Investigation on the course of complex processes carried out in vortex impinging jet reactors

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The paper presents an application of CFD simulations to predict a course of complex processes carried out in jet reactors. In this work the simulations results were verified with PIV and PLIF techniques and validated by comparing model predictions with experimental data for fast parallel chemical test reactions. Experimental results of $BaSO_4$ precipitation in jet reactors are also discussed.
Keywords: jet reactors, mixing, chemical reaction, precipitation, numerical simulation

Introduction

Obtaining chemical product with desired characteristics is often achieved by utilizing several complex chemical processes and physical operations. Only few of those give valuable products, while the other can lead to side products which could be harmful and difficult to utilization. Therefore, process should be carried out under expected conditions. The course of the chemical reactions is affected not only by the reaction kinetics, but also by method of contacting the reactants or mixing intensity. For that reason quality of the products produced can be affected by varying both the chemical parameters (concentration, temperature, pH, etc.), but also by the physical parameters (e.g. dosing rate, reactors type, mixing intensity, etc.). Proper selection of these parameters often allows not only achieving better efficiency of the process equipment, but also reducing the amount of side products (including those harmful to the environment) and decreasing the number of necessary manufacturing steps.

One of the ways of improving process parameters in reactor under consideration is to take advantage of the phenomenon of turbulent flow. Such flows are widely observed in nature, industry and laboratory, however it is very difficult to study them since they take place at very wide range of characteristic length scales [1]. Because of this fact, development of accurate numerical simulation methods for turbulent reacting liquid flows is necessary for effective design of chemical reactors. To predict, control and optimize mixing effects on processes involving chemical reactions one needs to apply so called micromixing models and closure methods, usually in combination with computational fluid dynamics (CFD) [2].

In recent years there has been an increase in interests of practical application of this type of reactors in particular in obtaining nanoparticles in pharmaceutical [3, 4] and catalytic processes [5]. Such complex chemical processes are difficult and expensive to study experimentally, so nowadays CFD simulations are employed very often for that purpose. It has started to be possible since computers computational power increased significantly in last 20 years. This is true especially for models such as large eddy simulations (LES) which require much more computational resources in comparison with widely used simpler Reynolds-Averaged Navier Stokes (RANS) models. LES models are considered as an attractive solution, because they can be applied in wide range of Reynolds and Schmidt numbers, which often occurs in engineering problems. LES are based on assumption that turbulent kinetic energy is dominated by large scale motions, which are dependent on systems geometry, while kinetic energy dissipation takes place within small scales range in energy spectrum, where effects of systems geometry are less significant.

Taking into account more and more frequent use of numerical simulations in engineering applications, investigations of velocity, concentration and other quantities distributions inside reactors are very important to validate CFD models, especially time-dependent models like LES.

Experimental system

In this work authors have investigated the course of chemical processes in impinging jet reactors (vortex T-mixers). Impinging jets are often used in process industry due to possible almost instantaneous mixing of contacting liquids. This happens due to formation of high values of the rate

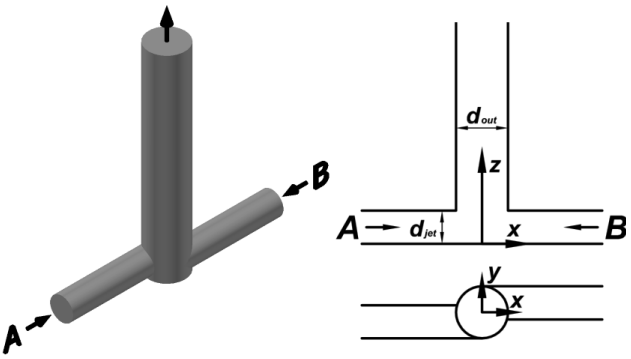

Figure 1. Geometry of vortex T-mixer

of energy dissipation in impingement zone of the inlet streams. Each fluids element passes through this region without a possibility of bypassing. The region of high energy dissipation is formulated since kinetic energy of inlet streams is being converted into highly turbulent flow caused by collisions and redirections of the flows in a very small space. In the scientific literature there are many interesting geometric solutions of jet reactors such as: Confined Impinging Jets [5] or Multi-Inlet Vortex Mixer [6].

Schematics of reactors considered in this study are shown in Figure 1. Both T-mixers are equipped with inlet pipes arranged tangentially to the outlet pipe. First studied reactor has inlet pipes diameter d_{jet} = 7 mm and second one d_{jet} = 4,6 mm. Outlet diameter is the same for both systems d_{out} = 11 mm. Lengths of inlet and outlet pipes were equal to 100 mm and 300 mm respectively.

Experiments

Local, instantaneous values of fluid velocity and passive tracer concentration were measured using two techniques: particle image velocimetry (PIV) and planar laser induced fluorescence (PLIF). Double-cavity Nd-YAG 532 nm laser with energy equal to 50 mJ per pulse was used. Laser beam was transformed to a collimated planar laser sheet of thickness about 150 µm. The laser sheet crossed the experimental system vertically through the axis of the outlet pipe. Glass spherical particles of average size equal to 10 µm were used as seeding particles for PIV measurements. Rhodamine B was used as a fluorescent tracer for PLIF measurements and its concentration in the inlet solution was equal to 0.2 g m^{-3}. Spatial resolution of PLIF measurements resulted from the resolution of digital images and the thickness of the laser sheet [7] and in our case it was 10x10x 150 μm^3. Spatial resolution of PIV measurements corresponded to approximately 150x150x150 μm^3.

Next, influence of mixing on two particular processes was investigated. The first one is the process of reactive flow involving two parallel chemical reactions which allows determining the quality and rate of mixing in the system. Following reactions were considered: neutralization of sodium hydroxide with hydrochloric acid and alkaline hydrolysis of ethyl chloroacetate:

$$NaOH + HCl \xrightarrow{k_1} NaCl + H_2O$$

$$NaOH + CH_2ClCOOC_2H_5 \xrightarrow{k_2} CH_2ClCOONa + C_2H_5OH$$

First reaction is infinitely fast compared to mixing and is limited by mixing, so final selectivity of second reaction corresponds to measure of mixing in the system (in this case lower selectivity means better mixing intensity).

The second process is precipitation of the sparingly soluble test material (barium sulphate) from two aqueous ionic solutions: sodium sulphate and barium chloride:

$$BaCl_2 + Na_2SO_4 \rightarrow BaSO_4 \downarrow + 2NaCl$$

$$Ba^{2+} + SO_4^{2-} \rightarrow BaSO_4 \downarrow$$

Analytical grade chemicals and doubly distilled water were used to prepare reactant solutions for both test processes. In case of precipitation, reactor was cleaned before each experiment with sulphuric acid to remove the barium sulphate from the reactor walls. Products samples were collected at the reactor outlet. Final selectivity of parallel chemical reactions was measured using HPLC technique. The crystal size distribution was measured directly after the experiment using a laser diffraction particle size analyser (Beckman Coulter LS 13320). Filtered and dried particles of barium sulphate were observed and photographed with the use of scanning microscope.

Simulations

Simulations of hydrodynamics were done using CFD code Ansys Fluent 14. The numerical grid consisted of about 800 000 hexahedral computational cells for both reactors. Such number of cells can be considered as satisfactory for LES calculations, since allows to resolve near-wall regions of the numerical domain [8]. Computations were regarded as satisfactory converged when the total normalized residuals were smaller than 10^{-6}. Two turbulence models were used: the realizable k-ε and LES models. Simulations in bigger vortex T-mixer (d_{jet} = 7 mm) were performed using k-ε and LES, while in second reactor only with k-ε model. In LES computations a dynamic stress model was employed to reflect effects of the small scale on large ones. The calculations were performed for several values of Reynolds numbers in range of 300 ≤ Re_{jet} ≤ 4000, where:

$$Re_{jet} = \frac{u_{jet} d_{jet} \rho}{\mu} \qquad (1)$$

where u_{jet} is mean velocity at the inlet pipe.

In case of LES modelling, closure problem occurs resulting from concentration fluctuations below numerical scale. Subgrid tracer mixing was modelled using concept of constant subgrid Schmidt number (Sc_{sgs} = 0.4). Mixture fraction variance in subgrid scale was determined with the assumption of fractal nature of turbulence, which means that the small-scale statistics can be inferred from larger-

scale statistics [9]. The subgrid modelling of fast parallel chemical reactions was based on a beta distribution of the mixture fraction in combination with a conditional moment closure based on linear interpolation of local instantaneous reactant concentration values. More details of numerical procedure and models were presenter in [10].

The RANS model was completed with the non-equilibrium multiple-time-diffusion model – turbulent mixer model (TMM) [11]. The TMM enables prediction of the distribution of the concentration variance, as well as its inertial-convective, viscous-convective and viscous-diffusive components within energy spectrum.

Results and discussion

Figures 2 and 3 present exemplary distributions of measured and predicted mean velocity and mixture fraction in the vortex T-mixer (inlet pipe diameter 7 mm) for Re_{jet} = 1000 and Re_{jet} = 2000. The most important region in jet reactors is region of impingement. One can see that agreement between experimental and simulation results of velocity values in this region is better for large eddy simulation than for the k-ε model especially, for lower value of Reynolds number. For high values of Re_{jet} both models predict good results, which are in good agreement with experiments. It results from theory of the k-ε model, that it was developed for fully turbulent flow. In case of tracer concentration distribution, the differences between models and experiments are smaller, which proves correctness of used

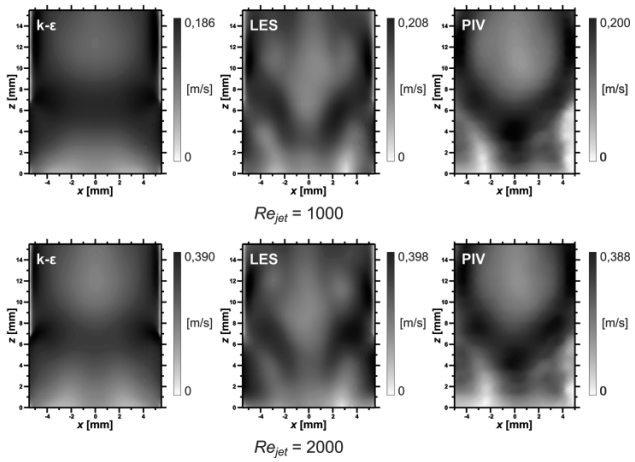

Figure 2. Distributions of the mean velocity in vortex T-mixer (inlet pipe diameter 7 mm)

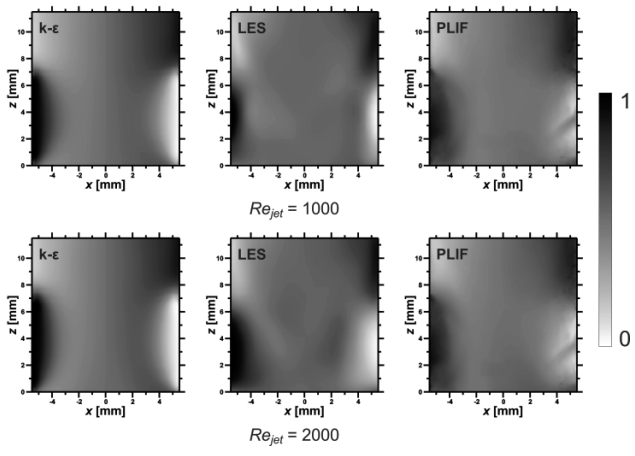

Figure 3. Distributions of the mean mixture fraction in vortex T-mixer (inlet pipe diameter 7 mm)

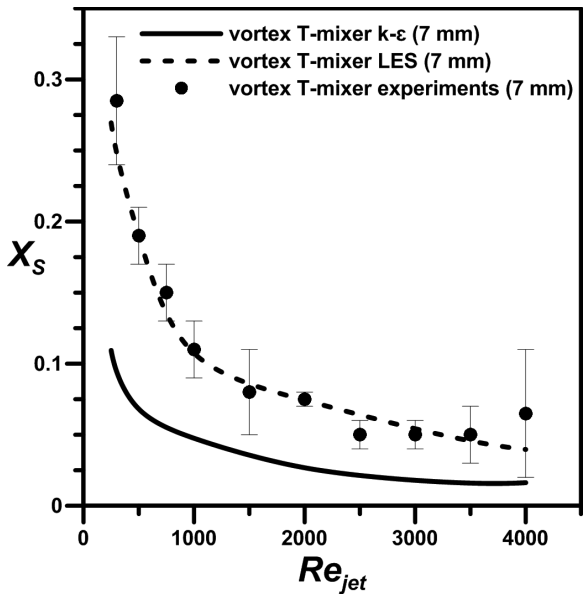

Figure 4. Effects of Reynolds number on final selectivity in vortex T-mixer (inlet pipe diameter 7 mm), $c_{A0} = c_{B0} = c_{C0}$ = 100 mol m^{-3}

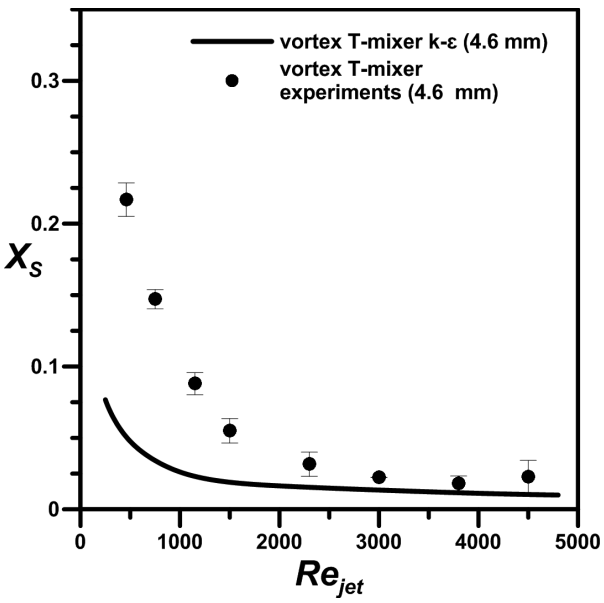

Figure 5. Effects of Reynolds number on final selectivity in vortex T-mixer (inlet pipe diameter 4.6 mm), $c_{A0} = c_{B0} = c_{C0}$ = 100 mol m^{-3}

numerical mesh and allows modelling of complex chemical processes in studied systems.

Effects of mixing on final selectivity of parallel chemical reactions are shown in Figure 4 and 5. Final selectivity has been defined as:

$$X_S = \frac{\langle c_{C0} \rangle - \langle c_C \rangle}{\langle c_{A0} \rangle} \qquad (2)$$

where c_{C0}, c_C - chloroacetate inlet and outlet concentrations accordingly, c_{A0} - sodium hydroxide inlet concentration.

Results show that increase of Reynolds number provides improvement in mixing, which means decrease in amount of side-product of the reaction. Also very good agreement of LES predictions with experimental data can be observed in whole range of tested values of Reynolds numbers, as can be expected basing on literature [12].

Predictions of k-ε model underestimates experimental data in whole range of Reynolds number for both reactors, which was already observed in literature [13]. In case of mixer with smaller inlet pipes agreement is better, because of higher linear velocities of colliding streams and resulting higher energy dissipation rates in impingement zone, which corresponds to higher intensity of turbulence.

Results of both models indicates that LES can be successfully applied to solve engineering problems, especially for lower Reynolds numbers and despite high computational costs, however k-ε can also be used in considered systems for fully turbulent flow.

Figure 6 shows effect of Reynolds number on mean precipitated $BaSO_4$ particle diameter size d_{43}. One can see that in this case we obtained almost the same mean diameter for both reactors. One can see differences only for smaller Reynolds number, less than $Re_{jet} = 1000$. Less mixing intensity caused large mean diameters of crystals.

Effects of inlet concentration for $Re_{jet} = 500$ on $BaSO_4$ particles diameter size are shown in figure 7. One can see that with increasing solutions concentration, we obtain smaller particles. Differences between results for both studied reactors are not significant, however they increase with increasing Reynolds number.

Photographed particles are shown in figures 8 and 9. It can be observed that for low concentration values, aggregation and breakage of particles are negligible. For higher concentrations both phenomena are taking place, therefore

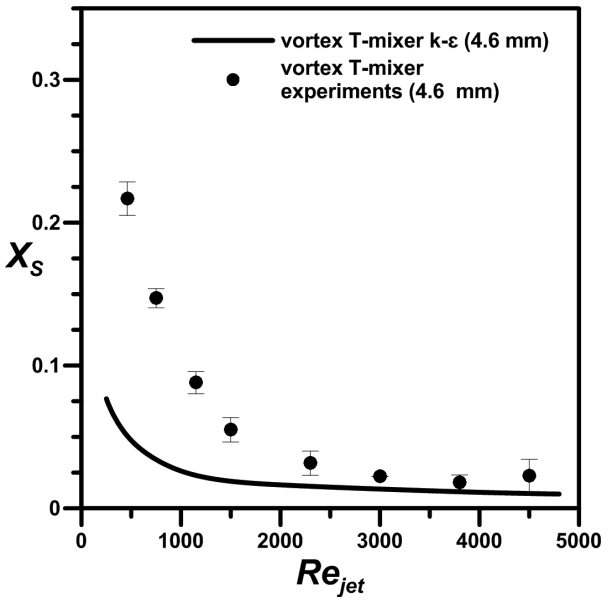

Figure 6. Effects of Reynolds number on mean precipitated $BaSO_4$ particle diameter size, experimental data, $c_{A0} = c_{B0} = 50$ mol m^{-3}

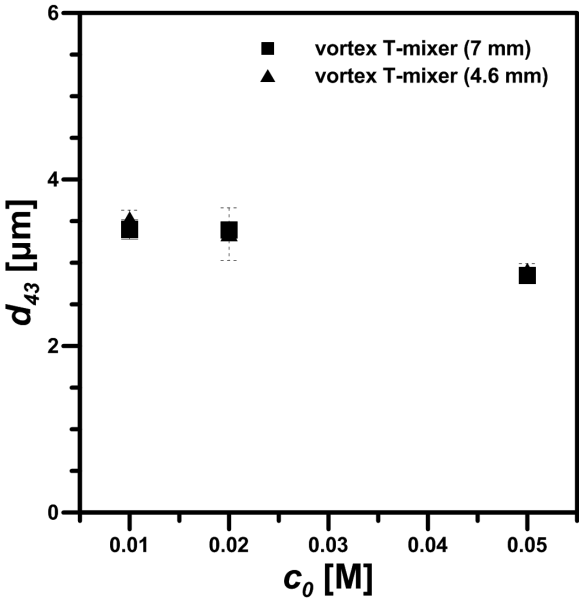

Figure 7. Effects of inlet concentration on mean precipitated $BaSO_4$ particle diameter size, experimental data, $c_0 = c_{A0} = c_{B0}$, $Re_{jet} = 500$

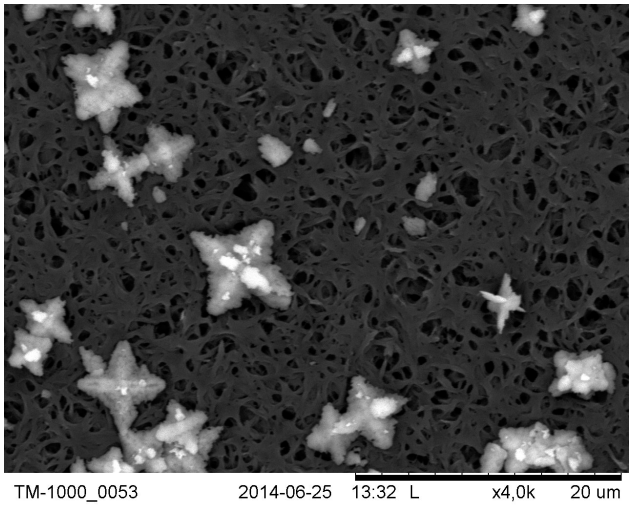

Figure 8. Image of obtained $BaSO_4$ particles in vortex T-mixer (inlet pipe diameter 4.6 mm), $c_{A0} = c_{B0} = 10$ mol m^{-3}, $Re_{jet} = 460$

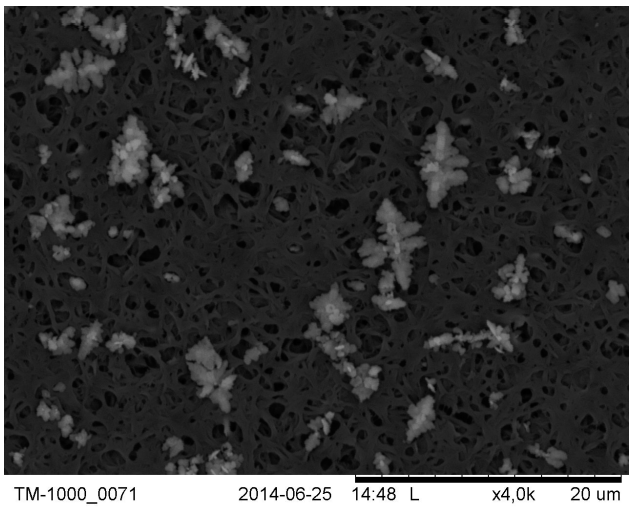

Figure 9. Image of obtained $BaSO_4$ particles in vortex T-mixer (inlet pipe diameter 4.6 mm), $c_{A0} = c_{B0} = 50$ mol m^{-3}, $Re_{jet} = 2000$

in CFD simulations one should properly model them in order to obtain satisfactory results, remembering that it can significantly increase computational time of solving the problem.

Conclusions

The LES and k-ε models were applied to simulate complex chemical processes (mixing and parallel chemical reactions) in two vortex T-mixers. Also analysis of experimental data of precipitation process has been performed. Amongst others PIV and PLIF techniques were applied to validate numerical results.

Presented results show that LES modelling gives better results especially for low Reynolds numbers. This illustrates importance of effect of large scale inhomogeneities that are predicted by LES and neglected by RANS. The second explanation for these differences results from theory of the k-ε model that was developed for fully turbulent flow. For higher Re_{jet} both models predict similar results.

Precipitation results indicates that analysis of mixing process in jet reactors is not sufficient to determine and predict reactors behaviour in case of particles production. It has been showed that detailed CFD simulations with kinetics of crystal growth, nucleation and aggregation applied are required in this case.

Collected experimental data can be used in next works for validation of improved and new CFD models of the complex processes, such as mixing or precipitation.

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