Analysis and modelling of the effective reaction rate in a developing mixing layer

Karol Wędolowski\textsuperscript{1,2}, Konrad Bajer\textsuperscript{1,2}, Kamil Kwiatkowski\textsuperscript{1,2}

\textsuperscript{1}Institute of Geophysics, Faculty of Physics, University of Warsaw, Poland
\textsuperscript{2}Interdisciplinary Centre for Mathematical and Computational Modelling, University of Warsaw, Poland

E-mail: kjwed@okwf.fuw.edu.pl

Abstract. Simulations of the chemically reacting mixing layer were performed. Especially the early stage of the flow transition, when the coherent Kelvin-Helmholtz vortices can be recognized, was studied. This is a physical problem of great importance in many industrial and environmental systems. Simple passive reaction of the second order is considered. Its effective rate is analysed for different values of the Damköhler number. The results are compared to the case of the steady, laminar shear flow. It is found that slow and fast reactions respond in different ways to the fluctuations of the reactant concentration. Faster reactions appeared to be much more sensitive to the character of mixing. Moreover a buffer layer of product of fast reactions forms in the system and separates the substrates what influence the effective reaction rate.

1. Introduction

Chemically reacting flows are ubiquitous in both environmental and industrial systems (Renard, 2000; Kwiatkowski \textit{et al.}, 2011; Crimaldi \textit{et al.}, 2008). Vortical structures occurring in a turbulent flow enhance mixing, which is of great importance in the case of non-premixed reaction (Renard, 2000; Crimaldi \textit{et al.}, 2008, 2006). The enhancement of diffusion and of the reaction rate was subject of many studies in two- and three- dimensional flows (Bajer \textit{et al.}, 2001; Riley \textit{et al.}, 1985). Equations governing the concentrations of the chemically reacting species are non-linear. Consequently, they cannot be simply averaged without producing additional terms that need to be modelled and parametrised. Moreover we should not assume that the solution of such equation is linearly dependent on the coefficients describing a particular reaction, e.g. the Damköhler number ($Da$).

A plane mixing layer is a special case in which accelerated mixing and enhanced reactions could be considered. This simple system was a subject of many studies. Considerable effort was focused on the development of the hydrodynamic instability itself (Rogers & Moser, 1992, 1993; Michalke, 1964). Especially, Rogers & Moser (1992, 1993) analysed thoroughly the issue of the three-dimensional Kelvin-Helmholtz instability. Numerical simulations described in this article are based on their work. The problem of a mixing layer may includes chemical reactions as well. It usually refers to the non-premixed case in which substrates are initially separated and mixing processes limit the overall reaction rate. Numerical simulations of the chemically reacting shear
layer were performed, for example by Riley et al. (1985). Experimental work on this issue was conducted by Bilger et al. (1991).

Even though the Kelvin-Helmholtz vortices could be properly resolved in many contemporary simulations, there are still areas of research in which this phenomenon is a sub-grid process, e.g. in the environmental flows. Therefore the analysis of the effective reaction rate in the vicinity of such vortical structures could ameliorate a parametrisation of reactions in a mixing layer. In this paper dependence of the effective reaction rate on the Damköhler number is examined. Description of important equations and the numerical model could be found in section 2. Evolution of the Kelvin-Helmholtz instability is presented in section 3 while the section 4 refers to the analysis of the effective reaction rate.

2. Mathematical and numerical model

In this section mathematical and numerical model of the three-dimensional temporal developing mixing layer is presented. The shear layer was simulated using the finite volume method implemented in the OpenFOAM® toolbox.

2.1. Equations of the fluid motion

In our study we used the Navier-Stokes equation formulated in terms of the vorticity $\omega$ and velocity $u$. This formulation is quite popular in numerical simulations of transitional flows (Rempfer, 2003). Taking the curl of the Navier-Stokes equation yields the prognostic equation for the vorticity

$$\frac{\partial \omega}{\partial t} + (u \cdot \nabla)\omega = (\omega \cdot \nabla)\nabla u + \nu \Delta \omega,$$

where $\nu$ denotes the kinematic viscosity. The first term on the RHS is responsible for the vortex stretching effect which occurs only in the three-dimensional flows. It leads to the transition to the turbulence and is exceedingly important in the analysed problem. In this approach assumption of the constant density is very important, since it allows us to remove the problematic pressure term from the Navier-Stokes equation. However, one must retrieve the velocity field from the vorticity distribution because it appears explicitly in the equation (1). It could be achieved using a simple identity coming from the vector analysis

$$\nabla (\nabla \cdot u) - \nabla \times (\nabla \times u).$$

(2)

Since the divergence of the velocity field vanishes in case of incompressible flow this identity could be simplified to the following form

$$\nabla u = -\nabla \times (\nabla \times u).$$

(3)

Equations (1) and (3) form a complete set of equations of the fluid motion. They need to be solved just once in each time step, i.e. no additional Piso-like loop is needed. On the other hand the suite of appropriate boundary conditions for the vorticity usually is not straightforward. Especially some difficulties occur when solid walls are present in the system (Rempfer, 2003). Fortunately, in the case of a temporal developing mixing layer periodic boundary conditions can be used in the streamwise and spanwise direction. Moreover irrotational flow could be assumed at the remaining sides.

2.2. Transport equations for species

We considered a simple passive irreversible reaction of the second order

$$A + B \rightarrow C.$$  

(4)
The law of the mass action is assumed and consequently the reaction rate is equal
\[ w = k Y_A Y_B , \] (5)
where \( Y_A \) and \( Y_B \) denote the mass fraction of species \( A \) and \( B \) respectively and \( k \) is a reaction constant which in general depends on the thermodynamic variables, e.g. through the Arrhenius law. In this work we assumed constant value of \( k \) depending on a particular simulation.

Three transport equations (one for each specie) need to be solved. They have following form
\[
\frac{\partial Y_A}{\partial t} + (\mathbf{u} \cdot \nabla) Y_A = \frac{\nu}{Sc} \Delta Y_A - k Y_A Y_B , \\
\frac{\partial Y_B}{\partial t} + (\mathbf{u} \cdot \nabla) Y_B = \frac{\nu}{Sc} \Delta Y_B - k Y_A Y_B , \\
\frac{\partial Y_C}{\partial t} + (\mathbf{u} \cdot \nabla) Y_C = \frac{\nu}{Sc} \Delta Y_C + k Y_A Y_B ,
\] (6)-(8)
where \( Sc \) denotes the Schmidt number which is the ratio of the kinematic viscosity of the fluid and the molecular diffusivity of a particular specie. We assumed equal diffusivities for all species.

When the equations (6)-(8) are averaged problem with non-linear terms appears. The averaged advection term produces additional diffusion which could be introduced into equations by using turbulence models. The source term is the another issue. Substrates within the computational cell usually are not homogeneously mixed. Deviation from the well mixed limit can be represented by the effective reaction rate as follows
\[ k \bar{Y}_A \bar{Y}_B = k_{eff} \bar{Y}_A \bar{Y}_B , \bar{Y}_A \bar{Y}_B \]
what yields
\[ k_{eff} = \frac{k \bar{Y}_A \bar{Y}_B}{Y_A Y_B} , \] (9)
where “bar” denotes an averaged variable. This quantity is the main subject of our study. It depends explicitly on the reaction constant \( k \). However implicit dependence appears in the correlation of the concentration fields as well.

2.3. Time scales
In the problem one can distinguish different time scales describing different processes. Given a length scale \( \delta \), defined below by the initial conditions, and a velocity scale \( U \) we can obtain advection and diffusion time scales as follows
\[ \tau_A = \frac{\delta}{U} , \tau_D = \frac{\delta^2}{D} , \tau_\nu = \frac{\delta^2}{\nu} , \] (10)
where \( \tau_A \), \( \tau_D \) and \( \tau_\nu \) denote advection, diffusion and viscous time scale respectively. In flows including chemical reactions there is additional time scale that refers to the reaction rate. It is defined by the value of the reaction constant and characteristic value of the substrates concentration \( Y_{AB}^0 \), e.g. given by the initial or boundary conditions
\[ \tau_R = k Y_{AB}^0 . \] (11)

Ratios of these time scales yield non-dimensional numbers describing different regimes of the flow
\[ Re = \frac{\tau_\nu}{\tau_A} , \quad Sc = \frac{\tau_\nu}{\tau_D} , \quad Da = \frac{\tau_D}{\tau_R} . \] (12)
In our work Reynold (\( Re \)) and Schmidt (\( Sc \)) numbers remain constant and equal 500 and 1 respectively. Damköhler number was the main factor analysed in this study. We considered four different value of \( Da \): 1, 10, 100 and 1000. There was not any significant difference between cases with \( Da \) less than one. Therefore they are not included in this paper.
2.4. Initial and boundary conditions
In all simulations the same suite of initial and boundary conditions were used. The initial conditions refer to the analytical solution of the one-dimensional diffusion equation (case of the one-dimensional mixing layer). They have the following form

\[ U_0 = U_0 \text{erf} \left( \sqrt{\frac{\pi y}{\delta}} \right) e_x, \]  
\[ \omega_0 = \frac{2U_0}{\delta} \exp \left( -\pi \frac{x^2}{\delta^2} \right) e_z, \]  
\[ Y_{A0} = \frac{1}{2} \text{erf} \left( 2\sqrt{\frac{\pi y}{\delta}} \right) + \frac{1}{2}, \]  
\[ Y_{B0} = 1 - Y_{A0}, \]  
\[ Y_{C0} = 0. \]  

where \( \text{erf} \) denotes the error function, \( U_0 \) is a characteristic velocity and \( \delta \) is a initial width of the mixed layer defining the characteristic length scale as well. Using this parameter as the length scale is reasonable since we are interested in the early stage of the instability growth when the turbulence is still not well developed. Given the length scale and velocity scale one can derive a vorticity scale equal \( \Omega = 2U_0/\delta \). A disturbance in the vorticity field was added to the initial profile presented above. There were two types of the disturbance tapped in simulations following the study of Rogers & Moser (1992)

\[ \omega'_z = \frac{15A_z U_0 k_z}{16 \left[ \cosh(k_z y) - 0.25 \cos(k_z x) \right]^2}, \]  
\[ \omega'_x = A_x \sin(k_x z) \exp \left( -\pi \frac{y^2}{\delta^2} \right), \]  

referring to the \( z \) and \( x \) component of the vorticity. \( A_z \) and \( A_x \) denote the amplitudes while \( k_z \) and \( k_x \) the wave numbers of the perturbations. Note that all initial conditions obey the symmetry according to the plane \( z = 0 \).

Simulations were performed in a rectangular domain with periodic boundary conditions in two (\( x \) and \( z \)) directions. Size of the domain was chosen in such a way that the most unstable modes of the Kelvin-Helmholtz instability could evolve (Michalke, 1964; Rogers & Moser, 1992). Thus \( \Delta x = 7.28 \delta \) and \( \Delta z = 4.37 \delta \). Boundary conditions were prescribed consistently with the initial profiles. The vorticity vanishes at both non-periodic sides while \( Y_B \) at the “top” and \( Y_A \) at the “bottom” side of the domain.

2.5. Numerics
How it was mentioned before, simulations were conducted using the finite volume method (FVM). It has numerous of advantages however, in its basic form FVM is limited to the second order of the spatial accuracy. One of goals of our study was to check the capability of using the FVM in the transitional flows modelling, since it requires a very careful treatment (Rempfer, 2003). A crucial element within the numerical schemes was the interpolation scheme of the advection term in equation (1). Linear upwind scheme was used for this purpose in order to ensure the second order of accuracy and the stability of the solution.

Two different approaches to the temporal discretisation of the vorticity equation were used. We employed both implicit schemes of the second order (backward and Crank-Nicholson) and explicit second and fourth order Runge-Kutta schemes. All second order schemes gave similar results while the fourth order RK scheme appeared to stabilise the flow so that the K-H instability evolved slower.
Simulations were performed with three different resolution of the mesh containing 32x48x19, 64x96x38 and 128x192x76 cells. Our analysis was meant to check which stages of the KH instability could be properly resolved with these grids.

3. Development of the Kelvin-Helmholtz instability

All simulations confirmed the main role which the two-dimensional mode plays in the early stage of the KH instability. Even in the presence of the three-dimensional perturbation the flow remained two-dimensional for a long time (up to $20-30\tau_A$). On the other hand three-dimensional flow finally evolves even from the plane initial conditions. Evolution of the spanwise vorticity in the symmetry plane ($z = 0$) is presented in the figure 1. Initially plane layer of mixed fluid abruptly rolls up. This process refers to the first stage of the KH instability. The roller does not grow permanently and at $t = 10\tau_A$ it reaches its maximal width. After this time vorticity slowly diffuses and the roller becomes flat again. It could be considered as the second stage of the instability. It is present in both two- and three-dimensional flows. However, in three-dimensional flows it is followed by the transition to turbulence. This transition may appear at different time depending on the type of initial conditions. The higher three-dimensionality is introduced at the beginning the sooner turbulence evolves. In our simulations turbulence developed even from the two-dimensional initial conditions. Therefore one can state that two-dimensional mixing layer is not a stable solution of the equation (1). Evolution of the vorticity field during the transition is presented in the figure 2. Very similar picture has been obtained by Rogers & Moser (1992).

![Figure 1. Cross-section of the domain in the plane $z = 0$ with isolines of the $z$-component of the vorticity. Difference between neighbouring isolines is equal $0.1\Omega$. Red colour denotes the irrotational flow while the blue refers to the negative value of $\omega_z$.](image)

4. Effective reaction rate

4.1. Mixing

Three stages of the K-H instability refer to different type of mixing in the system. In the first step molecular diffusion is accelerated by the vortex forming in the system (Bajer et al., 2001). However, due to limited size of the vortex effective transport takes place in the central area of the domain. Along the edges of this area high gradients of transported quantities occur. Molecular diffusion plays the main role in this place and therefore it limits the overall mixing rate. This is a reason of slowing down the mixing during the second part of the instability development. The
flow is then still two-dimensional however, three-dimensional instability is evolving continuously. When it becomes an important part of the flow trajectories of the fluid particles gain the chaotic features. It could be seen in the figure 3. When the flow comes into the turbulent regime the mixing accelerates again. Contrary to the first stage of the K-H instability turbulent mixing takes place in the whole domain. It is then influenced by the imperfect boundary conditions what significantly deteriorates the quality of the simulations. Even though the mixing processes seem to be properly modelled during most of the instability evolution. In the figure 4 averaged profiles of the passive scalar are presented. The averaging has been performed over the periodic directions of the domain. One can recognise changes in the character of the mixing during the simulation.

**Figure 2.** Cross-section of the domain in the plane \( z = 0.1 \) with isolines of the \( z \)-component of the vorticity. Difference between neighbouring isolines is equal 0.1Ω in figures (a), (b) and 0.2Ω in figures (c), (d). Presented colour scale has units equal 0.1Ω.

**Figure 3.** Stream lines starting from the plane \( z = 0.1 \) at \( t = 25\tau_A \) (a) and \( t = 35\tau_A \) (b). In the left picture all stream lines remain in the plane what is not the case in the picture (b). Colour refers to the magnitude of the velocity in units equal \( 2U_0 \).
4.2. Reaction

The nature of the mixing influences the character of the reaction present in the system. Very slow reactions do not change significantly concentration of substrates which could be treated as passive scalars. Therefore the source term is considerable only in the equation (8). During the whole simulation mixing is in progress what effects the increase in the effective reaction rate for slow reactions. Faster reactions could influence the mixing of substrates as well.

Evolution of the effective reaction rate (see equation (9)) for different value of the Damköhler number is presented in the figure 5. Slow reactions demonstrate the regular, monotonic changes in the effective reaction rate while fast reactions appear to be very sensitive to the character of mixing. After the first stage of the K-H instability a significant drop in the effective reaction rate occurs for Damköhler number equal 100 and 1000. This effect is caused by creation of a buffer layer of product in the area of well mixed fluid which separates the substrates and slows down the reaction. This layer could be broken only when the turbulent motions develop.

![Figure 5](image)

**Figure 5.** Evolution of the effective reaction rate normalised by the reaction constant for different value of the Damköhler number in the case of stable (a) and unstable (b) mixing layer.
In order to check the reliability of our simulations they were performed on three different meshes. In figure 6 comparison of results obtained from these simulations is presented. Shown is the concentration of product of the reaction averaged over the whole domain. What can be seen from this picture is that the largest difference between simulations occurs in cases of fast reactions when the vortex roller is forming. Moreover the coarsest mesh could not represent properly the turbulent mixing. In spite of these differences our modelling seems to be at least qualitatively reliable.

![Graphs showing concentration of product vs. Damköhler number](image)

Figure 6. Comparison of the averaged concentration of the product of the reaction obtained with three meshes with different resolution for different value of the Damköhler number.

5. Summary

The main goal of our work was to analyse the influence of the Kelvin-Helmholtz instability on the reaction in a mixing layer. We performed numerical simulations of the instability using the finite volume method. Evolution of the vorticity field was qualitatively compared with the work of Rogers & Moser (1992). Additional transport equations were included in the model so that we could simulate the reaction between two initially separated substrates. We considered reactions with different time scales in order to analyse influence of this parameter on the effective reaction rate. We obtained interesting results concerning the dependence of the effective reaction rate on the Damköhler number. Fast reactions appeared to be very sensitive to character of
the mixing. Creation of the buffer layer of product between the substrates seems to be very
important phenomenon which change the dynamic of fast reactions. This problem was considered
by Crimaldi et al. (2006) and needs to be analysed in detail for the case of flows with high
Damköhler numbers.

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