

# Study on the application of laser diagnosis technology in the rapid real time measurement of soot

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## Highlights

- A new method for characterizing soot formation using the LIBS technique is presented.
- Atomic intensity ratios of flames characterize initial and mature soot formation.
- The competing relationship between soot generation and chemiluminescence is elucidated.

## 1. Introduction

So far, LIBS technology has been mainly used to study the measurement of equivalence ratio and mixed components in flames<sup>[1]</sup>. However, there are few reports on the application of this technique to evaluate the atomic ratio and soot formation in flames, and the relationship between flame radical chemiluminescence and soot formation is not clear<sup>[2]</sup>. Therefore, on the basis of previous studies on soot, this paper proposes a new method to quantify the evolution of soot formation mechanism in flames. The relationship between atomic intensity ratio and soot formation in different regions of flame was studied. At the same time, a detailed chemical kinetic simulation of the formation process of soot was carried out. The key reactions of flame state and soot formation in different regions were analyzed. The influence of free radical chemiluminescence on the formation mechanism of soot was discussed and the relationship between free radical chemiluminescence and soot formation was determined.

## 2. Methods

A high power Q-Nd:YAG second harmonic laser beam at a wavelength of 532 nm is used in the LIBS system to generate a large amount of plasma under the continuous action of the laser. The released signal is collected by a lens and focused onto an optical fiber. Finally, the LIBS signal is converted to a spectrometer and recorded by a CCD camera to obtain a spectral signal.

The soot volume fraction was measured using the laser-induced incandescence (LII) technique. A Nd-YAG laser with a wavelength of 1064 nm and a repetition frequency of 10 Hz was used. The emitted laser beam was controlled by a series of beamers to create a vertical axis of laser light at the center of the burner. Detection of the LII signal was collected by an enhanced ICCD camera using a bandpass filter with a center wavelength of 440 nm.

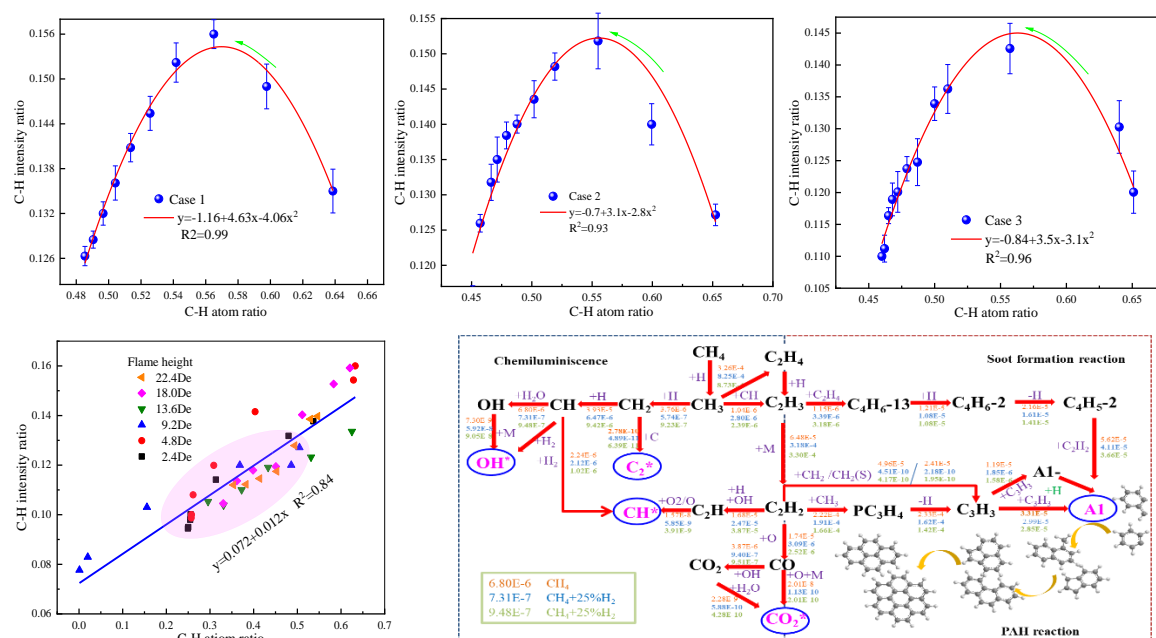
An ultraviolet (UV) camera equipped with a 50 mm 310 nm narrow band-pass filter and a lens 40 cm from the flame axis is used to measure OH\*. In addition, visible images of the flame, soot emission, and chemiluminescence (Swan bands: CH\*: 431 nm, C<sub>2</sub>\*: 516 nm, CO<sub>2</sub>\*: 350-600 nm) were obtained simultaneously using a multi-wavelength approach with a hyperspectral imager (PCO dimax S1) in the spectral range of 380-800 nm<sup>[3]</sup>.

## 3. Results and discussion

The C-H atomic ratio in the flame is a key factor to describe the chemical evolution of soot. It can be used to describe the maturity of soot. Figure.1 shows the relationship between atomic intensity ratio and soot formation and the relationship between soot formation and free radicals. The second peak range of the C-H atomic intensity ratio is the soot formation stage. Therefore, through the fitting analysis of the three cases, the polynomial fitting relationship is a quadratic function in the soot

formation stage. On the right side of the highest point of the polynomial, it is mainly the initial stage of soot formation, and then moves to the left side of the highest point, that is, the maturity of soot gradually increases. With the increase of methane velocity, the C-H atomic strength ratio gradually decreases in the soot formation stage, and the highest point decreases from 0.156 to 0.145. In addition, the error analysis of the three measurement results shows that the error changes little in the mature stage of soot, but the error line is larger in the initial soot formation process. It shows that the initial process of soot formation is unstable in the soot formation stage. The ratio of radial position at different heights is linearly distributed, and it is mainly distributed in the center of the flame, which is the main area of soot generation. It can be seen from the results that the position and trend of soot formation can be detected online by using the C-H atomic intensity ratio in soot flame.

The Rate of production (ROP) analysis visualises the difference between soot and radical chemiluminescence generation pathways, which compete at fuel pyrolysis product ( $\text{CH}_3$ ,  $\text{C}_2\text{H}_2$ , etc.) generation, as they follow opposite reaction paths. The pathway for soot formation is  $\text{CH}_4 \rightarrow \text{CH}_3$   $\text{C}_2\text{H}_3/\text{C}_2\text{H}_2 \rightarrow \text{A1} \rightarrow \text{PAHs} \rightarrow \text{soot}$ , which leads to carbon chain extension and PAH growth. Whereas the generation of radial chemiluminescence is clearly a conversion between C1/C2 radicals, e.g.  $\text{CH}_4 \rightarrow \text{CH}_3 \rightarrow \text{CH}_2/\text{C}_2\text{H} \rightarrow \text{OH}^*/\text{CH}^*/\text{C}_2^*/\text{CO}_2^*$ .



**Figure 1.** Relationship between C-H atomic ratio and intensity ratio during soot formation and the relationship between soot and free radical chemiluminescence.

## 4. Conclusions

The axial quadratic function distribution of C-H intensity ratio can clearly reflect the formation position of initial soot in the flame and the transition stage from young soot to mature soot. The starting position of the second peak of the atomic intensity ratio is the initial soot formation zone. The radial C-H ratio is linearly distributed, and the C-H ratio in the range of 0.10 ~ 0.13 can form the main ash nucleation zone. In the area filled with soot, the chemiluminescence of free radicals is inhibited, and a large number of pyrolysis products are used for the formation of soot. This indicates that the formation pathways of soot and chemiluminescence are competitive.

## References

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## Keywords

Inverse diffusion flame; soot; atomic intensity; Chemiluminescence