Extended abstract of PhD thesis entitled: ” CFD modeling of particle agglomeration in counter-current spray drying process” of Dr. Eng. Maciej Jaskulski.

Introduction

The thermal dehydration of wet materials is widely used in industry in order to decrease product weight and to prevent materials from undergoing biological deterioration. The form of the received material strongly depends on the chosen drying method. Dehydration by spray drying is one of the most common thermal methods to obtain dry loose material from a solution, slurry, suspension or paste. The method involves the atomisation of a liquid feed in a stream of hot drying agent which leads to the rapid extension of the phase contact area and the significant acceleration of moisture evaporation rate [1]. The evaporation process runs at a relatively low droplet temperature inside the spray envelope (wet-bulb temperature), which prevents against overheating of the dried material. Furthermore, particle drying and residence time inside the drying chamber is short which minimises the probability of product thermal degradation. Thus, spray drying is used in many industries, from pharmaceutical to food and chemical, also for drying of heat-sensitive materials [2]. Because of different methods of phase introduction in the drying chamber the spray-drying process can be carried out in three ways: co-current, counter-current and mixed. Each method results in different physical and morphological properties of the produced powder.

In co-current spray-drying towers, due to parallel phase flow, air and particle recirculation hardly occurs which limits particle agglomeration and wall deposition [3].

Counter-current spray dryers are used mainly in the production of detergents and chemical fertilisers. In terms of flow dynamics, the process is more complex than the co-current because of the intensive mixing of the phases. There are just few pilot-scale or industrial scale counter-current spray drying systems that enable the measurement of flow dynamics of a continuous phase and changes in the morphology and properties of dried materials during the process [4,5]. Currently, the most comprehensive source of experimental data for counter-current spray drying were obtained at the Faculty of Process and Environmental Engineering at the Lodz University of Technology, Poland [6].

In order to minimise operating costs, the counter-current spray drying systems are optimised in terms of energy consumption and required properties of the product. Industrial companies show growing interest in computer simulations of the spray-drying process in order to determine the impact operating parameters on energy consumption and product quality. This is simply because computer simulations are time and cost effective compared to experiments. Optimisation of the process based on computer simulations requires a detailed description of the parameters of heat and mass transfer between the phases and dryer geometry and operating parameters [7]. Additionally, in most models the effect of agglomeration process on drying kinetics is simplified or totally ignored [8,9].

The coalescence of droplets and the agglomeration of particles are phenomena that occur in many industrial processes. During counter-current spray drying, due to an intensive mixing of phases, particles and droplets can collide and form bigger agglomerates. Particle agglomeration is difficult to control and its effect on the drying kinetics, the temperature distribution of the two phases, and the flow dynamics of drying agent in the tower is complex. With the current state of the art of agglomeration model we cannot predict the size of the resulting product or optimise the process without experimental methods.

Particle agglomeration in spray drying

As a result of atomisation, liquid is disintegrated into droplets, which upon drying change into particles. Droplet size depends on the type of atomiser and the physicochemical properties of the liquid, and may vary from a few to several hundred microns. As a consequence of air recirculation and rapid changes of flow trajectory, the droplets/particles collide. During the spray-drying process, we can distinguish three types of collisions: the collision of two droplets, droplet-particle collision and the collision of two particles [10]. Each of these may lead to coalescence, but depending on the type of collision the particles may merge in different ways [11].

Inside the atomisation zone, the collisions between droplets can be observed. This process, called coalescence, may result in the formation of new and bigger droplets with a spherical shape. If the velocity of the droplets in the collision is high, the droplets may be fragmented into smaller fractions by the impact force [12]. Alternatively, if the impact force is not high enough to break the surface tension forces, the droplets bounce against each other and coalescence will not occur [13]. When dry particles are recycled back into the atomisation zone, a second type of collision can occur: the collision between dry particles and liquid droplets. Consequently, dry particles can be coated with a new layer of the solution and take a shape similar to the spherical ones. Collision between two wet particles can result in the formation of one irregular agglomerate. Particles are connected by surface moisture, which form a so-called ‘liquid bridge’ [14]. As a consequence of drying, the ‘liquid bridge’ will solidify and create a stiff junction between the two agglomerated particles. An additional parameter controlling the development of the liquid bridge is particle stickiness on the dry particle surface. Particle stickiness strongly depends on the particle surface temperature, moisture content and the individual properties of the dried material [15]. In the literature, there are several methods for determining a particle’s sticky point (temperature at which particles change from sticky to non-sticky), most of which are based on the calculation of glass transition temperature from the particle moisture content [16]. Particle sticky temperature is higher by approximately 10 to 30°C than glass transition temperature [17]. The determination of the time period when the particle is sticky allows the prediction of regions where particles can agglomerate. These properties are characteristic of materials containing sugars, especially encountered in the food industry [18]. Studies on particle stickiness are carried out to reduce the possibility of particle agglomeration during spray drying (e.g. with skimmed milk) [19]. Glass transition and particle sticky temperatures, however, are properties that differ for specific materials, and must be determined experimentally.

The last type of collision, contributing in agglomeration inside the drying tower, is the inter-collision of non-stick and dry particles. Those particles can agglomerate due to electrostatic or Van der Waals forces but resulting structures, however, are not stable because of the weakness of bonds between particles.

Changes in particle size and the resulting changes in the area of contact with the hot air have a significant impact on the process of drying and must therefore be taken into account in modelling counter-current spray drying [20].

Aim of the thesis

The objective of the thesis is to construct and verify the model of droplet coalescence and particle agglomeration during counter-current spray drying.

In the first part of the work a discrete phase model (DPM) is developed. The model includes

- Droplet coalescence and particle agglomeration.
- Momentum, mass and heat transfer between particles and drying air.
- Trajectory and position of particles.

In order to verify the proposed model of agglomeration, results of simulation of counter-current spray drying will be compared with experimental data [6] obtained in a pilot-plant spray drying tower. In the second stage, the proposed model of agglomeration will be used in the complex CFD simulation of counter-current drying on an industrial scale. The results will be compared with experimental data to finally confirm the correctness of the proposed model of particle agglomeration.

Particle agglomeration model

Commercially available software for CFD calculations will allow us to calculate the discrete phase on the basis of the PSI-Cell method [21]. The methodology to calculate the process of drying taking into account particle agglomeration, developed within this thesis proceeds according to the following scheme. After determining the initial and boundary conditions the airflow inside the drying chamber is calculated. The calculation is made using a standard CFD model built into the Fluent v.13 program. Then, the solver performs a so-called DPM iteration which consists of a number of calculation steps made on the basis of our own discrete phase model.

In the first step of the DPM iteration initial parameters of particle stream in the injection point are determined. The stream is divided into single streams corresponding to different particle fractions. Each stream has its own trajectory inside the computational domain. The trajectory is composed of segments with constant length (the length of segments should be equal to the length of computational mesh elements). In each DPM iteration step for each trajectory the position of one segment is calculated. The particle trajectory begins in the point defined in the previous step, while the position of the segment end is determined on the basis of particle velocity components calculated from the equation of balance of forces. Knowing the length of the segment and local particle velocity it is possible to determine the time which the particle needs to move from the beginning to the end of the segment (a so-called DPM time step). The total value of DPM time steps is used in the determination of particle residence time inside the drying chamber. During each DPM step a model of particle agglomeration is referred to. The result of calculation of changes in particle mass and diameter is taken into account in the calculation of the position of the end of the next segment of trajectory of a given stream. The streams of particles are tracked until they leave the computational domain, each of the trajectory may have a different number of segment components. After determining the trajectory of all streams of particles the mass and heat balance between particle stream and the surrounding air is solved according to the PSI-Cell method. After completing balance calculations of both phases the air velocity distribution is updated using a standard CFD solver. A newly obtained pressure, velocity and temperature profile is used in the next DPM iteration in which new particle trajectories are determined. The procedure is repeated until reaching a convergent solution.

One of the aims of the proposed model was to determine the frequency of inter-particle collisions on the basis of the current particle position. For this purpose the DPM model tracks the trajectory of all particle streams inside the computational domain and stores them in the cache. Agglomeration of particles can occur during the collision of two or more streams of particles, it is therefore necessary to define places of intersection of particle trajectories in order to identify the areas where particle agglomeration may take place. The model checks the distance between streams:

$$\sqrt{\left(x_{p,1}-x_{p,2}\right)^2+\left(y_{p,1}-y_{p,2}\right)^2+\left(z_{p,1}-z_{p,2}\right)^2} \le 0.75\left(d_{p1}+d_{p2}\right) \quad (1)$$

where x, y and z are the Cartesian coordinates of particle stream position inside the computational domain.

In the model of agglomeration and drying a spherical shape of particles is assumed. In fact the particles, especially their agglomerates, may have irregular shapes. In addition, the particles vibrate during flow due to local turbulence and microeddies omitted in calculations with the use of RANS-type turbulence models. Therefore, it is assumed that the effective distance between the centers of the particles at which the collision will occur is greater than the sum of particle radii. Based on the results of several test simulations we estimate that the distance between particles is $0.75(d_{p1}+d_{p1})$.

In order to count the number of collisions, the stochastic collision model was modified and implemented. In the literature, the frequency of collisions is calculated on the basis of the parameters of a real particle and a randomly generated virtual partner. In the calculations of the frequency of collisions in our DPM model, only real particles are considered and parameters such as velocity and diameter of the particles are taken from the CFD simulations.

In the place of collision of two streams, the number of particles N is equal to the sum of particles in both streams ($N_{p,1} + N_{p,2}$). This assumption was implemented in the stochastic collision model [22] in order to calculate the frequency of particle collisions:

$$P_{coll} = \frac{\pi}{4}\left(d_{p,1} + d_{p,2}\right)^2 \left|\vec{v}_{p,1} - \vec{v}_{p,2}\right| \left(N_{p,1} + N_{p,2}\right) \tag{2}$$

Not every collision may result in the agglomeration of particles: the smaller the moisture content of the particles, the lower the probability of agglomeration. As mentioned earlier particle collisions can be divided into three basic types: collisions of droplets, wet particles and dry particles. To determine how many of the collisions result in particle agglomeration the agglomeration correction factor γ was proposed. If two droplets collide it was assumed that each collision resulted in coalescence, and the correction factor γ took the value 1. For particles with moisture content lower than critical moisture content, the following function is proposed, according to which the agglomeration correction factor depends on the average particle moisture content $\gamma = X/X_{cr}$.

The DPM model checks the moisture content of the two colliding streams, and it chooses a higher value to determine the type of collision and calculate factor γ. The probability of agglomeration increases when the dry particle returns to the atomisation zone and collides with wet particles. The case when completely dry particles collide was also studied. The agglomeration of such particles takes place when the resultant velocity of colliding particles is smaller than the critical value:

$$\text{if} \quad \left|\vec{u}_{p1} - \vec{u}_{p2}\right| cos\phi \ \leq \ v_{cr} \quad \text{then} \quad \gamma = 1 \tag{3}$$

The critical collision velocity could be determined from the energy balance equations in which van der Waals forces are assumed to be the factor combining the particles [22].

When the number of collisions has been determined, the amount of particles in the stream can be recalculated.

$$N_{p,ac} = N_{p,bc} - P_{coll} \frac{l_p}{v_p} \gamma \tag{4}$$

Since the reduction of the number of particles within the stream results in an increase in particle mass, from calculation of the dry mass balance and considering moisture loss we can determine the mass of particles in the stream after the agglomeration:

$$m_{p,ac} = \left(\frac{N_{p,bc}}{N_{p,ac}} m_{p,s} \right) \left(1 + \bar{X} \right) \tag{5}$$

In the standard Fluent code mixture density is calculated on the basis of volume mixing law. During spray drying process the porosity of particles is rising so that the final particle density is lower than the density of solid material. In this case, it was assumed that up to the value of critical moisture content, particle density increases linearly. After exceeding the critical value particle shrinkage is stopped and particle density starts decreasing. The new diameter and particle mass are used to calculate mass, heat and momentum transfer between the phases in the next DPM iteration step. The change in diameter significantly affects the value of the Reynolds number, which determines not only the coefficients of heat and mass transfer between the particle and the drying air, but also the flow dynamics drag coefficient.

CFD simulation of counter-current semi industrial spray drying tower

The object of simulations was the counter-current spray drying tower constructed at the Lodz University of Technology [6]. The body of the experimental installation is a column with 0.5 m diameter and 8 m high. A hot air distribution system is located at a distance of 1 m from the column base. Drying air is introduced tangentially through twelve 10 mm narrow slots. A thermostated nozzle was set at the top of the dryer at the level of 6.7 m from the air inlet. Dry powder is received in the bottom of the dryer and also recovered from an air purifying system (cyclones and bag filter) on top of the column. Powder collected in cyclones is not recycled into the drying chamber not to influence agglomeration process in the tower. In order to reduce heat losses in cylindrical part of the column (above air inlet level) the tower is insulated with a 10 cm glass wool layer. A methodology for the measurement of properties and behavior of the continuous and discrete phase was described by Piątkowski and Zbicinski, 2006. To reduce wall deposition in the column, slurry was atomized using a full cone nozzle (Spray System, USA) with narrow spray angle, 13º. As a drying material 50% mass solution of starch maltodextrin in water was used. Initial PSD was determined from PDA measurements.

Large air temperature drop (even 60ºC) was observed during the operation of the tower without spray drying process. To ensure accurate temperature distribution inside the drying tower heat transfer coefficient from the tower wall to environment was calculated from the Grashoff equation. The drying column is insulated only in cylindrical part above the hot air inlet. The model of heat losses was tested in four different conditions (amount of air 200 Nm^3/h and 300 Nm^3/h, temperature 150ºC and 200ºC). Error of heat losses calculation was lower than 8%. To test the effect of various factors on the agglomeration process seven, 3D simulations with different initial drying conditions were carried out. Parameters that were changed were: air flow rate (200 and 300 Nm^3/h), air temperature (150 and 200°C), feed rate (6 and 12 kg/h) and the flow rate of atomizing air (0.6, 1.8 and 2.4 kg/h). After the tests, mesh with 1.5 million elements was chosen. Large number of mesh elements resulted from high mesh density in hot air inlet area. Due to rapid air velocity changes, Reynolds Stress Model (RSM) was chosen for turbulence calculations. Final simulations were performed in steady state conditions.

To check accuracy of the CFD model five parameters, i.e. temperature and humidity of both phases and average particle diameter were analyzed. The analysis of simulation results of air humidity and particle moisture content shows good agreement with experimental data (Fig. 1 top). Particle moisture content is decreasing with the distance from the nozzle. In bottom part of the drying tower particles reach equilibrium moisture value. At the same time air humidity is increasing from initial value to 18 g/kg for slurry feed rate = 6kg/h and 24 g/kg for slurry feed rate = 12 kg/h. Thus, the characteristic drying curve (CDC) [23] evaporation model can be successfully used in simulations where calculations of mass transfer resistance inside each single particle consume much computational power. Both graphs show the range of the nozzle operation and identify effective working area of the dryer which is crucial in the determination of an optimal tower length.

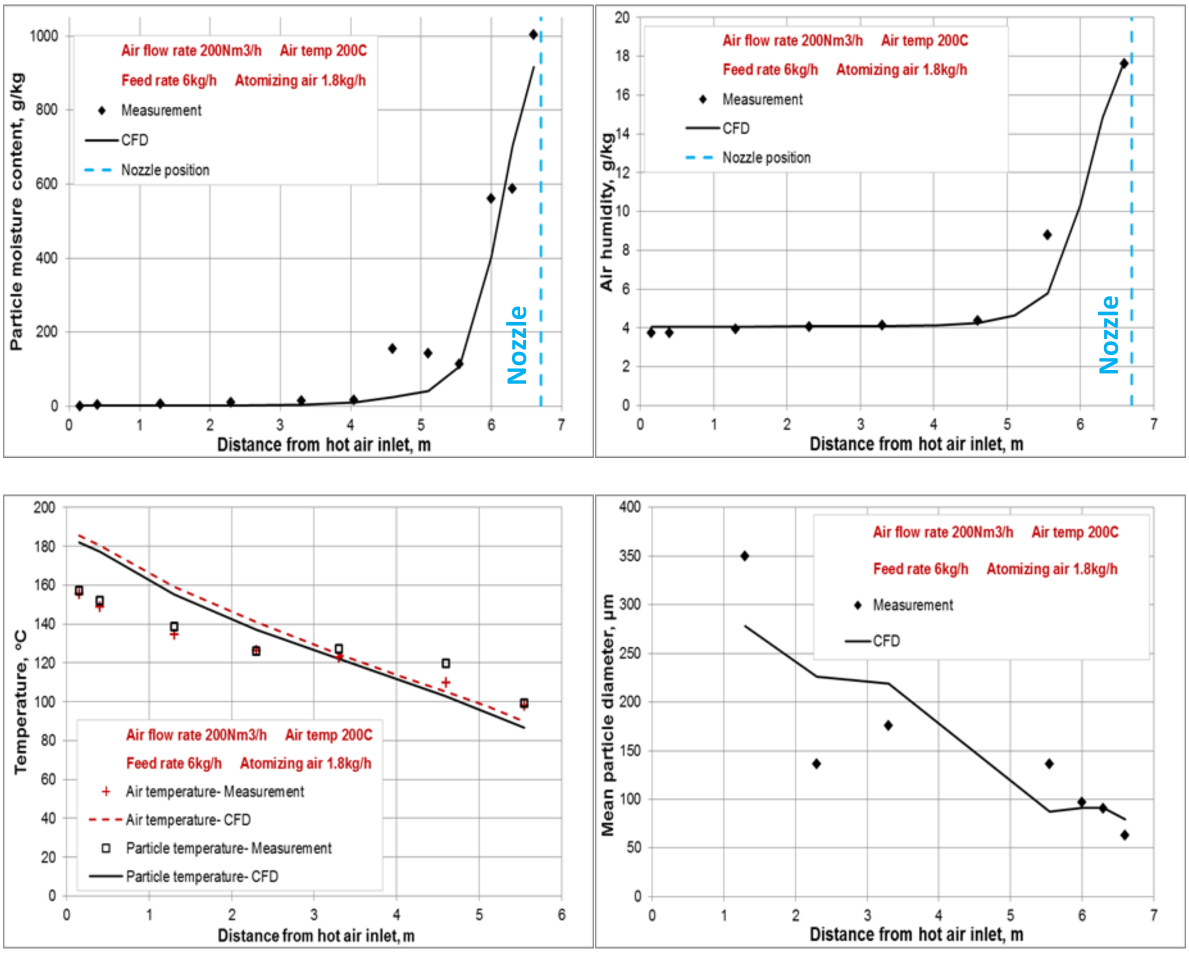

Fig. 1 Changes in air humidity, particle moisture content, air and particle temperature and particle diameter along dryer high calculated by the developed CFD model. Experimental data from Piątkowski (2011).

A comparison of CFD simulation results with measurement data for the temperature of both phases is shown in Fig. 1. Temperatures of particles and air are similar and decreasing towards the nozzle. Fig. 1 shows also comparison of changes of particle average diameters due to agglomeration and coalescence in the counter-current spray drying process with measurement data. In CFD simulations, the calculated diameters are bigger than average value obtained in the measurements because the model does not take into account degradation and abrasion of particles. In the real process, small particles and dust in the dryer recirculate intensively and can be counted many times by a PDA measuring system especially between 2 and 3 meters from the air inlet where small and light particles are unable to go through the drying air supply zone of the dryer and remain at the same level.

This test analysis of the developed CFD spray drying model with particle agglomeration shows high quality of the simulations and allows to calculate other, industrial spray tower.

CFD simulation of counter-current industrial spray drying tower

The object of simulation was the drying tower for the detergent, built at the beginning of the 70s and modernized in 1993, operated in Raciborz, Poland. The total tower height is 37 m (33 m height of drying chamber), and the inner diameter of the cylindrical part is 6 m. At the top of the tower there is a system of bag filters, which purify the outlet air. The filter consists of 720 bags, each 2.6 m long and 0.15 m in diameter. The walls of the tower were insulated with a layer of mineral wool of thickness 0.12 and 0.14 m.

Slurry is sprayed by two systems of nozzles located on two levels. Twelve nozzles are located at a height of 18 m and 10 nozzles at 10 m from the hot air inlets. In the analysed spray drying process 12 nozzles were used, six on each level. The spray angle of the nozzles (SH118) was 65°. The initial particle size distribution ranged from 20 μm to 600 μm. In order to determine initial atomisation conditions, the Rosin-Rammler distribution was fitted and a mean diameter $\bar{d}$ = 285 μm and spread parameter $\dot{n}$ = 3.783 were calculated. The initial velocity of particles was 50 m/s. The nozzles were directed to the dryer axis and inclined at an angle of 55°. At the higher level, the nozzles were mounted on shorter lances 650 mm long; on the lower level the lances were 1370 mm long. In order to obtain the data necessary to verify developed in this work drying and agglomeration model, the temperature distribution was measured in the drying chamber along the dryer radius at seven levels during drying of the washing powder. The measurements were made using a microseparator, whose construction was adapted for measurements on an industrial scale [24]. In each measuring point two temperatures were recorded, which are average values recorded by using two temperature sensors: internal which measures the air temperature, and external which measures the particle temperature [8].

Drying air is heated in a gas burner and supplied to the drying chamber through a distribution ring with a rectangular cross-section. Hot air from the distribution ring flows to the drying tower through 16 connection ducts perpendicular to the tower wall inclined at an angle of 45°. The inlets of the ducts are divided into three parts. In five connection ducts, the cross-sectional area was reduced locally in size to 2/3. The tangential supply of air and differences in the cross-sectional areas of the connection ducts cause uneven distribution of air supplied to the drying tower. This unevenness has a significant influence on airflow inside the tower which was confirmed on the basis of thermoanemometer measurements (AP471 DeltaOhm, Italy). Mass flow rates inside the connection ducts obtained from measurements were used as boundary conditions at the inlet to the drying tower in subsequent CFD simulations of the drying process. The total amount of drying air was equal to 21.9 kg/s with an initial temperature of 259°C.

Full 3D geometry of the drying tower was developed and subjected to the discretization process and series of numerical meshes differing in type and number of elements was tested. Finally, a dense, non-structural mesh consisting of 745k elements was used.

The upper air outlet was defined as a free flow (the *pressure outlet* condition). The lower outlet was defined as a wall constituting a barrier for the air but permeable to DPM particles. The coefficients of heat transfer from the walls to the surrounding air were determined from the heat balance of the tower. For insulated walls the heat transfer coefficient was 3.5 W/m^{2}K and for the cone chute 4.75 W/m^{2}K where wall was in direct contact with the distribution ring with no heat losses.

Large instabilities of airflow inside the tower required calculations under unsteady-state condition with a time step of 0.1 s. Tests of the turbulence models showed that, for such a dynamic airflow with swirls and recirculation, a convergent solution could be obtained using the Reynolds Stress Model [25], which, however, increased the computational time. The simulation covered 305 seconds of real operation of the drying tower. The initial 180 seconds were considered as stabilisation time, while the next 125 seconds were used to analyse the process.

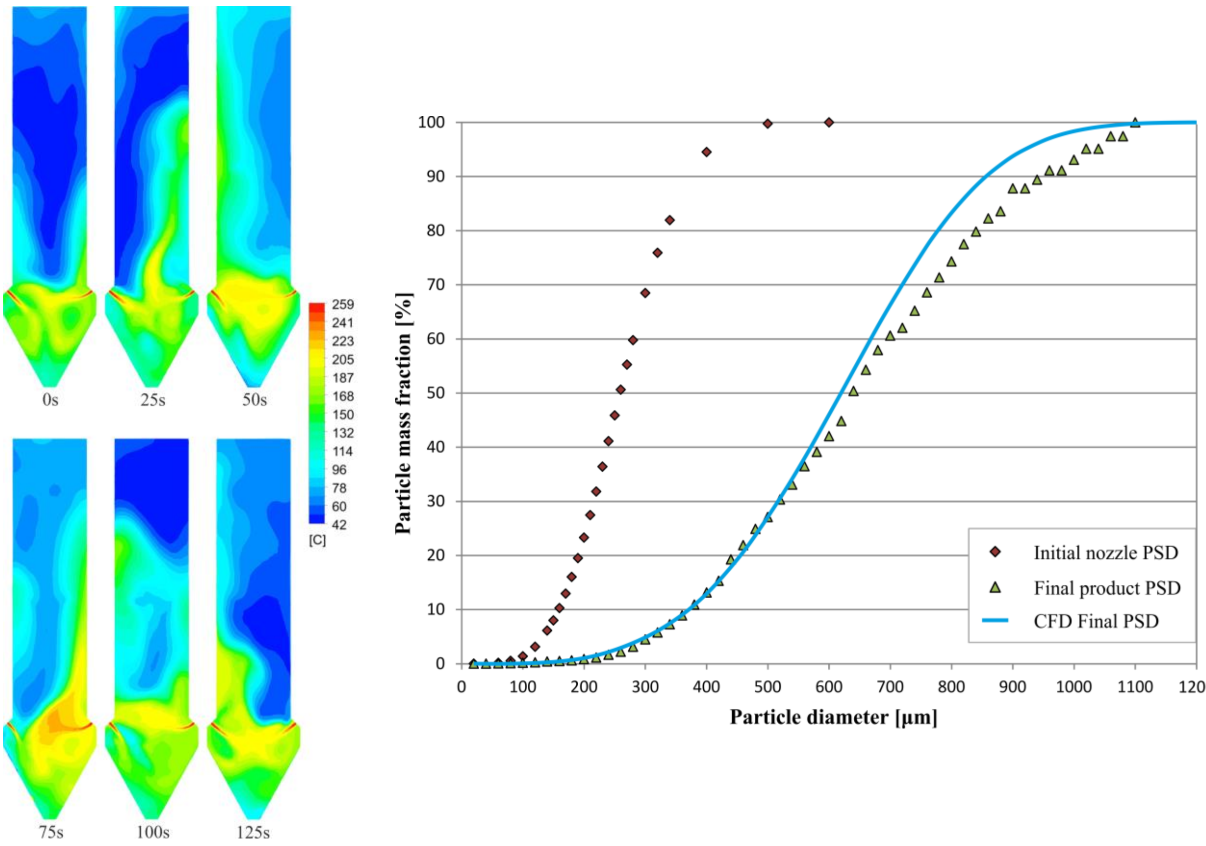

Fig. 2 Air temperature profiles in axial cross-sections of the dryer at consecutive time steps (left) and comparison of initial and final PSD and diameters calculated from the CFD agglomeration model (right).

Experimental temperature distributions were compared with time-averaged results of the CFD simulation. Comparison of the results confirms good agreement of temperature distributions obtained from simulations with temperature distributions of the real drying process on an industrial scale. Analysis of air flow trajectories shows not only changes in flow direction but also a change in the location of eddies in the horizontal axis at different heights of the drying tower. Swirl air motion can be observed in the cylindrical part. The changes in airflow have a significant influence on air temperature distribution during the drying process. Figure 2 (left) shows asymmetry and chaotic changes of air temperature pattern selected time steps.

The main aim of this part of the study was to verify the accuracy of the agglomeration model in the industrial spray dryer. For this purpose, the PSD of product particles measured experimentally was compared with the distribution obtained from the CFD simulations. For the analysis, the PSD of the particles collected at the bottom of the column at each time step of the simulation was determined, and one distribution curve was generated for the entire simulated time of the process. In Figure 2 right, comparison of initial and final PSD and diameters calculated from the CFD agglomeration model is shown. The final average particle diameter of the product obtained with microscopic measurements of powder was 725 µm; the value obtained in the CFD simulation was 665 µm. Difference between simulation results and measured values can be caused because the particle breakage and abrasion were not taken into account in calculations. Both curves, however, have a similar shape and overlap in the range of fractions from 20 to 500 µm, which proves that the proposed model of agglomeration during spray drying is accurate.

Summary

A survey of the literature on modeling the processes of droplet coalescence, particle agglomeration and frequency of inter-particle collisions is presented in the thesis. It was found that in the literature there are no verified models of the mechanism of particle agglomeration during counter-current spray drying process which could be implemented in CFD calculations by the Euler-Lagrange method. For the first time in the literature the following problems have been discussed in this thesis:

1. A model of particle agglomeration during spray drying process is proposed. The model takes into account particle trajectories, changes of particle morphology (resulting from the drying process) and the effect of changes in process parameters on the rate of agglomeration and particle diameter distribution. The proposed models of agglomeration and drying were implemented, along with the libraries of physicochemical properties of substances and auxiliary programs, as our own DPM model in the Fluent v13 program. The Ansys program was used in the hydrodynamic calculations of continuous phase and as a solver to calculate differential equations in the DPM model.

2. To verify the proposed model of particle agglomeration, selection of model parameters and determination of a method for implementation to the Fluent computing environment, 13 simulations of counter-current spray drying were made for different process parameters in the pilot-plant scale. Average values of moisture content and temperature of the continuous and disperse phase as well as mean particle diameters at various heights of the drying column obtained in the CFD simulation were analyzed and compared with experimental data.

3. The model of particle agglomeration was verified by a comprehensive simulation of counter-current drying of a detergent in an industrial installation. Comparison of the results with experimental values confirmed correctness of the agglomeration model, hydrodynamic calculations of air flow inside the drying tower and the model of momentum, mass and heat transfer between phases.

4. On the basis of the properties of powder obtained in the process of drying in an industrial plant, the final particle diameter distribution of the product was determined and compared with the values obtained in the proposed agglomeration model. Very good agreement of the results clearly proves that the proposed method to calculate changes in particle diameter distributions during spray drying is correct. Also the need for further extension of the model with calculation of the degradation and abrasion of particles during the process as well as implementation of the agglomeration mechanism to skin-forming materials for which agglomeration kinetics depends on moisture content on particle surface is shown.

Analysis of the results of CFD simulation allows us to determine the effect of process parameters and changes in drying tower geometry on drying rate, dried material properties (final moisture content, diameter distribution and density) and also on the determination of intensive evaporation and agglomeration zones. Results of the CFD simulation with the implemented DPM model can be used to optimize spray dryer operation in terms of energy consumption and to design new spray drying systems.

Results of the CFD simulation of spray drying presented in this thesis, which include the proposed model of particle agglomeration, are the first experimentally verified results available in the literature.

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