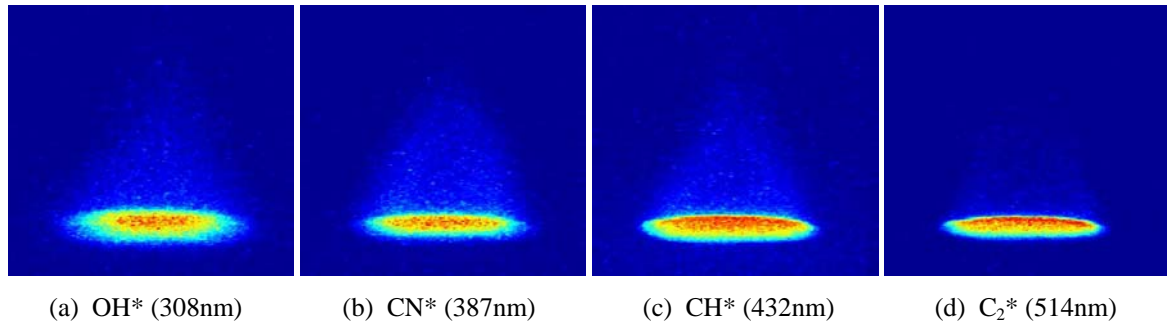


<div align="center">(a) OH* (308nm)    (b) CN* (387nm)    (c) CH* (432nm)    (d) $C_2$* (514nm)</div>

**Figure 2.** Images captured by the ICCD camera.



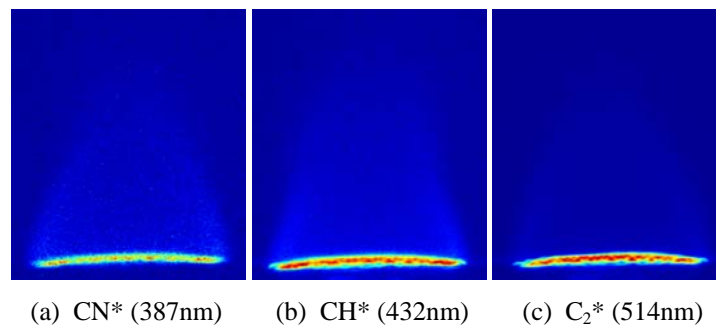<div align="center">(a) CN* (387nm)    (b) CH* (432nm)    (c) $C_2$* (514nm)</div>

**Figure 3.** Images captured by the EMCCD camera.

As previously described, the emission from radical species in the flame produces narrow bandwidth peaks which are superimposed over the underlying blackbody (Plank) spectrum (section 2.1). In order to isolate the background emission from the radical peaks, images from neighbouring wavelengths in an off-peak region of the emission spectrum can be subtracted from the on-peak image. For a premixed gaseous flame, the background emission is of negligible intensity. This is evident in Figure 4, where radiation intensities of the radicals can be compared to the intensity from neighbouring off peak bands, shown in a 3-D plot of the data. Comparing the emission from the $C_2$* peak at 514nm with that of a neighbouring off peak region at 490nm shows that the off peak contribution may be considered negligible. Similarly, the CH* radiation at 432nm is much stronger than the contribution from 410nm. For a coal-fired flame, however, the superimposition effect may not be negligible as there is a large amount of soot in the flame, emitting strong blackbody radiation through the UV to visible and extending to IR range. In this case, a technique that can minimise the disturbance from soot incandescence may have to be employed. The CN* at 387nm is relatively weak. However, the primary mechanism for CN* radical formation is through fuel nitrogen, and the fuel used (propane/butane mixture) has negligible nitrogen content to support CN* radical formation.
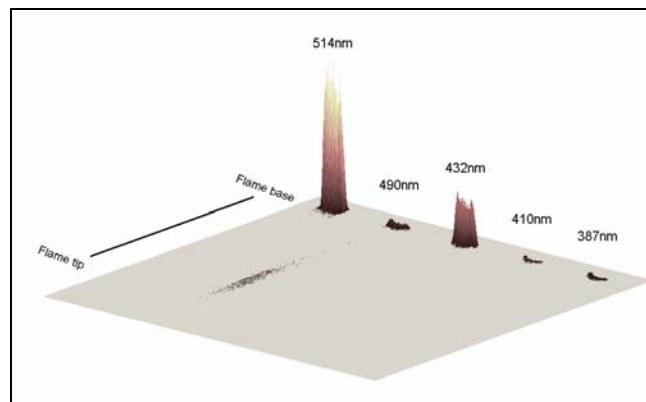
**Figure 4.** Flame radiation intensities.

## 4. Conclusion

An approach combining emission spectroscopy with intensified digital imaging technique has been described for visualising and quantifying the radiative characteristics of free radicals of combustion flames. Recent advances in CCD technology, especially in electron multiplication intensification, has established CCD sensing as a viable method of studying spectral emission from excited flame radicals. Preliminary results indicate that images obtained from electron multiplication intensification are of equal or better quality than those obtained using conventional photomultiplier tube intensification. Future work will focus on the characterisation of radical emission intensity and distribution, and using the information obtained to establish quantitative relationships between the radiative characteristics of the flame and flue gas composition.

## Acknowledgements

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