

Development of TDLAS-Based Diagnostics for Combustion of Preheated Lean Dimethyl Ether/Air Mixtures

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Introduction

Dimethyl ether (DME) represents a prototypical synthetic fuel and multi-purpose energy carrier being suitable for transportation sector as well as for electricity generation or heating purposes. Its future use is of remarkable importance in framework of the Methanol Economy [1] as it is considered as derived fuel produced by chemical recycling of carbon dioxide [2].

Detailed chemical kinetics of DME oxidation (both comprehensive and simplified schemes) including validated low-temperature mechanisms has been investigated extensively during previous two decades (e.g. [3-6] and references therein). Considerable improvement in predictive performance and reliability of relevant chemical kinetic models is thus inferred despite the presence of remaining uncertainties for the rate constants of some key elementary reactions in low-temperatures DME oxidation schemes [4,5].

Simultaneous progress in experimental studies of specific DME oxidation modes (e.g. [7]) promotes a need of combustion diagnostics suitable for observation of corresponding flame structures and their dynamics respectively. Quantitative measurement of species concentration profiles in laminar cool flames is still a challenging goal in given context.

Applicability of Tunable Laser Diode Absorption Spectroscopy (TDLAS) for non-invasive diagnostics in a harsh combustion environment was previously reviewed [8]. Wavelength Modulation Spectroscopy (WMS) has been demonstrated as feasible TDLAS technique for temperature and species concentration measurement in laminar reacting flows as well as turbulent flames.

Aim of the present study is twofold: (i) to explore the combustion regimes and structure of burner-stabilized laminar flames of preheated lean and ultra-lean DME/air mixtures at atmospheric pressure both experimentally and employing relevant chemical kinetic models; (ii) to select appropriate target species and their spectral features in order to characterize these flames using WMS measurements.

Methodology

Experimental setup for observing the structure of the given flames was based on calibration (flat-flame) burner [9], which was heated by stoichiometric ($\Phi = 1$) methane/air flame steadily burning in its co-flow section instead of application of cooling water circulation.



Fig. 1: Images of visually observed lean (left) and ultra-lean (centre) DME/air flames and N_2 /air flow (right) surrounded by CH_4 /air co-flow flame. Heated alumina tube (with thermocouple probe) seen as hot spot in post-flame region.

Preheated DME/air mixtures at equivalence ratio $\Phi = 0.5$ (referred as “lean”) and $\Phi = 0.25$ (referred as “ultra-lean”) were considered as representative cases for this study. Steady-state structure of burner-stabilized laminar flames corresponding to these experimental conditions was predicted by one-dimensional (1-D) numerical simulation taking into account multicomponent transport properties (incl. thermophoresis), radiative heat transfer and detailed chemical kinetics. The CANTERA software package [10] was utilized for performing this task. Near-infrared absorption spectra covering the narrow $6360 \pm 2 \text{ cm}^{-1}$ region were measured for these conditions using wavelength modulation spectroscopy with second-harmonic detection (i.e. $2f$ -WMS technique). Collimated beam from a current-modulated semiconductor (DFB) laser was doubly passed through the measurement section (flame) at selected height above burner ($HAB = 5 \text{ mm}$). Signal from an optical (InGaAs) detector was demodulated using lock-in amplifier and acquired on digital oscilloscope. Post-processing of $2f$ -WMS signal was performed employing the method and software tools, which have been developed recently in our group. Briefly, demodulated and averaged $2f$ -WMS signals were normalized using incident laser power approximated as being proportional to quadratic fit of the dc ($0f$) signal from InGaAs detector. Difference spectra and ratio of normalized ($2f/0f$) WMS signals acquired with and without the flow of DME/air mixture into the burner were extracted in the following step. Predictions of $2f$ -WMS signal in relevance to our measurements were carried out assuming concentration profiles obtained from 1-D model. Hydroxyl (OH) radical and water (H_2O) vapor were considered as target (measurable) quantities.

Results and discussion

Combustion of preheated DME/air mixtures surrounded by stoichiometric methane/air flame (in the co-flow section at $Q_{v,tot} = 10 \text{ l/min}$ for both cases) was visually observable (see Fig. 1). It is worth to note that the inlets of oxidizer (preheated air) and fuel (DME) were located approx. 20 cm upstream the heated ($t \approx 300^\circ\text{C}$) burner plate. Oxidation of DME/air mixture may thus eventually occur before it reaches the measurement section (at $HAB < 0 \text{ cm}$). This fact is important for interpretation of experimental observations and also implies safety issues, i.e. caution concerning a flashback phenomenon.

Chemical kinetic models (selected from recently available schemes of DME oxidation) having different level of complexity were employed in this study. Firstly, the compact mechanism [6] including 24 species and 76 elementary reactions was used for initial trial calculations and optimization of model setup (regarding the convergence of time-stepping and progress of iterations from initial guess to steady-state solution on refined grid). Resulting temperature and species concentration profiles were also obtained by entire model run with more detailed chemical kinetic scheme [5] (with 120 species and 711 elementary reactions), see Fig. 2.

Absorption transition of OH (at 6359.71 cm^{-1} and 6360.75 cm^{-1}) is expected to dominate the $2f$ -WMS signal measured in high-temperature flames. Moderate level of OH concentration ($X_{OH} \sim 10^{-3}$) is present in post-flame region according to calculated species profiles (see Fig. 2). On the other hand, missing or weak OH absorption features can be assumed as indirect indication of transition to cool flame regime or flame extinction/extinguishment.

Combustion of ultra-lean mixtures is also accompanied by significantly weaker hot water absorption (in the investigated spectral region) due to lower temperature and decreased H_2O concentration in cool flame.

These assumptions are consistent with $2f$ -WMS data obtained from predictions and measurements for both experimental cases. Nevertheless, the results are not conclusive for the case of ultra-lean ($\Phi = 0.25$) DME/air flames where the absorption of co-flow flame is much stronger than contribution from the central burner section, which is of interest. Important issue for future investigation is interaction of the given reactive/nonreactive flows with the surrounding (co-flow) flame.

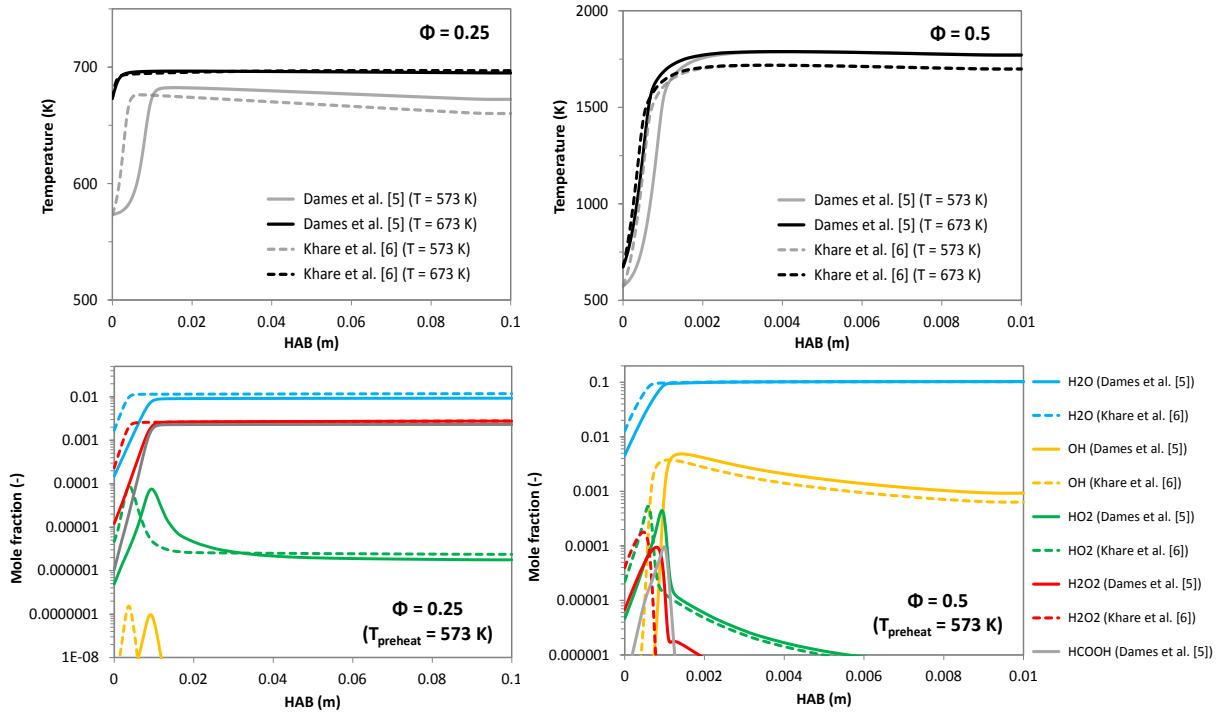


Fig. 2: Calculated temperature profiles (upper traces) and mole fractions of selected species (lower traces) in burner-stabilized ultra-lean and lean DME/air laminar premixed flame as predicted with two chemical kinetic schemes (described in text).

Conclusions

Recent findings concerning cool flame behavior and exceptional flammability of preheated DME/air mixtures need to be addressed in practical applications (incl. combustion, storage and handling). Computational part of this work is complementary to previous numerical studies performed using similar approach when studying preheated combustion of lean DME/oxygen systems [11] and rich DME/air mixtures [12]. Experimental study which indicates consistency with modelling results was accomplished. Development of $2f$ -WMS diagnostics for identifying different combustion regimes observed for lean ($\Phi = 0.5$) and ultra-lean ($\Phi = 0.25$) laminar premixed flames of preheated DME/air mixtures is in progress. High-temperature flame regime of DME/air combustion can be distinguished by OH radical and hot water lines present in absorption spectra of lean flames near 1572 nm investigated in this work. Following the modelling results, unambiguous identification of cool flames regime relevant to oxidation of preheated ultra-lean DME/air mixtures is attainable by the means of $2f$ -WMS measurement. Candidate species selected based on this study as targets for further analysis and experiments focusing different wavelength ranges include hydrogen peroxide (H_2O_2), hydroperoxyl (HO_2) radical and formic acid (HCOOH).

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