Influence of turbulent fluctuations on non-equilibrium chemical reactions in the flow

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Abstract. In chemically nonequilibrium flows the problem of calculation of sources (formation rates) in equations for chemical species is of utter importance. Formation rate of each component is a non-linear function of mixture density, temperature and concentration of species. Thus the suggestion that the mean rate may be determined via mean values of the flow parameters could lead to significant errors. One of the most accurate approaches here is utilization of probability density function (PDF). In this paper the method for constructing such PDFs is developed. The developed model was verified by comparison with the experimental data. On the example of supersonic combustion it was shown that while the overall effect on the averaged flow field is often negligible, the point of ignition can be considerably shifted up the flow.

1. Introduction
In high enthalpy turbulent flows the problem of calculation of sources (formation rates) in equations for chemical species is of utter importance. Formation rate of a component is a function of density, temperature and concentration of species:

\[ \dot{w}_i = \dot{w}_i (\rho, T, C_1, \ldots, C_N) \]  

This function of is appreciably nonlinear. Thus, the suggestion that its mean value depends only on mean values of the arguments \( \overline{\dot{w}_i} = \dot{w}_i (\rho, T, C_1 \ldots C_N) \) could lead to serious errors.

Actually, mean formation rate strongly depends on average gas-dynamic parameters, but also on fluctuations of these parameters. Correlation of temperature and concentration of species could lead to both increase and decrease of rate of chemical reactions.

In this paper a method is developed to take these effects into account within the standard Reynolds-averaged Navier-Stokes (RANS) approach.

2. Probability density function
As we are dealing with random fluctuations of the parameters, the most reliable way to consider the problem is to implement probability density function (PDF) for calculation of chemistry reaction rates. As it is known from the probability theory, the mean value of a random function could be calculated using the following formula:
\[
\overline{w}_i = \int_{0}^{\infty} \cdots \int_{0}^{\infty} \int_{0}^{\infty} \psi_i (\rho, T, C_1, \ldots, C_N) P (\rho, T, C_1, \ldots, C_N) d \rho d T C_1, \ldots, d C_N,
\]

where \( P (\rho, T, C_1 \ldots C_N) \) is a PDF of random variables \( \rho, T, C_1 \ldots C_N \).

The general form of the PDF for chemical processes is yet unknown.

In this paper a simplified approximate form of the PDF is introduced. It is supposed that PDF represented by a product of three independent PDFs:

\[
P (\rho, T, C_1 \ldots C_N) = P_\rho (\rho) P_T (T) P_C (C_1 \ldots C_N)
\]

The corresponding random variables are supposed to be statistically independent.

PDF for gas density is represented as Dirac’s delta \( P_\rho (\rho) = \delta (\rho - \bar{\rho}) \) [1]. PDF for gas temperature in this work is represented either as Gauss or as beta-function ([2] and [3]). PDF for concentrations of species is represented as multi-dimensional beta-function [4].

Integrals, which include PDF for temperature, could not be taken analytically. Thus, the following numerical algorithm is implemented. In order not to take the integral at each time step and at each point of the computational grid, mean reaction rates were computed for every possible value of mean temperature and temperature variance beforehand. This reaction rates were represented as a bilinear interpolation function. This reduced computation time significantly.

The main advantage of implementing multidimensional beta-function as a PDF for concentrations is that all the statistical moments of the random variables can be found analytically. Formation rates depend on product of concentrations of species. As a result mean rates depend on product of mean concentrations, variance of concentrations and their covariance (multivariate moments of the second order). Variance and covariance for multidimensional beta-function PDF are determined using the following formulae:

\[
\frac{C_m^s C_n^s}{C_m C_n} = \begin{cases} 
\frac{\sigma_C^{-1}}{1 - \sum_{m=1}^{N_c} C_m^2}, & m \neq n \\
\frac{\sigma_C C_m (1 - \sum_{n=1}^{N_c} C_n^2)}{1 - \sum_{m=1}^{N_c} C_m^2}, & m = n 
\end{cases}
\]

where \( \beta_m = C_m \left( \frac{1 - \sum_{m=1}^{N_c} C_m^2}{\sigma_C^2} - 1 \right) \), \( \sum_{m=1}^{N_c} \left( C_m^2 \right)^{2} \), \( \sigma_C = \sum_{m=1}^{N_c} C_m^2 \).

In order to define PDFs for temperature and concentrations, we are to determine their variance.

By implementing Reynolds decomposition and averaging to the corresponding scalar transport equation, general equation of the following form could be derived:

\[
\frac{\partial}{\partial t} \left( \bar{p} \vec{f}^2 \right) + \frac{\partial}{\partial x_j} \left( \bar{p} \mu_j \vec{f}^2 \right) = \frac{\partial}{\partial x_j} \left( \frac{\mu_T}{\sigma_{T,f}} \frac{\partial \vec{f}^2}{\partial x_j} \right) + 2 \frac{\mu_f}{\sigma_{f,f}} \left( \frac{\partial \vec{f}^2}{\partial x_j} \right)^2 - 2 \beta e_f + 2 \vec{f} \omega
\]

where \( \vec{f}^2 = C_m^2, T^2 \) represents squared variance of concentration or temperature; \( \sigma_{T,f}, \sigma_f \) are analogs of Prandtl/Schmidt numbers; \( e_f \) - is dissipation of fluctuations of the corresponding variable; \( \omega \) - is the source.

Dissipation could be determined from a separate differential equation similar to (5). However, for the sake of simplification the following supposition can be implemented:

\[
R = \frac{\vec{f}^2}{e_f} \left( \frac{K}{e_f} \right) \approx \text{const}
\]

(6)
From (6) we obtain:

\[ \frac{\varepsilon_f}{\varepsilon} = \frac{1}{R} \int \frac{x^2}{K} \, dx \]  

(7)

In [5] it is suggested that \( R = 1 \).

For the source term in (5) there are a lot of semiempirical formulae (e.g., see, [6]-[8]). These formulae are often self-contradictory. Due to this fact, in this paper it is supposed that source is determined by generation and dissipation.

3. Model verification

3.1. Subsonic hydrogen combustion[9]

Experimental work [9] was carried out in the 1.8 m wind tunnel with \( 305 \times 305 \) mm cross-section. Hydrogen is supplied at the initial section of the tunnel through the nozzle with outlet diameter of 7.62 mm. The nozzle is ideally profiled. Nozzle edge width is 0.1 mm. Hydrogen velocity at the outlet is 151 m/s, the co-flow velocity is 10% of hydrogen velocity = 15.1 m/s.

Initial hydrogen temperature was taken to be equal \( T_i = T_e = 293 \) K. Hydrogen combustion was modeled using system of reactions [10] introduced by Connaire et al.

On figure 1, axial distributions of temperature and concentration of water are presented. As it is seen on the picture, implementing chemistry model with PDF gives results being closer to the experimental data.

As it was already mentioned, turbulent fluctuations can both increase (temperature fluctuations) and decrease (fluctuations of concentrations) rates of chemical reactions. In the experiment under consideration, combustion is close to equilibrium, and thus fluctuations of temperature do not influence the reactions. Due to this fact, fluctuations of concentrations of species are predominant.

Cross profiles of temperature and concentration also show that PDF chemistry model allows to obtain more accurate simulation results (see figures 2-3).
Figure 2. Cross profiles of temperature in hydrogen jet reacting with air co-flow at $x/R_a=160$
1 – experimental study by Kent and Bilger [9]; 2 – simulation using quasi-laminar chemistry model; 3 – simulation using chemistry model with PDF

Figure 3. Cross profiles of molar fractions of species in hydrogen jet reacting with air co-flow at $x/R_a=160$
- dots – experimental study by Kent and Bilger [9];
- dashed line – simulation using quasi-laminar chemistry model;
- continuous line – simulation using chemistry model with PDF

3.2. Combustion of supersonic hydrogen jet
This test was performed using data of Evans et al. [11]. Experimental setup is presented in figure 4.[9]

Figure 4. Flow geometry. Outer nozzle diameter $D = 0.0653m$, inner nozzle diameter $d_j = 0.009525m$, inner nozzle edge width – $0.0015m$.

Figure 5 shows distribution of species along jet axis. As one can see, the difference between simulation results obtained via quasi-laminar and PDF chemistry models are quite similar and both are in good convergence with experimental data (taking into account that experimental accuracy in [11] was about 30%). However, when using PDF chemistry model, ignition occurs earlier up the flow (see figure 6).
Figure 5. Distribution of concentration of species.
(a) - \( H_2 \), (b) - \( H_2O \), (c) - \( N_2 \).
1 – simulation using quasi-laminar chemistry model;
2 – simulation using chemistry model with PDF;
3 – experimental data [11]

Figure 6. Spatial temperature distribution. (a) – quasi-laminar chemistry model; (b) – PDF chemistry model.

3.3. Wall jet of hydrogen in supersonic co-flow
Flows with supersonic combustion in wall jet could be found in different technical systems, e.g. in a scramjet.

This test was performed using experimental data from [12]. In [12] combustion of cool hydrogen injected through the slit into the supersonic heated air flow was investigated. Experimental setup is presented in figure 7.
Simulation was performed with the following inlet parameters. Hydrogen: Mach number \( M_{H2} = 1 \), static temperature \( T_{H2} = 242 \text{K} \), static pressure \( p_{H2} = 1 \text{ atm} \). Co-flow: Mach number \( M_{air} = 2.44 \), static temperature \( T_{air} = 1270 \text{K} \), static temperature \( p_{air} = 1 \text{ atm} \).

Axial distributions in this test were found to be practically independent on chemistry model, but the ignition point in case of PDF chemistry model was shifted up the flow (see figure 8). Qualitatively, the results of testing different chemistry models is similar to the one obtained in the previous section.

4. Conclusion

In this paper it was shown that for combustion near chemical equilibrium influence of turbulent fluctuations on chemistry rates manifests itself through the concentration pulsations, which leads to decrease in formation rates (so-called “unmixedness” effect).

In supersonic flows with pronounced non-equilibrium chemistry, effect of turbulent fluctuations on the developed regions of the flame is rather weak, however the position of the ignition point is influenced greatly. It is primarily connected with temperature fluctuations, because formation rates depend on temperature non-linearly and, as a result, formation rates computed via PDF model are much higher than the ones computed via quasi-laminar chemistry.

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