Laser-induced fluorescence measurements and modeling of absolute CH concentrations in strained laminar methane/air diffusion flames

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Abstract

We have applied linear laser-induced fluorescence to obtain spatially resolved profiles of CH radicals in laminar methane/air and methane/nitric oxide/air counterflow diffusion flames at atmospheric pressure. Excitation and detection of transitions in the A–X band and calibrating the optical detection efficiency via Rayleigh scattering allowed the determination of absolute radical concentrations. Flames at strain rates from 59 to 269 s^{-1} were studied to characterize the strain rate dependence of the CH concentration. The work shows that CH concentrations increase with increasing strain rate. Comparisons have been made with predicted CH levels obtained using two different chemical kinetic mechanisms (Lindstedt et al. and GRI-Mech. 3.0). Computed concentrations are shown to be in good agreement with experimental data. It was furthermore found that the addition of up to 600 ppm NO to the fuel did not have a measurable effect on the CH radical concentration. This is also in agreement with predictions from both mechanisms. The current work has shown that measurements of absolute CH radical concentrations are possible in non-premixed flames without the need for spatial temperature or quenching corrections.
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1. Introduction

Investigations of the performance of chemical kinetic mechanisms that are used to simulate the detailed structure of laminar flames need comparisons to high fidelity experimental measurements. In a recent work, we have measured temperature, NO [1] and O-atom [2] concentration profiles for laminar methane/air counterflow diffusion flames

at various strain rates. The purpose of this work was to experimentally determine quantitative CH concentrations in such flames and to investigate the ability of existing detailed chemical kinetic mechanisms to reproduce measured values. In the flames under consideration, a substantial amount of nitric oxide is formed via a prompt mechanism, and computed levels are very sensitive to the $CH + N_2$ initiation reaction. Therefore, measurements of CH radical profiles will provide important information for the assessment of kinetic mechanisms aimed at predicting the formation of nitric oxide. Furthermore, CH radicals

are involved in reactions that lead to the removal of nitric oxide. In a previous study [3], we concluded: "however, a central difficulty remains in as much as the chemistry of the CH radical needs to be further investigated along with NO destruction channels in the fuel rich part of diffusion flames. The latter has been clearly illustrated by computations and measurements of flames where nitric oxide and ammonia has been seeded in the fuel stream. It has also been shown that the chemistry of pollutant formation is significantly different in premixed and diffusion flames and that accurate quantitative determination of CH radical concentrations in diffusion flames remains imperative." Here, we report such quantitative measurements of CH radical profiles in laminar methane/air counterflow diffusion flames and NO-doped methane/air flames for a range of strain rates along with comparisons to predictions obtained using detailed chemical kinetic modeling.

Simulations that address CH profiles in a variety of flames of different fuels with and without additional dopants are reported in the literature. Williams and Fleming [4] have measured relative LIF CH profiles and made comparisons to simulations for low-pressure $CH_4/O_2/Ar$ flames doped with N_2O, NO, and NO_2 to address reburn-related questions. Quantitative CH profiles in low-pressure methane and propane flames were obtained by Luque et al. [5] and found to be in good agreement with simulations using GRI-Mech 2.11. In the context of reburning, Juchmann et al. [6] and Mercier et al. [7] investigated low-pressure NO-doped CH_4/O_2 flames and reported absolute CH profiles along with model comparisons. For atmospheric pressure flames, the work of Walsh et al. [8] describes quantitative CH measurements and two-dimensional computations of a laminar jet diffusion flame.

Substantial progress has been made on techniques for quantitative measurements of CH radicals in flames over the past few years. Most of these studies addressed low-pressure premixed flames where adequate spatial resolution and direct measurements of effective fluorescence lifetimes with ns-pulsed lasers are feasible, e.g. [6,9]. Non-premixed and atmospheric pressure flames were examined early-on by Norton and Smyth [10], and more recently by Carter et al. [11], Mercier et al. [12], Klein-Douwel et al. [13], Moreau et al. [14], and Luque et al. [15]. A characteristic of CH profiles in premixed flames is the rapid spatial change of temperature, the associated change in gas composition, and the resulting effective collisional environment. The result is that CH measurements in premixed flames require high spatial resolution and knowledge of the local temperature and collisional environment if CH radicals are to be measured with laser-induced fluorescence. De-convolution of measured profiles can help one to enhance the spatial resolution; see

e.g. [15]. The CH layer in non-premixed flames is embedded between the oxidizer and the fuel very close to the peak temperature region. In atmospheric pressure flames, the predicted width of the CH layer (FWHM) is ~0.2 mm. Over the width of the profile, temperature and collisional environment change only very little as will be shown below. This can potentially simplify the interpretation of measured LIF profiles, even for flames of different strain rates due to the reduced overall sensitivity to temperature and quenching effects. This does not, however, eliminate the need for high spatial resolution and calibration to absolute concentration profiles for which information on collisional quenching is needed. Data for such corrections can be found from Tamura et al. [16] who provide a comprehensive list of quenching cross-sections for CH and Renfro et al. [17] who measured time-resolved CH fluorescence for 77 methane-based counterflow diffusion flames at atmospheric pressure and reported a wide range of fluorescence lifetimes and revised temperature dependences for N_2 and H_2O as colliders. Absolute CH concentration measurements using different strategies have been reported. Bonczyk and Shirley [18] used saturated laser-induced fluorescence, while Norton and Smyth [10] applied chemical path analysis to deduce CH concentrations in laminar CH_4/air flames. Walsh et al. [8] studied a laminar jet diffusion flame and used Rayleigh scattering for calibration of CH LIF signals. Klein-Douwel et al. [13] measured absolute number densities of the CH radical in a partially premixed methane/air bunsen flame ($\phi = 1.36$) at atmospheric pressure by exciting to a predissociating level via the CH B–X $(1,0)$ transition using a quasi-linear LIF scheme and Rayleigh scattering for calibration of optical efficiency. Mercier et al. [12] and Moreau et al. [14] combined cavity-ring-down-spectroscopy (CRDS) and linear LIF detection of CH radicals using C–X $(0,0)$ excitation in flames stabilized on a Wolfhard–Parker burner (including a sooting methane/air diffusion flame) to measure absolute CH concentrations. Luque et al. [19] combined CRDS with excitation in the B–X $(1,0)$ band for spatially resolved CH profile measurements in low-pressure flames. Evertsen et al. [20] also used CRDS for CH measurements in an atmospheric pressure premixed flat flame. CRDS for calibration will not work for the flame geometry in our experiment. The edges of the flame at the ends of the cylinder are bent upwards; this will lead to beam deflection and thus spatial averaging in a CRDS (multiple beam passages) experiment. For calibration to absolute CH concentrations, the strength of Rayleigh scattering signals is compared to the strength of LIF signals [9]. This approach allows the determination of the overall optical detection efficiency, and subsequently, absolute concentration is obtained, given that experimental parameters,