LARGE EDDY SIMULATION OF MIXING PROCESSES IN TURBULENT LIQUID FLOWS WITH CHEMICAL REACTIONS

Ying Huai and Amsini Sadiki
Department of Mechanical Engineering,
Institute for Energy and Powerplant Technology,
Technical University Darmstadt
Petersenstraße 30, D-64287 Darmstadt, Germany
sadiki@ekt.tu-darmstadt.de

ABSTRACT

In this work, a new SGS model for the filtered scalar flux vector is presented to develop Large Eddy Simulation (LES) of non-premixed, turbulent reacting liquid flows. To cover different reaction regimes, this work uses different ways to approach the chemical source term. To assess the accuracy of the SGS models proposed, the results are first provided for a non-reacting jet in (water) channel flow showing a very good agreement with experimental data. Then the LES was applied to a gas mixing layer flow with chemical reactions. LES computations are well compared with DNS data. Thereafter the first results in a confined impinging jets reactor are presented and discussed.

INTRODUCTION

There has always been a considerable interest in turbulent mixing as it is one of the operations applied most frequently in the chemical industry, both as an independent operation with the objective of obtaining homogeneity of a mixture, and as a way of increasing the contact surface in other operations (such as absorption, extraction or drying) (see e.g. Villermaux, 2002; Baldyga & Bourne, 1999; Fox, 2003). In recent years this has further increased due to the need of a reliable prediction of mixing either in reacting environments or non-reacting applications. In turbulent reacting flows scalar mixing is particularly important, for in many instances it is the rate determining process. To model the turbulent reactive mixing, it is useful to check which of the sequence of mixing processes can directly or indirectly affect the course of the chemical reactions. Comparison of the characteristic times for turbulent mixing and reaction (through the so called Damkhöler number) can provide this information.

For complex configurations of technical importance in which experimental investigations are difficult to be accomplished, a comprehensive knowledge of phenomena can well be achieved only by solving the equations governing the processes involved in the frame of Computational Fluid Dynamics (CFD) (Baldyga & Bourne, 1999, Fox, 2003) .

Large Eddy Simulation (LES) is carried out in this work. Compared with Reynolds Averaging based Numerical Simulation (here, RANS), which wipes out most of the important characteristics of a time-dependent solution, and Direct Numerical Simulation (DNS) which resolves all turbulent scale structures and remains still computationally unrealistic for turbulent flows of high Reynolds number, LES emerges as a suitable tool for such flow systems governed by unsteady large scales structures. Classical LES allows to compute the resolved large scales structures whilst the non resolved small scales structures are modeled by means of so called subgrid scale (SGS) models, Smagorinsky (1963).

The difficulties encountered in modeling the mixing processes in reacting turbulent liquid flows can be primarily attributed to the closure problems of SGS-scalar flux in higher Schmidt number fluids and of filtered chemical source term. Michioka et al. (2004) presented LES of turbulent liquid flows with chemical reaction based on a presumed (beta) probability density function model without particular emphasis of the SGS scalar flux effect as they used the linear eddy diffusivity model. However, it is well known that this simple model has a limited validity in turbulent reacting flows (Peng et al., 2002; Jaberi et al., 2003). Especially it is not valid for high Schmidt number fluid transport processes (Fox, 2003, Huai, 2005, Huai et al., 2006).

With regard to SGS scalar flux models, different model approaches have been evaluated in the past (see in Jaberi et al, 2003, Peng et al., 2002, Kang et al., 2001, Huai, 2005). Toward the development and assessment of the SGS model for the filtered reaction source term, most of previous research work focused on fast reaction systems related to combustion, especially dealing with gaseous flows. However, for slow or moderate chemical reactions that are mostly related to process engineering applications, the investigations are very few, Fox (2003), Michioka et al., (2004).

In this work, a new SGS model for the filtered scalar flux vector is presented along with a filtered reaction term to develop Large Eddy Simulation (LES) of non-premixed, turbulent reacting liquid flows. In its cubic form in terms of the gradient of the filtered scalar field, this model combines the conventional linear Eddy diffusivity model with two additional new terms. The first contribution accounts for the cubic dependency in terms of the gradient of the filtered scalar field and the second couples the (deviatoric) SGS stress tensor and the gradient of the filtered scalar field. Thus they involve a non-linear Eddy diffusivity tensor in terms of the gradient of the filtered scalar field. To cover different reaction regimes, this work uses different ways to approach the chemical source term. A (beta) presumed probability density function (pdf) and a dynamic scale similarity approach will be used.

To assess the accuracy of the SGS models proposed, the results are first provided for a non-reacting (water) jet in channel flow achieving a very good agreement with experimental data (Meyer et al, 2001). Then the LES is applied to a gas mixing layer flow with chemical reactions.

The results are compared with DNS data by Michioka et al, 2004. To confirm whether the present SGS models are applicable for a practical turbulent reacting liquid flow, a confined impinging jets reactor is investigated. It consists of two high-velocity, coaxial liquid jets as experimentally investigated by Johnson and Prud'homme (2003). First numerical results are provided and discussed.

MATHEMATICAL MODELS AND NUMERICAL PROCEDURE

By applying a filter operation, the filtered continuity, momentum, conserved scalar and species concentration equations emerge for LES computations. The transport equation of a reactive concentration can be written as:

$$\frac{\partial}{\partial t}\bar{\rho}\bar{\Gamma}_K + \frac{\partial}{\partial x_i}\left(\bar{\rho}\bar{u}_i\bar{\Gamma}_K\right) = \frac{\partial}{\partial x_i}\left(\bar{\rho}D_{\Gamma_K}\frac{\partial\bar{\Gamma}_K}{\partial x_i}\right) - \frac{\partial}{\partial x_i}\left(\bar{\rho}J_{Ki}^{sgs}\right) + \bar{S} \quad (1)$$

where the overbar denotes a filtered value except for the density. The quantity $\bar{\Gamma}_K$ is the filtered concentration of reactant K ($K=1,\ldots,N$), u_i ($i=1,2,3$) denotes the velocity components at x_i-direction, ρ the density, D_K the molecular diffusivity coefficient.

While the effect of the small scales in the flow field appears through the unknown SGS stress tensor, it is accounted for in scalar field through the SGS scalar flux vector, J_i^{SGS}. The latter must be modeled together with the SGS stress tensor in order to obtain a closed system of equations. The quantity S in eq. (1) represents the chemical reaction source term.

We do not focus especially on the flow field, rather concentrate on an accurate representation of the SGS term in the scalar field. We therefore apply the simple Smagorinsky model (Eddy viscosity model) with a standard Germano dynamic procedure for determining the model coefficient to close the SGS stress tensor in the motion equation.

When a second-order, irreversible, and isothermal reaction (A + B -> P) is considered, the filtered reaction source term is expressed as $\bar{S} = Da\overline{\Gamma_A\Gamma_B}$ where Da is the Damkoehler number. In a rapid reaction case, the chemical timescale is far smaller than that of the turbulent diffusion. Since such a small time step cannot be set even for the fastest supercomputer, a conserved scalar approach found therefore usual applications in describing turbulent liquid flows. Thereby the SGS scalar flux vector appearing in the transport equation of the conserved scalar (here the mixture fraction) was approximated by means of the Eddy diffusivity model. In the present work an approximation will be achieved following the new model to be introduced later on. Assuming the equilibrium chemistry, the concentrations of all species in a non-premixed, single-step reaction can be related to the conserved scalar (Fox, 2003, Michioka et al., 2004). The filtered concentration values including SGS mixing should be computed from the SGS probability density function of the conserved scalar. A presumed ansatz has been used for this purpose.

In a moderately fast or slow reaction case, the timescale of the chemical reaction is equivalent to that of the turbulent diffusion. In the transport equation (1), the filtered reaction source term can be decomposed into two terms, as:

$$\bar{S} = Da\overline{\Gamma_A\Gamma_B} = Da\overline{\Gamma_A\Gamma_B} = \left(\bar{\Gamma}_A\bar{\Gamma}_B + \overline{\gamma'_A\gamma'_B}\right) \quad (2)$$

where γ_i' are the concentration fluctuation of the chemical species, *i,* at subgrid scale level. In Michioka et al. (2004) the model that involves the mixing of the chemical species at the SGS by means of joint probability density function of species concentrations has been employed. In the case a slow chemistry is considered, the fluctuation part in (2) can also be modeled through a dynamic scale similarity approach following Vinuesa et al., (2005). This approach is used in this work.

The SGS scalar flux model is expressed as an explicit anisotropy-resolving algebraic model derived from the transport equation of the SGS scalar flux vector, such that the irreversibility requirements of the second law of thermodynamics are automatically fulfilled by the suggested parameterization (Sadiki, 2005). In its at least cubic form, the chosen new model combines the conventional linear eddy diffusivity model with two additional terms. The first term involves the gradient of the filtered scalar field in cubic form and the second couples the (deviatoric) SGS stress tensor and the gradient of the filtered scalar field as given in equation

$$J_i^{SGS} = (D_{ed} + \lambda\frac{\partial\bar{f}}{\partial x_k}\frac{\partial\bar{f}}{\partial x_k})\frac{\partial\bar{f}}{\partial x_i} + D_{dev}T_{SGS}\tau_{ij}^{SGS(dev)}\frac{\partial\bar{f}}{\partial x_j} \quad (3)$$

where $D_{(\cdot)}$ and λ are the model coefficients. This model involves a nonlinear tensor of diffusivity

$$D_{ij}^{SGS} = (D_{ed} + \lambda\frac{\partial\bar{f}}{\partial x_k}\frac{\partial\bar{f}}{\partial x_k})\delta_{ij} + D_{dev}T_{SGS}\tau_{ij}^{SGS(dev)}, \quad (4)$$

in terms of the quadratic of the gradient of the filtered scalar field. That term may be related to the scalar dissipation rate which can be expressed in terms of the time-scale-ratio that is in turn function of the Re- and Sc-numbers (Fox, 2003). So the new model is designed to account well for flow and scalar anisotropy effects. According to the modeling level used for the deviatoric part of the SGS stress tensor (linear, non-linear, anisotropic) this model may lead to various special models that have been proposed in the literature. A detailed analysis of this consideration can be found in (Sadiki et al., 2007).

Restricted ourselves in this paper to Smagorinsky type model and to linear terms in scalar gradient in eq. (3), the simplest model case to be considered can be derived by expressing the SGS time scale in (3) in terms of the filter size and the SGS viscosity defined in the Smagorinsky model. Equation (3) then reduces to

$$J_i^{SGS} = D_{ed}\frac{\partial\bar{f}}{\partial x_i} + D_{an}\Delta^2\bar{S}_{ij}\frac{\partial\bar{f}}{\partial x_j} = D_{ij}^{Smag}\frac{\partial\bar{f}}{\partial x_j}; \quad D_{ij}^{Smag} = D_{ed}\delta_{ij} + D_{an}\Delta^2\bar{S}_{ij} \quad (5)$$

where $D_{(\cdot)}$ are the model coefficients and D_{ij}^{Smag} the reduced eddy diffusivity tensor. In particular

$$D_{ed} = \frac{\upsilon_t}{\sigma_t},\ \sigma_t \equiv Sc_t, \mathrm{Pr}_t \quad (6)$$

expresses the well known Eddy diffusivity coefficient. All the model parameters in (3) or (5) have to be determined dynamically according to the requirements along the entropy inequality treatment (Sadiki et al., 2007).

The SGS scalar flux model (5) has been successfully validated in different configurations along with a jet in cross flow as experimentally investigated by Andreapoulos et al., 1984, a mixing layer without chemical reactions (Huai et al., 2007) and a jet in channel water flow (Meyer et al., 2001). Thereby the mixing processes associated with jets in cross and channel flows could be quantified by using the so called mixedness parameters. According to the nature of turbulent mixing, three main characteristic scales have been individuated (macro-, meso- and micro-mixing) and quantified by means of three different parameters. For details, see Huai, 2005. Here focus is put on the ability of the new models to well capture the effect of high Schmidt number in reacting environments based on their proved performance in non-reacting flows.

All the filtered continuity, momentum, conserved scalar and species concentration equations were implemented into the FASTEST-3D CFD code. It features geometry-flexible block-structured, boundary fitted grids with collocated variable arrangement. Second-order central schemes are used for spatial discretization except for the convective term in the scalar transport equation. Here, a flux-limiter with TVD (total variation diminishing) properties is employed to ensure bounded solutions for the mixture fraction. Pressure-velocity coupling is achieved via a SIMPLE similar procedure. As time integration scheme the second-order implicit Crank-Nicolson method is used. The resulting set of linear equations is solved iteratively using a SIP-solver. The code is parallelized based on domain decomposition using the *MPI* message passing library.

CONFIGURATIONS, RESULTS AND DISCUSSION

To assess the prediction ability of the proposed models two configurations have been investigated in this work.

1. Non-reacting Jet in channel flow (water): The reference geometry consists of a straight duct with a square cross section of *40*40 mm* as experimentally investigated by Meyer et al. (2001). The jet, emerging from the mid-point of one side wall and perpendicular to this wall, is created from a pipe with an inner diameter of *D=4* mm. Both channel and jet consists of clean water. The water temperature is *26°C*. The Reynolds number *Re = 33750* is based on hydraulic diameter and bulk mean velocity U_{cf} of the duct flow. The velocity ratio *R=2*.

The coordinate system is centered at the jet axis at the entrance to the duct, *x*-axis *being* in the direction of duct flow and *z*-axis in the direction of the jet axis as depicted in Figures 1a. Experimental data are available in the x-z plane centered in the duct, yielding the *U* and *W* components of velocity in *x* and *z* directions, respectively. Values of the mean mixture fraction and corresponding fluctuations are also provided.

To make a compromise between the real CV requirement for mixing process simulation using LES (see in Huai, 2005) and the available computational capacity the total control volumes of *415788* cells have been found to be acceptable (Figure 1b).

In order to achieve a fair assessment for the SGS scalar flux models for this high Schmidt number case, simulation results with different SGS scalar flux models are compared to the experimental data. The original eddy diffusivity model is used with a constant model parameter. Then, its modified version by means of a dynamic procedure for determining the model coefficient is used. For comparison a scale similarity model and the thermodynamically consistent anisotropy model (5) are also implemented with dynamical procedure to compute the model coefficients. Figure 2 (top) shows the calculated values of mean mixture fraction against experimental data. It can be seen that the difference between the eddy diffusivity model and anisotropy model becomes larger in the wake the flow (not shown). The same behaviour is observed with the scale similarity model. The mixture fraction seems to be under-predicted while a qualitative agreement is quite good. This error may be ambilateral. In their paper, Meyer et al. specified the error for the mean mixture fraction as high as *3.5%*. On the other hand, numerical error may mainly depend on the veracity of SGS scalar flux models. In fact the results of the scale similarity model and the eddy diffusivity model with/without dynamic procedure are almost the same results for the mean quantity while the anisotropy SGS model appreciably improves the prediction.

Concentrated on the fluctuation quantity, the results obtained by different models are presented in Figure 2 (bottom). The eddy diffusivity model with dynamic procedure reproduces the experimental observations poorly in comparison to the scale similarity model. In contrast, the anisotropy model is still more recommendable as it retrieves clearly well both the position and the maximum values of scalar fluctuations. This better prediction ability in terms of qualitative and quantitative statements is however appended by more computational cost. Comparatively, all other models over-predict the scalar flux at the near wall region and under-predict this quantity in the mixing region (shear layer of the jet flow), as a consequence of a non satisfactory wall treatment. Proving its ability in predicting mixing systems characterized by high Schmidt numbers, let us now focus on gas systems.

2. Reacting gas mixing layer: The next configuration is a reacting mixing layer that has been first investigated by Michioka et al., 2004. It consists in a sheared reacting gas mixing layer in which chemical species A and B were introduced separately into lower and upper parts of the computational domain at the initial time as shown in figure 3. In their work, Michioka et al., carried out DNS and LES calculations as mentioned above. Accordingly the numbers of the computational grid points were 256 * 256 * 256 in the *x*, *y*, and *z* directions for the DNS and 32 * 32 * 32 for the LES. The Reynolds number based on the initial velocity difference and vorticity thickness at $x = 0$ was 3350.

Both fast reaction and moderate fast reaction cases have been investigated. Figure. 4 shows the time evolutions of the mean concentration of chemical product P at $y = 0$ for a fast reaction and moderate fast reaction case. Although the LES overestimates the amount of product P for the fast reaction case, the LES based on the present similarity models well agrees with the DNS. Figure. 4 also shows the time evolution of the mean-squared concentration fluctuation of species A at $y = 0$ for both fast reaction and moderate fast reaction cases. The LES predictions based on Eq.2 are also in good agreement with the DNS predictions of gas flows characterised by low Schmidt numbers. The next step is to perfom quantitative predictions for a complex reacting liquid flow systems.

3. Confined Impinging-Jets Reactor: The geometry used is identical to the experiments by Johnson and Prud'homme as depicted in figure 5. The diameter of the

impinging jets is 0.5mm. Having in mind the quantities D, H, Z and delta as the chamber diameter, height, length, and the outlet diameter, respectively, their scaled values are D=4.76d, H=0.8D, Z=1.2D and delta=2d. Detailed information on this configuration can be obtained from Johnson and Prud'homme, 2003.

A pair of second-order parallel reactions is used to evaluate the extent of mixing in the experiments. The reaction stoichiometry can be expressed as

$$OH^- + H^+ \rightarrow H_2O$$

$$CH_3C(OCH_3)_2CH_3 + H^+ (+H_2O) \rightarrow CH_3COCH_3 + 2CH_3OH + H^+$$

or in the form:

B+A → P1 and D+A+H2O → P2+P3+A

Where A, B and D are reactants and P1, P2 and P3 are products. As noted by Liu et al., 2006 the second reaction is catalytic so that A appears as both a reactant and a product with no net consumption. Non-premixed feed conditions are used and the reactor is operated in continuous mode with mass flow rates as given in figure 5. As pointed out by Liu et al., (2006) and Johnson et al., (2003), since the first reaction is very rapid, when excess B is present thze second reaction will take place under conditions where mixing is slow compared to its reaction rate. The conversion of D is thus a sensitive measure of the extent of mixing in the reactor. In this work we aim at comparing the predicted conversion to experimental data using LES in contrast to Liu et al., (2006) who used RANS.

The computational grid consists of about 500.500 CV. The inflow conditions are taken according to experimental data.

Some first results are provided in figure 6. It displays the distributions of the Reynolds-average species of A, B and D in comparison with experimental data when the jet Reynolds number is 400. An encouraging agreement with experimental data is achieved demonstrating the ability of the numerical model to satisfactorily capture the mixing processes under study. Advanced models for the description of the reaction source term that well account for the turbulence-chemistry interaction and a consistent consideration of chemical reaction contribution in the SGS scalar flux model could improve the prediction results. This task is left for future work.

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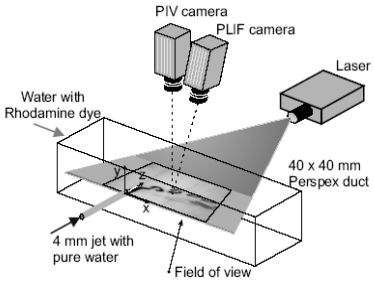

Figure 1a: Schematic drawing of the flow and measurement system setup of jet in channel flow (Meyer et al., 2001)

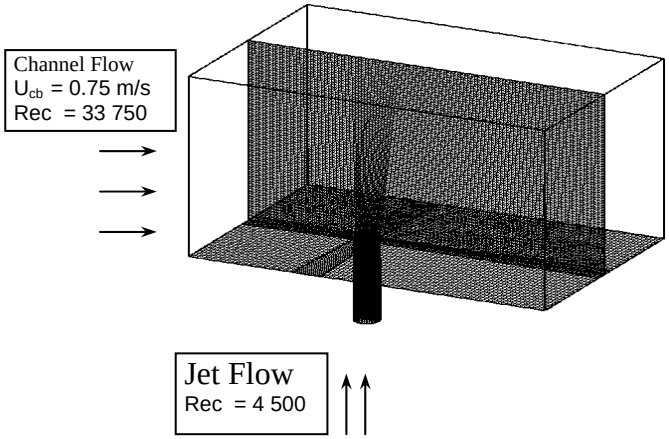

Figure1b: Numerical Setup of Jet in Channel Flow (CV: 415788)

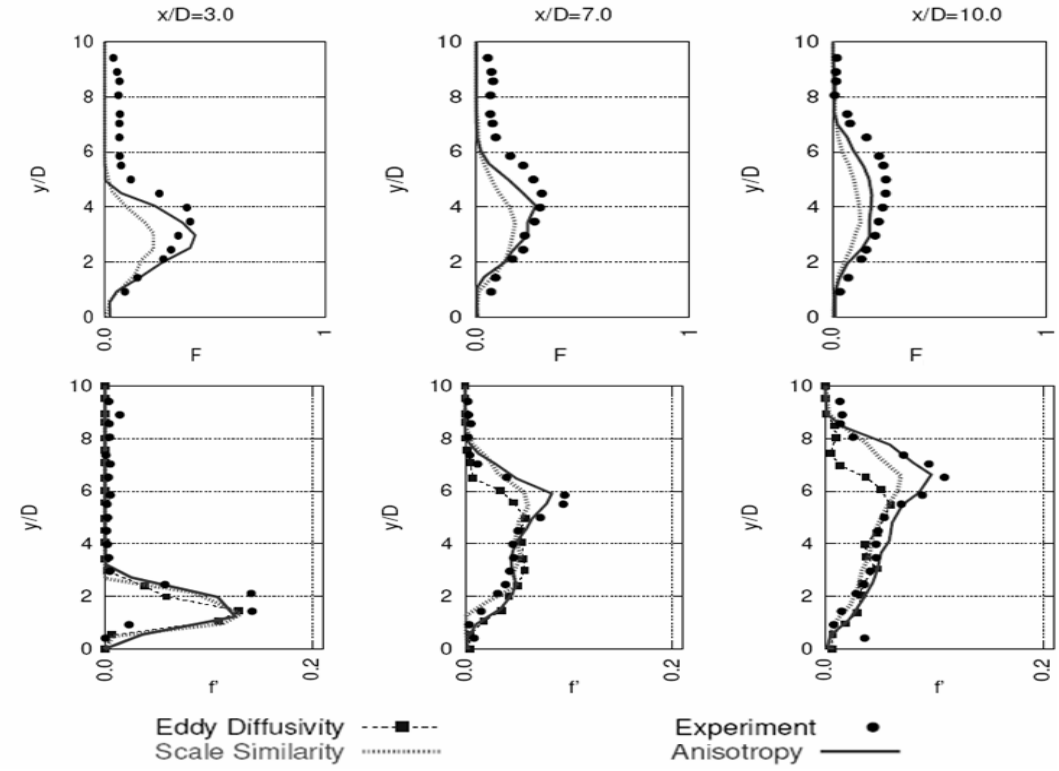

Figure.2: Comparison of mixture fraction quantities with experimental data for the jet in channel flow configuration Mean mixture fraction (*F*); fluctuation of mixture fraction (*f'*)

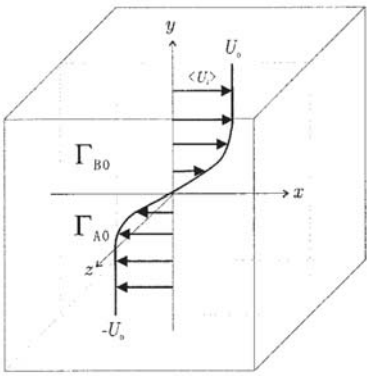

Figure.3 Numerical setup of reaction mixing layer

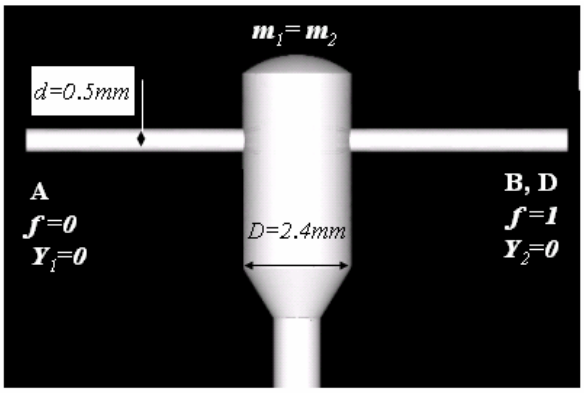

Figure 5: Confined impinging-jets reactor

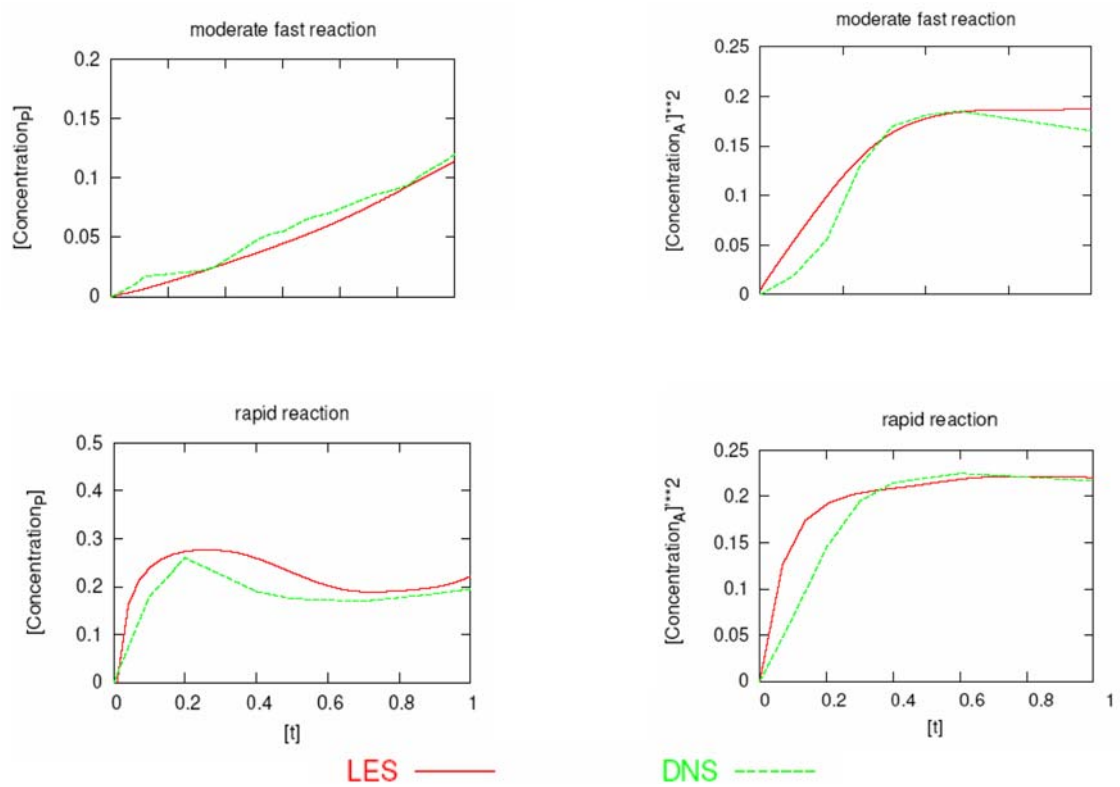

Figure.4 Time evolution of the mean and squared concentration fluctuation of chemical species (top: Moderate fast reaction case; bottom: Fast reaction case)

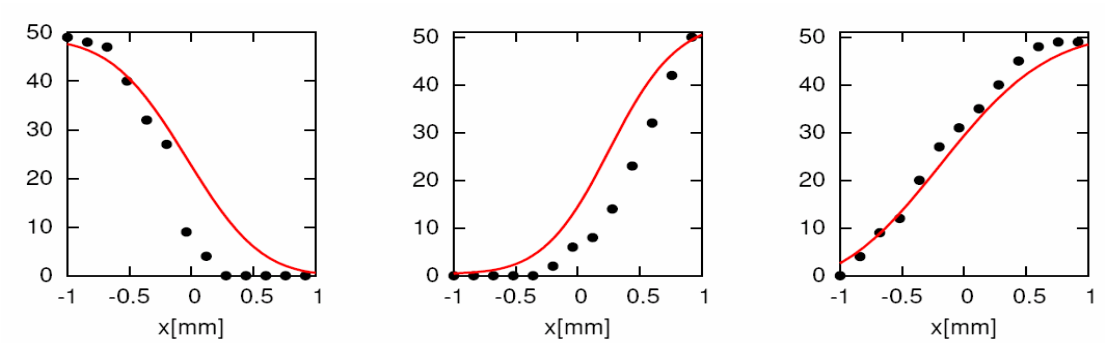

Figure 6: Axial distributions of the Reynolds-average species of A (left), B (middle) and D (right) in comparison with experimental data, respectively, when the jet Reynolds number is 400.