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Effects of H_2 and H preferential diffusion and unity Lewis number on superadiabatic flame temperatures in rich premixed methane flames

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Abstract

The structures of freely propagating rich CH_4/air and CH_4/O_2 flames were studied numerically using a relatively detailed reaction mechanism. Species diffusion was modeled using five different methods/assumptions to investigate the effects of species diffusion, in particular H_2 and H, on superadiabatic flame temperature. With the preferential diffusion of H_2 and H accounted for, significant amount of H_2 and H produced in the flame front diffuse from the reaction zone to the preheat zone. The preferential diffusion of H_2 from the reaction zone to the preheat zone has negligible effects on the phenomenon of superadiabatic flame temperature in both CH_4/air and CH_4/O_2 flames. It is therefore demonstrated that the superadiabatic flame temperature phenomenon in rich hydrocarbon flames is not due to the preferential diffusion of H_2 from the reaction zone to the preheat zone as recently suggested by Zamashchikov et al. [V.V. Zamashchikov, I.G. Namyatov, V.A. Bunev, V.S. Babkin, Combust. Explosion Shock Waves 40 (2004) 32]. The suppression of the preferential diffusion of H radicals from the reaction zone to the preheat zone drastically reduces the degree of superadiabaticity in rich CH_4/O_2 flames. The preferential diffusion of H radicals plays an important role in the occurrence of superadiabatic flame temperature. The assumption of unity Lewis number for all species leads to the suppression of H radical diffusion from the reaction zone to the preheat zone and significant diffusion of CO_2 from the postflame zone to the reaction zone. Consequently, the degree of superadiabaticity of flame temperature is also significantly reduced. Through reaction flux analyses and numerical experiments, the chemical nature of the superadiabatic flame temperature phenomenon in rich CH_4/air and CH_4/O_2 flames was identified to be the relative scarcity of H radical, which leads to overshoot of H_2O and CH_2CO in CH_4/air flames and overshoot of H_2O in CH_4/O_2 flames.

Keywords: Rich premixed flames; Preferential diffusion; Superadiabatic flame temperature; Chemical kinetics; Chemical equilibrium

1. Introduction

The phenomenon of superadiabatic flame temperature (SAFT) in rich hydrocarbon premixed flames

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is of great fundamental and practical interest. It has been observed experimentally and predicted numerically [1–6]. In a numerical study of diamond chemical vapor deposition (CVD) using a strained, rich premixed $C_2H_2/H_2/O_2$ flame, Meeks et al. [1] found that the flame temperature exceeds the adiabatic temperature, which is somewhat unusual in premixed hydrocarbon flames. Their study is perhaps the first numerical work to reveal the phenomenon of SAFT in rich premixed hydrocarbon flames. The appearance of superadiabatic temperature in the flame numerically studied by Meeks et al. [1] was subsequently confirmed experimentally by Bertagnolli and Lucht [2] and Bertagnolli et al. [3]. Meeks et al. [1] suggested that the primary reason for the occurrence of SAFT in the rich $C_2H_2/H_2/O_2$ premixed flame is that unreacted acetylene requires a relatively long time to dissociate to its equilibrium concentration. Another explanation of SAFT in this flame was later presented by Bertagnolli et al. [3], who believed that the presence of superequilibrium concentrations of CO_2 and H_2O was the primary cause of the superadiabatic flame temperatures. The superequilibrium concentrations of C_2H_2 and H_2O are consistent with the subequilibrium concentrations of H and H_2. Later in a numerical study of CVD of diamond films using rich acetylene–oxygen flames, Ruf et al. [4] also noted the significantly subequilibrium concentration of H and suggested that the slow and endothermic reaction $H_2 + M \Leftrightarrow H + H + M$ is responsible for the subequilibrium concentration of H and SAFT. More recently, the nature of SAFT has been explored by Liu et al. [5] and Zamashchikov et al. [6] in their numerical studies of freely propagating rich hydrocarbon premixed flames. Through a systematic investigation of the structure and maximum flame temperature in CH_4/air, CH_4/O_2, $C_2H_2/H_2/O_2$, C_2H_4/O_2, C_3H_8/O_2, and H_2/O_2 flames, Liu et al. [5] found that SAFT occurs only in rich hydrocarbon flames but not in hydrogen flames and suggested that the nature of SAFT is chemical kinetics. However, Zamashchikov et al. [6] concluded, through a careful examination of the numerically predicted structures of rich C_3H_8/air and CH_4/air flames, that the nature of SAFT in rich hydrocarbon flames is the diffusion of molecular hydrogen from the reaction zone to the preheat zone and its preferential oxidation compared to hydrocarbons. While the superadiabatic flame temperature phenomenon is clearly a nonequilibrium process, the brief review of the literature discussed above on this topic reveals that our current understanding of the nature of SAFT remains unclear given the two completely different views presented in Refs. [5,6].

Although it has been made clear that the mechanism of superadiabatic temperature in diffusion flames, such as the numerical study of Takagi and Xu [7], is a direct consequence of the preferential diffusion effects of H_2 and H, the effects of preferential diffusion of H_2 and H and the Lewis number in general on the occurrence of SAFT in rich hydrocarbon premixed flames have yet to be explored. This study is motivated by the lack of understanding of the effects of preferential diffusion of H_2 and H and the species Lewis number on SAFT in rich premixed hydrocarbon flames. Although the nature of SAFT was previously speculated to be chemical kinetics [1,3–5], the question of why SAFT occurs when a hydrocarbon mixture is sufficiently rich has not been adequately answered. An attempt was also made in this study to provide further insight into the nature of SAFT. In the present study, the structures of atmospheric freely propagating planar rich premixed flames in mixtures of CH_4/air and CH_4/O_2 were numerically computed using detailed chemistry and complex thermal and transport properties. To investigate the effects of preferential diffusion of H_2 and H from the reaction zone to the preheat zone and the Lewis number on SAFT, numerical calculations were carried out using five different treatments of species diffusion. Reaction flux analyses were performed to reveal the chemical nature of SAFT.

2. Numerical model

The conservation equations of mass, momentum, energy, and chemical species for steady planar freely propagating premixed flames were solved using a CHEMKIN-based code [8]. The thermochemical and transport properties of species were obtained from CHEMKIN [8] and TPLIB [9,10] database. Since our previous study found that radiation heat loss has negligible impact on the flame temperature in the reaction zone and the immediate postflame zone [5], all the calculations were conducted without radiation heat loss. At a spatial location of $x = 0.05$ cm, the mixture temperature is fixed at 400 K. In all the calculations, the upstream location (fresh mixture) is always kept at $x = -2.5$ cm. The location of downstream (reacted combustion products) specified in the calculations varies with the gas mixture. In all the calculations, however, it was checked that the computational domain was sufficiently long to achieve adiabatic equilibrium. The gas mixture temperature at the upstream boundary was kept at 300 K and zero-gradient conditions were specified at the downstream boundary. All the calculations were performed at atmospheric pressure. The GRI Mech 3.0 reaction mechanism [11], which was optimized for methane combustion, was used to model the chemical kinetics in this work. The only modification made to this mechanism is the re-