NO concentrations in NH3-doped CH4+air flames measured using saturated LIF and probe sampling

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Due to massive research efforts made in the last two-three decades, major reactions of nitrogen chemistry in flames are now well understood. Considerable progress has been achieved in the development of detailed kinetic mechanisms describing NO_x formation and reburning in natural gas combustion, for instance GRI-mechanism [1]. In most cases the GRI-mech. predictions are in good quantitative or at least in qualitative agreement with new experimental results obtained since its last release. However, some experimental data on ammonia conversion in premixed flames cannot be reproduced by contemporary kinetic schemes. Konnov et al. [2] measured concentrations of NO in $(CH_4+NH_3) + O_2 + N_2$ flames and found satisfactory agreement between experiments and modelling in lean flames. In rich mixtures, however, NO measurements were significantly over-predicted by the mechanisms of Konnov [3] or of Skreiberg et al. [4]. It was concluded that significant qualitative discrepancy between the experiments and modelling is probably due to missing (unknown) reactions and not due to wrong rate constants.

To narrow the search for deficiencies in the kinetic (sub-)mechanisms, experimental data on flame structure of rich hydrogen flames doped with NH_3 at atmospheric pressure could be most helpful. Therefore flat premixed burner-stabilized $H_2 + O_2 + N_2$ flames, neat or doped with 300-1000 ppm of NO or NH_3, were studied experimentally using molecular-beam mass-spectrometry and simulated numerically [5]. Spatial profiles of temperature and concentrations of stable species, H_2, O_2, H_2O, NO, NH_3, and of H and OH radicals obtained at atmospheric pressure in lean, near-stoichiometric and rich flames have been reported. Good agreement between measured and calculated structure of lean and near-stoichiometric flames was found. Significant discrepancy between simulated and measured profiles of NO concentration was observed in the rich flames doped with NO. Modification to the model was proposed to improve an overall agreement with the experiment. Experiments in the rich hydrogen flames doped with NH_3, however, did not indicate the need for the model modification.

To find the reason of the intriguing discrepancy between the modelling and experimental data in rich $(CH_4+NH_3) + O_2 + N_2$ flames, Laser-saturated fluorescence (LSF) was adopted to measure NO concentrations. A Heat Flux method was used for stabilization of non-stretched flames on a perforated plate burner at atmospheric pressure. The concentrations of NOx were also measured by means of a non-cooled quartz probe at different axial distances from the burner. These measurements were compared to predictions of the detailed kinetic model [3]. The modelling is found in good agreement with the experiments in lean, stoichiometric and rich flames. Even more surprisingly, present probe sampling measurements significantly differ from the previous results at very similar conditions [2]. Possible experimental peculiarities explaining these findings are discussed. The role of reliable experiments in the model development and validation is emphasized.

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