



# Efficient calculation of multicomponent diffusion fluxes based on kinetic theory



Manuel Arias-Zugasti\*, Pedro L. Garcia-Ybarra, Jose L. Castillo

Departamento de Física Matemática y de Fluidos, Facultad de Ciencias UNED, Senda del Rey 9, 28040 Madrid, Spain

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## ABSTRACT

The calculation of the diffusion matrix in mixtures of dilute gases with large numbers ( $N$ ) of components is revisited. An extremely simple relation providing the multicomponent diffusion matrix as a power series in terms of the  $N - 1$  independent mole fractions in the mixture is analytically derived from the kinetic theory of gases. This power series provides a convergent scheme with high convergence rate in the case of a gas mixture with one major component in which the remaining  $N - 1$  species are diluted. However, the convergence rate of this power series is lower if a certain number ( $M$ ) of these species are far from the dilute limit. In that case we show that a straightforward modification of the former scheme leads to a relation providing the diffusion matrix as a power series in terms of a subset of  $N - 1 - M$  mole fractions, which are assumed to be dilute, keeping full dependence on the mole fractions of the remaining  $1 + M$  major species. This relation takes full advantage of the usual situation found in combustion, where there is a relatively small number of major species (here  $1 + M$ ), with the remaining  $N - 1 - M$  chemical components (often radicals) being in trace amounts. The error found in the diffusion fluxes with each one of these new methods in a typical combustion scenario is analyzed as a function of the number of terms included in the expansion. It is found that the second method (based on a power series in terms of the dilute species), truncated at the linear term, produces relative errors less than 1% in all the cases considered—including cases far from the dilute limit. Hence, this method provides an efficient tool for the accurate calculation of multicomponent diffusion fluxes in combustion.

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## 1. Introduction

The structure and stability of flame fronts in multi-component gas mixtures depend, to a great extent, on the diffusion properties of the chemical species at the flame front, as emphasized in earlier works [1–9] and more recently by [10–16]. In particular multicomponent transport may lead to substantial local modifications of turbulent flame properties [13]. These modifications are especially noticeable on the propagation velocity and flame stretch in flames with high curvature regions or when quenching phenomena arise.

As is well known, a proper knowledge of the multicomponent diffusion matrix  $D_{ij}$  is essential for accurate calculations of multicomponent diffusion fluxes. At moderate pressures, the ensemble of major chemical species, as well as the radicals, behaves to a very good approximation as a multi-component mixture of dilute gases. In that case the kinetic theory of gases (KTG) provides a general framework for the calculation of the multicomponent diffusion coefficients  $D_{ij}$ , through the Chapman–Enskog expansion [17–19]. However, in the

general case of a multicomponent mixture with a possibly large number of components ( $N$ ), the multicomponent diffusion matrix is provided by the KTG in the form of a linear system of  $N$  algebraic equations, which has to be solved. In principle there is no difficulty in solving that linear system. However, if the number of components in the mixture is high it becomes computationally expensive to calculate the exact solution for the multicomponent diffusion matrix. This problem is particularly important in numerical simulations of unsteady flames, where the diffusion matrix has to be re-calculated at every time step and every node in the computational mesh. This difficulty has hampered the accurate evaluation of multicomponent transport properties in numerical simulations of unsteady (2D and even more 3D) flames with complex kinetic mechanisms.

In combustion, depending on the fuel involved the number of species usually found in most detailed kinetic schemes ranges between  $\mathcal{O}(10)$  to several tens of species for light fuels as hydrogen, CO and light hydrocarbons (see, e.g., [20]), and several hundreds of species for heavy fuels as longer chain hydrocarbons or usual engine fuels (see, e.g., [21]). However, in some extreme cases the realistic chemistry of practical fuels requires consideration of several thousands of species. For instance this is the case of methyl decanoate, used as a biodiesel fuel surrogate, with a detailed kinetic mechanism

\* Corresponding author. Fax: +34 91 398 7628.

E-mail address: [maz@dfmf.uned.es](mailto:maz@dfmf.uned.es), [maz@ccia.uned.es](mailto:maz@ccia.uned.es) (M. Arias-Zugasti).

involving  $N = 3034$  chemical species [22]. In such extreme cases the evaluation of transport properties becomes problematic even for simple approximations as the *mixture averaged* transport properties [14,23–25], which has motivated diffusion coefficient reduction schemes through *species bundling* [26]. The difficulties inherent to the implementation of large kinetic schemes in numerical computations have been the subject of several reviews (see, e.g., [27] and references therein).

Whereas chemical kinetics effects have been extensively analyzed in earlier combustion studies, the detailed diffusive transport which is equally important regarding flame front properties has received comparatively less attention. An exception to the former general rule is provided by the works of Giovangigli (see, e.g., [10,12,16,28–31] and references therein). For instance [28] proves the convergence of earlier iterative methods of Jones and Boris [3] (see also Oran and Boris [4]) for the calculation of the diffusion matrix. On the other hand, projected iterative algorithms for the calculation of transport properties of multicomponent mixtures are provided in [12,29–31] and references therein. A different strategy is that adopted by Xin et al. in a recent work [32]. In this case Xin et al. [32] perform a sensitivity analysis to determine the group of species which diffusive transport has the strongest impact on the flame dynamics. This group, typically formed by the major species plus a few specific radicals, is referred to as the *critical-diffusivity-species* (CDS), and the remaining species are termed as the *non-critical-diffusivity-species* (NCDS). Then, the diffusive transport of the CDS is computed solving the corresponding KTG multicomponent linear system, while the diffusive transport of the NCDS is computed by means of the mixture averaged approximation.

The purpose of this work is to provide two new simple and accurate analytical expressions for the evaluation of the multicomponent diffusion matrix and fluxes of typical combustion gas mixtures, that can be easily handled in future numerical as well as theoretical studies. The method to achieve this goal is to take full advantage of the usual scenario often found in multicomponent mixtures with large numbers of components, especially in combustion, i.e.: *there is a subset of a relatively small number of major species, in which the remaining species are dilute*. This is a simplification not taken into account in existing approximate methods for the calculation of transport properties of multicomponent mixtures (loc. cit.). The present work is based on the way devised in [7] for the evaluation of transport properties of multicomponent systems. In the methodology introduced by [7] the pertinent KTG results are expanded in terms of the diluted species mole fractions, and then the corresponding linear systems of equations are obtained and solved to each order. In [7] this method was pursued up to first order and the expressions of the diffusive fluxes were fully worked out and used in an analytical study of the dynamics of curved flame fronts.

Two new analytical formulae for the calculation of  $D_{ij}$  are provided here: Model 1 (see Eq. (23)) and Model 1+ $M$  (see Eqs. (39) and (40)). Although both models, 1 and 1+ $M$ , are analytical results, practical and efficient application of each model (i.e., fast convergence rate) depends on the number of major species in the mixture. Model 1 is extremely simple and has fast convergence rate in the case of a single major component in which the remaining  $N - 1$  species are diluted. Model 1+ $M$ , on the other hand, considers the case in which there is a small number ( $M$ ) of additional major species besides species  $N$ th, in which the remaining  $N - 1 - M$  species are dilute.

The paper is organized as follows. Section 2 summarizes the subject of Fick diffusion in multicomponent gases, as well as the notation used. The linear system that determines the diffusion matrix according to the kinetic theory of gases is presented in Section 3. This section also contains the two new algorithms proposed here to solve that linear system (see Sections 3.2 and 3.3). The details needed for the derivation of the former new algorithms and the evaluation of matrix  $A$  (Eq. (21)) are left for Appendix A and Appendix B respectively. The

performance of these two new algorithms is illustrated in Section 4 for the particular case of premixed hydrogen combustion as a function of dilution. In this section the results found with Model 1 and Model 1+ $M$  are compared to the corresponding results found with: the familiar mixture averaged approximation; Giovangigli's iterative method (see, e.g., [12]); and with the KTG *exact* results found using the exact solution for the multicomponent diffusion matrix. Finally, Section 5 summarizes the main conclusions derived from the former results.

## 2. Fick diffusion in multicomponent gases

Let us consider a multicomponent ideal gas mixture with  $N$  different chemical species and assume that all state variables of the mixture are given quantities (mass density  $\rho$ , molecular number density  $n$ , pressure  $p$ , absolute temperature  $T$ ). Using the already classical notation defined in, e.g., [19], we denote the *mole fraction* of species  $i$  by  $x_i = n_i/n$ , and the *mass fraction* of species  $i$  by  $y_i = \rho_i/\rho$ , where  $n_i$  is the *molecular* number density of species  $i$  and  $\rho_i$  is the local mass density of species  $i$ . The mean molecular mass of the mixture,  $m = \rho/n$ , is given by the relations

$$m = \sum_{i=1}^N m_i x_i = \left( \sum_{i=1}^N \frac{y_i}{m_i} \right)^{-1} \quad (1)$$

where  $m_i$  is the *molecular mass* of component  $i$  (equal to the corresponding mole mass over Avogadro's number), which together with the normalization relations

$$\sum_{i=1}^N x_i = \sum_{i=1}^N y_i = 1 \quad (2)$$

provide the conversion between mole and mass fractions

$$m_i x_i = m y_i \quad (3)$$

As is well known [9,12,17–19,25,33–35] the contribution to the mass diffusion flux of species  $i$  ( $\mathbf{j}_i$ ) owed to composition inhomogeneities in the mixture, barodiffusion and differential external forces, is given by the generalized Fick's law, which in the present notation reads as

$$\mathbf{j}_i = -\rho y_i \sum_{j=1}^N D_{ij} \mathbf{d}_j, \quad i = 1, \dots, N \quad (4)$$

where  $D_{ij}$  is the multicomponent diffusion matrix and  $\mathbf{d}_i$  are the mass diffusion driving force vectors, given by ([19] p. 171)

$$\mathbf{d}_i = \nabla x_i + (x_i - y_i) \nabla \ln p - \frac{\rho y_i}{p} \left( \mathbf{F}_i - \sum_{j=1}^N y_j \mathbf{F}_j \right) \quad (5)$$

where  $\mathbf{F}_i$  is the body force per unit mass acting on species  $i$ .

Because of the normalization relations Eq. (2) the mass diffusion driving force vectors  $\mathbf{d}_i$  are linearly dependent

$$\sum_{i=1}^N \mathbf{d}_i = 0 \quad (6)$$

as can be easily checked. As a consequence Eq. (4) can be re-written as

$$\mathbf{j}_i = -\rho y_i \sum_{j=1}^{N-1} (D_{ij} - D_{iN}) \mathbf{d}_j, \quad i = 1, \dots, N \quad (7)$$

in terms of the first  $N - 1$  diffusion driving force vectors, which are independent. On the other hand, because of mass conservation the sum of all mass diffusion fluxes vanishes

$$\sum_{i=1}^N \mathbf{j}_i = 0 \quad (8)$$

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