

TURBULENT TRANSPORT OF CHEMICALLY REACTING GASEOUS ADMIXTURES

Tov Elperin¹, Nathan Kleorin¹, M. Liberman^{2,3} & Igor Rogachevskii¹

¹ *Department of Mechanical Engineering, Ben-Gurion University of the Negev, Beer-Sheva, Israel*

² *Nordita, KTH Royal Institute of Technology and Stockholm University, Stockholm, Sweden*

³ *Moscow Institute of Physics and Technology, Dolgoprudnyi, 141700, Russia*

Abstract We study turbulent diffusion of chemically reacting gaseous admixtures in a developed turbulence. In our previous study [Phys. Rev. Lett. **80**, 69 (1998)] using a path-integral approach for a delta-correlated in time random velocity field, we demonstrated a strong modification of turbulent transport in fluid flows with chemical reactions or phase transitions. In the present study we use the spectral tau approximation, that is valid for large Reynolds and Peclet numbers, and show that turbulent diffusion of the reacting species can be strongly depleted by a large factor that is the ratio of turbulent and chemical times (turbulent Damköhler number). We have demonstrated that the derived theoretical dependence of turbulent diffusion coefficient versus the turbulent Damköhler number is in a good agreement with that obtained previously in the numerical modelling of a reactive front propagating in a turbulent flow and described by the Kolmogorov-Petrovskii-Piskunov-Fisher equation. We have found that turbulent cross-effects, e.g., turbulent mutual diffusion of gaseous admixtures and turbulent Dufour-effect of the chemically reacting gaseous admixtures, are less sensitive to the values of stoichiometric coefficients. The mechanisms of the turbulent cross-effects are different from the molecular cross effects known in irreversible thermodynamics. In a fully developed turbulence and at large Peclet numbers the turbulent cross-effects are much larger than the molecular ones. The obtained results are applicable also to heterogeneous phase transitions.

TURBULENT TRANSPORT OF ADMIXTURES AND TEMPERATURE

Turbulent transport in flows with chemical reactions is of great interest in various applications, ranging from combustion to physics of turbulent atmosphere of the Earth. During the decades turbulent transport of passive scalar and particles has been subject of an active research in analytical, numerical and laboratory studies. However, impact of chemical reactions on turbulent transport have been studied mainly numerically and in the context of turbulent combustion.

Combustion process is the chemical reaction accompanied by heat release. Turbulent combustion can proceed as volume distributed chemical reaction (e.g., as a homogeneous burning of the turbulent premixed gaseous mixture) or propagate as a flame front in a turbulent flow separating fresh unburned fuel and combustion products. Turbulence in these systems is created by an external forcing and can be enhanced by intrinsic instability of the flame front. In a turbulent atmosphere the most common are the volume distributed chemical reactions, while during wild fires propagation of the turbulent flame front is of particular interest in the atmospheric and industrial applications.

For the first time the effect of chemistry on turbulent diffusion was studied analytically in [1] by means of a path-integral approach for the Kraichnan-Kazantsev model of the random delta-correlated in time velocity field, and it was found that turbulent diffusion can be strongly depleted by chemical reactions or phase transitions. It was also shown in [1] that there exists an additional non-diffusive turbulent flux of number density of gaseous admixture (proportional to the mean temperature gradient multiplied by the number density of gaseous admixture) and additional turbulent heat flux (proportional to the gradient of the mean number density of gaseous admixture) in flows with chemical reactions or phase transitions. In the present study (for details see [2]) we investigate turbulent transport of chemically reacting gaseous admixtures in a developed turbulence using a spectral tau approach (high-order closure procedure). We have derived the following equation for the turbulent flux of reacting admixtures, $\langle n'_\beta \mathbf{u} \rangle$, and the turbulent heat flux, $\langle \theta \mathbf{u} \rangle$:

$$\langle n'_\beta \mathbf{u} \rangle = -D_\beta^T \nabla \bar{N}_\beta + \sum_{\lambda=1; \lambda \neq \beta}^m D_\lambda^{\text{MTD}}(\beta) \nabla \bar{N}_\lambda + \mathbf{V}_{\text{eff}} \bar{N}_\beta; \quad \langle \theta \mathbf{u} \rangle = -D^T \nabla \bar{T} - \sum_{\lambda=1}^m D_\lambda^{\text{TDE}} \nabla \bar{N}_\lambda, \quad (1)$$

where using a mean-field approach, we decomposed the number density of the admixtures n_β and the fluid temperature T into the mean quantities, \bar{N}_β and \bar{T} , and fluctuations n'_β and θ . We decompose the velocity field in a similar fashion, and assume for simplicity vanishing mean fluid velocity, $\bar{\mathbf{U}} = 0$. In Eqs. (1) we used the following notations: D_β^T is the coefficient of turbulent diffusion of the number density of admixtures:

$$D_\beta^T = D_0^T \left(1 - \frac{\nu_\beta^2}{\bar{N}_\beta \alpha_n} \Phi(\text{Da}_\tau) \right), \quad \Phi(\text{Da}_\tau) = 1 - \frac{1}{\text{Da}_\tau} \left(1 - \frac{\ln(1 + 2\text{Da}_\tau)}{2\text{Da}_\tau} \right); \quad (2)$$

$\text{Da}_\tau = \tau_0/\tau_c$ is the turbulent Damköhler number, $\tau_0 = \ell_0/u_0$ is the characteristic turbulent time, u_0 is the characteristic turbulent velocity in the integral scale ℓ_0 of turbulence, τ_c is the characteristic chemical time, $D_0^T = \tau_0 u_0^2/3$, ν_β is the stoichiometric coefficient that is the order of the reaction with respect to species β , $\sum_{\beta=1}^m \nu_\beta$ is the overall order of the

reaction, $\alpha_n = \sum_{\beta=1}^m \nu_\beta / \bar{N}_\beta$ and m is total number of species, $D_\lambda^{\text{MTD}}(\beta)$ is the coefficient of the mutual turbulent diffusion of the number density of admixtures, $\mathbf{V}_{\text{eff}} = -D_\beta^{\text{TTD}} \nabla \ln \bar{T}$ is the effective velocity of the number density of admixtures due to the turbulent thermal diffusion, and D_β^{TTD} is the coefficient of turbulent thermal diffusion:

$$D_\lambda^{\text{MTD}}(\beta) = D_0^T \frac{\nu_\beta \nu_\lambda}{\bar{N}_\lambda \alpha_n} \Phi(\text{Da}_T); \quad D_\beta^{\text{TTD}} = D_0^T \left[1 - \frac{\nu_\beta}{\bar{N}_\beta \alpha_n} \Phi(\text{Da}_T) \left(\sum_{\lambda=1}^m \nu_\lambda + \gamma \frac{E_a}{RT} \right) \right]; \quad (3)$$

$\gamma = c_p/c_v$ is the ratio of specific heats, E_a is the activation energy, R is the universal gas constant; D^T is the coefficient of turbulent diffusion of the temperature and D_λ^{TDE} is the coefficient that describes the turbulent Duffor effect:

$$D^T = D_0^T \gamma \left[1 + \frac{q}{\gamma \bar{T} \alpha_n} \Phi(\text{Da}_T) \left(\sum_{\beta=1}^m \nu_\beta + \gamma \frac{E_a}{RT} \right) \right]; \quad D_\lambda^{\text{TDE}} = D_0^T \frac{q \nu_\lambda}{\bar{N}_\lambda \alpha_n} \Phi(\text{Da}_T). \quad (4)$$

To elucidate physics of the obtained results we consider examples of chemical reactions proceeding in a stoichiometric mixture. For a small concentration of reactive admixtures, $\bar{N}_\beta \ll \bar{N}_f$, the characteristic chemical time τ_c varies from 10^{-3} s to 10^{-2} s, where \bar{N}_f is the ambient fluid number density. For typical values of turbulent velocity in atmospheric flows $u_0 = 1$ m/s and integral scale $\ell = 100$ m, we obtain characteristic turbulent time $\tau_0 = \ell/u_0 = 10^2$ s, so that the case of large turbulent Damköhler numbers, $\text{Da}_T = \tau_0/\tau_c \gg 1$ is of the main physical interest.

The stoichiometric coefficient ν_β is known as the order of the reaction with respect to species β . For a simplified model of a single-step reaction the overall order of the reaction is the molecularity of the reaction, indicating the number of particles entering the reaction. 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.

Let us consider first the simplest chemical reaction $A \rightarrow B$, assuming a large turbulent Damköhler numbers, $\text{Da}_T \gg 1$. An example of such chemical reaction is the dissociation: $O_2 \rightarrow O + O$. As follows from Eqs. (2), turbulent diffusion of the number density of admixtures is $D_\beta^T = D_0^T / \text{Da}_T = \tau_c u_0^2 / 3$, which means that the turbulent diffusion of admixture is determined by the chemical time. This is in agreement with the result obtained in [1]. The underlying physics of this phenomena is quite transparent. For a simple first-order chemical reaction $A \rightarrow B$ the species A of the reactive admixture are consumed and their concentration decreases much faster during the chemical reaction, so that the usual turbulent diffusion based on the turbulent time $\tau_0 \gg \tau_c$, does not contribute to the mass flux of a reagent A . The turbulent diffusion during the turnover time of the turbulent eddies is effective only for the product of reaction, B . Applicability of the obtained results requires the condition $\text{Pe} / \text{Da}_T \gg 1$ to be satisfied, where $\text{Pe} = u_0^2 \tau_0 / 3$ is the Peclet number.

Let us determine the turbulent diffusion coefficients for the second-order chemical reaction that is determined by the following equation: $A + B \rightarrow C + D$. An example of such chemical reaction is $H + O_2 \rightarrow OH + O$. The numbers of species in this reaction are stoichiometric coefficients, which define number of moles participating in the reaction. The stoichiometric reaction whereby the initial substances are taken in a proportion such that the chemical transformation fully converts them into the reaction products, can proceed as the inverse reaction also. For this reaction we obtain $\alpha_n = 2/\bar{N}$, where $\bar{N}_A = \bar{N}_B \equiv \bar{N}$. On the other hand, using Eq. (2) for the turbulent diffusion coefficients of species A and B , we obtain $D_{A,B}^T = \frac{1}{2} D_0^T (1 + \text{Da}_T^{-1})$. Correspondingly for the third-order reaction, $A + B + C \rightarrow D + E$, we find $D_{A,B,C}^T = \frac{2}{3} D_0^T (1 + \text{Da}_T^{-1})$, where we have taken into account that in this case $\alpha_n = 3/\bar{N}$.

Consider the stoichiometric third-order reaction with different stoichiometric coefficients of the reagents $2A + B \rightarrow 2C$ (for example, the chemical reaction $2C + O_2 \rightarrow 2CO$ or $2H_2 + O_2 \rightarrow 2H_2O$). In this case $\alpha_n = 2/\bar{N}$ (where we took into account that for the stoichiometric chemical reaction with optimum amounts of reagents $\bar{N}_C = 2\bar{N}_{O_2}$), and the turbulent diffusion coefficients of species A and B are as follows: $D_A^T = D_0^T / \text{Da}_T$, $D_B^T = \frac{1}{2} D_0^T (1 + \text{Da}_T^{-1})$. Since the species A have a larger stoichiometric coefficient 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 . The derived in the present study theoretical dependence of turbulent diffusion coefficient versus the turbulent Damköhler number is in a good agreement with that obtained in [3] using mean-field simulations of a reactive front propagating in a turbulent flow and described by the Kolmogorov-Petrovskii-Piskunov-Fisher equation.

References

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