The impact of thermal diffusion on the structure of non-premixed flames

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ABSTRACT

It is well understood how the flame structure of laminar non-premixed counterflow flames is influenced by differential diffusion, which is driven by species gradients, and corresponding models are available for physical and flamelet space. Thermal diffusion, driven by temperature gradients, is another process that leads to changes in the flame structure and can cause species separation due to differences in the thermal diffusion coefficients. In the present work, a systematic study quantifying the impact of thermal diffusion on the flame structure of non-premixed flames is carried out for five fuel-diluent-oxidizer systems. We also investigate the influence of curvature and strain and present a method for easily incorporating differential diffusion and cross-diffusional effects in flamelet modeling. The study is carried out for one-dimensional counterflow flames, tubular flames and a laminar two-dimensional coflow flame. Examples of flame structures are examined and compared to results obtained with an extended flamelet model, accounting for thermal diffusion and non-unity Lewis number diffusion.

1. Introduction

Diffusion is a transport process that has a substantial influence on the inner structure of laminar and turbulent flames. On a molecular level, it transports reactants to the reaction zone and products and chemical heat away from it, thereby significantly influencing the strength and structure of the flame. The various physical effects causing chemical species to diffuse [1,2] are comprised in the diffusion velocity. The diffusion velocity of a chemical species incorporates a term that is linked to molecular diffusion, mainly driven by species gradients (contained in the diffusion driving force $\mathbf{d}_j$), and a term that is called thermal diffusion (Soret effect), driven by temperature gradients

$$\mathbf{V}_i = -\underbrace{\sum_{i=1}^{N_s} D_{ij}\mathbf{d}_j}_{\text{Molecular}} - \underbrace{\theta_i \nabla(\ln T)}_{\text{Thermal}} . \tag{1}$$

While the effects of detailed molecular transport models in combustion have been well studied (see e.g., [3] and references therein), much less effort has been spent on studying the effects of thermal diffusion. Although diffusion due to species gradients is the dominant mode of transport [4], the extent to which Soret diffusion may alter flame physics can be quite substantial. The calculation of thermal diffusion coefficients is computationally expensive. Therefore, even if detailed molecular diffusion is considered, Soret diffusion is often neglected.

A previous study already suggested that *although thermal diffusion is not a primary process of combustion phenomena, it is often quantitatively significant* [5]. Hence, a tradeoff has to be made between physical accuracy and computational effort. For example, the results in [6] suggest that thermal diffusion reinforces systematic shifts that are introduced by detailed molecular diffusion models, like mixture-averaged or multicomponent diffusion. Investigating dilution effects on laminar diffusion flame temperatures and positions, for light (e.g., hydrogen) and heavy (e.g., n-heptane, n-dodecane and n-eicosane) fuel vapors, the results in [6] suggest that for fuels significantly lighter or heavier than the host gas, Soret diffusion introduces significant shifts in both the flame temperature and position. The influence of thermal diffusion on the flame structure was also confirmed in [7].

Soret diffusion has also been investigated in various other studies, e.g., its influence on extinction strain rates [8,9], its impact on wall quenching [10,11], changes in burning velocities [9,12], sooting flames [13,14] and oxy-fuel combustion [15] and in all cases it was found to have a certain, non-negligible, impact.

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Although the physical effect of thermal diffusion itself is well understood, there is no general consensus on when the Soret effect is of major or minor importance and, hence, it is often neglected in numerical studies. In the literature, only a limited number of fuel-oxidizer mixtures are usually discussed in detail, but a broad overview for many different mixtures does not exist to our knowledge. Due to the computational costs associated with the inclusion of thermal diffusion, knowledge of this kind would be beneficial, e.g., before setting up computationally expensive, large-scale flame computations. Thus, it is the scope of this work to present a systematic approach facilitating a quantitative assessment of thermal diffusion. Therefore, we studied five different fuel-diluent-oxidizer systems, namely H_2–N_2, H_2–He, CH_4–N_2, CH_4–CO_2 and C_2H_6O–N_2, in which the oxidizer is O_2 in all cases. While we focus on the influence of the Soret effect on flame structures for hydrogen and small hydrocarbon fuels, the approach can easily be extended to other fuel-diluent-oxidizer systems, e.g., for large hydrocarbon fuels or PAHs. In this context, we investigate the influence of the mixture by varying its composition systematically and further address curvature and strain effects. Moreover, we show how differential diffusion and cross-diffusional effects can be consistently incorporated in flamelet modeling and compare the flamelet results to exemplary 1D and 2D flame data.

2. Numerical approach

The Soret effect in non-premixed flames is assessed in a systematical manner using laminar counterflow flame simulations and a flamelet model. In all models we use an in-house chemistry library [16] to evaluate the chemical source terms and the EGlib library [17,18] for transport properties. Diffusive fluxes are modeled using the Curtiss–Hirschfelder approximation [19], including a correction velocity and the Soret effect. For H_2 flames we utilize a recently developed mechanism from the ELTE research group [20], for CH_4 flames the GRI mechanism by Smith et al. [21] is employed and for dimethyl ether flames we use a mechanism developed by Zhao et al. [22].

2.1. Flamelet model

Here, we outline how generic flamelet diffusion modeling can be facilitated in composition space. The common balance equations for the species mass fractions Y_i and the temperature T are

$$\rho \frac{\partial T}{\partial t} + \rho \mathbf{u} \cdot \nabla T$$
$$= \frac{1}{c_p}\left[\nabla \cdot (\lambda \nabla T) - \rho \nabla T \cdot \left(\sum_{i=1}^{n} c_{p,i} Y_i \mathbf{V}_i \right) - \sum_{i=1}^{n} h_i \dot{\omega}_i \right], \qquad (2)$$

$$\rho \frac{\partial Y_i}{\partial t} + \rho \mathbf{u} \cdot \nabla Y_i = -\nabla \cdot (\rho Y_i \mathbf{V}_i) + \dot{\omega}_i, \qquad (3)$$

where ρ is the density, $\mathbf{u}$ is the velocity vector, c_p and λ are the heat capacity and thermal conductivity of the mixture, h_i and $\dot{\omega}_i$ are the enthalpy and the chemical source term of species i, respectively. Eqs. (2) and (3) can be transformed into composition space, applying the transformation rules

$$\frac{\partial \phi}{\partial t} = \frac{\partial \phi}{\partial \tau} + \frac{\partial Z}{\partial t}\frac{\partial \phi}{\partial Z} \quad \text{and} \quad \nabla \phi = \nabla Z \frac{\partial \phi}{\partial Z}, \qquad (4)$$

where ϕ represents a general scalar quantity and Z denotes the mixture fraction, governed by the transport equation [23]

$$\frac{\partial \rho Z}{\partial t} + \nabla \cdot (\rho \mathbf{u} Z) = \nabla \cdot (\rho D_Z \nabla Z). \qquad (5)$$

Here, D_Z is determined based on the unity Lewis number diffusion model, i.e., $D_Z = \lambda/(\rho c_p)$. By this means, the term representing species diffusion can be reformulated as

$$Y_i \mathbf{V}_i = Y_i \tilde{V}_i \nabla Z, \qquad (6)$$

where $\tilde{V}_i$ represents a diffusion velocity in mixture fraction space. The above expression is motivated by the fact that diffusion models generally involve a product of diffusivity and scalar gradients, the latter being split in accordance with Eq. (4). Thus, $Y_i \tilde{V}_i$ contains products of the diffusivity and gradients with respect to Z instead of physical space coordinates. In the following, the approach can be utilized to easily transfer any transport model from physical space into mixture fraction space using conventional transport libraries. This includes e.g., the Curtiss–Hirschfelder diffusion model which was previously transformed into mixture fraction space [23] because of the necessity to model differential diffusion in turbulent flames and also models which account for the Soret effect. Applying Eqs. (4) and (6) to the balance equations for species and temperature, the flamelet equations are obtained as

$$\rho c_p \frac{\partial T}{\partial \tau} = \tilde{\chi}_Z \frac{\partial}{\partial Z}\left(\lambda \frac{\partial T}{\partial Z} \right)$$
$$- \tilde{\chi}_Z \frac{\partial \rho D_Z}{\partial Z}\frac{\partial T}{\partial Z} - \rho \tilde{\chi}_Z \sum_{k}^{N} c_{p,k} Y_k \tilde{V}_k \frac{\partial T}{\partial Z} - \sum_{k}^{N} h_k \dot{\omega}_k, \qquad (7)$$

$$\rho \frac{\partial Y_i}{\partial \tau} = -\tilde{\chi}_Z \frac{\partial}{\partial Z}(\rho Y_i \tilde{V}_i) - \tilde{\chi}_Z \frac{\partial \rho D_Z}{\partial Z}\frac{\partial Y_i}{\partial Z} + \dot{\omega}_i$$
$$- \left(\rho Y_i \tilde{V}_i + \rho D_Z \frac{\partial Y_i}{\partial Z} \right)\left(\frac{1}{2}\frac{\partial \tilde{\chi}_Z}{\partial Z} - \kappa_Z \sqrt{\tilde{\chi}_Z} \right). \qquad (8)$$

The quantity $\tilde{\chi}_Z$ represents the squared gradient of the mixture fraction, which is related to the scalar dissipation rate $\chi = 2D_Z \tilde{\chi}_Z = 2D_Z(|\nabla Z|)^2$. The quantities $\tilde{\chi}_Z$ and κ_Z are flamelet input parameters which are either modeled or extracted from physical space solutions.

2.2. Systematic quantification of thermal diffusion

In this work, the influence of thermal diffusion is assessed by comparing it to mixture-averaged diffusion modeling without thermal diffusion. Hence, a computation with and without the Soret effect is carried out for every flame setup under consideration. The effect of thermal diffusion on the flame structure is then quantified based on the relative deviation of the maximum of the OH radical mass fraction between both simulations

$$OH_{rel} = \frac{OH_{max}^{Mix+Soret} - OH_{max}^{Mix}}{OH_{max}^{Mix+Soret}}. \qquad (9)$$

Other quantities could also be used as observables, such as heat release rate (HRR), a differential diffusion parameter or the maximum temperature. However, we chose to use OH which besides serving well as a flame marker in a broad range of flames has two advantages:

1. **Locality** – OH is a radical species which plays an important role in the flame chemistry. It exists in a small layer in the reaction zone and quickly vanishes outside the reaction zone. Its concentration profile indicates the strength and location of a diffusion flame, which is why it is often used as a flame marker.
2. **Sensitivity** – the OH-radical mass fraction reacts very sensitively to a higher availability of fuel or oxidizer species with respect to diluents. Hence, it also indicates how particular transport effects promote or impede the transport of reactants to the reaction zone. Due to its direct connection to the diffusion of the species contributing to production and destruction of OH