Turbulent Diffusion of Combustion Gaseous Admixtures

T. Elperin¹, N. Kleeorin¹, M. Liberman², and I. Rogachevskii¹ ¹Department of Mechanical Engineering, Ben-Gurion University of the Negev P. O. Box 653, Beer-Sheva, Israel ²Nordita, KTH Royal Institute of Technology and Stockholm University Stockholm, Sweden

1 Introduction

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Turbulent transport in flows with chemical reactions is of great interest in various applications, ranging from combustion physics to the turbulent atmosphere. During the decades turbulent transport of passive scalar and particles has been fore-front topic in analytical, numerical and laboratory studies. However, impact of chemical reactions on turbulent transport have been studied mainly numerically and/or in the context of turbulent combustion. Turbulent combustion can proceed as a homogeneous burning of the turbulent gaseous mixture or propagate as a flame front separating unburned fuel and combustion products in a turbulent flow [1]. Typically turbulence is created by an external forcing and can be enhanced by intrinsic instability of the flame front (see, e.g., [1-3]). The effect of chemistry on turbulent diffusion was studied by means of a path-integral approach for the Kraichnan-Kazantsev model of the random velocity field and it was shown that the turbulent diffusion can be strongly depleted by chemical reactions or phase transitions [4]. In the present study we investigate turbulent transport of chemically reacting gaseous admixtures in a developed turbulence using a spectral tau approximation for high-order closure procedure [5, 6]. Impact of the chemical reaction on turbulent diffusion is investigated using a global one-step chemical reactions taking into account the reaction order. It is shown that for a large turbulent Damköhler number turbulent diffusion of the admixtures can be strongly reduced by a large factor Da_T , depending on the stoichiometric coefficients of chemical species.

2 Governing equations and turbulent flux

Advection-diffusion equations for the number density $n_{\beta}(t, \mathbf{r})$, the temperature field $T(t, \mathbf{r})$ of the chemically reacting species in a turbulent flow of the gas (fluid) of the density ρ and velocity \mathbf{v} are:

$$\frac{\partial n_{\beta}}{\partial t} + \nabla \cdot (n_{\beta} \mathbf{v}) = -\nu_{\beta} \mathcal{W}(n_{\beta} T) + \hat{D}(n_{\beta}), \tag{1}$$

$$\frac{\partial T}{\partial t} + (\mathbf{v} \cdot \nabla)T + (\gamma - 1)T(\nabla \cdot \mathbf{v}) = q \mathcal{W}(n_{\beta}T) + \hat{D}(T), \qquad (2)$$

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) = 0, \tag{3}$$

Correspondence to: mliber@nordita.org (M. Liberman)

$$\frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v} \cdot \nabla)\mathbf{v} = -\frac{1}{\rho}\nabla P + \hat{D}_{\mathbf{v}}(\mathbf{v}), \qquad (4)$$

where v is the gas velocity, $\hat{D}(n_{\beta}) = \text{div}[\rho D_{\beta} \nabla (n_{\beta} / \rho)]$ is the linear diffusion operator of n_{β} , D_{β} is the coefficient of the molecular diffusion based on molecular Fick's law, T is the gas temperature, $\mathbf{v}_{\beta} \mathcal{P}(n_{\beta}T)$ is the source term, \mathbf{v}_{β} is the stoichiometric coefficient, $\sum_{\beta=1}^{m} \mathbf{v}_{\beta}$ is the overall order of the reaction, *m* is total number of species, $\hat{D}(T) = \rho^{-1} \text{div}[\rho \chi \nabla(T)]$ determines the molecular diffusion of the gas temperature, χ is the coefficient of molecular diffusion of temperature, $\hat{D}_{y}(\mathbf{v})$ is the viscous term, P is the gas pressure, $\gamma = c_p/c_v$. We use a one-step reaction model described by the Arrhenius kinetics $\mathcal{W}(n_{\beta},T) = A \exp(-E_a / RT) \prod_{\beta=1}^{m} (n_{\beta})^{\nu_{\beta}}$, where A is the pre-exponential factor, E_a is the activation energy, Q is the reaction energy release, and $q = Q/\rho c_p$. Notice that the equilibrium composition and temperature of the combustion products are purely thermodynamic characteristics determined by thermodynamic equilibrium laws yielding a relation between the initial and final states. In this case, chemical kinetics described by a one-step Arrhenius model provides results which usually are in good agreement with experimental data. On the contrary, in order to reproduce transient processes that are accompanied by compression and shock waves, it is necessary to take into account the detailed chemical reaction mechanisms. In the latter case the chemical time scales, such as induction time and period of exothermal reaction, compete with transport time scales in the formation of energy release zone and the propagating flame evolution [7].

Using a mean-field approach, we average Eqs. (1, 2) over an ensemble of turbulent velocity fields, assuming for simplicity the mean fluid velocity $\mathbf{U} = 0$, decompose the number density of the admixtures and the gas temperature into the mean quantities, \overline{N}_{β} and \overline{T} , and obtain for fluctuations

$$n'_{\beta} = n_{\beta} - N_{\beta} \text{ and } \theta = T - T:$$

$$\frac{\partial n'_{\beta}}{\partial t} + \nabla \cdot (n'_{\beta} \mathbf{u} - \langle n'_{\beta} \mathbf{u} \rangle) = -\nu_{\beta} (\mathcal{W} - \mathcal{W} - \mathcal{W}) - \nabla \cdot (\bar{N}_{\beta} \mathbf{u}) + \hat{D}(n'_{\beta})$$

$$\frac{\partial \theta}{\partial t} + \nabla \cdot (\theta \mathbf{u} - \langle \theta \mathbf{u} \rangle) + (\gamma - 2) [\theta (\nabla \cdot \mathbf{u}) - \langle \theta (\nabla \cdot \mathbf{u}) \rangle] =$$

$$q(\mathcal{W} - \mathcal{W}) - (\mathbf{u} \cdot \nabla)T - (\gamma - 1)\overline{T} (\nabla \cdot \mathbf{u}) + \hat{D}(\theta)$$
(5)

where \boldsymbol{u} is the gas velocity fluctuations, $\langle n'_{\beta} \mathbf{u} \rangle$, $\langle \theta \mathbf{u} \rangle$, $\langle \theta (\nabla \cdot \mathbf{u}) \rangle$ are the corresponding turbulent fluxes, $\overline{W} = \langle W(n_{\beta}, T) \rangle$, and the angular brackets imply the averaging over the statistics of turbulent velocity field. Taking into account that the fluctuations of temperature and the number density of admixtures are much smaller than the corresponding mean values, we can write

$$\mathcal{W}^{\mathbf{x}} - \bar{\mathcal{W}} = \sum_{\beta=1}^{m} \left(\frac{\partial \mathcal{W}^{\mathbf{x}}}{\partial n_{\beta}} \right)_{\bar{N}_{\beta}} n'_{\beta} + \left(\frac{\partial \mathcal{W}^{\mathbf{x}}}{\partial T} \right)_{\bar{T}} \theta = \bar{\mathcal{W}}C', \tag{7}$$

where: $C'_{n} = \sum_{\beta=1}^{m} v_{\beta} \frac{n'_{\beta}}{\bar{N}_{\beta}}, C'_{T} = \frac{E_{a}}{R\bar{T}^{2}} \theta, C' = C'_{n} + C'_{T}.$

Using the Navier-Stokes equation we obtain from Eqs. (5-7) a system of equations for the second-order moments $\langle n'_{\beta} \mathbf{u} \rangle$ and $\langle \theta \mathbf{u} \rangle$:

$$\frac{\partial (n'_{\beta} u_i)}{dt} = -\mathbf{v}_{\beta} \overline{W} (\langle C' u_i \rangle - \langle u_i u_j \rangle \nabla_j \overline{N}_{\beta} - \overline{N}_{\beta} \langle u_i (\nabla \cdot \mathbf{u}) \rangle + \hat{\Re} \langle n'_{\beta} u_i \rangle,$$
(8)

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Turbulent diffusion

$$\frac{\partial(\boldsymbol{\theta}\boldsymbol{u}_{i})}{dt} = q \,\overline{W} \langle C'\boldsymbol{u}_{i} \rangle - \langle \boldsymbol{u}_{i}\boldsymbol{u}_{j} \rangle \nabla_{j} \overline{T} - (\gamma - 1) \overline{T} \langle \boldsymbol{u}_{i} (\nabla \cdot \mathbf{u}) \rangle + \hat{\Re} \langle \boldsymbol{\theta}\boldsymbol{u}_{i} \rangle. \tag{9}$$

Here $\hat{\Re}\langle n'_{\beta}u_i \rangle = -\langle [\nabla \cdot (n'_{\beta}\mathbf{u})u_i \rangle + \langle \hat{D}(n'_{\beta})u_i \rangle - \langle n'_{\beta}[(\mathbf{u} \cdot \nabla)u_i + \rho^{-1}\nabla_i p'] \rangle + \langle n'_{\beta}\hat{D}_{\nu}(u_i) \rangle$ include the third-order moments caused by the nonlinear terms and the second-order moments due to the molecular dissipative terms. The expression for $\hat{\Re} \langle \Theta u_i \rangle$ is similar to $\hat{\Re} \langle n'_{\beta} u_i \rangle$.

To close the system of equations (8-9) we use the spectral τ -approximation (see e.g. [5, 6]) which postulates that the deviations of the third-order moments from the contributions of the terms afforded by the background turbulence can be expressed through the similar deviations of the second-order moments, e.g.:

$$\hat{\Re} \left\langle n'_{\beta} u_{i} \right\rangle_{\mathbf{k}} - \hat{\Re} \left\langle n'_{\beta} u_{i} \right\rangle_{\mathbf{k}}^{0} = -\frac{1}{\tau(k)} \left\{ \left\langle n'_{\beta}(\mathbf{k}) u_{i}(-\mathbf{k}) \right\rangle - \left\langle n'_{\beta}(\mathbf{k}) u_{i}(-\mathbf{k}) \right\rangle^{0} \right\}$$
(10)

For the low-Mach-number and for an ideal gas the continuity equation is $\nabla \cdot \mathbf{u} = (\mathbf{u} \cdot \nabla) ln\rho$ and we obtain: $\overline{T}\langle u_i(\nabla \cdot \mathbf{u})\rangle = \langle u_i u_i \rangle (\nabla_i \ln \overline{T} - \nabla_i \ln \overline{P})$. Thus, if there is no external pressure gradient, and correspondingly there is no mean flow, $\overline{T} \langle u_i (\nabla \cdot \mathbf{u}) \rangle = \langle u_i u_j \rangle \nabla_j \ln \overline{T}$. If the gradients of the mean temperature and the mean number density vanish, then also vanish the turbulent fluxes of the heat and chemical admixtures, and Eq. (11) reduces to $\hat{\Re} \langle n'_{\beta} u_i \rangle_{\mu} = - \langle n'_{\beta} (\mathbf{k}) u_i (-\mathbf{k}) \rangle / \tau(k)$.

Coefficients of turbulent diffusion 3

Taking into account that the characteristic time scale of variation of the second moments is much larger than the correlation time $\tau(k)$ for all turbulence scales, the steady-state solutions of Eqs. (8-9) for the turbulent fluxes written in the Fourier space are:

$$\left\langle n'_{\beta}(\mathbf{k})u_{i}(-\mathbf{k})\right\rangle = -\tau(k)\left\{\nu_{\beta}\overline{W}\left\langle C'(\mathbf{k})u_{i}(-\mathbf{k})\right\rangle + \left\langle u_{i}(\mathbf{k})u_{j}(-\mathbf{k})\right\rangle\left(\nabla_{j}\overline{N}_{\beta} + \overline{N}_{\beta}\nabla_{j}\overline{T}\right)\right\}$$
(11)

$$\left\langle \Theta(\mathbf{k})u_i(-\mathbf{k})\right\rangle = -\tau(k) \left\{ -q \overline{W} \left\langle C'(\mathbf{k})u_i(-\mathbf{k})\right\rangle + \gamma \left\langle u_i(\mathbf{k})u_j(-\mathbf{k})\right\rangle \nabla_j \overline{T} \right\},\tag{12}$$

$$\left\langle C'(\mathbf{k})u_i(-\mathbf{k})\right\rangle = -\tau_{eff}(k)\left\langle u_i(\mathbf{k})u_j(-\mathbf{k})\right\rangle \times \left[\sum_{\beta=1}^m \nu_\beta \nabla_j \left(\ln \bar{N}_\beta + \ln \bar{T}\right) + \frac{\gamma}{q}\alpha_T \nabla_j \bar{T}\right].$$
(13)

Here $\tau_{eff}^{-1} = \tau_c^{-1} + \tau^{-1}(k)$, and $\tau_c^{-1} = \overline{W}\left(\sum_{\beta=1}^m (\nu_\beta / \overline{N}_\beta) - \frac{qE_a}{R\overline{T}^2}\right)$ is the characteristic inverse chemical

time.

To distinguish the effects of turbulent transport caused by the chemistry from those caused by inhomogeneity of turbulence, we consider isotropic and homogeneous background turbulence [8]:

$$\left\langle u_i(\mathbf{k})u_j(-\mathbf{k})\right\rangle = \frac{u_0^2 E_T(k)}{8\pi k^2} \left[\delta_{ij} - \frac{k_i k_j}{k^2}\right],\tag{14}$$

where $E_T(k) = (\mu - 1)k_0^{-1}(k / k_0)^{-\mu}$ is the energy spectrum function, $1 < \mu < 3$, $\tau(k) = 2\tau_0(k / k_0)^{1-\mu}$ is the turbulent correlation time, $\tau_0 = l_0 / u_0$ is the characteristic turbulent time, u_0 is the characteristic turbulent velocity in the integral scale l_0 . Integrating (11-12) in k space and taking into account (13) we arrive to the equations for the turbulent flux of reacting admixtures and the turbulent heat flux:

$$\left\langle n'_{\beta} \mathbf{u} \right\rangle = -D_{\beta}^{T} \nabla \overline{N}_{\beta} + \sum_{\lambda=1,\lambda\neq\beta}^{m} D_{\lambda}^{MTD}(\beta) \nabla \overline{N}_{\lambda} - D_{\beta}^{TTD} \overline{N}_{\beta} \nabla \ln \overline{T}$$
(15)

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$$\left\langle \boldsymbol{\Theta} \mathbf{u} \right\rangle = -D^T \nabla \overline{T} - \sum_{\lambda=1}^m D_\lambda^{TDE} \nabla \overline{N}_\lambda, \tag{16}$$

Turbulent diffusion

where:

$$D_{\beta}^{T} = D_{0}^{T} \left[1 - \frac{\mathbf{v}_{\beta}^{2}}{\bar{N}_{\beta}(\alpha_{n} - \alpha_{T})} \Phi(Da_{T}) \right]$$
(17)

is the coefficient of turbulent diffusion of the number density of admixtures,

$$D_{\lambda}^{MTD}(\beta) = D_0^T \frac{\mathbf{v}_{\beta} \mathbf{v}_{\lambda}}{\bar{N}_{\lambda} (\alpha_n - \alpha_T)} \Phi(Da_T)$$
(18)

is the coefficient of the mutual turbulent diffusion of the number density of admixtures,

$$D_{\beta}^{TTD} = D_{0}^{T} \left[1 - \frac{\nu_{\beta}}{\bar{N}_{\beta}(\alpha_{n} - \alpha_{T})} \Phi(Da_{T}) \left(\sum_{\lambda=1}^{m} \nu_{\lambda} + \gamma \frac{E_{a}}{R\bar{T}} \right) \right]$$
(19)

is the coefficient of turbulent thermal diffusion,

$$D^{T} = D_{0}^{T} \gamma \left[1 + \frac{q}{\gamma \overline{T}(\alpha_{n} - \alpha_{T})} \Phi(Da_{T}) \left(\sum_{\beta=1}^{m} \nu_{\beta} + \gamma \frac{E_{a}}{R\overline{T}} \right) \right]$$
(20)

is the coefficient of turbulent diffusion of the temperature,

$$D_{\lambda}^{TDE}(\beta) = D_0^T \frac{q \mathbf{v}_{\lambda}}{\bar{N}_{\lambda}(\alpha_n - \alpha_T)} \Phi(Da_T)$$
(21)

is the coefficient that describes the turbulent Duffor effect, $Da_T = \tau_0/\tau_c$ is the turbulent Damköhler

number,
$$D_0^T = u_0^2 \tau_0 / 3$$
, $\alpha_n = \sum_{\beta=1}^m (v_\beta / \overline{N}_\beta)$, $\alpha_T = \frac{qE_a}{R\overline{T}^2}$, $\Phi(Da_T) = 1 - \frac{1}{Da_T} \left(1 - \frac{\ln(1 + Da_T)}{2Da_T} \right)$

The effect of chemical reaction on turbulent diffusion and the reactive front propagation has been studied in [9] using the direct numerical simulations as well using the mean-field approach for the Kolmogorov-Petrovskii-Piskunov equation [10]/



Figure 1. Comparison of the theoretical dependence of turbulent diffusion coefficient D_{β}^{T}/D_{0}^{T} versus turbulent Damköhler number with the corresponding results of the mean-field approach [9].

In the mean-field approach [9], which was amended by an advection term to describe the interaction with a turbulent velocity field, the memory effects of turbulent diffusion was taken into account to determine the dependence of the turbulent diffusion coefficient versus the turbulent Damköhler number, while the reaction front speed, s_T , was determined by differentiating the concentration

integrated over the whole computational domain. Fig. 1 shows the theoretical prediction (circles) for the turbulent diffusion coefficient $D_{\beta}^{T}(Da_{T})$ in comparison with the corresponding numerical results from [9] (solid line), for $s_{T} = 2\sqrt{D_{\beta}^{T}/\tau_{c}}$, $v_{\beta} = 1$, α_{n} ? α_{T} .

4 Discussion

To elucidate physics of the obtained results let us consider examples of chemical reactions proceeding in a stoichiometric mixture. For a small concentration of reactive admixtures \overline{N}_{β} compared to the ambient gas number density, \overline{N}_0 , the characteristic chemical time, which is about $\tau_c : 10^{-3}s$, is smaller than the characteristic turbulent time, $\tau_0 = l_0/u_0$. Indeed, for typical turbulent velocity $u_0 \approx 1$ m/s, and for integral scale $l_0 \approx (0.1 \div 1)$ m, we have $\tau_0 = l_0/u_0 : (10^{-1} \div 1)s$. So that the case of large turbulent Damköhler numbers, $Da_T = \tau_0/\tau_c >> 1$, is of the main physical interest. It can be seen, that also $\alpha_n/\alpha_T \approx (R\overline{T}/E_a)(\overline{N}_0 / \overline{N}_{\beta}) >> 1$ for a small values \overline{N}_{β} of the diluted reactive admixtures.

In general the overall order of most chemical reactions is 2 or 3, though for complex reactions the overall order of the reaction can be fractional one (for example, it is 2.74 for H₂/O₂). For the simplest first-order reaction $A \rightarrow B$ and for $Da_T >> 1$, the turbulent diffusion of admixtures is determined by the chemical time, $D_{\beta}^{T} = D_{0}^{T} / Da_{T} = \tau_{c} u_{0}^{2} / 3$ (see Eq. 17), so that usual turbulent diffusion based on the turbulent time $\tau_0 >> \tau_c$ does not contribute to the mass flux of a reagent A. In this case the species A are consumed by the reaction and their concentration decreases before they were carried away by turbulent diffusion. Thus, the turbulent diffusion in this case is effective only for the reaction product B. For the second-order reaction $A + B \rightarrow C + D$ (for example, $H + O_2 \rightarrow OH + O$) we obtain $\alpha_n = 2/\bar{N}$, where $\bar{N}_A = \bar{N}_B = \bar{N}$, and from Eq. (18) $D_A^T = D_B^T = 0.5D_0^T(1+1/Da_T)$. For the thirdorder reaction, $A + B + C \rightarrow D + E$, we obtain $D_A^T = D_B^T = D_C^T = 2D_0^T (1 + 1/Da_T)/3$. For different stoichiometric coefficients of the reagents for the third-order reaction, the coefficients of turbulent diffusion for the species are different (for example: $2H_2 + O_2 = 2H_2O$). Since the species A have a larger stoichiometric coefficient (2A + B) and, correspondingly, larger number of moles participating in the chemical reaction, they are consumed more effectively in the reaction and the turbulent diffusion coefficient for the species A decreases much stronger than that for species B: $D_A^T = D_0^T / Da_T$, while $D_B^T = 0.5 D_0^T (1 + 1 / Da_T)$. It should be noticed that in the case of nonstoichiometric reactions, when the species with higher molecularity are excessive in the initial mixture, e.g., appears in an amount larger than that required according to the stoichiometric equation, the turbulent diffusion coefficient of the species A tends to zero, so molecular diffusion can be important.

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