



Application of the Monte Carlo technique in simulation of flow and modeling the residence time distribution in a continuous two impinging liquid–liquid streams contactor

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ABSTRACT

In the present study, a stochastic model for the residence time distribution (RTD) in a coaxial counter current liquid–liquid impinging streams system has been developed.

Simulations of droplets movements in the impinging spray system determine the residence time distribution. Droplets dynamics has been formulated from the Boltzmann equation using direct simulation Monte Carlo (DSMC) method. The data predicted from the model has been correlated with the experimental results obtained from a coaxial counter current two impinging liquid–liquid streams apparatus. The degree of agreement was within 85%.

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1. Introduction

One of the important operations in chemical engineering is the mass transfer between immiscible phases. In an impinging streams apparatus a unique flow behavior is utilized by which the transfer processes in heterogeneous systems are intensified [1]. In such systems two feed streams, flowing parallel or counter currently collides with each other at a zone in which the two streams impinge.

Impinging streams systems, first proposed by Elperin [2], have been applied as some suitable devices for enhancing mass and heat transfer processes in flowing gas–liquid, gas–solid and liquid–liquid emulsions. In such apparatus, two droplet-laden gaseous jets flowing in opposite direction are allowed to impinge. Some of the multiphase reactions carried out in two impinging streams reactors include two-phase mono-nitration of toluene [3], chemical absorption of CO₂ gas in both sodium hydroxide [4] and monoethanolamine solutions [1] and isomerization of glucose to fructose by an immobilized enzyme [5]. Impinging streams systems have been successfully applied to other chemical processes such as dissolution and mixing as well as mass and heat transfer operations [6–9].

In the present study a stochastic model for the residence time distribution (RTD) of the aqueous phase within a coaxial counter current two liquid–liquid impinging streams contactor (TISC) has been carried out. In addition a comparison has been made between the experimentally determined RTD data and those predicted from the model.

2. Contactor system

The experimental apparatus used in the present study is shown in Fig. 1. The contactor consists of a cylindrical vessel made of Pyrex glass, length 60 cm and internal diameter 14 cm. The contactor is equipped with two circular plates made of “Teflon” placed at the two ends of the contactor. Thus the length of the contact compartment can be varied by moving the plates away from or towards each other.

The two immiscible liquids (toluene and water) were kept in separate glass containers and fed to the contactor via four identical feed nozzles made of glass, situated at the two ends of the vessel. Liquids were transported to the nozzles using metering pumps. The liquid streams were sprayed into the contactor by applying pressurized nitrogen via special ducts, placed behind the feed nozzles. In Fig. 2, the diagram of a feed nozzle is shown. As it may be observed from this figure, there is no mixing of fluids within the nozzle. The mixing is occurred only at the exit of the system. In each end of the

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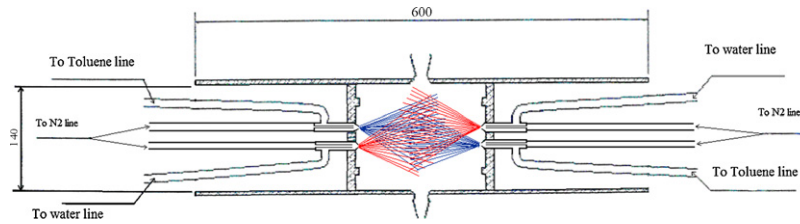


Fig. 1. Contactor dimensions (figures are in mm).

contactor two identical feed nozzles were installed spraying two different liquids towards the impingement zone.

Operating conditions are shown in Table 1.

In order to determine the suitable positions and angles for the feed nozzles at which stable jets of liquids would be established, a number of experiments were performed.

This experimental setup allows consideration of the effects of changing certain pertinent parameters of the system on the residence time distribution of materials and hence on the extent of the heterogeneous liquid–liquid mixing and reaction.

3. Results and discussion

3.1. Measurement of the residence time distribution of aqueous phase

To determine the residence time distribution of the aqueous phase within the contactor, the following experiment was performed. Toluene and distilled water were fed to the contactor via spray nozzles. At a time, a change from the water stream to one of water containing a mineral salt (potassium dichromate) with known concentration and colour intensity (C_0) was rapidly performed (step input). Samples at the outlet of the contactor were collected, using a circular vessel divided into 24 segments with equal volumes, placed under the exit port and rotated at a pre-

set speed applying an electric motor. Successive samples were obtained at equal time intervals by this method. Change in the rotating speed altered the time intervals between the sample collections. The content of each segment was transferred to a separating funnel. The aqueous phase was separated from the organic layer and the concentration (colour intensity) of the former was measured (C), using a UV spectrophotometer. From a plot of C/C_0 versus time, the RTD data were determined.

3.2. Developing a stochastic model for the residence time distribution of the aqueous phase in TISC

3.2.1. Gas flow simulation

Gas flow pattern is required to simulate the droplet movement. By the velocity pattern, the drag force which affects the droplet velocity may be estimated.

Complete solution of Navier–Stokes equation for such a system can provide the necessary data, for this pressure, although the approximate solution of this equation (analytical relations) for free jet may be also used.

In the present study the Navier–Stokes equation has been solved, using the SIMPLE method and a mesh system shown in Fig. 3.

The gas stream is assumed to be an incompressible flow (this assumption is valid for the fluid flows having Mach number lower than 0.3).

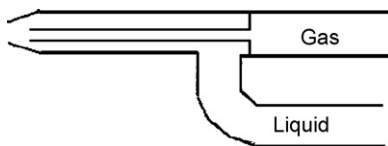


Fig. 2. Diagram of a feed nozzle.

Table 1
Operating conditions of the impinging streams contactor

Temperature ($^{\circ}\text{C}$)	20.1
Distance between the two feed nozzles (cm)	25
Flow rate of nitrogen (cm^3/min)	1300
Toluene flow rate (cm^3/min)	320
Water flow rate (cm^3/min)	325

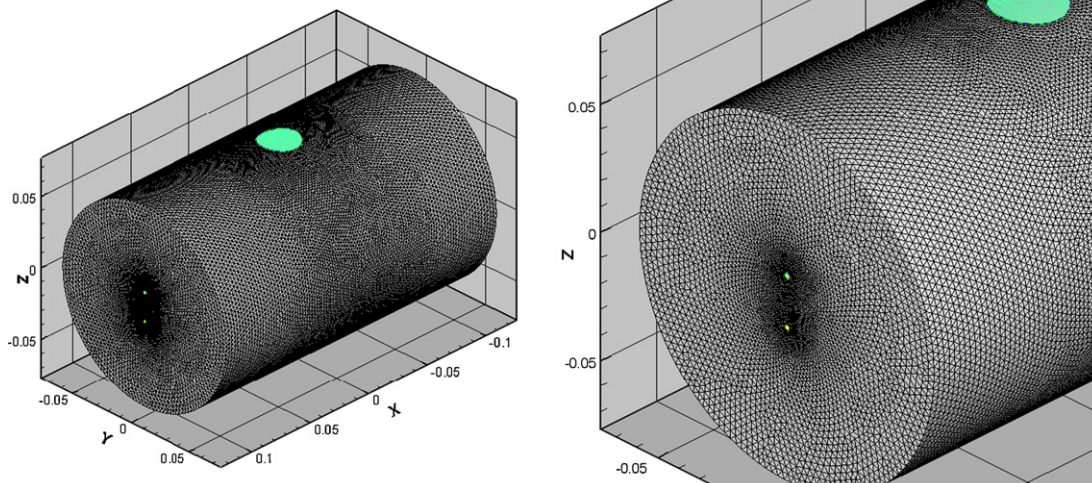


Fig. 3. Pyramid mesh for the contactor.

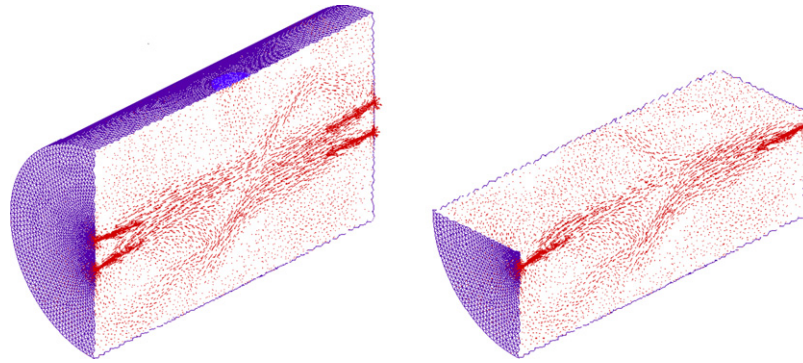


Fig. 4. Velocity vectors and streamlines in the two impinging streams contactor.

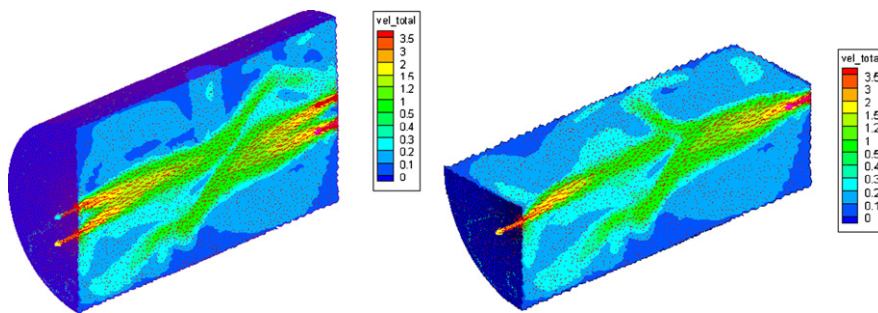


Fig. 5. Gas velocity vectors in the contactor with 1 m/s velocity at the entrance nozzle.

Calculation of pressure contours is the main problem in this kind of fluid flow and the SIMPLE method seems to be a way by which the pressure profile under these conditions may be estimated [10]. The results are shown in Figs. 4 and 5. In Fig. 5 the gas velocity vectors in three dimensions have been presented from which, the overall velocity has been determined using the relation, $V_{overall} = \sqrt{v_x^2 + v_y^2 + v_z^2}$.

3.2.2. Droplet simulation

An analogy between the droplet collisions in emulsions and the molecular collisions, described in the kinetic theory of gases, enables the application of the Boltzmann equation to the droplets, as first suggested by Pai [11].

In this work, the direct simulation Monte Carlo (DSMC) method, first proposed by Bird [12] for solving the Boltzmann equation in molecular gas dynamics, is used for modeling the droplet interactions in dense sprays.

3.3. Mathematical formulations

The Boltzmann equation may be written as

$$\frac{\partial}{\partial t}(nf) + V \frac{\partial}{\partial r}(nf) + F \frac{\partial}{\partial v}(nf) = \int_{-\infty}^{+\infty} \int_0^{4\pi} n^2 (f^* f_1^* - ff_1) V_r \sigma d\Omega dv_1 \quad (1)$$

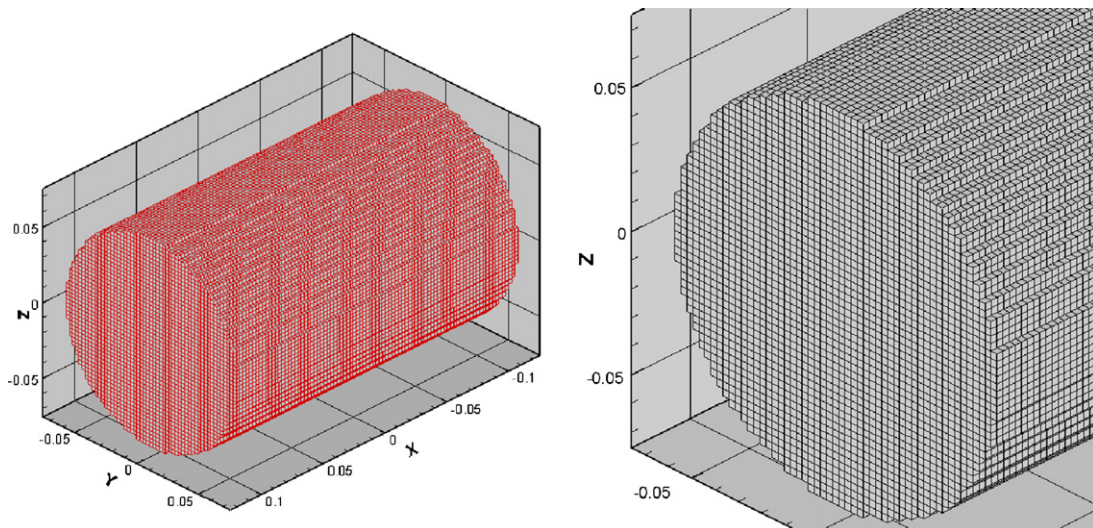
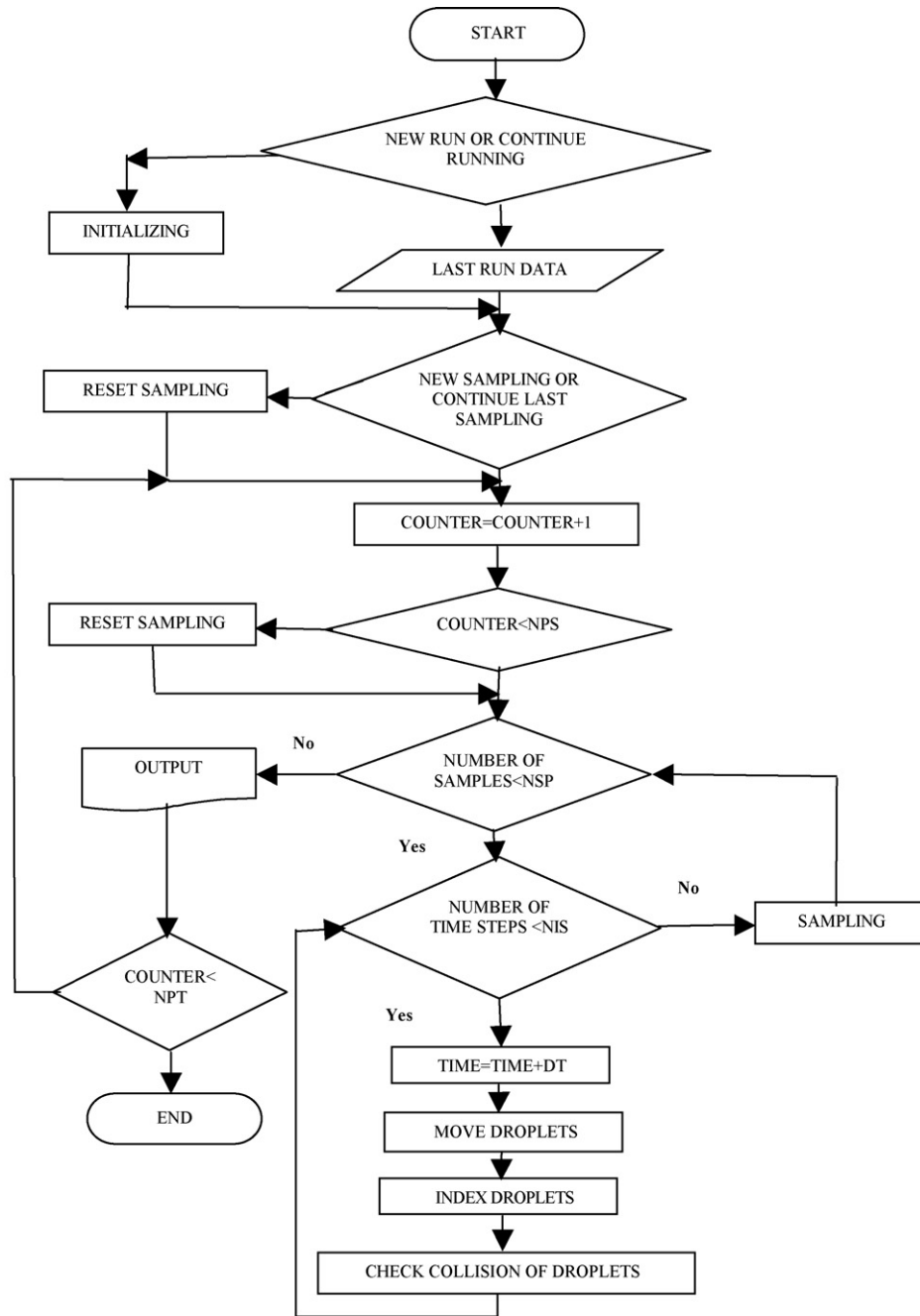


Fig. 6. Contactor mesh for DSMC simulation.



NPS is the estimated number of samples to steady flow.

NIS is the number of time steps between samples.

NSP is the number of samples between restart and output file updates.

NPT is the number of file updates to stop.

Fig. 7. Algorithm for DSMC.

In this equation, n is the number density of droplets, f is the velocity probability distribution function of droplet of class having the velocity V , f_1 is the velocity distribution function of the droplet of class having the velocity V_1 , V_r is the velocity of a test droplet in the class of droplets having the velocity V_1 , f^* is the post-collision velocity probability distribution function and f_1^* is the post-collision velocity probability distribution function of the droplet of class hav-

ing the velocity V_1 , F is the external force per unit mass and Ω is the angle in the spherical coordinates.

The key ideas of the DSMC method are: (a) the uncoupling of droplet motions and collisions during a time step Δt_m , i.e. the use of the operator-splitting technique, (b) the simulation of droplet collisions by disregarding droplet position coordinates within spatial coils and (c) the simulation of fewer droplets than those present in

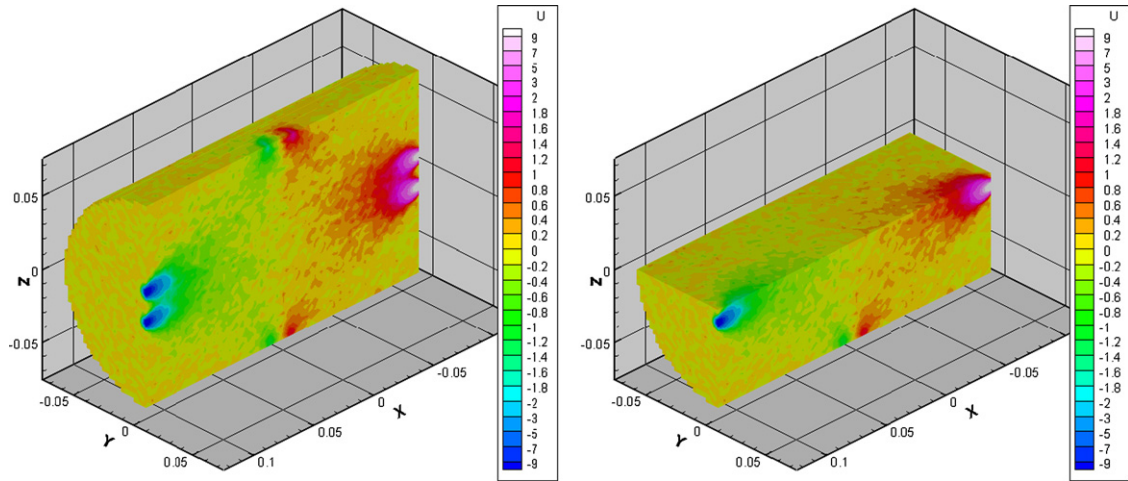


Fig. 8. Droplets velocities in X direction.

the real flow, while normalizing the collision cross-section so that the collisions rate is not changed. Assumption (a) is valid when Δt_m is smaller than the time between collisions, and larger than that of collision duration, and assumption (b) is valid provided that the cell is so small that the spatial variation of flow variables in the cell is negligible. Assumption (c) may not be necessary for dilute emulsion flow, due to relatively small droplet number densities; in the present work, however, due to the formation of large number of fragments, the implementation of this procedure is required.

Under the assumptions listed above the DSMC method for the solution of Boltzmann kinetic equation, describing the flow of gas and droplets emulsions can be formulated as follows. The flow system is divided into equal-volume cells. Simulated droplets are distributed in the system, with their positions, sizes and velocities sampled from the initial distribution function. When a stationary kinetic equation is solved, the initial distribution function is chosen arbitrarily. The droplets population is normalized such that each K_f droplets of identical size in the real system is substituted by a single droplet in the simulation, having the same diameter as these droplets. The collision cross-section for each such simulated droplet is accordingly increased by the factor K_f so as to preserve the true collision rate. Provided that the droplet distribution function at time $(n - 1)\Delta t_m$ is determined, the distribution function at time $n\Delta t_m$ may be calculated as follows. Droplets are allowed to move in the system, without colliding with each other, for a time interval

Δt_m , with each droplet's subsequent position, velocity and diameter are determined from the droplet equation of motion. Within the time interval Δt_m , a droplet may encounter a boundary either an open boundary, through which it leaves the system or a wall, onto which it sticks. Following the collisionless flow, droplets are allowed to collide with each other. The droplet population is discredited by location and size such that the number of droplets of type k in cell m , $N_{m,k}$ having a volume V_m , is given as

$$N_{m,k}(t) = \int_{V_m} dr \int_0^\infty dv \int_{\delta_k^s}^{\delta_k^l} d\delta f(r, v, \delta, t) \quad (2)$$

where δ_k^s and δ_k^l are the lower and upper diameter limits, respectively, for droplets of type k . The total number of droplets in cell m is

$$N_m = \sum_{k=1}^s N_{m,k} \quad (3)$$

In the stochastic model it has been assumed that collision durations are negligible, so that droplet motion can be described as a free motion, disrupted instantaneously by collisions. For high collision velocity, which is characteristic of impinging streams systems, shorter collision durations may be expected.

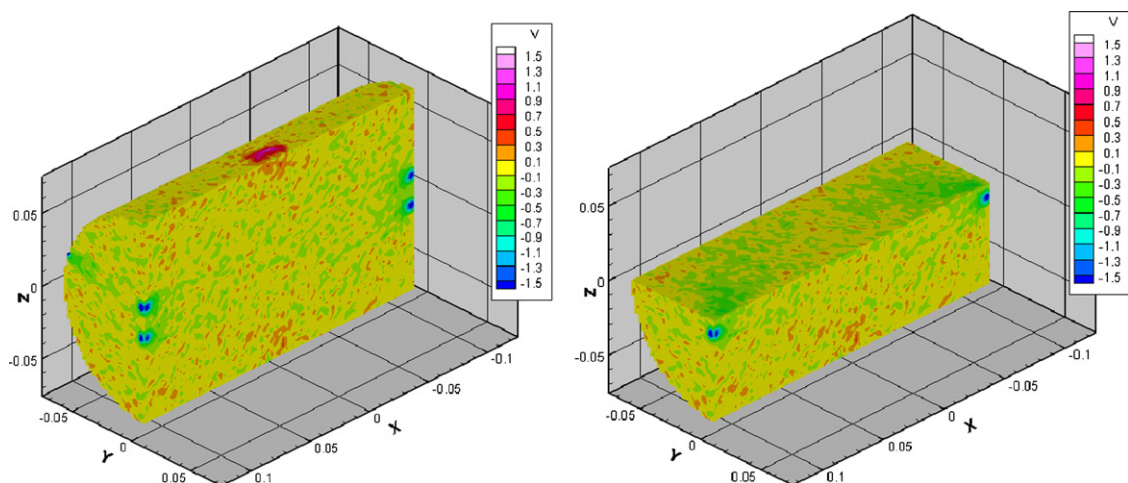


Fig. 9. Droplets velocities in Y direction.

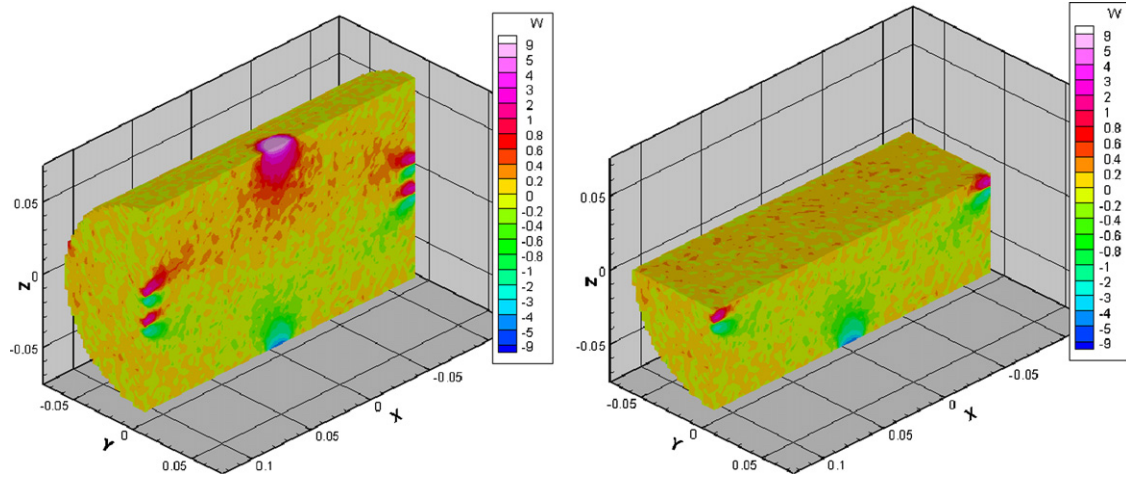


Fig. 10. Droplets velocities in Z direction.

Another assumption made was that droplets are spherical and the aerodynamic breakup due to gas shear during free droplet motion is negligible. Borisov et al. [13] investigated experimentally the breakup of single droplets moving in a gas flow. Their results show that droplet breakup does not occur when Weber number < 6.

When only fluid drag acts upon droplets during Δt_m , droplet trajectory is calculated by integrating the following ordinary differential equation:

$$\frac{d^2 r}{dt^2} = 0.75 C_D [\rho_g (\rho_p \delta)] |U - v| (U - v) + g \quad (4)$$

where, U is the gas velocity at position r that is determined according to the procedure presented in gas flow simulation section, g is the gravity acceleration and C_D is the gas drag coefficient for liquid droplet flowing in a gas and may be determined from the following relation proposed by Hestroni [14]:

$$C_D = \begin{cases} 8(3\Theta + 2)/Re_p(\Theta + 1) & \text{for } Re_p < 2 \\ [14.9/Re_p^{0.78} + \Theta(24/Re) + (4/Re_p^{1/5})]/(\Theta + 1) & \text{for } 2 < Re_p < 500 \end{cases} \quad (5)$$

$Re_p = \rho_g \delta |U - v| / \mu_g$, $\Theta = \mu_p / \mu_g$

In this model the droplet were considered to be spherical and collide with each other at different speeds.

3.4. The numerical procedure

In simulation of processes, using the DSMC, two grid systems are normally considered. The first grid system is used to calculate the averages of flow properties. This grid system is chosen to be fine enough in order to increase the computational accuracy. The grid system is refined until the variations of the flow properties are not substantial (the variations of the flow properties should be less than 2%). The second grid system (Fig. 6) is selected to be extremely fine (the mesh size is equal to 0.2 times of the mean free path of the droplets) so that the collision of droplets could be controlled within each mesh with high accuracy. The grid system chosen in this study consisted of 88 divisions in X direction, 54 divisions in Y direction, 54 divisions in Z direction (totally 256,608 meshes) and the total number of model droplets was 3,800,000.

Each model droplet consists of $K_f = 70$ real droplets. The size of the mesh is in order of the mean free path of droplets and the time step in the simulation process is chosen to be 0.2 times the collision time [12]. The droplets are distributed in the mesh system according to the normal distribution. The initial velocity of the droplets is chosen based upon the velocity of gas jets in the flow field. Then as time passes the new position of the droplets is designated. The collisions of the model droplets are occurred based on the pattern put

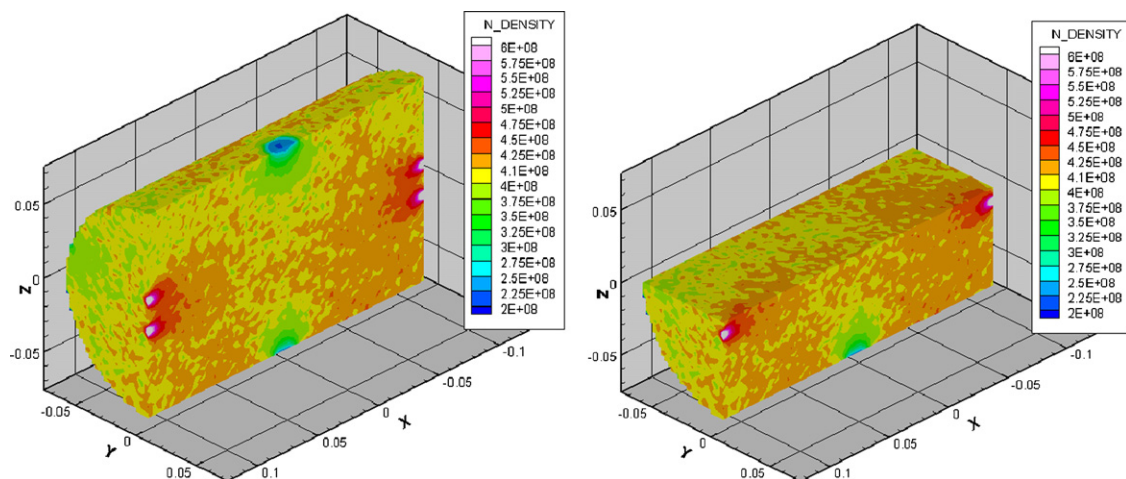


Fig. 11. N-density toluene droplets N/m^3 .

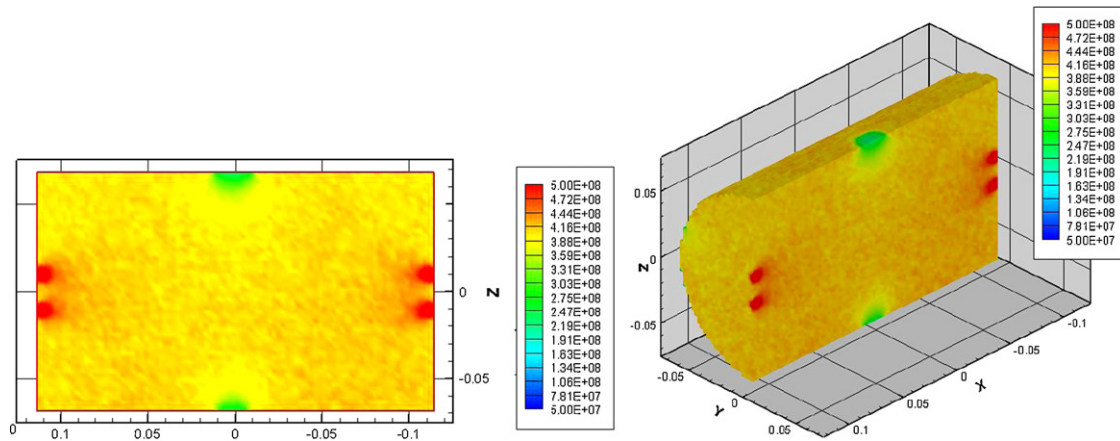


Fig. 12. N -density water droplets N/m^3 .

forward by Bird [12]. Passage of time is continued until the statistical fluctuations of the flow properties attain minimum values.

To implement the DSMC method, spatial cells have to be produced in the first step. the model droplets are placed in the cells with appropriate distribution. With regard to local gas velocity, a normal-distributed velocity is supposed for droplets. Without any statistical sampling, droplets are moved (NPS times repetition), until the latter attain suitable conditions for statistical sampling. For large domain simulations, this step can be ignored. NPS value definition is dependent on cells conditions; however optimization cannot be approached without sufficient experience.

At the next time step, according to each droplet velocity and the extended time step, the droplets are moved and so their new coordinates will be attained. If a droplet passes cell boundary, it will be eliminated from the cell, and a new droplet would be entered to the domain. Furthermore, if in this replacement, the droplet collides with a physical surface, it will be reflected from the surface according to the collision of “surface-droplet model”. The index number of cell and sub-cell containing the droplet is defined; and then the

probability droplet collision is investigated based on the model and if there is any collision, the velocity and location of the droplet will be modified. After NIS times repetition of movements, indexing, and collision investigations, statistical sampling should be done, the output of the program will be provided after NPT times repetition of sampling. This output is corrected up to NPT times. Flowchart of this algorithm is shown in Fig. 7.

3.5. Collision sampling techniques

The expected number of collisions in a cell, during a time interval Δt_m is given as

$$N_c = 1/2 n^2 \overline{\sigma_T C_r} \Delta t_m \quad (6)$$

The probability P of collision between two simulated droplets over the time interval Δt_m is equal to the ratio of the volume swept out by their total cross-section moving at the relative speed between the droplets to the volume of the cell, i.e.

$$P = F_N \sigma_T C_r \frac{\Delta t}{v_c} \quad (7)$$

Maximum efficiency is achieved if the fraction is such that the maximum probability becomes unity. The fraction is given by

$$P_{max} = F_N (\sigma_T C_r)_{max} \frac{\Delta t}{v_c} \quad (8)$$

In the above equation σ_T is collision cross-section and v_c is the relative droplet velocity.

The average number of real droplet in the cell is $n v_c$ and the average number of simulated droplets is $N = n v_c / F_N$, where, n , is the number density in liquid phase.

The full set of collisions could be calculated by selecting in turn, all $N(N-1)/2$ pairs in the cell and by computing the collision with probability P .

Pairs are selected from the cell at the time step, and the collision is computed using the following probability relation given by Bird [12].

$$\frac{\sigma_T C_r}{(\sigma_T C_r)_{max}} \quad (9)$$

In the present work two different liquids (water and toluene) have been used. Therefore, the collisions were between similar or unlike droplets so the method has to be slightly modified.

A pair of colliding droplets of types p and q is sampled from the possible pairs of these types with the probability of $(\sigma_T C_r / (\sigma_T C_r)_{max})_{pq}$. This term has been substituted for $\sigma_T C_r / (\sigma_T C_r)_{max}$ in all related equations.

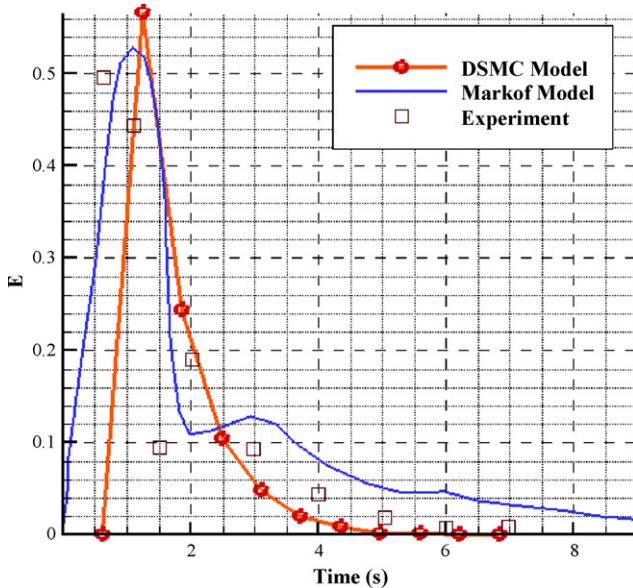


Fig. 13. A typical residence time distribution data for the continuous two impinging streams contactor. Internozzle distance: 25 cm; mean residence time: 2.2 s; sampling period in each segment: 1.4 s; nitrogen flow rate = 1300 cm^3/min ; toluene flow rate = 320 cm^3/min ; water flow rate: 325 cm^3/min .

Simulation results for these droplets are shown in Figs. 8–12.

A typical experimental residence time distribution curve obtained for the continuous two impinging streams contactor and those predicted applying the Markov chain model and DSMC technique are shown in Fig. 13.

4. Conclusion

The direct simulation Monte Carlo (DSMC) technique was applied to predict the motion of liquid droplets and residence time distribution in a two impinging streams contactor. This method was found to be more accurate and flexible in prediction of RTD data compare to the Markov chain discrete time formulation applied by Sohrabi et al. [3,5,15] in some previous studies. The degree of agreement between the data estimated from the present model and those determined experimentally was within 85%. While in case of Markov formulation the degree of agreement was lower than 75%.

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