

# SIMULATION OF CHEMICAL PROCESSES IN TURBULENT FLOW REACTORS BY MONTE CARLO METHOD

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**Abstract:** A coupled RANS – Monte Carlo method is developed for computational studies of chemical processes in industrial turbulent flow reactors. The method is validated against simple diffusion and reaction-diffusion problems.

## Introduction

Mixing and reactivity in turbulent reactive flows is the issue of major importance for many industrial applications including chemical technology [1]. Computational Fluid Dynamics (CFD) is currently considered as a powerful tool for chemical plant design and optimization both to ensure efficient operation process and to avoid uncontrolled energy release. Nowadays, CFD based engineering calculations most often use the simplest models of turbulence (e.g., k-epsilon model) and turbulence – chemistry interaction (e.g., perfect stirring reactor). Such models do not take into account that mixing of reactants can occur at the rates comparable with the rates of chemical reactions. Also, they neglect the existence of a whole spectrum of residence times for different flow elements in reactor, which is caused by turbulent velocity fluctuations.

It is well accepted that the local mean reaction rate in the turbulent reactive flow must be calculated with due regard for local instantaneous reaction rates. In the presence of turbulent velocity fluctuations all flow parameters should be treated as stochastic. In view of it a local instantaneous state of the matter in any flow location can be considered as one of multiple stochastic realizations. Then the mean reaction rate in this location can be found by averaging over all possible realizations. This approach implies the use of the probability theory formalism, in particular, probability density functions.

This paper briefly describes the approach and provides several computational examples.

## Formulation

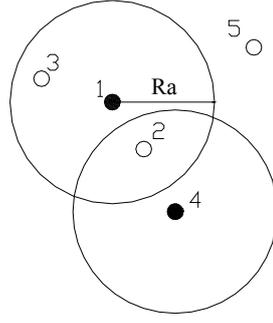
The conservation equations for turbulent reactive flow can be formulated either in Eulerian or in Lagrangian form [2, 3]. The Lagrangian formalism requires consideration of multiple realizations in each flow location. Evolution of each realization (“Lagrangian element”) is governed by the set of equations with certain initial and boundary conditions. Each element possesses its own instantaneous velocity vector, density, and enthalpy. Lagrangian elements interact with each other via mass, momentum, and energy transfer only within a so called “action sphere” of radius  $R_a$  (Fig. 1). Introduction of the action sphere is grounded on the fact that scalar diffusion and momentum fluxes exhibit finite penetration rate. In Fig. 1, element 1 interacts with elements 2 and 3 inside its action sphere, whereas it does not interact with elements 4 and 5 located outside.

The set of governing equations for each Lagrangian element is presented below [2, 3].

*ith element trajectory equation:*

$$\frac{dx_k^i}{dt} = u_k^i, \quad (1)$$

where  $x_k^i$  is the element coordinate ( $k=1,2,3$ ) and  $u_k^i$  is its velocity component;



**Fig. 1.** Interaction of Lagrangian elements in turbulent flow.

*ith element mass conservation equation:*

$$\frac{d(\rho_l^i V^i)}{dt} = \nabla \vec{J}_l^i, \quad (2)$$

where  $\rho_l^i$  is the partial density of the  $l$ th species in the element,  $V^i$  is the element volume,  $\nabla \vec{J}_l^i$  is the diffusion flux of the  $l$ th species to/from the element.

*ith element momentum conservation equation:*

$$\rho^i \frac{d\vec{u}_k^i}{dt} = \frac{\partial P}{\partial x_k} - \nabla(p\vec{E} - \tau), \quad (3)$$

where  $\rho^i$  is the element density ( $\rho^i = \sum_1^{\text{nsp}} \rho_l^i$ ),  $\nabla(p\vec{E} - \tau)$  is the term representing the effects of molecular viscosity and pulsating pressure gradient on element momentum,  $P$  is the local mean pressure;

*ith element energy conservation equation:*

$$\rho^i \frac{dh^i}{dt} = -\nabla \vec{q}^i + h_{\text{hom}}^i, \quad (4)$$

where  $h^i$  is the element enthalpy,  $\nabla \vec{q}^i$  is the heat flux to/from the element, and  $h_{\text{hom}}^i$  is the heat effect of chemical reactions.

Note that element motion in Eq. (1), as well as the effect of mean pressure gradient in Eq. (3) and chemical energy release in Eq. (4) are treated exactly, i.e., do not require modeling. However the effects of molecular diffusion (term  $\nabla \vec{J}_l^i$  in Eq. (2)), molecular thermal conductivity (term  $\nabla \vec{q}^i$  in Eq. (4)), and term  $\nabla(p\vec{E} - \tau)$  in Eq. (3) require modeling. For modeling these terms the standard models [4] were used:

*Diffusion flux:*

$$\nabla \vec{J}_l^i = -0.5C(Y_l^i - \bar{Y}_l^i)\rho^i V^i \omega, \quad (5)$$

where  $C \approx 2.0$  is the coefficient,  $Y_l^i$  is the concentration of the  $l$ th species in the  $i$ th element,  $\bar{Y}_l^i$  is the local mean concentration of the  $l$ th species at the position of the  $i$ th element, and  $\omega$  is the local turbulence frequency determined by the local turbulent kinetic energy  $k$  and its dissipation  $\varepsilon$ ;

*Heat flux:*

$$\nabla \vec{q}^i = -0.5C(h^i - \bar{h}^i)\rho^i \omega, \quad (6)$$

where  $\bar{h}^i$  is the local mean enthalpy at the position of the  $i$ th element;

*Viscous fluxes and pulsating pressure gradient:*

$$(\rho^i)^{-1} \nabla(\rho E - \tau) = -\zeta(\mathbf{u}_k^i - \bar{\mathbf{u}}_k^i) + \mathbf{A}(t), \quad (7)$$

where  $\bar{\mathbf{u}}_k^i$  is the local mean velocity component at the position of the  $i$ th element,  $\zeta \approx 2.075\omega$  is the coefficient,  $\mathbf{A}(t)$  is the stochastic function.

The equations are subject to boundary conditions at rigid walls, symmetry planes, and open boundaries. At reactor walls the tangential velocity component, turbulent kinetic energy and its dissipation have to vanish. At symmetry walls, mirror conditions are applied. At open boundaries inlet/outlet conditions are used.

The solution algorithm including the interpolation procedure is described elsewhere [2, 3]. For the sake of computational efficiency the problem is solved by coupling the finite volume solution of Reynolds Averaged Navier – Stokes (RANS) equations with the Monte Carlo (MC) solution of Eqs. (1)-(7). The finite volume method provides the fields of turbulent kinetic energy and its dissipation, as well as the field of mean pressure gradient. Using this information, the MC method provides information on the motion of all elements and on the evolution of their properties. In general, RANS–MC coupling requires iterations to obtain the same fields of mean flow variables from both sides.

## Validation

The RANS–MC method was validated against well-known solutions of relatively simple diffusion and reaction-diffusion problems.

### TEST 1: 1D DIFFUSION IN ISOTROPIC HOMOGENEOUS TURBULENCE

In this test, the following diffusion problem was solved with the stepwise initial distribution of passive scalar concentration  $Y$ :

$$\begin{aligned} \frac{\partial Y}{\partial t} - D^2 \frac{\partial^2 Y}{\partial x^2} &= 0 \\ Y(x,0) &= \begin{cases} 1, & x \in (0, 0.5l) \\ 0, & x \in (0.5l, l) \end{cases} \\ Y(0,t) &= Y(l,t) = 0 \end{aligned} \quad (11)$$

where  $D = \text{const } k^2 / \varepsilon$  is the turbulent diffusion coefficient, and  $l$  is the domain size. The analytical solution of this problem is well known:

$$Y(x,t) = 0.5 + \sum_{n=1}^{\infty} \frac{2}{\pi n} \cos\left(\frac{\pi x}{l}\right) \sin\left(\frac{\pi n}{2}\right) \exp\left(-\frac{(\pi n a)^2}{l^2} t\right) \quad (12)$$

Figure 2 compares the MC and analytical solutions. The error of the numerical solution depends on the number of Lagrangian elements and for the case of Fig. 2 does not exceed 3%.

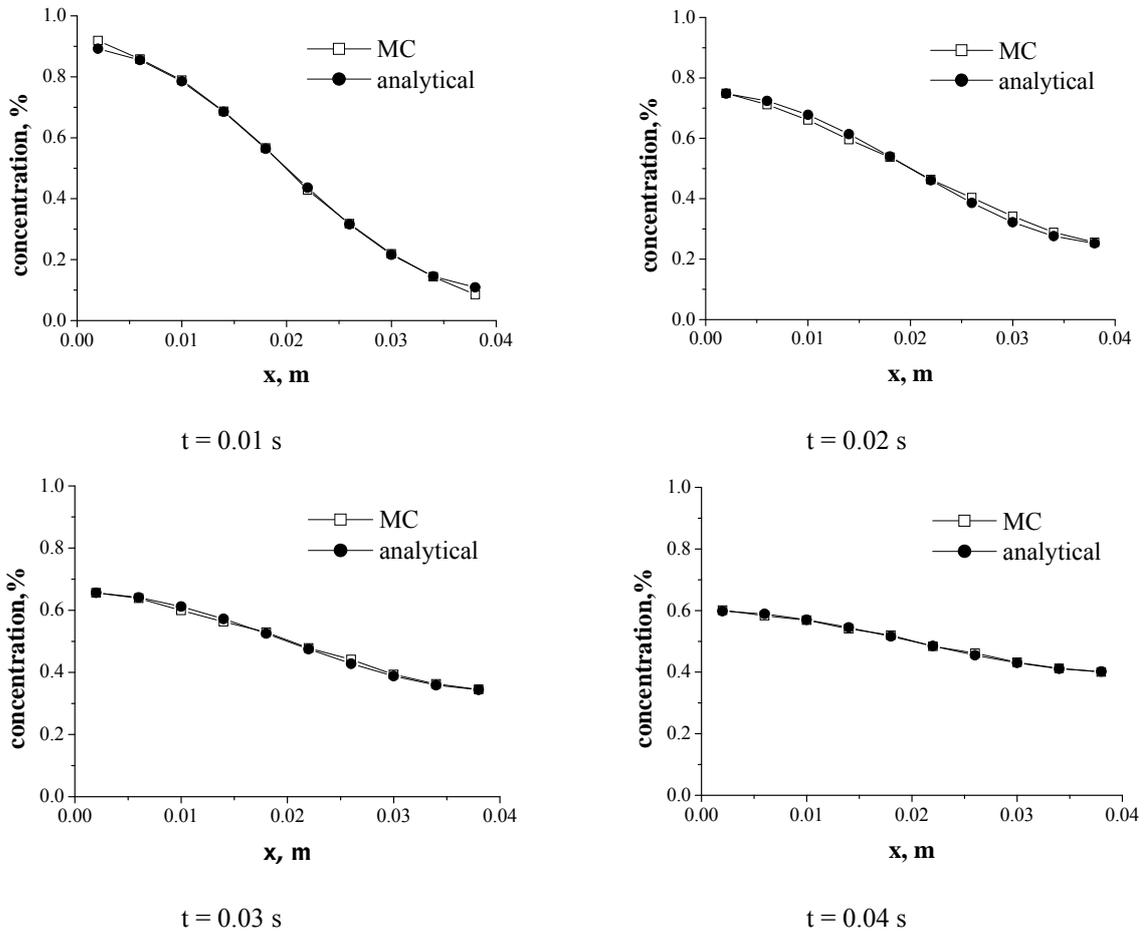
### TEST 2: 2D DIFFUSION IN TURBULENT FLOW

Figure 3 shows the schematic of the turbulent flow reactor with two turbulent inlet streams: one with the mean velocity 1 m/s and the other with the mean velocity 10 m/s. The concentration of passive scalar in the first stream was zero ( $Y = 0$ ), whereas in the second stream  $Y = 1$ . The shear between the streams resulted in turbulent mixing.

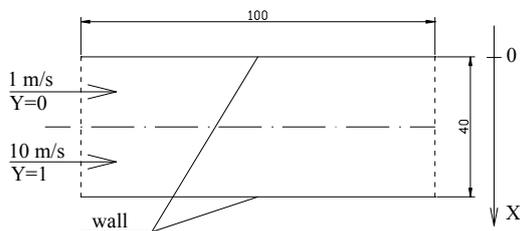
First, steady-state RANS solution of the problem was obtained using a finite volume code. Then the steady-state fields of pressure gradient, turbulent kinetic energy  $k$  and its dissipation  $\varepsilon$  were used for obtaining the steady-state MC solution. Figure 4 compares the results of RANS and RANS-MC calculations in terms of the transversal profile of passive scalar concentration at the reactor outlet.

Figure 5 shows the calculated trajectories of Lagrangian elements in the flow reactor. Initially, all the elements were located in the same computational cell at the reactor inlet, i.e. represented possible

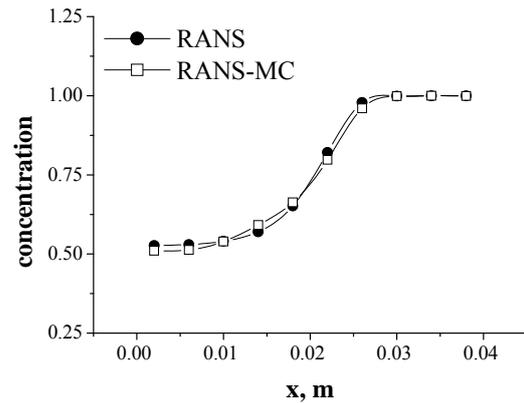
turbulent flow realizations at fixed mean values of flow variables (only the trajectories of 6 out of 40 elements are shown in Fig. 5). It is seen that element trajectories differ considerably from the mean “streamline” shown by the thick curve. This means that the RANS-



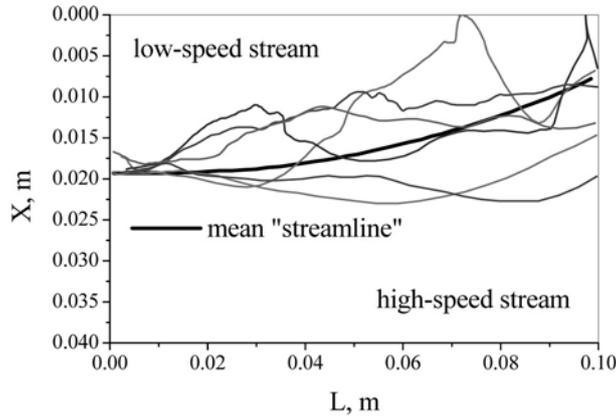
**Fig. 2.** Comparison of MC and analytical solutions of simple 1D diffusion problem



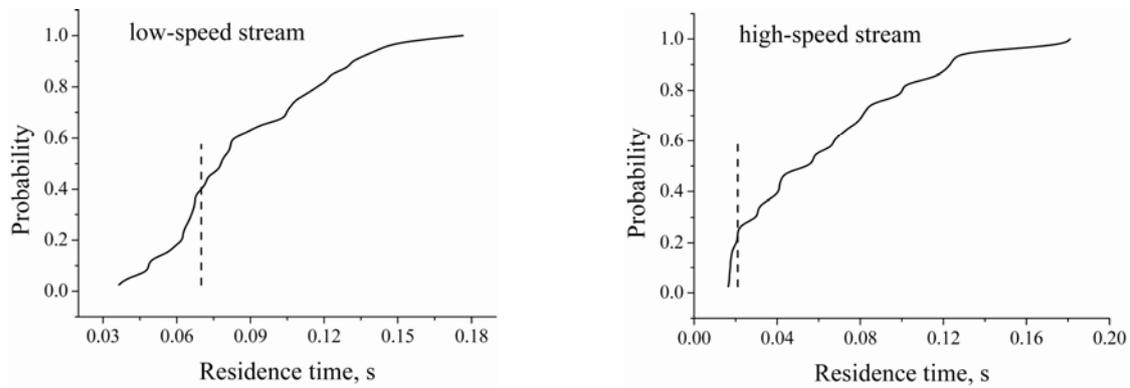
**Fig. 3.** Schematic of the turbulent flow reactor with two inlet streams. Dimensions are in millimeters



**Fig. 4.** Comparison of predicted RANS and RANS-MC profiles of passive scalar concentration at reactor outlet



**Fig. 5.** Calculated trajectories of 6 of 40 Lagrangian elements initially located in the same computational cell at reactor inlet



**Fig. 6.** Residence time distribution in low-speed (left) and high-speed (right) streams in the reactor of Fig. 3

MC method provides residence time distribution which differs from that predicted by the RANS method. As a matter of fact, the curves in Fig. 6 show the calculated integral residence time distributions for low-speed (left) and high-speed (right) streams in the reactor of Fig. 3. Vertical bars correspond to the residence time predicted by the RANS method.

### TEST 3: NITROGEN OXIDE FORMATION IN 2D TURBULENT FLOW REACTOR

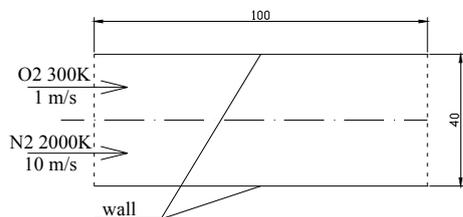
Figure 7 shows the schematic of the reactor used for testing the RANS-MC method for turbulent reacting flow. Similar to the flow configuration in Fig. 3, there are two streams at the reactor inlet: cold (300 K) oxygen stream with the velocity of 1 m/s and hot (2000 K) nitrogen stream with the velocity of 10 m/s. Oxygen and nitrogen are allowed to react according to the simplified kinetic mechanism presented in Table 1.

**Table 1.** Mechanism of thermal NO formation

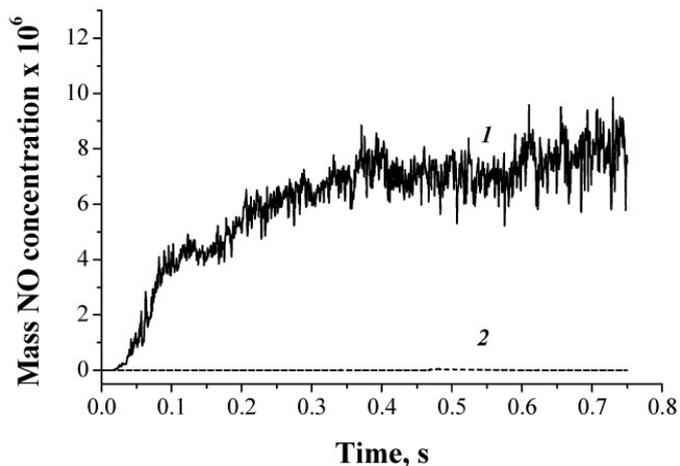
Reaction	Heat effect kcal/mol	A Mol, l, s	$\alpha$	$E_a$ kcal/mol
$O_2 + N_2 \rightarrow NO + NO$	-43.0	$1.7 \cdot 10^{17}$	-0.5	136.0
$NO + NO \rightarrow O_2 + N_2$	43.0	$4.1 \cdot 10^{15}$	-0.5	93.3

Reaction rate constant is determined as  $k=AT^{\alpha} \cdot \exp(-E/RT)$

Figure 8 shows the results of calculations in terms of the overall mass concentration of NO at the reactor outlet. Curve 1 corresponds to the RANS-MC calculations applying the Lagrangian element temperature and species concentrations in the reaction rate of NO formation whereas curve 2 corresponds to the RANS-MC calculations based on the local mean temperature, other conditions being equal. When the computation time exceeds 3-4 maximal residence times in the flow reactor, the



**Fig. 7.** Schematic of flow reactor with cold oxygen and hot nitrogen streams. Dimensions are in millimeters



**Fig. 8.** Predicted overall mass NO concentrations at the flow reactor outlet. 1 – RANS-MC method applying the Lagrangian element temperature and species concentrations in the reaction rate of NO formation, 2 - RANS-MC method applying the local mean temperature in the reaction rate of NO formation

levels of NO at the reactor outlet attain the quasi-steady values. The predicted quasi-steady NO-emission levels in Fig. 8 differ by 3 – 4 orders of magnitude. This example clearly demonstrates the importance of turbulence – chemistry interaction in the flow reactor.

## Conclusion

This paper presents the RANS-MC methodology for simulating industrial turbulent reactive flows. The methodology provides extensive statistics of the turbulent reactive flow, including distributions of residence times, and allows for modeling turbulence – chemistry interaction. The computational code has been developed and tested against relatively simple diffusion and reaction-diffusion problems. The analysis of the computational example indicates that the account for turbulence – chemistry interaction and distributed residence time can lead to considerable differences in the predicted mean reaction rates as compared to standard RANS-based calculations.

## Acknowledgements

This work was supported by the Russian Foundation for Basic Research (grant 08-08-00068).

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