Time-resolved LII in comparison with mass spectrometry measurements in a premixed ethylene/air flame

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The measurement of combustion generated nano- and sootparticles with sizes up to 20 nm remains a challenge as all current size measurement techniques do not cover the whole range of particle sizes. Therefore it is necessary to combine different methods. In addition it is essential to find the overlapping detectable regions for comparison of the different sizing methods.

Introduction

We have used two alternative methods, LII and photoionization mass spectrometry to obtain particle sizes under well defined experimental conditions. To this end a McKenna burner was used with an ethylene/air flame stabilized by a horizontal stainless steel plate located 21 mm above the burner surface. Measurements were carried out for equivalence ratios ranging from Φ=1.8 to 2.4. Furthermore the particle sizes have been determined as a function of the height above burner (HAB) between 6 and 16 mm. Additionally first LII measurement at elevated pressures up to 3 bar with a similar burner configuration where performed.

Laser-induced Incandescence

The laser-induced incandescence measurements were conducted with a Nd:Yag laser emitting at 1064 nm. The LII signal was detected at two different wavelengths around 400 nm and 700 nm using fast photomultipliers in combination with interference filters. Fluorescence of PAH´s is very unlikely to be induced, because of the low fluences and the high wavelength of the laser beam. Based on a LII model [1] accounting for absorption, conduction, sublimation, annealing, oxidation and radiation the particle sizes were derived.

Photoionization Mass Spectrometry

For MS measurements sample gas was drawn via a short quartz probe (50 cm, 6 mm id) equipped with a nozzle at its tip. The nozzle provided a pressure drop by a factor of about 60. The resulting drop together with short residence times of about 5 ms helped to minimize coagulation in the sampling line. Introduction to the MS was carried out via a pulsed valve. For photoionization a photon fluence of 3×10^5 W/cm² at a wavelength of 193 nm (ArF) was used. This ensures negligible fragmentation.

Results

The nanoparticle mass spectra show a bimodal behaviour in the HAB range covered, the lower mode with mean diameters between 2 and 3 nm (large circles), the upper mode around 5 nm (small circles) and more pronounced at larger heights. For LII only the upper mode is detectable (dots). In the overlapping region around Φ=1.9 a good match is found between LII and MS. Unfortunately, for larger Φ, that means higher masses, there is a cut-off in the MS mass range so that the high mode peak cannot be reliably measured. The low mode peaks, on the other hand, are observed throughout our stoichiometry range.

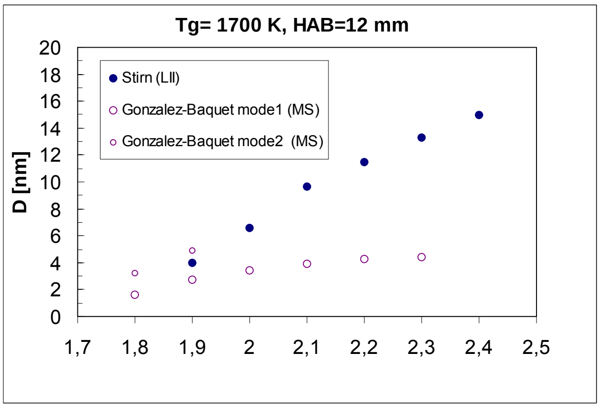

Fig. 1: Mean particle diameters of bimodal size distributions determined by mass spectrometry and laser-induced incandescence in a premixed ethylene air flame

Conclusion

We conclude that both methods show a complementary behaviour. Low mode sizes which are undetectable for LII are readily observable by MS. Conversely, at higher masses when the MS range tends to be limited, LII is particularly sensitive. In addition, it is obvious that for particles as small as 4 nm reliable data from LII can be extracted. MS has the potential to become a reliable sizing technique for small particles up to 10 nm provided the mass cut-off can be shifted to higher masses.

[1] H.A. Michelsen, Understanding and predicting the temporal response of laser-induced incandescence from carbonaceous particles, J. Chem. Phys. 118, 7012 (2003).

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