# Fast and Accurate Multicomponent Transport Property Evaluation

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We investigate iterative methods for solving linear systems arising from the kinetic theory and providing transport coefficients of dilute polyatomic gas mixtures. These linear systems are obtained in their naturally constrained, singular, and symmetric form, using the formalism of Waldmann and Trübenbacher. The transport coefficients associated with the systems obtained by Monchick, Yun, and Mason are also recovered, if two misprints are corrected in the work of these authors. Using the recent theory of Ern and Giovangigli, all the transport coefficients are expressed as convergent series. By truncating these series, new, accurate, approximate expressions are obtained for all the transport coefficients. Finally, the computational efficiency of the present transport algorithms in multicomponent flow applications is illustrated with several numerical experiments. © 1995 Academic Press, Inc.

## 1. INTRODUCTION

In gaseous multicomponent flow calculations, transport property evaluation is an important and often time-consuming task. Indeed, the governing equations for these flows contain the transport fluxes, i.e., the pressure tensor $\Pi$, the species diffusion velocities $V_i$, for $i \in \mathcal{S}$, and the heat flux vector $q$. Using the kinetic theory of dilute polyatomic gas mixtures [1], these fluxes may be written to a first approximation in the Enskog–Chapman expansion in terms of various transport coefficients, i.e., the volume viscosity $\kappa$, the shear viscosity $\eta$, the diffusion matrix $D = (D_{ij})_{i,j \in \mathcal{S}}$, the thermal diffusion vector $\theta = (\theta_i)_{i \in \mathcal{S}}$, and the partial thermal conductivity $\lambda'$. More specifically, we have

$$\Pi = \bar{p}I - (\kappa - \tfrac{2}{3}\eta)(\nabla \cdot v)I - \eta(\nabla v + (\nabla v)^t), \quad (1.1)$$

$$V_i = -\sum_{j \in \mathcal{S}} D_{ij}d_j - \theta_i \nabla \log \bar{T}, \quad i \in \mathcal{S}, \quad (1.2)$$

$$q = \sum_{i \in \mathcal{S}} \rho h_i Y_i V_i - \lambda' \nabla \bar{T} - \bar{p} \sum_{i \in \mathcal{S}} \theta_i d_i, \quad (1.3)$$

where $\bar{p}$ is the thermodynamic pressure, $I$ the identity matrix, $v$ the mass averaged flow velocity, $d_i$ the diffusion driving force

of the $i$th species, $\bar{T}$ the absolute temperature, $\mathcal{S} = [1, n]$ the set of species indices, $n$ the number of species, $\rho$ the density, $h_i$ the enthalpy per unit mass of the $i$th species, and $Y_i$ the mass fraction of the $i$th species. The vectors $d_i$ incorporate the effects of various state variable gradients and external forces and are given by

$$d_i = \nabla X_i + (X_i - Y_i)\frac{\nabla \bar{p}}{\bar{p}} + \frac{\rho}{\bar{p}}\sum_{j \in \mathcal{S}} Y_i Y_j (b_j - b_i), \quad i \in \mathcal{S}, \quad (1.4)$$

where $X_i$ denotes the mole fraction of the $i$th species and $b_i$ is the external force per unit mass on the $i$th species. Alternatively, the diffusion velocities and the heat flux vector may be written in terms of the thermal diffusion ratios $\chi = (\chi_i)_{i \in \mathcal{S}}$ and the thermal conductivity $\lambda$ as follows [1]

$$V_i = -\sum_{j \in \mathcal{S}} D_{ij}(d_j + \chi_j \nabla \log \bar{T}), \quad (1.5)$$

$$q = \sum_{i \in \mathcal{S}} \rho h_i Y_i V_i - \lambda \nabla \bar{T} + \bar{p} \sum_{i \in \mathcal{S}} \chi_i V_i. \quad (1.6)$$

For a reactive mixture in the tempered reaction regime, the transport fluxes due to macroscopic variable gradients are also given by (1.1)–(1.6) [2].

It follows from the expressions (1.1)–(1.6) that detailed modeling of a polyatomic gas mixture requires the evaluation of its transport coefficients. These coefficients, in turn, are functions of the state variables $\bar{p}$, $\bar{T}$, and $Y_1, \dots, Y_n$. The evaluation of the transport coefficients, however, requires solving linear systems [1, 2]. Since the size of these systems can be relatively large and since transport properties have to be evaluated at each computational cell in space and time, transport property evaluation by direct numerical inversions [3–7] may become computationally expensive. As a consequence, the use of iterative techniques constitutes an interesting and appealing alternative. Moreover, analytic approximate expressions for the transport coefficients can then be obtained by truncation.

For the shear viscosity and the thermal conductivity of monatomic gas mixtures, the associated linear systems are naturally nonsingular, and iterative algorithms have been implicitly considered in [8–10]. For diffusion velocities, which involve the solution of a constrained singular system, iterative schemes have been introduced in [11]. In order to select the proper diffusion velocities, a corrector term needs to be added after convergence [11, 12]. The convergence of the Jones–Oran–Boris algorithm has been proven rigorously by Giovangigli [13] who also established that the corresponding iteration matrix has a spectral radius unity. Additional algorithms for which the iteration matrix has a spectral radius strictly lower than unity are also introduced in [13] for multicomponent diffusion matrices. These algorithms are obtained from the theory of iterative methods for singular systems and require the application of a projection matrix at each step

A systematic development of a mathematical and numerical theory of iterative algorithms for evaluating all the transport coefficients of dilute polyatomic gas mixtures is given in [2]. Various iterative schemes are proven to be convergent by using the theory of iterative methods for constrained singular linear systems and symmetric positive semi-definite matrices [2]. These convergence results strongly rely on the mathematical properties of the Boltzmann equation and the structure of the variational space selected for the species perturbed distribution functions. As a result, all the transport coefficients are expressed as convergent series, for which all the partial sums satisfy the mathematical properties that are important from a thermodynamic viewpoint, i.e., symmetry, mass conservation, and positive entropy production. Rigorously derived, analytic, approximate expressions are then obtained for all the transport coefficients by truncating convergent iterative methods.

The transport linear systems, i.e., the linear systems associated with the evaluation of all the transport properties, are obtained in [2] in their naturally constrained, singular, and symmetric form, using the formalism of Waldmann and Trübenbacher [1]. On the contrary, Monchick, Yun, and Mason [14] have systematically eliminated the singularities arising in the linear systems, by explicitly using the linear constraints and zeroing the diagonal coefficients of the system matrices, following a procedure introduced by Curtiss and Hirschfelder [8, 15]. This formulation of the linear systems presents three important drawbacks. First, the authors have obtained in [2] symmetric positive definite forms of the singular systems which can be inverted at a lower computational cost than that required for nonsymmetric systems. Furthermore, the original constrained singular symmetric systems are preferable for iterative techniques. Finally, the original systems have simpler analytic expressions so that they are better suited for analytic approximations of the transport coefficients. Note also that symmetric diffusion coefficients, which are formally compatible with Onsager reciprocal relations, have been used in [1, 2, 13, 16–19] at variance with [8, 14, 15], where this symmetry was artificially destroyed [20]. Since Waldmann and Trübenbacher [1] only

derived a formal theory, a calculation of the system coefficients was needed and has been performed in [2] for all the transport linear systems. In addition, two misprints have been identified in the paper of Monchick, Yun, and Mason.

The purpose of this paper is now to derive accurate analytic approximations for all the transport coefficients and to investigate the associated computational costs in multicomponent flow calculations. Using the theory of [2] as a starting point, various strategies designed to optimize transport property evaluation are described. Truncation of convergent series then results from a compromise between computational cost and accuracy of the corresponding expressions of the transport coefficients. The theoretical basis for the transport algorithms used in this paper is briefly presented in Sections 2 and 3. In Section 2 we describe the transport linear systems and summarize their mathematical properties. Various convergence theorems are stated in Section 3 and we refer to [2] for their proof. Finally, practical, accurate approximations for all the transport coefficients are derived in Section 4. Numerical experiments are performed illustrating the high convergence rate and the low computational cost of the present algorithms for typical multicomponent flow applications.

## 2. TRANSPORT LINEAR SYSTEMS

### 2.1. Notation

The transport linear systems are indexed with the superscript $\mu$, where $\mu = \eta$ for the shear viscosity, $\mu = \kappa$ for the volume viscosity, $\mu = D_k$, $k \in \mathcal{S}$, for the diffusion matrix, $\mu = \lambda'$ for the partial thermal conductivity and the thermal diffusion vector, and $\mu = \lambda$ for the thermal conductivity and the thermal diffusion ratios. The transport linear systems considered in this paper take on either the nonsingular form

$$G\alpha^\mu = \beta^\mu, \tag{2.1}$$

where $G$ denotes the system matrix and $\beta^\mu$, the right-hand side, or the constrained singular form

$$G\alpha^\mu = \beta^\mu,$$
$$\langle \mathcal{G}, \alpha^\mu \rangle = 0, \tag{2.2}$$

where $\mathcal{G}$ denotes the constraint vector and $\langle , \rangle$ the scalar product. Both systems are typically associated with the evaluation of the transport coefficient $\mu = \langle \alpha^\mu, \overline{\beta}^\mu \rangle$, where $\overline{\beta}^\mu$ is a given vector.

The transport linear systems are derived from a variational procedure by considering polynomial expansions of the species perturbed distribution functions. The finite dimensional functional space used in the variational procedure is denoted by $\mathcal{A}^\mu = \text{span}\{\xi^{rk}, (r, k) \in \mathcal{B}^\mu\}$, where $\xi^{rk}, (r, k) \in \mathcal{B}^\mu$, are basis functions. Here, $\mathcal{B}^\mu$ is the set of basis function indices which has $\omega$ elements. In the notation $(r, k)$, the index $k$ refers to the species and the index $r$ refers to the function type that is

considered. The basis functions $\xi^{rk}$ are generally expressed in function of the Laguerre–Sonine polynomials and the Wang Chang and Uhlenbeck polynomials in the internal energy, thus accounting for the polyatomic nature of the molecules [1].

For each transport coefficient, various transport linear systems can be considered, corresponding to different choices for the variational space $\mathcal{A}^\mu$. The standard choice yields the systems presented in Table I, where $p$ denotes the number of polyatomic species in the mixture. New transport linear systems associated with the use of variational spaces of lower dimension [2, 21, 22] are briefly discussed in Appendix A. These reduced systems yield new approximations for the transport coefficients which are generally within a few percentages of accuracy of the transport coefficients obtained with the standard systems [2]. However, since the reduced systems are of smaller size, they constitute in some cases a computationally interesting alternative to the standard systems [2, 22]. In Table I, the first column contains the system $G\alpha^\mu = \beta^\mu$; the second, the size of the system $\omega$; the third, the constraint $\langle \mathcal{G}, \alpha^\mu \rangle = 0$; and the last, the expression for the associated transport coefficient $\mu$. For the thermal diffusion ratios, the matrix $[L^{0010}, L^{0001}]$ is an $n \times (n + p)$ rectangular matrix. The explicit expressions for all of the system matrices, right-hand sides, and constraint vectors can be found in [2].

## 2.2. Block Structure of the Transport Linear Systems

The set $\mathcal{B}^\mu$, ordered with the lexicographical order, can be used as a natural indexing set. The components of any vector $x \in \mathbb{R}^\omega$ are then denoted by $x = (x_k^r)_{(r,k)\in\mathcal{B}^\mu}$. For $x, y \in \mathbb{R}^\omega$, the scalar product $\langle x, y \rangle$ is given by $\langle x, y \rangle = \sum_{(r,k)\in\mathcal{B}^\mu} x_k^r y_k^r$. Furthermore, for $x \in \mathbb{R}^\omega$, $x \neq 0$, we denote $x^\perp = \{y \in \mathbb{R}^\omega; \langle x, y \rangle = 0\}$.

We denote by $\mathbb{R}^{\omega,\omega}$ the set of square matrices of size $\omega$, and for $G \in \mathbb{R}^{\omega,\omega}$, we write $G^t$ the transpose of $G$ and $G = (G_{kl}^{rs})_{(r,k),(s,l)\in\mathcal{B}^\mu}$ the coefficients of the matrix $G$. The nullspace and range of $G$ are denoted by $N(G)$ and $R(G)$, respectively, and $I$ is the identity matrix. For $x \in \mathbb{R}^\omega$, $\text{diag}((x_k^r)_{(r,k)\in\mathcal{B}^\mu})$ denotes the diagonal matrix of $\mathbb{R}^{\omega,\omega}$ whose diagonal elements are $x_k^r$, $(r, k) \in \mathcal{B}^\mu$, ordered as $\mathcal{B}^\mu$. For $y, z \in \mathbb{R}^\omega$, the matrix $y \otimes z$ is given by $y \otimes z = (y_k^r z_l^s)_{(r,k),(s,l)\in\mathcal{B}^\mu}$.

For any function type $r$, we consider the subset $\mathcal{S}_r \subset \mathcal{S}$ given by $\mathcal{S}_r = \{k \in \mathcal{S}, (r, k) \in \mathcal{B}^\mu\}$ and we denote by $\omega_r$ the number of elements of $\mathcal{S}_r$. Note that $\mathcal{S}_r$ may differ from $\mathcal{S}$ since some types of functions do not appear for certain species. For instance, functions in the internal energy must not be considered for the monatomic species. The transport linear system matrix $G = (G_{kl}^{rs})_{(r,k),(s,l)\in\mathcal{B}^\mu}$ in $\mathbb{R}^{\omega,\omega}$ can then be partitioned into the blocks $G^{rs} = (G_{kl}^{rs})_{k\in\mathcal{S}_r, l\in\mathcal{S}_s}$ of size $\omega_r * \omega_s$. For instance, for the volume viscosity, the indexing set is given by $\mathcal{B}^\kappa = \{10\} \times \mathcal{S} \cup \{01\} \times \mathcal{P}$, where $\mathcal{B}$ is the set of polyatomic species indices, and $\mathcal{B}^\kappa$ has $\omega = n + p$ elements. Thus, the system matrix $K \in \mathbb{R}^{n+p,n+p}$ admits the block-decomposition

$$K = \begin{bmatrix} K^{1010} & K^{1001} \\ K^{0110} & K^{0101} \end{bmatrix}, \quad (2.3)$$

### TABLE I

Standard Transport Linear Systems

| System | Size | Constraint | Evaluation |
|---|---|---|---|
| $H\alpha^\eta = \beta^\eta$ | $n$ | — | $\eta = \langle \alpha^\eta, \beta^\eta \rangle$ |
| $K\alpha^\kappa = \beta^\kappa$ | $n + p$ | $\langle \mathcal{K}, \alpha^\kappa \rangle = 0$ | $\kappa = \langle \alpha^\kappa, \beta^\kappa \rangle$ |
| $L\alpha^{D_i} = \beta^{D_i}$ | $2n + p$ | $\langle \mathcal{L}, \alpha^{D_i} \rangle = 0$ | $D_{kl} = \langle \alpha^{D_k}, \beta^{D_l} \rangle$ |
| $L\alpha^{\lambda'} = \beta^{\lambda'}$ | $2n + p$ | $\langle \mathcal{L}, \alpha^{\lambda'} \rangle = 0$ | $\lambda' = (\bar{p}/T)\langle \alpha^{\lambda'}, \beta^{\lambda'} \rangle$ $\theta_k = -\langle \alpha^{\lambda'}, \beta^{D_k} \rangle$ |
| $\Lambda\alpha^\lambda = \beta^\lambda$ | $n + p$ | — | $\lambda = (\bar{p}/T)\langle \alpha^\lambda, \beta^\lambda \rangle$ $\chi = [L^{0010}, L^{0001}]\alpha^\lambda$ |

with $K^{1010} \in \mathbb{R}^{n,n}$, $K^{1001} \in \mathbb{R}^{n,p}$, $K^{0110} \in \mathbb{R}^{p,n}$, and $K^{0101} \in \mathbb{R}^{p,p}$.

The matrix $\text{diag}(G)$ is simply the diagonal of $G$ and, similarly, $\text{diag}(G^{rs})$ is the diagonal of the rectangular block $G^{rs}$, i.e., $(\text{diag}(G^{rs}))_{kl} = G_{kl}^{rs}\delta_{kl}$, for $(r, k), (s, l) \in \mathcal{B}^\mu$. The matrix formed by the diagonal of all the rectangular blocks $G^{rs}$ of $G$ plays a fundamental role in the theory of iterative algorithms for the transport linear systems [2]. It is denoted by $db(G) \in \mathbb{R}^{\omega,\omega}$ and is given by

$$db(G)_{kl}^{rs} = G_{kl}^{rs}\delta_{kl}, \quad (r, k), (s, l) \in \mathcal{B}^\mu, \quad (2.4)$$

where $\delta_{kl}$ is the Kronecker symbol. For instance, for the matrix $K$, we have

$$db(K) = \begin{bmatrix} \text{diag}(K^{1010}) & \text{diag}(K^{1001}) \\ \text{diag}(K^{0110}) & \text{diag}(K^{0101}) \end{bmatrix}. \quad (2.5)$$

The matrix $db(G)$ has very general structure properties that are given in the next section.

## 2.3. Mathematical Structure of the Transport Linear Systems

The following fundamental results are obtained directly from the Boltzmann equation and under very general assumptions on the functional space $\mathcal{A}^\mu$ [2]. For the nonsingular systems, the following property holds

(G1) The matrices $G$, $2db(G) - G$, and $db(G)$ are symmetric positive definite for $n \geq 1$.

On the other hand, for the singular systems, we have

(G2) For $n \geq 1$, the matrix $G$ is symmetric positive semi-definite and positive definite on $\mathcal{G}^\perp$, its nullspace is a one-dimensional space denoted by $N(G) = \mathbb{R}\mathcal{G}$, and we have $\langle \mathcal{G}, \mathcal{G} \rangle \neq 0$ and $\langle \mathcal{G}, \beta^\mu \rangle = 0$.

(G3) The matrix $2db(G) - G$ is symmetric positive semi-definite for $n \geq 1$; this matrix is positive definite for

$n \geq 3$; for $n = 2$ and $\mathcal{S} = \{1, 2\}$, its nullspace is given by $N(2db(G) - G) = \mathbb{R}\mathfrak{X}^*$, where $\mathfrak{X}_k^{*r} = (-1)^k\mathfrak{X}_k^r$, $(r, k) \in \mathfrak{B}^\mu$; for $n = 1$, its nullspace is given by $N(2db(G) - G) = \mathbb{R}\mathfrak{X}$.

(G4) The matrix $db(G)$ is symmetric positive semi-definite for $n \geq 1$; this matrix is positive definite for $n \geq 2$, whereas its nullspace is given by $N(db(G)) = \mathbb{R}\mathfrak{X}$ for $n = 1$.

We deduce from (G1) and (G2) that the transport linear systems are well posed; i.e., they admit a unique solution $\alpha^\mu$. Furthermore, all the singular systems can also be cast into a nonsingular form, since we have

$$\alpha^\mu = (G + a\mathcal{G} \otimes \mathcal{G})^{-1} \beta^\mu, \qquad (2.6)$$

$$\mu = \langle (G + a\mathcal{G} \otimes \mathcal{G})^{-1} \beta^\mu, \overline{\beta}^\mu \rangle.$$

Indeed, we deduce from (G2) that the matrix $G + a\mathcal{G} \otimes \mathcal{G}$ is symmetric positive definite for any real number $a > 0$ [2]. In addition, this matrix can be directly inverted at a lower computational cost than that required for the nonsymmetric forms obtained in [14], as further discussed in Section 4.1. Note, however, that iterative methods applied to the matrix $G + a\mathcal{G} \otimes \mathcal{G}$ generally converge more slowly than those applied to the original system matrix $G$ [2].

### 2.4. The Singular Limit of Vanishing Mass Fractions

In practical applications, it is important, from a computational viewpoint, to understand the mathematical and numerical behavior of the transport coefficients and the iterative algorithms when some mass fractions become arbitrarily small. Zero mass fractions lead to artificial singularities in the transport linear systems which are eliminated by considering rescaled versions of the original systems [2]. In particular, provided the diffusion matrix is replaced by the flux diffusion matrix $\tilde{D}_{kl} = Y_k D_{kl}$, $k$, $l \in \mathcal{S}$, it is proven in [2] that all the transport coefficients are smooth rational functions of the mass fractions and admit finite limits when some mass fractions become arbitrarily small. Moreover, the iterative algorithms obtained for positive mass fractions can be rewritten in terms of a rescaled system matrix that is still defined for nonnegative mass fractions [2]. For positive mass fractions, all the iterative algorithms then yield the same sequence of iterates, whether applied to the original transport linear system or to the rescaled one. This result establishes rigorously the validity of a common practice in numerical calculations, which consists in evaluating transport properties of a given gas mixture by first adding to all the species mass fractions a very small number, typically lower than the machine precision.

### 3. CONVERGENT ITERATIVE METHODS

#### 3.1. Standard Iterative Methods

We refer to [23–26] for an introduction to the solution of singular consistent linear systems by standard iterative methods.

For a matrix $G \in \mathbb{R}^{\omega,\omega}$, the decomposition $G = M - Z$ is a splitting if the matrix $M$ is invertible. In order to solve the linear systems (2.1) or (2.2), this splitting induces the iterative scheme

$$x_{i+1} = Tx_i + M^{-1} \beta^\mu, \quad i = 0, 1, ..., \qquad (3.1)$$

where $T = M^{-1}Z$. With the terminology of [24], the matrix $T \in \mathbb{R}^{\omega,\omega}$ is said to be convergent when $\lim_{i\to\infty} T^i$ exists, not necessarily being zero. When $\beta^\mu \in R(G)$, one has then the property that the iterative scheme (3.1) converges for any $x_0 \in \mathbb{R}^\omega$ if and only if the matrix $T$ is convergent.

In the following discussion we assume that the matrix $T$ is convergent. When the matrix $G$ is nonsingular, we then have $\rho(T) < 1$, where $\rho(T)$ denotes the spectral radius of the matrix $T$, so that $\lim_{i\to\infty} T^i = 0$, and for any $x_0 \in \mathbb{R}^\omega$, the iterates (3.1) converge towards $\lim_{i\to\infty} x_i = G^{-1} \beta^\mu = \alpha^\mu$. This case is considered in Theorem 1 which applies to all the nonsingular transport linear systems. On the other hand, when the matrix $G$ is singular, the spectral radius of the iteration matrix $T$ is unity, and the limit of (3.1) then explicitly depends on the initial value $x_0$ [24]. In order to obtain an iteration matrix of spectral radius strictly lower than unity, the projected version of the iterative scheme (3.1) is considered in Theorem 2 which applies to all the singular transport linear systems. Projected iterative schemes have been introduced in [13] when studying convergent iterative methods for multicomponent diffusion coefficients, and systematically used in [2] for all the singular transport linear systems. For a more mathematical discussion of projected iterative algorithms, we also refer to [26].

THEOREM 1. *Assume that* (G1) *holds and let* $M \in \mathbb{R}^{\omega,\omega}$ *be the matrix* $M = db(G) + \mathrm{diag}((\sigma_k^r)_{(r,k)\in\mathfrak{B}^\mu})$, *where the coefficients* $\sigma_k^r$, $(r, k) \in \mathfrak{B}^\mu$, *are nonnegative. Consider the splitting* $G = M - Z$, *let* $T = M^{-1}Z$, $x_0 \in \mathbb{R}^\omega$, *and consider, for* $i \geq 0$, *the iterates* (3.1). *Then the matrix* $T$ *is convergent,* $\rho(T) < 1$, *and we have the limits*

$$\lim_{i\to\infty} x_i = \alpha^\mu,$$

$$\lim_{i\to\infty} \langle x_i, \overline{\beta}^\mu \rangle = \mu, \qquad (3.2)$$

*where* $\alpha^\mu$ *is the unique solution of* (2.1). *Moreover, for* $i \geq 1$, *the quantities*

$$\mu^{[i]} = \left\langle \sum_{j=0}^{i-1} T^j M^{-1} \beta^\mu, \overline{\beta}^\mu \right\rangle, \qquad (3.3)$$

*are positive if* $\overline{\beta}^\mu = \beta^\mu \neq 0$, *and we have*

$$\lim_{i\to\infty} \mu^{[i]} = \left\langle \sum_{j=0}^{\infty} T^j M^{-1} \beta^\mu, \overline{\beta}^\mu \right\rangle = \mu. \qquad (3.4)$$

THEOREM 2. *Assume that* (G2), (G3), *and* (G4) *hold and let* $M \in \mathbb{R}^{\omega,\omega}$ *be the matrix* $M = db(G) + \mathrm{diag}((\sigma_k^r)_{(r,k)\in\mathcal{B}^\mu})$, *where the coefficients* $\sigma_k^r$, $(r, k) \in \mathcal{B}^\mu$, *are nonnegative and, in the particular cases* $n = 1$ *or* 2, *such that* $\sigma_k^r > 0$ *for at least one* $(r, k) \in \mathcal{B}^\mu$ *such that* $\mathcal{X}_k^r \neq 0$. *Consider the splitting* $G = M - Z$, *let* $T = M^{-1}Z$, *and let also* $P = I - \mathcal{X} \otimes \mathcal{G}/\langle\mathcal{X}, \mathcal{G}\rangle$ *be the oblique projector onto the hyperplane* $\mathcal{G}^\perp$ *along* $\mathbb{R}\mathcal{X}$. *Let* $x_0 \in \mathbb{R}^\omega$, $y_0 = Px_0$, *and consider, for* $i \geq 0$, *the iterates* (3.1) *and also the iterates*

$$y_{i+1} = PTy_i + PM^{-1}\beta^\mu. \tag{3.5}$$

*Then* $y_i = Px_i$ *for all* $i \geq 0$, *the matrices* $T$ *and* $PT$ *are convergent*, $\rho(T) = 1$, $\rho(PT) < 1$, *and we have the limits*

$$\lim_{i\to\infty} y_i = P(\lim_{i\to\infty} x_i) = \alpha^\mu,$$

$$\lim_{i\to\infty} \langle y_i, \overline{\beta}^\mu \rangle = \mu, \tag{3.6}$$

*where* $\alpha^\mu$ *is the unique solution of* (2.2). *Moreover, for* $i \geq 1$, *the quantities*

$$\mu^{[i]} = \left\langle \sum_{j=0}^{i-1} (PT)^j PM^{-1} P^t \beta^\mu, \overline{\beta}^\mu \right\rangle, \tag{3.7}$$

*are positive if* $\overline{\beta}^\mu = \beta^\mu \neq 0$, *and we have*

$$\lim_{i\to\infty} \mu^{[i]} = \left\langle \sum_{j=0}^{\infty} (PT)^j PM^{-1} P^t \beta^\mu, \overline{\beta}^\mu \right\rangle = \mu. \tag{3.8}$$

Note that a key point in establishing that $\rho(PT) < 1$ is that $2db(G) - G$ is positive definite for $n \geq 3$, and this property is deduced from the Boltzmann equation and the structure of the variational space $\mathcal{A}^\mu$, as stated in Section 2.3. Furthermore, the projector $P$ is needed for the convergence of the series (3.8) since the series $\Sigma_{j=0}^i T^j M^{-1}$ has no limit. In addition, since $\rho(T) = 1$, the components of $x_0$ in $N(G)$, according to the decomposition $N(G) \oplus R(I - T) = \mathbb{R}^\omega$, remain undamped in (3.1). Such difficulties are avoided with the projected algorithm involving the matrix $PT$ of spectral radius strictly lower than unity.

Finally, we deduce from Theorems 1 and 2 that all the transport coefficients can be expressed as convergent series. In the case of projected series, the projector matrix ensures that the partial sums satisfy the mathematical properties that are important from a thermodynamic viewpoint [2, 26]. In particular, the diffusion matrix can be written as a projected convergent series for which all the partial sums are symmetric, conserve mass, and yield a positive entropy production on the physical hyperplane of zero sum diffusion driving forces [2, 13, 26].

## 3.2. Conjugate Gradient Methods

Recalling that within the Waldmann and Trübenbacher formalism [1, 2], the transport linear systems are symmetric, it is also possible to use a conjugate gradient method for their solution. Therefore, we now consider conjugate gradient methods for symmetric positive definite systems [27] and positive semi-definite systems [28]. For the former systems, the convergence theorem is omitted for brevity [2]. For the latter systems, the projected conjugate gradient method, introduced in [2], is now presented in the following theorem. Note also that the use of the matrix $db(G)$ in the preconditioner arises from the theoretical results of Section 2.3.

THEOREM 3. *Assume that* (G2), (G3), *and* (G4) *hold and let* $M \in \mathbb{R}^{\omega,\omega}$ *be the matrix* $M = db(G) + \mathrm{diag}((\sigma_k^r)_{(r,k)\in\mathcal{B}^\mu})$ *or* $M = \mathrm{diag}(G) + \mathrm{diag}((\sigma_k^r)_{(r,k)\in\mathcal{B}^\mu})$, *where the coefficients* $\sigma_k^r$, $(r, k) \in \mathcal{B}^\mu$, *are nonnegative and, in the particular case* $n = 1$, *such that* $\sigma_k^r > 0$ *for at least one* $(r, k) \in \mathcal{B}^\mu$ *such that* $\mathcal{X}_k^r \neq 0$. *Let also* $P = I - \mathcal{X} \otimes \mathcal{G}/\langle\mathcal{X}, \mathcal{G}\rangle$ *be the oblique projector onto the hyperplane* $\mathcal{G}^\perp$ *along* $\mathbb{R}\mathcal{X}$. *Let* $x_0 \in \mathbb{R}^\omega$, $y_0 = Px_0$, $r_0 = \beta^\mu - Gx_0$, $p_0 = 0$, $\xi_0 = 0$, *and consider, for* $i \geq 1$, *the iterates*

$$
\begin{aligned}
p_i &= M^{-1}r_{i-1} + \xi_{i-1}p_{i-1}, \\
\zeta_i &= \langle r_{i-1}, M^{-1}r_{i-1}\rangle/\langle p_i, Gp_i\rangle, \\
x_i &= x_{i-1} + \zeta_i p_i, \\
y_i &= y_{i-1} + \zeta_i Pp_i, \\
r_i &= r_{i-1} - \zeta_i Gp_i, \\
\xi_i &= \langle r_i, M^{-1}r_i\rangle/\langle r_{i-1}, M^{-1}r_{i-1}\rangle.
\end{aligned}
\tag{3.9}
$$

*Then* $y_i = Px_i$ *for all* $i \geq 0$, *the sequence of iterates* $y_i$ *converges towards the unique solution of* (2.2) *in at most* $\omega$ *steps, and the quantities*

$$\mu^{[i]} = \langle y_i, \overline{\beta}^\mu \rangle, \tag{3.10}$$

*converge towards* $\mu = \langle \alpha^\mu, \overline{\beta}^\mu \rangle$ *in at most* $\omega$ *steps. Furthermore, if* $x_0 = 0$ *and* $\overline{\beta}^\mu = \beta^\mu \neq 0$, *we have* $\mu^{[i]} > 0$ *for all* $i \geq 1$.

## 4. PRACTICAL, ACCURATE APPROXIMATIONS FOR THE TRANSPORT COEFFICIENTS

In this section we derive accurate approximations for all the transport coefficients and investigate their computational cost. We first discuss the use of empirical expressions versus expressions rigorously derived from the kinetic theory for evaluating transport coefficients. We then perform numerical experiments illustrating the high convergence rate of the iterative methods derived in Section 3 for typical gas mixtures. Truncation to a desired accuracy then yields explicit analytic expressions for all the transport coefficients. Conjugate gradient methods usually have a higher convergence rate and should generally be

preferred. However, as opposed to standard iterative methods, they do not yield a linear dependency between the iterates and the system right members, and this dependency is of fundamental importance in some cases as for instance with the diffusion matrix. Finally, we investigate the computational cost of the present approximate expressions for the transport coefficients in multicomponent flow applications.

## 4.1. *Empirical versus Rigorously Derived Transport Algorithms*

Expressions for the transport coefficients that are rigorously derived from the kinetic theory require us to solve the associated transport linear system, by using either a direct numerical inversion or an iterative method. In order to compare these approaches, we evaluate their computational cost. Since transport property evaluation only depends on the local state of the mixture and, therefore, parallelizes and vectorizes perfectly, it is sufficient to use an operation count. As usual, we define an operation to be one multiplication plus one addition. We assume that the number of species $n$ is large, keeping in mind that this condition is met in typical multicomponent flow calculations, where $n \geq 10$. For simplicity, we assume that $p = n$ so that the transport linear systems are of size $\omega = \gamma n$ with $\gamma = 1, 2$, or 3, as indicated in Table I. We will also write $\varphi(n) = \mathcal{O}(\psi(n))$ in order to indicate that $\varphi(n)/\psi(n)$ is bounded when $n$ is large.

First, it is well known that Gaussian elimination yields the LU decomposition of a matrix in $C_{\mathrm{LU}} = \omega^3/3 + \mathcal{O}(n^2)$ operations [27]. In the particular case of symmetric positive definite matrices, one can construct in approximately half as many operations the LDL$^t$ decomposition of the system matrix or its Cholesky LL$^t$ decomposition in $C_{\mathrm{LDL}'} = \omega^3/6 + \mathcal{O}(n^2)$ operations [27]. Note, however, that this estimate for $C_{\mathrm{LDL}'}$ is only observed in practice for very large systems or when solving a family of linear systems with the family indices used in the inner loops. A typical example for the latter case is provided by the transport linear systems at each grid node. In the other cases, forming the LDL$^t$ decomposition of the system matrix is computationally cheaper than forming its LU decomposition, but not by a factor of two.

Furthermore, the cost of performing $m$ steps of any of the iterative methods described in Theorems 1–3 is $C_{m,\mathrm{IM}} = m\,\omega^2 + \mathcal{O}(n)$ operations [2]. It is, therefore, expected that an iterative method will be more cost-effective than a direct numerical inversion provided that the first few iterations already yield approximate expressions with a high enough level of accuracy. These issues will be discussed into more detail in Sections 4.2 and 4.3.

On the other hand, it is important to note that transport algorithms rigorously derived from the kinetic theory necessarily require at least $\mathcal{O}(n^2)$ operations, since each gas interacts with all the other gases present in the mixture. Consequently, it is impossible to obtain approximate expressions rigorously

derived from the kinetic theory at a cost of $\mathcal{O}(n)$ operations. We still note that it is possible to obtain *empirical* expressions at a cost of $\mathcal{O}(n)$ operations, and these expressions are usually referred to as mixture-averaged formulas. Since they are often considered as an economical alternative for transport property evaluation in multicomponent flow calculations, mixture-averaged formulas are also considered in [2]. Widely used approximations in numerical simulations are discussed and new mixture-averaged formulas are introduced, in particular the average formulas of order $t$ given by

$$\mathcal{M}_t(\mu) = \left( \sum_{k \in \mathcal{S}} X_k(\mu_k)^t \right)^{1/t}, \qquad (4.1)$$

for $t \neq 0$, and for $t = 0$, by

$$\mathcal{M}_0(\mu) = \exp\left( \sum_{k \in \mathcal{S}} X_k \log(\mu_k) \right), \qquad (4.2)$$

where the quantities $\mu_k$, $k \in \mathcal{S}$, denote the transport coefficient obtained for each pure species [2]. These low-cost expressions are not as accurate as the analytic expressions rigorously derived from the kinetic theory, but still constitute an alternative strategy which is not further considered in this paper.

## 4.2. *Accuracy*

The numerical experiments presented in this section are performed for a nine species mixture used in hydrogen–air flame computations and a 26 species mixture used in methane–air flame computations [29–31], at temperature $\overline{T} = 1000$ K and pressure $\overline{p} = 1$ atm. The chemical system used in the hydrogen–air flame is composed of the $n = 9$ species, $H_2$, $O_2$, $N_2$, $H_2O$, $H$, $O$, $OH$, $HO_2$, and $H_2O_2$, and is referred to as "the hydrogen mixture." The chemical system used for the methane–air flame is composed of the $n = 26$ species, $CH_4$, $CH_3$, $CH_2$, $CH$, $N_2$, $H_2$, $O_2$, $H_2O$, $H$, $O$, $OH$, $HO_2$, $H_2O_2$, $C_2H_6$, $C_2H_5$, $C_2H_4$, $C_2H_3$, $C_2H_2$, $C_2H$, $CHO$, $CH_2O$, $CH_3O$, $CH_2CO$, $CO_2$, $CO$, and $C_2HO$, and is referred to as "the methane mixture."

We consider three test mixtures referred to as mixtures 1, 2, and 3. Mixture 1 is an hydrogen mixture with all the mass fractions set equal to $1/n = 1/9$. In mixture 2, the mass fractions of $H_2$, $O_2$, and $N_2$ are set equal to $1/3 - 2\varepsilon$ and the remaining ones set to $\varepsilon = 1\mathrm{E}{-}4$. Finally, mixture 3 is a methane mixture with all the mass fractions set equal to $1/n = 1/26$.

The transport linear systems are evaluated using approximate collision integrals [32,33], as detailed in [2]. It is worthwhile to point out that by using approximate collision integrals, the linear system coefficients are no longer "noble" constants from theoretical physics, but merely "ordinary" numerical parameters. As a consequence, it is necessary to verify that the structure properties (G1)–(G4) are still valid for the approximate system

matrix. This has been done systematically in [2] for all the transport linear systems in the framework of the practical approximations [32, 33] that are used in this paper.

Finally, all the iterative algorithms considered in this section have been initialized by taking $x_0 = 0$ in Theorems 1–3.

*The Shear Viscosity.* We consider the linear system $H\alpha^\eta = \beta^\eta$ preconditioned by the matrix $db(H)$. Conjugate gradient methods yield the iterates $\eta^{[i]}$ for which the reduced errors

$$e_\eta^{[i]} = \frac{|\eta - \eta^{[i]}|}{\eta}, \quad i = 1, 2, 3, 4, \tag{4.3}$$

are presented in Table II for mixtures 1, 2, and 3. The first iteration yields the analytic expression

$$\eta^{[1]} = \frac{(\sum_{k \in \mathscr{S}} X_k^2 / H_{kk})^2}{\sum_{k,l \in \mathscr{S}} X_k X_l H_{kl} / (H_{kk} H_{ll})}, \tag{4.4}$$

within 2E-3 accuracy. We remind that the matrix elements $H_{kl}$, $k, l \in \mathscr{S}$, are given in [2] and are omitted for brevity. The new expression (4.4) for the shear viscosity is more accurate than the empirical Wilke formula often used in computer calculations [6, 34], and is also more cost-effective, as further discussed in Section 4.3.

*The Volume Viscosity.* We consider the linear system $K\alpha^\kappa = \beta^\kappa$ preconditioned by the matrix $\mathrm{diag}(K)$. Conjugate gradient methods yield the iterates $\kappa^{[i]}$ for which the reduced errors

$$e_\kappa^{[i]} = \frac{|\kappa - \kappa^{[i]}|}{\kappa}, \quad i = 1, 2, 3, 4, \tag{4.5}$$

are reported in Table III for the test mixtures. In particular, we can see that two iterations yield a new analytic expression for the volume viscosity within 1E-3 accuracy.

*The Partial Thermal Conductivity.* We consider the linear system $L\alpha^{\lambda'} = \beta^{\lambda'}$ preconditioned by the matrix $db(L)$. Conju-

### TABLE III

Volume Viscosity

| | Mixture 1 | Mixture 2 | Mixture 3 |
|---|---|---|---|
| 1 | 1.24E-4 | 4.91E-2 | 1.84E-2 |
| 2 | 1.04E-4 | 7.41E-6 | 9.32E-4 |
| 3 | 3.11E-6 | 3.14E-8 | 4.80E-5 |
| 4 | 1.69E-8 | 2.00E-10 | 8.89E-7 |

*Note.* Reduced errors for various mixtures with the preconditioner diag($K$).

gate gradient methods yield the iterates $\lambda'^{[i]}$ for which the reduced errors

$$e_\lambda^{[i]} = \frac{|\lambda' - \lambda'^{[i]}|}{\lambda'}, \quad i = 1, 2, 3, 4, \tag{4.6}$$

are reported in Table IV for mixtures 1, 2, and 3. After one iteration, we obtain the new analytic expression

$$\lambda'^{[1]} = \frac{\bar{p}}{T} \frac{\langle \beta^{\lambda'}, db(L)^{-1}\beta^{\lambda'}\rangle^2}{\langle db(L)^{-1}\beta^{\lambda'}, Ldb(L)^{-1}\beta^{\lambda'}\rangle}, \tag{4.7}$$

within 2E-2 accuracy. Recalling that the matrix $db(L)$ is formed by the diagonal of the nine blocks of the matrix $L$, the vector $db(L)^{-1}\beta^{\lambda'}$ is readily evaluated in $\mathcal{O}(n)$ operations by directly solving $n$ linear symmetric systems of size 2 or 3, as discussed in [2, 22]. Another expression for the partial thermal conductivity can be obtained from (4.7) by just replacing the matrix $db(L)^{-1}$ by $\mathrm{diag}(L)^{-1}$. Finally, we deduce from Table IV that two conjugate gradient iterations yield an expression for the partial thermal conductivity with an accuracy below 1E-4.

*The Thermal Conductivity.* We consider the linear system $\Lambda\alpha^\lambda = \beta^\lambda$ preconditioned by the matrix $db(\Lambda)$. Conjugate gradient methods yield the iterates $\lambda^{[i]}$ for which the reduced errors

$$e_\lambda^{[i]} = \frac{|\lambda - \lambda^{[i]}|}{\lambda}, \quad i = 1, 2, 3, 4, \tag{4.8}$$

### TABLE II

Shear Viscosity

| | Mixture 1 | Mixture 2 | Mixture 3 |
|---|---|---|---|
| 1 | 4.60E-4 | 4.93E-4 | 1.68E-3 |
| 2 | 4.50E-7 | 4.88E-9 | 1.30E-6 |
| 3 | 1.78E-10 | 4.27E-13 | 7.60E-10 |
| 4 | 4.64E-16 | — | 1.34E-14 |

*Note.* Reduced errors for various mixtures with the preconditioner $db(H)$.

### TABLE IV

Partial Thermal Conductivity

| | Mixture 1 | Mixture 2 | Mixture 3 |
|---|---|---|---|
| 1 | 1.22E-2 | 1.51E-3 | 9.02E-3 |
| 2 | 7.92E-5 | 1.85E-6 | 8.23E-5 |
| 3 | 1.88E-7 | 1.47E-7 | 3.23E-7 |
| 4 | 9.98E-9 | 2.29E-11 | 6.06E-10 |

*Note.* Reduced errors for various mixtures with the preconditioner $db(L)$.

## TABLE V

### Thermal Conductivity

|   | Mixture 1 | Mixture 2 | Mixture 3 |
|---|-----------|-----------|-----------|
| 1 | 1.17E-2   | 1.53E-3   | 8.78E-3   |
| 2 | 7.25E-5   | 1.13E-6   | 8.07E-5   |
| 3 | 5.32E-8   | 8.97E-11  | 3.00E-7   |
| 4 | 3.57E-12  | 4.36E-14  | 1.33E-12  |

*Note.* Reduced errors for various mixtures
with the preconditioner $db(\Lambda)$.

are reported in Table V for mixtures 1, 2, and 3. One iteration
yields the new analytic expression

$$\lambda^{[1]} = \frac{\overline{p}}{T} \frac{\langle \beta^\lambda, db(\Lambda)^{-1}\beta^\lambda \rangle^2}{\langle db(\Lambda)^{-1}\beta^\lambda, \Lambda db(\Lambda)^{-1}\beta^\lambda \rangle}, \qquad (4.9)$$

within 2E-2 accuracy. Recalling that the matrix $db(\Lambda)$ is formed
by the diagonal of the four blocks of the matrix $\Lambda$, the vector
$db(\Lambda)^{-1}\beta^\lambda$ is evaluated in $\mathcal{O}(n)$ operations by directly solving
$n$ linear symmetric systems of size 1 or 2, as discussed in [2,
22]. Another expression for the thermal conductivity can be
obtained from (4.9) by just replacing the matrix $db(\Lambda)^{-1}$ by
$\text{diag}(\Lambda)^{-1}$. Finally, we deduce from Table V that two conjugate
gradient iterations yield an expression for the thermal conduc-
tivity with an accuracy below 1E-4.

*The Diffusion Matrix.* Only the numerical experiments con-
cerning projected standard iterative methods will be discussed
here. Indeed, the resulting diffusion matrices satisfy the mathe-
matical properties that are important from a thermodynamic
viewpoint, that is, symmetry, mass conservation, and positive
definiteness on the physical hyperplane of zero sum gradients
[2, 13]. On the other hand, conjugate gradient methods can
also be considered for the diffusion matrix, since the iterates
converge in very few iterations [2]. However, these iterates are
not guaranteed to be symmetric nor to be positive definite on
the physical hyperplane of zero sum gradients.

Therefore, we consider projected standard iterations for the
$n$ systems $L\alpha^{D_l} = \beta^{D_l}$, $l \in \mathcal{S}$, with the splitting $L = M - Z$,
where the matrix $M$ is given by

$$M_{jk}^{rs} = \frac{L_{jk}^{rs}}{1 - Y_j}\delta_{jk}, \quad rs = 0000, j, k \in \mathcal{S},$$
$$M^{rs} = \text{diag}(L^{rs}), \quad rs \neq 0000. \qquad (4.10)$$

This yields the iterates $D^{[i]}$ for which the reduced errors

$$e_D^{[i]} = \frac{\|D - D^{[i]}\|_\infty}{\|D\|_\infty}, \quad i = 1, 2, 3, 4, \qquad (4.11)$$

where $\|D\|_\infty = \max_{k,l \in \mathcal{S}} |D_{kl}|$, are reported in Table VI. The
spectral radius of the projected iteration matrix is also included

## TABLE VI

### Diffusion Matrix

|   | Mixture 1 | Mixture 2 | Mixture 3 |
|---|-----------|-----------|-----------|
| 1 | 6.06E-2   | 6.01E-5   | 2.34E-2   |
| 2 | 1.28E-2   | 2.57E-5   | 2.42E-3   |
| 3 | 4.25E-3   | 4.69E-6   | 4.34E-4   |
| 4 | 1.73E-3   | 9.92E-7   | 8.72E-5   |
| $\rho$ | 3.90E-1 | 2.07E-1   | 2.00E-1   |

*Note.* Standard iterative methods with $M$
given by (4.10). Reduced errors and spectral
radius for various mixtures.

in this table. The first iterate yields the matrix $D^{[1]}$ within 6E-
2 accuracy. The matrix $D^{[1]}$ may be expressed as a projected
diagonal matrix

$$D^{[1]} = P \,\text{diag}(D_1^*/X_1, ..., D_n^*/X_n)P^t, \qquad (4.12)$$

where the coefficients $D_k^*$ generalize the ones arising in the
Hirschfelder–Curtiss approximate diffusion velocities [13, 35].
In addition, the projector matrix $P$ has coefficients given by $P_{kl}$
$= \delta_{kl} - Y_l/\Sigma_{i \in \mathcal{S}} Y_i$, $k, l \in \mathcal{S}$. Note that, although the mass
fractions should sum up to unity, omitting factors such as
$\Sigma_{i \in \mathcal{S}} Y_i$ may modify Jacobian matrices of discretized governing
equations when all the mass fractions are considered as indepen-
dent unknowns [36]. It is also important to observe that no
dense matrix multiplications are needed to form the projected
expressions in (4.12). Therefore, $D^{[1]}$ is evaluated in $\mathcal{O}(n^2)$ opera-
tions, although $n$ linear systems are solved.

Two iterations yield the diffusion matrix $D^{[2]}$ within 1E-2
accuracy. The matrix $D^{[2]}$ may be expressed as

$$D^{[2]} = P\mathfrak{R}M^{-1}(2M - L)M^{-1}\mathfrak{R}^t P^t, \qquad (4.13)$$

where $\mathfrak{R} \in \mathbb{R}^{n,2n+p}$ is the rectangular matrix formed by the
blocks $\mathfrak{r} = [I, 0, 0]$. Since the matrix $M$ consists of nine diagonal
blocks, the product of $M^{-1}$ with a given matrix only requires
$\mathcal{O}(n^2)$ operations, so that the computational cost of $D^{[2]}$ is still
$\mathcal{O}(n^2)$ operations. Finally, an additional iteration for the $n$ sys-
tems $L\alpha^{D_l} = \beta^{D_l}$, $l \in \mathcal{S}$, yields the matrix $D^{[3]}$ which is accurate
to 4E-3, but requires $\mathcal{O}(n^3)$ operations for its evaluation.

*The Thermal Diffusion Vector.* A new analytic expression
is obtained after two projected standard iterations for the system
$L\alpha^{\lambda'} = \beta^{\lambda'}$. The resulting thermal diffusion vector is within a
few percentages of accuracy and may be expressed as

$$\theta^{[2]} = -P\mathfrak{R}M^{-1}(2M - L)M^{-1}\beta^{\lambda'}. \qquad (4.14)$$

Note that (4.14) requires, for its evaluation, the same matrices
as the ones considered for the diffusion matrix $D^{[2]}$. Therefore,
the thermal diffusion vector $\theta^{[2]}$ and the diffusion matrix $D^{[2]}$
can be evaluated simultaneously, as needed, for instance, in

chemical vapor deposition problems [29]. We can also consider conjugate gradient methods for the system $L\alpha^{\lambda'} = \beta^{\lambda'}$ preconditioned by the matrix $db(L)$. This yields the iterates $\theta^{[i]}$ for which the reduced errors

$$e_\theta^{[i]} = \frac{\|\theta - \theta^{[i]}\|_\infty}{\|\theta\|_\infty}, \quad i = 1, 2, 3, 4, \tag{4.15}$$

where $\|\theta\|_\infty = \max_{k \in \mathcal{S}} |\theta_k|$, are reported in Table VII for mixtures 1, 2, and 3. Three iterations yield a new approximation for the thermal diffusion vector, $\theta^{[3]}$, within 5E-3 accuracy.

*The Thermal Diffusion Ratios.* We consider the linear system $\Lambda\alpha^\lambda = \beta^\lambda$ preconditioned by the matrix $db(\Lambda)$. Conjugate gradient methods yield the iterates $\chi^{[i]}$ for which the reduced errors

$$e_\chi^{[i]} = \frac{\|\chi - \chi^{[i]}\|_\infty}{\|\chi\|_\infty}, \quad i = 1, 2, 3, 4, \tag{4.16}$$

are reported in Table VIII. Three iterations yield a new approximate expression for the thermal diffusion ratios, $\chi^{[3]}$, within 4E-4 accuracy.

### 4.3. *Computational Cost*

In the previous section we have examined the accuracy of the approximate expressions derived for all the transport coefficients by truncating convergent iterative methods. One to three iterations have generally yielded analytic expressions for the transport coefficients with an accuracy between 1E-3 and 1E-2. Indeed, for the shear viscosity, the volume viscosity, the partial thermal conductivity, and the thermal conductivity, the first conjugate gradient iterate already yielded excellent accuracy. For the diffusion matrix, the thermal diffusion vector, and the thermal diffusion ratios, two or three iterations were needed, using either conjugate gradient or standard iterative methods. Note that more accurate expressions are not appropriate since the transport linear systems are derived by keeping only the first terms in the polynomial expansions of the species perturbed distribution functions.

The next issue that needs to be addressed is the computational cost of the present transport algorithms in practical implementa-

**TABLE VII**

Thermal Diffusion Vector

| | Mixture 1 | Mixture 2 | Mixture 3 |
|---|---|---|---|
| 1 | 2.61E-1 | 5.81E-1 | 1.66E-1 |
| 2 | 2.62E-2 | 9.85E-3 | 6.97E-3 |
| 3 | 2.38E-3 | 5.10E-3 | 1.53E-3 |
| 4 | 4.19E-4 | 6.81E-5 | 1.47E-4 |

*Note.* Reduced errors for various mixtures with the preconditioner $db(L)$.

**TABLE VIII**

Thermal Diffusion Ratios

| | Mixture 1 | Mixture 2 | Mixture 3 |
|---|---|---|---|
| 1 | 9.35E-2 | 6.12E-3 | 5.51E-2 |
| 2 | 5.10E-3 | 5.45E-4 | 7.86E-3 |
| 3 | 7.52E-5 | 2.95E-6 | 4.17E-4 |
| 4 | 3.05E-7 | 8.79E-8 | 2.73E-7 |

*Note.* Reduced errors for various mixtures with the preconditioner $db(\Lambda)$.

tions of multicomponent flow models. Therefore, we now compare direct numerical inversions versus iterative methods in the framework of a two-dimensional methane–air diffusion flame model [37]. We first restate that transport property evaluation only depends on the local state of the mixture. Optimal vectorization of transport property evaluation thus requires multiple input data subroutines that compute simultaneously transport properties over a wide number of grid nodes [38]. This approach has been implemented for instance in [29–31, 37] for various combustion problems. Vectorization is hence straightforward for the iterative algorithms as well as for the LDL' and LL' decompositions. For the LU decomposition, however, difficulties arise with the pivoting part of the algorithm. On the other hand, parallel optimization of transport property evaluation depends on the problem granularity [38]. For coarse-granularity parallel computations in which each processor is responsible for a different portion of the computational domain, multiple input data subroutines are preferable [38]. For fine-grained distributed parallel architectures, simple input data subroutines that only consider one state of the mixture must be used. In this paper only single input data subroutines will be investigated. The implementation of multiple input data subroutines is in progress and goes beyond the scope of this paper.

We now investigate the computational efficiency of iterative algorithms versus direct numerical inversions for a two-dimensional methane–air diffusion flame model involving 16 chemical species and $N = 7.5 \times 10^3$ mesh nodes [37]. For this problem, all the transport coefficients have been evaluated by using direct numerical inversion and also by truncating an iterative method. Apart from the diffusion matrix for which projected standard iterative methods were used, the conjugate gradient method has been considered for all the other transport coefficients. Based on the results of Section 4.2, we have evaluated the approximate expressions $\eta^{[1]}$, $\kappa^{[1]}$, $D^{[2]}$, $\lambda'^{[3]}$, $\theta^{[3]}$, $\lambda^{[3]}$, and $\chi^{[3]}$. Note that in applications where thermal diffusion is not included, it may be more efficient to evaluate the approximate thermal conductivity $\lambda^{[1]}$ or the partial thermal conductivity $\lambda'^{[1]}$ since these expressions already yield excellent accuracy.

The computations for the single input data subroutines were carried out on an IBM Risc System 6000 (model 560) workstation and on a CRAY C98 computer in scalar mode. The numerical results are reported in Tables IX and X, respectively. In

## TABLE IX

Comparison of Iterative Methods versus Direct Numerical Inversion for Transport Property Evaluation Applied to a Two-Dimensional Flame Problem

| Coefficient | $\eta^{[1]}$ | $\kappa^{[1]}$ | $\lambda^{[3]}, \chi^{[3]}$ | $\lambda'^{[3]}, \theta^{[3]}, D^{[2]}$ | All |
|---|---|---|---|---|---|
| Accuracy | 3.56E-3 | 4.76E-3 | 5.88E-9 ($\lambda$) 2.31E-4 ($\chi$) | 1.09E-7 ($\lambda'$) 1.14E-3 ($\theta$) 8.83E-4 ($D$) | — |
| $S_{LU}$ | 6.21 | 13.3 | 4.28 | 3.99 | 4.55 |
| $S_{LDL'}$ | 5.35 | 10.8 | 3.47 | 3.77 | 4.19 |
| $S'_{LU}$ | 1.14 | 1.61 | 1.33 | 2.20 | 2.12 |
| $S'_{LDL'}$ | 1.12 | 1.49 | 1.25 | 2.12 | 2.01 |

*Note.* Results obtained on the IBM workstation using single input data subroutines.

Table IX, the first line contains the relative error for the transport coefficient obtained by truncation averaged over the number of nodes. For instance, for the shear viscosity, we have reported

$$e_\eta = \left( \sum_{\nu=1}^{N} \frac{|\eta_\nu - \eta_\nu^{[1]}|^2}{\eta_\nu^2} \right)^{1/2}, \tag{4.17}$$

where $\eta_\nu$ and $\eta_\nu^{[1]}$ denote the shear viscosity at node $\nu$ obtained with direct numerical inversion and with one conjugate gradient iteration, respectively. As illustrated in Table IX, excellent accuracy is obtained for all the transport coefficients.

The last four lines in Tables IX and X indicate the speedups obtained when using an iterative method rather than a direct numerical inversion. Direct numerical inversion was performed by forming either the LU or the LDL' decomposition of the system matrix. For very large linear systems or when using multiple input data subroutines, the latter method requires approximately half the computational cost of the former. For the present linear systems the differences observed between both methods are not as large, but in all cases the LDL' decomposition is computationally cheaper. The evaluation of the transport linear system is not included in the CPU times when computing the speedups $S_{LU}$ and $S_{LDL'}$, but it is in the speedups $S'_{LU}$ and

## TABLE X

Comparison of Iterative Methods versus Direct Numerical Inversion for Transport Property Evaluation Applied to a Two-Dimensional Flame Problem

| Coefficient | $\eta^{[1]}$ | $\kappa^{[1]}$ | $\lambda^{[3]}, \chi^{[3]}$ | $\lambda'^{[3]}, \theta^{[3]}, D^{[2]}$ | All |
|---|---|---|---|---|---|
| $S_{LU}$ | 5.50 | 13.8 | 4.32 | 4.19 | 4.86 |
| $S_{LDL'}$ | 3.76 | 9.53 | 2.91 | 3.57 | 3.97 |
| $S'_{LU}$ | 1.37 | 2.80 | 1.74 | 2.85 | 3.01 |
| $S'_{LDL'}$ | 1.23 | 2.20 | 1.43 | 2.49 | 2.55 |

*Note.* Results obtained on the CRAY C98 computer in scalar mode using single input data subroutines.

## TABLE XI

Comparison of Various Strategies for Evaluating the Shear Viscosity

| Coefficient | $\theta$ | $\theta_{app}$ | $\theta^{[1]}$ | $\theta^{[1]}_{app}$ | Wilke |
|---|---|---|---|---|---|
| Accuracy | 0 | 3.29E-3 | 3.56E-3 | 6.08E-3 | 8.20E-3 |
| Speedup | 1.00 | 1.60 | 1.12 | 1.92 | 0.71 |

*Note.* The accuracies and the speedups are evaluated with respect to $\eta$.

$S'_{LDL'}$. When the evaluation of the transport linear system is not included, the speedups range between 2.91 and 13.8. When it is included, speedups between 1.12 and 3.01 are obtained. It is also interesting to note that speedups obtained on the CRAY C98 are generally higher than on the IBM Risc System 6000.

In the last column of Tables IX and X, we have evaluated the transport coefficients $\eta^{[1]}$, $\kappa^{[1]}$, $D^{[2]}$, $\lambda'^{[3]}$, and $\theta^{[3]}$. This choice is motivated by the fact that in most multicomponent flow applications, all the transport coefficients are needed. In this case, several collision integrals need to be evaluated only once and can be subsequently used in various transport linear systems, thereby reducing the computational cost of the system evaluation. As indicated in Tables IX and X for the present multidimensional flame problem, iterative methods yield an overall speedup of a factor of three with respect to direct numerical inversions. This, in turn, may result in significant savings in the total time needed to solve multidimensional flow problems.

Since the evaluation of the transport linear system accounts for a significant amount of the CPU time required to implement the iterative algorithms, it is interesting to consider some simplifications in the modeling of collision integrals. This would indeed yield transport linear systems that can be evaluated faster and thus more cost-effective transport algorithms. Consider, for instance, the shear viscosity $\eta$. As discussed in [2], the associated transport linear system may be expressed in terms of a ratio of collision integrals, denoted by $\bar{A}_{kl}$, $k, l \in \mathcal{S}$. In the previous numerical experiments, this quantity was evaluated as a function of the logarithm of a reduced temperature [2]. This has yielded, after one conjugate gradient iteration, the approximation $\eta^{[1]}$. An interesting simplification in the transport

## TABLE XII

Reduced Transport Linear Systems

| System | Size | Constraint | Evaluation |
|---|---|---|---|
| $K_{[01]}\alpha^\kappa_{[01]} = \beta^\kappa_{[01]}$ | $p$ | — | $\kappa_{[01]} = \langle \alpha^\kappa_{[01]}, \beta^\kappa_{[01]} \rangle$ |
| $L_{[e]}\alpha^{D_i}_{[e]} = \beta^{D_i}_{[e]}$ | $2n$ | $\langle \mathcal{L}_{[e]}, \alpha^{D_i}_{[e]} \rangle = 0$ | $D_{[e]kl} = \langle \alpha^{D_k}_{[e]}, \beta^{D_l}_{[e]} \rangle$ |
| $L_{[00]}\alpha^{D_i}_{[00]} = \beta^{D_i}_{[00]}$ | $n$ | $\langle \mathcal{L}_{[00]}, \alpha^{D_i}_{[00]} \rangle = 0$ | $D_{[00]kl} = \langle \alpha^{D_k}_{[00]}, \beta^{D_l}_{[00]} \rangle$ |
| $L_{[e]}\alpha^{\lambda'}_{[e]} = \beta^{\lambda'}_{[e]}$ | $2n$ | $\langle \mathcal{L}_{[e]}, \alpha^{\lambda'}_{[e]} \rangle = 0$ | $\lambda'_{[e]} = (\bar{p}/T)\langle \alpha^{\lambda'}_{[e]}, \beta^{\lambda'}_{[e]} \rangle$ |
| | | | $\theta_{[e]k} = -\langle \alpha^{\lambda'}_{[e]}, \beta^{D_k}_{[e]} \rangle$ |
| $\Lambda_{[e]}\alpha^\lambda_{[e]} = \beta^\lambda_{[e]}$ | $n$ | — | $\lambda_{[e]} = (\bar{p}/T)\langle \alpha^\lambda_{[e]}, \beta^\lambda_{[e]} \rangle$ |
| | | | $\chi_{[e]} = [L^{00e}_{[e]}]\alpha^\lambda_{[e]}$ |

linear system is obtained with the approximation $\overline{A}_{kl} = 1.1$, $k$, $l \in \mathcal{S}$. A direct numerical inversion of this approximate system yields a shear viscosity coefficient denoted by $\eta_{app}$. Similarly, one conjugate gradient iteration for this simpler system yields a new approximation, $\eta_{app}^{[1]}$. In Table XI we have considered these various strategies for evaluating the shear viscosity. We have computed $\eta$, $\eta_{app}$, $\eta^{[1]}$, $\eta_{app}^{[1]}$, and also the shear viscosity resulting from the Wilke approximation. The quantities $\eta$ and $\eta_{app}$ have been computed by forming the $LDL^t$ decomposition of the system matrix. As illustrated in Table XI, the new approximation $\eta_{app}^{[1]}$ is within 6E-3 accuracy of $\eta$ and can be evaluated almost three times faster than the Wilke approximation. We can also see that $\eta^{[1]}$ is twice as accurate as the Wilke approximation and is also computationally more effective. Finally, we point out that similar simplifications for collision integrals yield accurate approximations for the volume viscosity and the diffusion matrix, but not for thermal diffusion and thermal conduction transport coefficients.

## 5. CONCLUSION

The general theory of iterative methods for multicomponent transport algorithms [2] has been used systematically in order to obtain new, rigorously derived, approximate expressions for all the transport coefficients of dilute polyatomic gas mixtures. For each transport coefficient, approximate expressions have been discussed from a practical viewpoint by considering the trade-off between computational cost and accuracy. One to three conjugate gradient iterations generally yield excellent accuracy at a moderate computational cost. Projected standard iterative methods are preferable for the diffusion matrix and, in some cases, for the thermal diffusion vector also. The high convergence rates observed in all cases indicate that iterative methods constitute an appealing alternative to direct inversions in multicomponent flow calculations and a very general, rigorous, and efficient technique for multicomponent transport property evaluation.

## APPENDIX A: REDUCED TRANSPORT LINEAR SYSTEMS

In this appendix we briefly discuss the use of reduced transport linear systems for multicomponent transport evaluation. These reduced systems are obtained by using smaller variational spaces which reduces the size of the resulting linear systems and hence simplifies the transport algorithms. The reduced systems yield new transport coefficients which are generally within 1E-3 to 5E-2 accuracy of the transport coefficients obtained with the standard systems [2, 22]. However, since the simplified systems are of smaller size, they constitute in some cases a computationally interesting alternative to the standard systems.

In this paper, the transport coefficients corresponding to the largest variational space $\mathcal{A}^\mu$ have been denoted by $\mu$, and the ones associated with a reduced variational space will then be denoted by $\mu_{[x]}$, where x stands for a simple symbol associated

with the reduced variational space. The reduced transport coefficients introduced in [2, 22] are:

1. The volume viscosity $\kappa_{[01]}$ associated with a linear system of size $p$ instead of $n + p$;

2. The first-order diffusion matrix $D_{[00]}$ associated with a linear system of size $n$, instead of $2n + p$;

3. The partial thermal conductivity $\lambda'_{[e]}$, the thermal diffusion vector $\theta_{[e]}$, and the diffusion matrix $D_{[e]}$ associated with a linear system of size $2n$ instead of $2n + p$;

4. The thermal conductivity $\lambda_{[e]}$ and the thermal diffusion ratios $\chi_{[e]}$ associated with a linear system of size $n$, instead of $n + p$.

The linear systems associated with these reduced transport coefficients are summarized in Table XII.

It is interesting to note that the volume viscosity $\kappa_{[01]}$ can be evaluated explicitly in the framework of practical approximations for collision integrals [2, 32, 33]. Furthermore, the thermal conductivity $\lambda_{[e]}$ and the partial thermal conductivity $\lambda'_{[e]}$ are generally very accurate [2, 22]. Finally, we point out that iterative methods yield approximate expressions for all the reduced transport coefficients in the same form as those presented in Section 4. These expressions are given in [2, 22] and are omitted for brevity.

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