Fundamental limits of NO formation in fuel-rich premixed methane-air flames

van Essen, Vincent Martijn

Introduction

Combustion has always played an important role in the history of mankind: from prehistoric times when fire was used for cooking food and keeping wild animals at bay, to modern times in which combustion is used in industrial processes, power generation and space heating in buildings. However, beside the benefits combustion offers, it also has a downside: producing pollutants such as nitrogen oxides, sulfur oxides and soot.

Once released into the atmosphere, NO_x plays an important role in the formation of photochemical smog and acid rain [1]. The global emission of NO_x since 1900 has increased by a factor of ten, and from 1950s even with a factor of three [2]. A significant amount (~70%) of this increase can be attributed to combustion sources [3], while the rest of the NO_x emission comes from natural sources.

Since the early 1970s, many countries have implemented NO_x emissions regulations by law. The progressively stringent regulations for NO_x emission [4] stimulated designers of natural-gas-fired combustion equipment to search for techniques of limiting NO_x formation. It became increasingly clear that a more detailed understanding of the mechanism of NO_x formation is needed to assess and predict the effect of NO_x control strategies such as flue gas recirculation, use of radiant burners and water injection [2]. In the past 30 years, significant progress has been made in determining the chemical mechanisms by which NO_x is formed; however, despite this progress, the extent to which these mechanisms contribute to NO_x formation in many practical systems is still not known. As a result, the actual effect of NO_x control strategies in practice is difficult to predict and the development of combustion equipment is still based on 'trial and error' methods. The development process would be more efficient and effective with correct insight in the mechanism of NO_x formation and it response to various control strategies.

Whereas most studies have focused on understanding NO_x control strategies in fuel-lean premixed flames, the objective of this thesis is to provide insight in the effect of two NO_x control strategies on the formation of NO_x in laminar premixed fuel-*rich* methane-air flames: flue-gas recirculation (essentially the dilution of the fuel-air mixture with N_2+CO_2) and burner stabilization (leading to upstream heat loss, the operating principle of radiant burners). Towards this end, the distributions of temperature and concentrations of key chemical species important for NO formation have been measured in low-pressure flames while varying the exit velocity and composition of the unburned fuel/air mixture. At reduced pressures, the reaction zone broadens to a size larger than the typical spatial resolution of optical diagnostic techniques, facilitating the measurements (see for example [5]). For this reason, a low-pressure setup was constructed to perform these measurements. In this thesis, laser-induced fluorescence (LIF) is used for determining flame temperatures and absolute NO, OH and CH concentrations. To assist in the analysis of the underlying physical and chemical processes, and to assess the predictive power of current chemical mechanisms describing NO_x formation, the experimental observations are compared with the results of one-dimensional flame calculations. Furthermore, the experimental results can be utilized to benefit the design of low-NO_x combustion systems by not only showing the most promising conditions for minimizing NO_x formation, but also by improving the accuracy of NO_x predictions of detailed numerical models describing methane-air flames.

Scope of this thesis

In this thesis, a laser-diagnostic study of the effect of flue-gas recirculation and upstream heat-loss on NO_x formation in laminar low-pressure premixed fuel-rich methane-air flames is presented. In Chapter 1, basic information on the physics and chemistry of one-dimensional laminar flames, a short discussion of current insight into chemical processes leading to NO formation and NO_x emission reduction techniques are presented. Chapter 2 gives details of the low-pressure setup and the gas-handling system that are used in the experiments throughout this thesis. Chapter 3 presents the physical principles and experimental details of laser-induced fluorescence (LIF). Chapter 4 is devoted to a study of the effect of burner stabilization on Fenimore NO formation in low-pressure fuel-rich premixed $CH_4/O_2/N_2$ flames, while Chapter 5 considers the effects of flue-gas recirculation. Finally, in Chapter 6 the results from measuring NO mole fraction as function of pressure are discussed with the aim of investigating how the low-pressure results can be translated to atmospheric conditions.

Bibliography

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