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A computational study of preferential diffusion and scalar transport in nonpremixed hydrogen-air flames

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ABSTRACT

The nonpremixed hydrogen-air reacting flow is simulated using three-dimensional direct numerical simulation coupled with flamelet generated manifolds based on detailed chemical kinetics. From the comparisons between one computational case taking into account preferential diffusion and another case with unity Lewis number assumption, the instantaneous results show that the flow is more vortical in the absence of preferential diffusion. This indicates that preferential diffusion may smooth the flame under certain circumstances when coupled with the intrinsic hydrodynamic instability. The flame compositional structures are also influenced by preferential diffusion in a significant manner. Further, the statistical information suggests that turbulent scalar flux is affected by preferential diffusion. The phenomenon of counter-gradient diffusion of both the conserved and non-conserved scalars can be detected for the two cases. The gradient model for scalar closure is found to be incapable of accurately predicting the scalar transport in nonpremixed hydrogen flames.

Introduction

Emissions of pollutants from the combustion of conventional fossil fuels in energy utilization applications have caused major environmental concerns and the massive CO_2 emissions have been regarded as an important contributor to global climate change [1]. There is no doubt that clean energy solutions such as hydrogen-enriched fuels may play an increasingly important role in future energy supply. Hydrogen with a very large mass diffusivity and low molecular weight is a very promising energy carrier. The benefits of hydrogen call for further studies of the combustion of H_2, in particular, for the development of an advanced model for simulating turbulent nonpremixed flames such as jet flames of hydrogen or hydrogen-enriched fuels in future low emission combustion devices and engines.

Because hydrogen is highly diffusive, it may have impacts not only on the chemical process but also on the stability of flame. The phenomenon associated with the fact that each

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species has a different mass diffusion velocity in a multi-species flow system, is referred to as differential diffusion. According to Graham's law [2], the diffusion rate of a gas is inversely proportional to the square root of its molecular weight, which can be stated as $D_1/D_2 = \sqrt{W_2/W_1}$. Therefore, hydrogen molecules diffuse four times faster than those of oxygen. Differential diffusion exists in multi-species flows. When considering the combustion heat, the mass diffusivity of hydrogen is much faster than the thermal diffusivity. This is attributed to preferential diffusion, which is usually discussed in the flow considering combustion. An important parameter in characterizing preferential diffusion effects is the Lewis number *Le*, defined as the ratio of thermal diffusivity to mass diffusivity.

In many earlier studies, single one-step chemistry and unity Lewis number assumptions were used in the computational studies but they were not able to accurately predict the combustion process [3–5]. In the last few decades, there have been an increasing number of studies investigating preferential diffusion, including studies using laminar flamelet approach reported by Pitsch and Peters [6], Nilsen and Kosály [7], while experimental investigations [8–10] were also carried out. It was verified that the preferential diffusion effects in the premixed bluff body flames initially increased with Reynolds number, and then saturated and persisted at higher Reynolds numbers. In addition, the influences of preferential diffusion on the maximum flame temperature and burning rate were investigated using two-dimensional direct numerical simulations (DNS) [11,12], where the details of turbulence and vortex shedding were missing. Preferential diffusion could result in a shift in the equivalence ratio to leaner conditions and was sensitive to different compositions of fuel [13–16]. Furthermore, the probability density function mixing models [17,18], the effective Lewis numbers [19] and the conserved mixture fraction models [20,21] were developed and applied for preferential diffusion in computational studies. Since the differences between preferential diffusion and homogeneous effects are strong in such flames, a predictive model cannot be developed without understanding of the effects of preferential species transport on turbulent nonpremixed flames.

In the context of either large eddy simulation or Reynolds averaged Navier–Stokes approach, it is necessary to provide closure modeling for turbulent scalar flux terms. The gradient hypothesis is often used in the turbulent scalar transport in conjunction with turbulent eddy viscosity concepts for premixed flames [22,23]. If c represents a composition variable, according to the gradient transport hypothesis, the corresponding turbulent scalar transport $\widetilde{u_i''c''}$ in the ith direction is modeled as

$$\overline{\rho}\widetilde{u_i''c''} = -\overline{\rho}D_t\partial\tilde{c}/\partial x_i \quad (1)$$

here ρ is the density and the overbar represents the Reynolds averaged operation, and D_t is the turbulent eddy diffusivity. For a general quantity q, Favre averaged quantity $\tilde{q}$ is defined as $\tilde{q} \equiv \overline{\rho q}/\overline{\rho}$ and $\tilde{q} = q - q''$. Under some conditions the gradient hypothesis fails to predict either the magnitude or the direction of turbulent scalar transport, and indeed the existence of counter-gradient diffusion (CGD) in premixed flames has been detected from experimental data [24,25], theoretical analysis and numerical simulations [26–31]. CGD refers to the phenomenon of turbulent scalar fluxes moving in the same direction as the local mean gradient of the scalar, in violation of the conventional but most widely used gradient diffusion (GD) model. This was found in the early analyses of Bray [32] for premixed flames. Later, advanced closures for the turbulent flux terms were proposed by Veynante et al. [33] and Zimont et al. [34]. It has been found the occurrence of CGD is closely related to the pressure distribution across the flame brush due to the heat release, and the gradient model fails to accurately predict the scalar transport of turbulent premixed flames. Robin et al. [35] proposed an extended closure of the reactive scalar fluxes with one contribution that involved the flame wrinkling factor, and the corresponding closure has been then successfully applied to impinging flames [36]. The premixed flame is developed when oxidizer has been mixed well with fuel, which can create a thin flame front as all of the reactants are readily available. However, nonpremixed flame is mainly controlled by the fuel/oxidizer mixing and diffusion with combustion mainly taking place in the shear layer of the jet flame. Since the conditions corresponding to nonpremixed and premixed combustion mechanisms are somehow different, could the gradient model be available for predicting the turbulent scalar transport in nonpremixed flames? It has been discussed in the early study by Luo and Bray [37] in supersonic diffusion flames, and has been verified in the experimental investigation of Caldeira Pires and Heitor [38]. CGD is found to be related to the importance of the heat release in the vicinity of stoichiometric conditions which affects the normalized specific volume variations, as recently emphasized by Serra et al. [39]. Although advanced closures are generally proposed for the premixed turbulent flames, there is still a lack of detailed investigations on the closure modeling for nonpremixed flames.

This study was aimed to investigate the effects of preferential diffusion on nonpremixed hydrogen flames, and more importantly, to evaluate the gradient model of turbulent transport using fully three-dimensional (3D) DNS results based on detailed chemistry mechanism and transport. Two simulations have been carried out, with one simulation based on the realistic preferential diffusion and the other one based on the unity Lewis number assumption. The rest of the paper is organized into four sections: governing equations and chemistry mechanism are given in Section 2, flame configuration and numerical approach are presented in Section 3, results and discussion are described in Section 4 and conclusions are briefly summarized in Section 5.

Governing equations and chemistry mechanism

In this study, the nonpremixed hydrogen-air reacting flow is considered as an unsteady compressible viscous fluid with buoyancy effects and chemical reactions. Three-dimensional governing equations for the reacting flow field, including mass conservation, momentum transfer, energy conservation, and transport equations for progress variable and mixture fraction, are solved in their non-dimensional form: