Multicomponent transport in laminar flames

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Abstract

The transport fluxes and transport coefficients derived from the kinetic theory of polyatomic gas mixtures are discussed. The mathematical structure of transport linear systems and numerical algorithms for fast evaluation of accurate transport coefficients are presented. The impact of thermal diffusion and volume viscosity on flame structures and of thermodynamic nonidealities on cold mixing layers are investigated. © 2014 The Combustion Institute. Published by Elsevier Inc. All rights reserved.

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1. Introduction

Combustion models used to investigate pollutant emission, soot formation, ignition phenomena, strained or chemically controlled extinction limits combine complex chemistry with detailed transport phenomena [1–11]. The evaluation of accurate transport coefficients is therefore an important modeling and computational task. The purpose of this paper is to review the expression of transport fluxes, the mathematical structure of transport linear systems, the fast evaluation of accurate multicomponent transport coefficients as well as the impact of thermal diffusion and volume viscosity on flame structures and of thermodynamic nonidealities on cold mixing layers.

The expression of transport fluxes derived from the kinetic theory of polyatomic gas mixtures is first presented [12–22]. The transport fluxes are notably expressed in terms of transport coefficients and macroscopic variables gradients, generalizing expressions previously derived empirically. The transport coefficients for gas mixtures are still not given explicitly by the kinetic theory of gases but require solving transport linear systems. These linear systems arise from Galerkin solution of systems of linearized Boltzmann equations obtained with the Chapman–Enskog method.

The mathematical structure of the transport linear systems, that are typically singular constrained positive semi-definite systems, is discussed [23]. Symmetry properties of the linear systems and the transport coefficients are consequences of symmetry properties associated with Boltzmann collision operator [18–20]. The fast evaluation of accurate transport coefficients using either conjugate gradient methods or stationary iterative algorithms is addressed [24–30]. The relevant transport coefficients for flame calculations are reviewed and a library of fortran routines freely distributed for research purposes has been written [31].

For supercritical flames, the situation is far less satisfactory since there is no general kinetic theory of dense polyatomic fluid mixtures although some

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formal theories have been developed. The transport fluxes may notably be obtained from statistical mechanics, statistical thermodynamics, or the kinetic theory of dense mixtures of rigid spheres [32–43]. We notably discuss the thermodynamic nonidealities associated with multicomponent transport fluxes.

The importance and/or influence of multicomponent transport for computing flame structures has been emphasized by many authors [44–50]. Numerical investigations have brought further support for the importance of accurate transport property in various multicomponent reactive flows. The impact of Soret effect and multicomponent diffusion coefficients on flame structures is first investigated [51–73]. The influence of the volume viscosity coefficient—which is of the same order than the shear viscosity for polyatomic gases—is also discussed [74–85]. Finally, the impact of nonidealities on mixing layers of cold H_2 and O_2 at elevated pressure arising in supercritical flames [86–98] is studied.

2. Transport linear systems

2.1. Kinetic theories

The fundamental equations governing flames are derived from the kinetic theory of dilute polyatomic reactive gas mixtures [12–22]. The situation of mixtures of monatomic gases and single polyatomic species was first investigated [13,14,18,19], then generalized the situation of polyatomic gas mixtures [16,17] as well as reactive mixtures of polyatomic gases [7,21,22] and we refer to [7] for a summary. The situation of multitemperature flows or plasmas lay out of the scope of the present article [22,30]. The governing equations, the thermodynamic properties, the chemical production rates, the multicomponent transport fluxes, are obtained as well as a definition of transport coefficients in terms of linear systems naturally arising with the Chapman–Enskog expansion.

There are still differences in the structure of the transport linear systems and transport coefficients that may be obtained. Following Hirschfelder, Curtiss, and Bird, various authors have considered nonsymmetric coefficients as well as nonsymmetric transport linear systems [13,17] hereby destroying the natural symmetries associated with kinetic processes [99]. We consider in this article symmetric transport linear systems and diffusion coefficients—more interesting both theoretically and numerically—that have been obtained by many authors [14,16,99,18–20,7]. The symmetric diffusion coefficients have been introduced by Waldmann [14] and used in particular by Chapman and Cowling [18] and Ferziger and Kaper [19]. After the remarks of Van de Ree [99] symmetric coefficients have also been used by Curtiss [100].

2.2. Transport fluxes

The transport fluxes appearing in the multicomponent flow governing equations are the viscous tensor $\mathbf{\Pi}$, the heat flux $\boldsymbol{Q}$, and the species mass fluxes $\mathcal{F}_k = \rho_k \mathbf{v}_k, k \in S$, where $\mathbf{v}_k, k \in S$, denote the species diffusion velocity, $\rho_k, k \in S$, the species mass densities, $S = \{1, \ldots, n\}$ the set of species indices, and n the number of species. These fluxes may be written in the form [7,14,16,18–20]

$$\mathbf{\Pi} = -\kappa \mathbf{\nabla} \cdot \boldsymbol{v} \boldsymbol{I} - \eta \left(\mathbf{\nabla} \boldsymbol{v} + \mathbf{\nabla} \boldsymbol{v}^t - \frac{2}{3} (\mathbf{\nabla} \cdot \boldsymbol{v}) \boldsymbol{I} \right), \quad (1)$$

$$\mathbf{v}_k = -\sum_{l \in S} D_{kl} \boldsymbol{d}_l - \theta_k \mathbf{\nabla} \log T, \quad k \in S, \quad (2)$$

$$\boldsymbol{Q} = -\widehat{\lambda} \mathbf{\nabla} T - p \sum_{k \in S} \theta_k \boldsymbol{d}_k + \sum_{k \in S} h_k \mathcal{F}_k, \quad (3)$$

where $\boldsymbol{v}$ denotes the flow velocity, $\mathbf{\nabla}$ the space derivative operator, κ the volume viscosity, η the shear viscosity, $\boldsymbol{I}$ the three dimensional identity tensor, $D_{kl}, k, l \in S$, the multicomponent diffusion coefficients, $\boldsymbol{d}_k, k \in S$, the species diffusion driving forces, $\theta_k, k \in S$, the species thermal diffusion coefficients, T the absolute temperature, $\widehat{\lambda}$ the partial thermal conductivity, p the pressure, $h_k, k \in S$, the species enthalpy per unit mass, and ^{t} the transposition operator. The first term in the expression of the diffusion velocity $\mathbf{v}_k$ yields diffusion effects due to species driving forces while the second arising from temperature gradients corresponds to the Soret—or Ludwig-Soret—effect. The first term in the expression of the heat flux $\boldsymbol{Q}$ represents Fourier's law, the second corresponds to the Dufour effect which is symmetric of the Soret effect, and the third to the transfer of energy due to species diffusion. Letting $\mathsf{y} = (\mathsf{y}_1, \ldots, \mathsf{y}_n)^t$ where y_k is the mass fraction of the kth species, $\theta = (\theta_1, \ldots, \theta_n)^t$, $D = (D_{kl})_{k,l \in S}$, and $\langle, \rangle$ the scalar product, the diffusion matrix D and the thermal diffusion coefficients θ satisfy the mass conservation constraints $D\mathsf{y} = 0$ and $\langle \theta, \mathsf{y} \rangle = 0$. The matrix of diffusion coefficients D is symmetric, positive semi-definite with nullspace $\mathbb{R}\mathsf{y}$ and the entropy production due to diffusive processes reads $(p/T) \langle D\boldsymbol{d}, \boldsymbol{d} \rangle$ with $\boldsymbol{d} = (\boldsymbol{d}_1, \ldots, \boldsymbol{d}_n)^t$.

Assuming that gravity is the only force acting on the mixture, the species diffusion driving force $\boldsymbol{d}_k, k \in S$, may be written

$$\boldsymbol{d}_k = \mathbf{\nabla} \mathsf{x}_k + (\mathsf{x}_k - \mathsf{y}_k) \mathbf{\nabla} \log p, \quad (4)$$

where $\mathsf{x}_k, k \in S$, denote the species mole fractions. One may equivalently use the unconstrained diffusion driving forces $\widehat{\boldsymbol{d}}_k = \mathbf{\nabla} p_k / p$ where p_k denotes the partial pressure of the kth species, since

$$\boldsymbol{d}_k = \widehat{\boldsymbol{d}}_k - \mathsf{y}_k \langle \mathsf{u}, \widehat{\boldsymbol{d}} \rangle = \widehat{\boldsymbol{d}}_k - \mathsf{y}_k \mathbf{\nabla} \log p \quad \text{where}$$

$\widehat{\boldsymbol{d}} = (\widehat{\boldsymbol{d}}_1, \ldots, \widehat{\boldsymbol{d}}_n)^t$ and $\mathsf{u} = (1, \ldots, 1)^t$.

The multicomponent transport fluxes may also be written in terms of the species thermal diffusion ratios $\chi_k, k \in S$. The vector $\chi = (\chi_1, \ldots, \chi_n)^t$ is