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Hydrodynamics and Heat Transfer in Fluidized Bed with Liquid Spray: Experimental Visualization and Mathematical Modelling

Abstract: Fluidized beds with liquid spray have been widely applied in various industrial processes due to efficient heat transfer ability. In this work, the liquid-containing particles and the liquid dispersion pathway are visualized by a unique reversible thermochromic material (RTM) tracking method in an external-heated fluidized bed loaded with polypropylene particles. The effects of evaporative liquid spray on bubble density, particle motion and heat transfer are systematically investigated combined with PIV and DIA. The liquid injection decreases particle fluctuation, however, enables the uniform distribution of particle vorticity and promotion of bubble generation. A stable liquid-containing zone is observed under the top spray nozzle configuration, whereas liquid disperses rapidly under the side-wall spray mode. A multi-zone heat transfer model (MHTM) is further established based on the bubble growth and inter-layer exchange models. The experimental and simulated results agree well and indicate shallow insert of nozzle leads to larger temperature differences. The methods proposed are applicable for the analysis of the complex multiphase system and the optimization of the nozzle configuration.

1. Introduction

Gas-solid fluidized beds with liquid spray and injection have been widely applied in the many industrial processes, such as granulation and coating (Duangkhamchan et al., 2015), fluid catalytic cracking (Gao et al., 2001), polymerization in condensed mode (Jiang et al., 1997) in order to meet various industrial needs, for instance, efficient heat removal, good contact between catalyst and reactant. Furthermore, the penetration and
mixing of liquid within fluidized beds affected the hydrodynamics and particle fluctuation. For example, the agglomerates were formed instantly after liquid was injected into the fluidized bed (Stefan and Joachim, 2005). The study of Book et al. (Book et al., 2011) showed that liquid affected the bubble formation and channels via avalanche testing by means of acoustic and vibrometric methods. Fan et al. (Fan et al., 2001) reviewed the development progress of detection methods with liquid injection and the study indicated that the hydrodynamics and heat transfer behavior were closely related for the evaporative liquid injection system.

It is known that liquid transfer, coating process on the particles and liquid evaporation process are essential to the scale-up, optimization, avoiding hot spot (Briens et al., 2003) and increasing product output of the fluidized bed reactor. In industrial applications, the presence of the liquid in the gas-solid fluidized bed influences the hydrodynamics significantly and makes the fluidity behavior more complex and unpredictable. During the liquid injection process, the change of hydrodynamics and the evaporation of the liquid also affect the heat transfer behaviors, further altering the surrounding gas environment. Therefore, the temperature measurement is usually used together with the hydrodynamic measurement to study the heat transfer and fluidization process (Kolkman et al., 2017; Sutkar et al., 2015). Sun et al. (Sun et al., 2018) measured the spray zone by thermocouples and conductivity probe and found that the flow coherence and energy distribution shifted to higher frequencies. However, the current measurement methods are mainly focused on the detection of the liquid distribution or temperature distribution, and it is difficult to detect
and visualize simultaneously liquid dispersion on particles and how it affects the particle fluctuation.

The two-phase model is used for the description of the bubble phase and emulsion phase solid exchange, heat and mass transfer and thermal stability. With proper implementation of bubble growth model and inter-phase exchange model, the modified two-phase model is applicable for simulating the long-time running process of industrial scale units (Hatzantonis et al., 1998; Ibrehem et al., 2009; Shamiri et al., 2010). Many efforts have been made in order to predict the processes mentioned above by numerical investigations (Chen et al., 2016; Heinrich et al., 2003; McMillan et al., 2005; Nagaiah et al., 2007; Patel et al., 2013; Sutkar et al., 2016). For the industrial scale reactors involving chemical reaction, numerical simulations concerning two-phase model has been widely used in the polyethylene industry (Choi and Ray, 1985; McAuley et al., 1994; Ray and Villa, 2000), which have the capability of predicting the reaction kinetics and the heat transfer mechanism.

Therefore, to fully understand the change of the hydrodynamics on the resulting change of the heat transfer in the fluidized bed with liquid spray, the method of Fan et al. (Fan et al., 2018) was adapted, who developed an evaporation model for condensed mode polymerization and considered multicomponent liquid evaporation mechanism.

In this work, to study the liquid dispersion and the effect of the liquid injection on the hydrodynamics and heat transfer behavior, a novel thermosensitive color-changing coating (TSCC) is used in a polypropylene fluidized bed with continuous liquid spray. This kind of coating is made of reversible thermochromic material (RTM), which
displays different colors under different temperatures, and therefore the liquid dispersion pattern and wet particle distributions are visualized considering different nozzle configurations. Then, the bubble properties and the liquid-containing area are calculated through digital image analysis (DIA).

The results revealed that bubble generation effect caused by liquid introduction does not necessarily promote particle fluctuation. Nozzle configuration affects liquid aggregation form and therefore affects the liquid evaporation rates. The particle vortex is clearly enhanced with liquid introduction; however, the particle velocity is more sensitive to the circulation path. A refined multi-zone heat transfer model (MHTM) is developed for simulating the heat transfer behavior for the fluidized bed with liquid spray, where both the start-up process and the stable state are well predicted and agree well with experimental results. Therefore, the effect of the nozzle configuration on the liquid diffusion pathway and heat transfer are systematically investigated and the results can be applied for the guidance of the stable operation and nozzle optimization configuration of fluidized bed with liquid spray.

2. Experimental set-up and material

2.1 Experimental set-up

The experiments were performed in an external heated fluidized bed where the background is a heating panel with 5 heating rods inside. The background temperature inside the fluidized bed and the inlet gas temperature were controlled by a PID system developed by Zhejiang University (Zhou et al., 2016b). The outer main body was made of stainless steel and the inner part was filled with PTFE as shown in Figure 1. The
view-window was a transparent quartz glass, coated with the antifogging agent and of the same width of the fluidized bed. The fluidized bed was 100 mm in width and 16 mm thickness, filled with polypropylene particles of 1.1 mm diameter (SINOPEC, Tianjin Petrochemical). The plenum chamber was filled with ballotini balls of 1 cm in diameter to ensure the uniform gas distribution. The gas distributor was a perforated plate with XX holes of XX mm and the opening ratio was XX. Detailed experimental parameters are summarized in Table 1.

The typical flow pattern in the bubbling fluidized bed exhibits upward particle motion near the bed center and downward particle motion near walls due to strong bubble coalescence near the bed center line. And particle velocities and volume fraction near the bed center and walls will lead to different liquid dispersion pattern. Furthermore, the collision direction between liquid droplet and polypropylene particle will also affect the liquid film formation on particles, and eventually affect heat transfer. Thus, three kinds of nozzle configurations were constructed by comparison with no spray cases (NS). The liquid injected into the fluidized bed was alcohol analytically pure (XX Company) and it was injected into the heated fluidized bed at ambient temperature.

Top spray (TS). The nozzle position of TS was 1 cm above the material surface, and therefore liquid droplets collide with particles face-to-face, as shown in Figure 1. And the liquid-covered particles will spout first and then mixed with particles within the fluidized bed.

Deep-insert spray (DS) on the side wall. For DS, the insert depth was 2 cm from the
right-side wall, and thus the droplets will collide with downward moving particles.

**Shallow-insert spray (SS) on the side wall.** For SS, the insert depth was 3.5 cm from the right-side wall. Under SS configuration, the liquid droplets will collide with the upward moving particles and can be sprayed into bubbles. The liquid nozzle height for both DS and SS configurations are 10 cm above the gas distributor.

![Schematic diagram of the experimental apparatus](image)

**Figure 1.** Schematic diagram of the experimental apparatus

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2.2 Reversible thermochromic material (RTM) colored particles

To visualize the liquid dispersion path and the evaporation effect on the heat transfer and the liquid dispersion process, a unique reversible thermochromic material (RTM) was used in this work and it was coated on polypropylene particles uniformly with a spray gun and dried for 48 hours.

RTM (Kingcai Pigment Co., Ltd, China) is a kind of microcapsule that repeatedly changes color as the temperature rises or falls and was prepared from an electron transfer type organic compound system. The color turns at a specific temperature due to electron transfer, changing the molecular structure of the organic. The RTM microcapsule particles are spherical and have an average diameter of 2-7 μm and the outside shell is a transparent with a thickness of about 0.2-0.5 μm that neither dissolves nor melts. It is important to avoid damaging this layer during use because of its sensitivity to the illumination. Therefore, the coating particles were prepared separately for each experiment to avoid color fading.

In the present study, two kinds of third-order color reduction RTMs were tested, of which the colors fade to lighter ones under high temperature were tested. The first RTM changes its color from brown to green and then to fluorescent yellow (B-G-F) when rising temperature, corresponding to the threshold value of 45 °C and 65 °C as shown in Figure 2(a). The transition thresholds of the other RTM, as shown in Figure 2(b) are the same and it changes from dark violet to red and then to transparent (D-R-T). The color change of RTM responds quickly within 3 s and RTM was tested to be stable for around 3000-4000 times of color transition in the environment higher than
130 °C or 2 days’ soaking in alcohol. In the experiments, due to images taken by the high-speed camera were grey scales and the color of D-R-T was more convenient for DIA. Thus, D-R-T was chosen for all the experiments in this work.

![Image](image.png)

(a) B-G-F (b) D-R-T

Figure 2. Zoom-in color visualization for two kinds of RTM coated particles (2.5u_{mf}, 75 °C, Q_L=9.6 ml/min)

3. Experimental procedure and data analysis methods

3.1 Experimental procedure

Experiments were carried under different background temperatures. The temperature heating panel was adjusted by the PID system and the inlet gas temperature was kept to be 60 ºC under each experimental condition. After stable fluidization when the temperature of coated PP was heated to the set background temperature, alcohol with ambient temperature was injected and the temperatures and pressure fluctuations were recorded. Due to the injection of low temperature alcohol and the evaporation of alcohol led to the decreasing of the temperatures, the heating panel would adjust the heating power accordingly.

The pressure signals were recorded by a data acquisition system (National Instrument) and two pressure sensors (CYG1219, Baoji Sensor Institute) with 400 Hz sampling frequency, located at 3 cm and 8.5 cm above the gas distribution plate. Temperatures were sampled every 15 s for at least 5 min for each experiment by
thermocouples and were located 4.5 cm and 14 cm above the gas distribution plate. The experimental parameters are listed in Table 1.

3.2 DIA method

Based on the performance of the above-mentioned RTM, low temperature particles can be recognized. Afterwards, it is important to analyze the bubble generation process and bubble diameters under different nozzle configuration and temperatures. Moreover, quantification of liquid-containing zone is useful for studying the liquid dispersion and evaporation pathways. As depicted in Figure 1, there were two cameras applied during the experiment, where the high-speed camera was for the PIV analysis and bubble diameter calculation because the images recorded were grey-scale. And the color camera was used for recording color images and the visualization of the color transition process after liquid injection.

**Bubble recognition by high-speed camera.** Bubble boundaries were obtained by IMAGE J. As shown in Figure 3(a) and Figure 3(b), the original image was converted from gray-scale to threshold-adjusted image. Then the bubble boundaries are identified. The bubble equivalent diameters were calculated based on the ellipse areas and more than 2000 images were analyzed for each experiment for about 10 s.

![Figure 3. Bubble identification procedure](image-url)
adjustment (c) bubble boundary recognition (d) bubble area calculation

**Liquid dispersion visualization by color camera.** The examples of color image are shown in Figure 4. In Figure 4, the liquid was sprayed from the side-wall into D-R-T particles. The relative volume fraction of low-temperature particles was calculated by Equation (1), where $S_{m,k}$ is the instantaneous marked area of $k$th particle, $N_m$ is number of the marked area, $\sum S_p$ is the total area of all the particles in the fluidized bed.

$$W_c = \frac{\sum_{k=1}^{N_m} S_{m,k}}{\sum S_p}$$  \hspace{1cm} (1)

![Figure 4. Quantification of RTM coated PP (a) original image (b) marked image (c) marked particles](image)

3.3 PIV method

Particle image velocimetry (PIV) system was applied for the analysis particle velocity and vorticity, consisted of a high-speed camera (WX100, Photron) and the light source. The shutter speed was adjusted to the optimized value of 8000 to give clear visualization of polypropylene particles TSCC. For PIV analysis, the frame rate was set 750 per second and both the instantaneous and time-averaged velocity field were obtained.
3.4 Characterization of RTM coated particles

The bubble-dispersion effect for liquid and localized mixing of wet particles are important for studying the liquid dispersion and resulting change of heat transfer behaviors. By using the RTM coated particles, the temperature change of particles and the liquid dispersion pathway can be therefore recognized.

Figure 5 and Figure 6 showed the color change process under different static bed heights, where Figure 5 was the original image and Figure 6 was the marked image by the method demonstrated in Figure 4. As particles were hypothesized to form agglomerates instantaneously at the exit of the nozzle when liquid penetrates into the fluidized bed and the particles were partially mixing with liquid, the evaporation mechanism was different between the porous particles and non-porous particles (Stefan and Joachim, 2005). Therefore, agglomeration breakup was one of the important ways to transport liquid and the agglomerates also proved to have relative low temperature in the experiment. As shown in Figure 5, at the bubble boundary or the color transition boundary, there were dark violet areas which were the small agglomerates. When the liquid droplets collided with the particles near the central line, the particles were wetted immediately and the temperature dropped fast due to liquid evaporation. However, the low temperature area mainly floated on the material level despite the downward particle circulation near side walls. There were changes when some low temperature particles moved to the gas distributor, but were nevertheless have strong influence on the surrounding particles. Periodical mixing process of particles with different temperatures can also be observed in Figure 5.
Figure 5. Instantaneous snapshots of the color change process and the liquid dispersion path (85 °C, $Q_L=13.6$ mL/min, $3u_{mf}$, $H_{ob}=17.5$ cm)

For lower static material level as shown in Figure 6(a)-(f), it can be found that liquid penetrates particles first and particle temperature decreased along the penetration path until the color changed. Although the liquid atomization was sufficient in the air, however, Fan et al. (Fan et al., 2001) found that in cross-flow fluidized bed with evaporative jet, the vaporization region took up the major portion of jet region while dispersed vapor-droplet region was between core region and jet boundary. Therefore, in Figure 6(a)-(f), particles around nozzle exit were more easily to carry liquid and then dispersed by the bubbles. The periodical bubble generation and eruption generate large voidage fraction around nozzle and therefore dispersed droplets area extended. In Figure 6(a')-(f'), it can be observed that for DS case, low temperature particle clusters were formed instead of uniformly dispersed in the bulk flow in Figure 6(a'')-(f''). It was observed from experimental results that the material level, background temperature and the nozzle configuration affected the hydrodynamics significantly. First, the porous surface and the circulation path increased in high material level cases. Also, the heat introduced by the heating panel into the system was higher, resulting in sufficient liquid evaporation.
The complicated flow patterns resulting from the presence of liquid and its evaporation influenced the heat transfer mechanism and the particle mixing in the fluidized bed. Therefore, proper mathematical descriptions need to be clarified to give guidance for the scale-up of fluidized beds with liquid injections.

![Figure 6. Effect of nozzle configuration on particle temperature and dispersion process with Δt=1 s, where (a)-(f) are for TP (85 °C, \( Q_L=9.6 \text{ mL/min}, 3u_{mf}, H_{sb}=11.5 \text{ cm} \)), (a')- (f') are for DS(85 °C, \( Q_L=9.6 \text{ mL/min}, 3u_{mf}, H_{sb}=17.5 \text{ cm} \)), (a'')- (f'') are for SS (85 °C, \( Q_L=9.6 \text{ mL/min}, 3u_{mf}, H_{sb}=17.5 \text{ cm} \))](image)

In Figure 7 (a), the marked regions are quantified by Equation (1). It is indicated in Figure 7 for SS configuration under 90 °C, the liquid was rapidly dispersed in the bulk flow. For DS configurations, lower temperature lead to larger marked zones because of less liquid evaporation. Therefore, it can be concluded that in the fluidized
bed with side-wall liquid spray, the liquid first penetrated the bulk flow and formed small agglomerates. Because the agglomerates contained high liquid content, the dark violet color was observed. The agglomerates were then broken and particles with liquid film were then distributed. However, the nozzle position was essential to the liquid dispersion and liquid-containing particles distribution. DS nozzle configuration lead to the low temperature particle aggregation and the influence of DS to the bulk flow was not as significant as SS. Moreover, the bubbles were important for the liquid distribution. At the bubble boundaries, the low temperature particle aggregation were dispersed whereas the particles are entrained by the vortices.

![Figure 7. Comparison of the marked relative low temperature zones (3umf, Hsb=11.5 cm, QL=9.6 mL/min)](image)

4. Result and discussion

4.1 Bubble analysis

In fluidized beds with liquid spray, the liquid dispersion and particle mixing are influenced by the bubble dynamics, and the liquid evaporation will also increase bubble generation vice versa. In addition, under the same liquid feed flowrate, increasing
particle numbers affects the liquid film thickness and the liquid dispersion pathway. To investigate the overall impact of the liquid spray on bubbles, the influence of nozzle configuration and initial material height on the bubble number, after reaching equilibrium between liquid evaporation and feed is shown in Figure 8. Figure 8 shows the bubble number at different axis heights, where Figure 8(a) is for the initial material height of 11.5 cm and Figure 8(b) is for the initial material height of 17.5 cm.

In Figure 8(a), side-wall liquid spray increased the bubble number in all the experimental conditions with lower initial material level. For the side-wall liquid spray under 85 °C, deep-insert spray (DS) configuration showed strong bubble generation promotion effect. The liquid was easier to be disperse by the bubble motion and then circulated all over the fluidized bed. The liquid covered on particles did not yet evaporate and therefore, the liquid bridge force weakened the bubble motion above the nozzle height. As a result, the bubble number reduces. Compared to DS, shallow-insert (SS) configuration under 85 °C showed moderate promotion effect. It can be observed that near the side-wall, there were few bubbles and the particle circulation was relatively weak. However, Figure 8(a) indicated that top spray (TS) largely weakened the bubble generation. In this case, liquid penetrated vertically among particles and formed a stable liquid-containing zone on the top of the material level. This quasi-steady-state could be found at moderate liquid feed velocity, whereas higher feed velocity lead to the rapid uniform liquid dispersion and accumulation. In Figure 8(b), after increasing the static material level, the evaporation time of liquid, including discrete liquid droplets and liquid films, is prolonged. Therefore, bubble density near
the material surface also increased. Increasing liquid feed velocity in Figure 8, bubble generation effect was enhanced for TS cases with higher material level.

![Figure 8](image)

**Figure 8.** Influence of the nozzle configuration on the bubble number within 45 s (a) side-wall liquid spray, $3u_{mf}$, $Q_L=9.6$ ml/min, $H_{ab}=11.5$ cm (b) top spray, $H_{ab}=17.5$ cm (NS: no spray, DS: deep-insert side-wall spray; SS: shallow-insert side-wall spray; TS: top spray)

The liquid spray changed the bubble diameter distribution and the bubble number density. Nevertheless, the trend of change of these variables were not necessarily the same. STD is usually used as an indicator of the change of bubble sizes. The effect of the material level on the fluidization behavior was found to have a significant influence on the natural oscillation frequency of fluidized bed (Bi, 2007). Furthermore, former study found that the localized mixing and aggregation of particles were more uniform for deep fluidized bed under $3u_{mf}$ (Escudero and Heindel, 2011).

In Figure 9(a), the standard deviation (STD) of pressure drop between 3 cm-8.5 cm is compared for different nozzle configurations. In Figure 9(a), the liquid injection reduces bubble sizes below nozzle position. Figure 9(b) shows the power spectrum density (PSD) of the pressure drop between 8.5 cm and 3 cm. Main frequencies in liquid-spray cases increase compared to the dry case, indicating that the bubble densities increased between 3-8 cm, the in accordance with the results in Figure 8(a).
Two peaks are observed in DS cases and this might be relevant to the appearance of the agglomerates. The slope of in the higher frequency bend is found to decrease in the liquid-spray cases, which could be explained by the change of the particle fluctuations and the dissipation of bubbles.

Figure 9. (a) standard deviation of pressure drops and (b) power spectrum density of pressure drops with different nozzle configurations ($3\mu_m, H_{sh}=11.5 \text{ cm}, Q_L=9.6 \text{ mL/min}$)

Although STD can be used for comparison of the change of bubble sizes, quantitative analysis of bubble diameter is necessary because the continuous liquid injection affects the bubble generation differently along axial direction. Figure 10 shows the variation of bubble diameters along the vertical direction. Combined with Figure 8, the appearance of large bubble was clearly seen in Figure 10(b) at 4 cm for TS. For DS in Figure 10(c), bubble diameters below nozzle height were decreased. SS configuration in Figure 10(d) and Figure 10(f) leads to the increase in bubble size below 16 cm and rising temperature promoted the possibilities of generation of large bubbles.
Figure 10. Effect of nozzle configure on bubble distributions ($3u_{mf}, H_{th}$=11.5 cm), where red lines in each box represent medians, upper and bottom edges of the thick bars represent 25th ($q_{25}$) and 75th ($q_{75}$) percentiles, discrete points are extreme values and circles are mean values.

The liquid droplets collide instantaneously with particles after exiting the nozzle and form liquid film on particles (Stefan and Joachim, 2005). The increase of material level changes the thickness of liquid films because of the change of the particle circulation velocity and the particle surface as it is shown in Figure 11, and it also changes the evaporation path. Figure 11 shows bubble diameter distributions for an initial static bed height of 17.5 cm. As the liquid evaporation pathway was increased
compared to low static material height, the bubble growth was observed from 16 cm-24 cm. In this case, TS with 9.6 mL/min above 16 cm indicated strong bubble growth effect and the median increased from around 15 mm to 50 mm, although as shown in Figure 11(b) the bubble number decreased. Further increment of the liquid feed velocity leads to deeper penetration of liquid, however decreases the bubble diameters due to accumulation of liquid on particles.

**Figure 11.** Boxplot of variation of bubble diameter along vertical direction (3\(\mu_{mf}\), \(H_{ab}=17.5\) cm)

### 4.2 Flow pattern analysis

The presence of the evaporative liquid alters the bubble number, bubble diameter and inter-particle force, and finally changes the particle motions and circulation pattern. Figure 12 and Figure 13 show the velocity filed analysis of no spray (NS) and deep-insert (DS) side-wall liquid spray after 5 minutes’ injection time, respectively. In Figure 12, the particle circulation and entrainment ability of vortices were strong, where small
stray bubbles quickly coalesced with the main bubble. Instead of steadily moving upwards together as in Figure 13(c)-Figure 13(f), bubble collision and breakage were severe in Figure 12.

![Figure 12](image)

**Figure 12.** Velocity vector fields of instantaneous snapshots with the time interval $\Delta t=32$ ms from (a)-(g) (NS, 85 °C, $3\mu_{mf}$, $H_{sb}=11.5$ cm, relative 1.4 cm/magnitude in vector)

In Figure 13(a), after bubble passing by the nozzle position, there were strong vortices formed. As bubbles rising and colliding, vortices moved apart, resulting in particle-liquid mixing as shown in Figure 13(b)-Figure 13(d). In Figure 13(e)-Figure 13(g), it is indicated that the bubble near the right wall erupted more frequently. Due to the saturated liquid content covered on particles, there were some liquid-containing particles circulated with bulk flow, which are the dark areas in Figure 13.
The time-averaged velocity magnitude is shown in Figure 14. It is clearly seen that the liquid injection affects the particle fluctuation, especially near the material level. However, the introduction of the liquid decreases the elutriation comparing to NS case in Figure 14(a). Also, due to the accumulation of wet particles, the particle velocity decreases near the gas distributor. Better liquid dispersion are observed for SS as shown above, resulting in localized turbulence in Figure 14(c) and Figure 14(d). The change of hydrodynamics is affected by the mixing and evaporation of the liquid, and will inevitably influence the heat transfer behavior, which will be discussed in the following section.
4.3 Heat transfer model composition

As shown above, the color change of particles not only indicates the liquid dispersion and wet particle trajectory, but also shows the temperature change of the whole field. The change of the bubble sizes will affect the heat and mass transfer ability in the fluidized bed and the liquid evaporation will also enhance and change the heat transfer efficiency. The influence of hydrodynamics and heat transfer are mutual. Therefore, based on the hydrodynamic models and heat transfer models, the following work developed a multi-zone heat transfer model (MHTM).

In the present study, based on the two-phase model, the emulsion phase (Choi and Ray, 1985) and the bubble phase were divided into Nth segments, and each segment was regarded as complete mixing. The liquid entering the fluidized bed was in rapid contact with the emulsion phase particles, formed liquid films coated on the surface of particles, and dispersed to other areas of the reactor as the particles move. The evaporation process of the liquid in the fluidized bed, the thickness of the liquid film on the surface of the polypropylene particles, the mass transfer and heat transfer
mechanism were then coupled in the model. Figure 15 is a schematic diagram of a tank-in-series model, The model used the following basic assumptions:

(1) The reactor consisted of an emulsion phase of Nth total mixing tanks and a bubble phase of Nth total mixing tanks, and the voidage of the emulsion phase was the voidage at the minimum fluidization velocity;

(2) The rate of particle exchange between the segments in the emulsion phase was equal to the axial exchange rate of the bubble wakes;

(3) The gas in the emulsion phase was completely mixed in each tank, and there was no back-mixing of gas between the segments;

(4) The volume of the bubble phase of each layer was the same, and the average diameter of the bubble was used when calculating the bubble phase volume fraction;

(5) The liquid in the Nth emulsion phase completely entered the Nth bubble phase after evaporation;

(6) The heat required for the evaporation of the liquid was provided by the emulsion phase;

(7) When the liquid presented in the Nth layer, a liquid film of the same thickness was formed on the surface of all the particles;

(8) The change in gas volume flow rate in the reactor was negligible.
The evaporation model was based on the gas-liquid equilibrium and the simplified multi-component method was applied to calculate the liquid evaporation rate as shown in Equation (3). It was assumed that there was a vapor layer on the liquid surface and the concentration of alcohol in the vapor was calculated by Equation (4). In the dew point evaporation process when Lewis number was 1 indicated the heat and mass transfer were balanced in the fluidized bed, as assumed in current experimental conditions. The main physical parameters applied in the model are listed in Table 2. The system established in this paper consisted of a series of differential algebraic equations. Since the variables such as temperature were coupled and nonlinear, the equations were very sensitive to the initial values, which might lead to the failure of the solution. The solution was based on the general four- and fifth-order Runge-Kutta method. Details can be referred the work of Fan et al. (Fan et al., 2018)

The condensate alcohol in mole of the Nth emulsion phase is calculated by:
\[
\frac{dn_{C_2H_5OH,N}}{dt} = \frac{\dot{m}_d,N}{W_{C_2H_5OH}} + \frac{Q_s}{V_N}(n_{C_2H_5OH,N+1} - n_{C_2H_5OH,N-1} - 2n_{C_2H_5OH,N})
\]  
(2)

Where \(w_{C_2H_5OH}\) is molecular weight, \(Q_s\) is the particle interchange between dry particles and wet particles, \(V_N\) is volume of the Nth segment, \(\dot{m}_d\) is total evaporation rate of a certain droplet and is calculated by (Miller et al., 1998):

\[
\dot{m}_d = -\eta_c m_d \left( \frac{Sh}{3Sc} \right) \ln(1 + B_d)
\]  
(3)

where \(B_d = \frac{\sum Y_{v,s,C_2H_5OH} - \sum Y_{v,s,C_2H_5OH}}{1 - \sum Y_{v,s,C_2H_5OH}}\) is the mass transfer number, \(\tau_d = \frac{\rho \ddot{d}_p}{18\mu}\) is the particle response time, \(Sc = \frac{\mu_s}{\rho \sum Y_{v,s,C_2H_5OH} D_{C_2H_5OH}}\) Schmidt number, \(Sh = 2 + 0.552Re_d^{1/3}Sc^{1/3}\) is the Sherwood number, \(Re_d = \frac{\rho |u_x - u_e| \ddot{d}_p}{\mu}\) and \(\eta_c\) depends on the nozzle configuration. The surface vapor mass fraction is defined in Equation (4), where \(X_{v,s,C_2H_5OH} = \frac{P_{C_2H_5OH}}{P}\) is the surface vapor mole fraction.

\[
Y_{v,s,C_2H_5OH} = \frac{X_{v,s,C_2H_5OH}}{X_{v,s,C_2H_5OH} + (1 - X_{v,s,C_2H_5OH})\dot{W}/W_{C_2H_5OH}}
\]  
(4)

The mass balance of the liquid in the Nth emulsion phase segment is shown in Equation (5), where the first and the second term on the right-hand side are the convective mass transfer and the bubble phase to emulsion phase mass transfer respectively:

\[
\frac{dC_{CH_3OH,e,N}}{dt} = \frac{Q_s(C_{CH_3OH,e,N+1} - C_{CH_3OH,e,N})}{V_e} + \frac{K_b(C_{CH_3OH,b,N} - C_{CH_3OH,e,N})V_b}{V_e}
\]  
(5)

The mass balance in the Nth bubble phase is shown in Equation (6):

\[
\frac{dC_{C_2H_5OH,b,N}}{dh} = \frac{K_b(C_{C_2H_5OH,b,N} - C_{C_2H_5OH,e,N})}{V_b}
\]  
(6)

The emulsion phase temperature was affected by the gas convection, emulsion phase to bubble phase heat transfer, the background heat power, the liquid evaporation and back-mixing of particles. Equation (7) is the heat balance of the Nth layer emulsion
phase, where the heat loss of the experimental set-up was neglected:

\[
\left( V_c \rho C_{p,c} + V_e \rho C_{p,e} + V_i \rho C_{p,i} \right) \frac{dT_e}{dt} = Q_c \rho C_{p,c} \left( T_{e,n+1} - T_{e,n} \right) + H_m V_i \left( T_{i,n+1} - T_{i,n} \right) + V_e \Delta H_k
\]  

(7)

Bubble phase temperature was also affected by the gas convection and the emulsion phase to bubble phase heat transfer and defined as:

\[
V_c \rho C_{p,c} \frac{dT_e}{dt} = Q_c \rho C_{p,c} \left( T_{e,n+1} - T_{e,n} \right) + H_m V_i \left( T_{i,n+1} - T_{i,n} \right)
\]  

(8)

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Correlations</th>
<th>Parameters</th>
<th>Correlations</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bubble velocity</td>
<td>( u_b = u_e - u_{mf} + 0.711 (gD_b)^{0.8} )</td>
<td>Emulsion phase gas velocity</td>
<td>( u_e = \frac{u_{mf}}{\varepsilon_{mf}} (1 - \varepsilon_{mf}) )</td>
</tr>
<tr>
<td>Bubble phase fraction</td>
<td>( \delta_b = \frac{u_b - u_{mf}}{u_b} )</td>
<td>Voidage at minimum fluidization</td>
<td>( \varepsilon_{mf} = 0.586 \left( \frac{\mu^2}{\rho_g (\rho_e - \rho_g) D_b^{0.3}} \right)^{0.21} )</td>
</tr>
<tr>
<td>Bubble diameter</td>
<td>( D_b = D_{mf} - (D_{mf} - D_{bo}) e^{-0.13 (u_b)} )</td>
<td>Mass transfer coefficient</td>
<td>( K_{bc} = 4.5 \frac{D_{mf}}{D_b} + 5.85 \frac{D_{mf}}{D_b^{0.8}} )</td>
</tr>
<tr>
<td></td>
<td>( D_{mf} = 0.652 \left( A_b (u_b - u_{mf}) \right)^{1.1} )</td>
<td></td>
<td>( K_{pe} = 6.78 \left( \frac{D_{mf}}{D_b} \right)^{1.1} )</td>
</tr>
<tr>
<td></td>
<td>( D_{bo} = 0.00376 (u_b - u_{mf})^2 )</td>
<td>Heat transfer coefficient</td>
<td>( Q_e = \alpha \rho_g (1 - \varepsilon_{mf}) (u_b - u_{mf}) A )</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Solid exchange between dry particles and wet particles</td>
<td></td>
</tr>
<tr>
<td></td>
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</tr>
</tbody>
</table>

The experimental results measured by thermocouples and model calculated results are shown in Figure 16, where the total segment is 15. Here, the simulation results are the mixing temperature of bubble phase and emulsion phase.

It is observed from Figure 16(a) and Figure 16(c) that the increase of the initial static material level lead to higher stable temperatures for DS cases. In Figure 16(a), the stable temperatures at 4.5 cm and 14 cm were around 50°C and 47°C respectively for SS, and for DS are 57°C and 53°C respectively. Similar results were observed in Figure 16(c), the liquid injection and evaporation were balanced after 100 s and the
stable temperature at 4.5 cm and 14 cm were around 55.9°C and 48.9°C for SS cases, and for DS cases are 66.9°C and 61.9°C respectively for the high static material level case. It is clearly seen that increasing the static bed height resulted in the lower mass inter-segment particle exchange for SS cases, leading to the increase of the temperature difference. As for DS configuration, the particle exchange efficiency remained almost unchanged for all cases in Figure 16(a)-Figure 16(c). In Figure 16(b), it can be seen that the liquid evaporation for SS cases was largely promoted and the inter-segment exchange of particles was also enhanced, resulting in the stable temperature of 79.9°C and 56.8°C at 4.5 cm and 14 cm respectively. However, for TP cases in Figure 16(d), a low temperature zone was formed at the top of the material level.

\[ H_{sb} = 11.5 \text{ cm}, \quad Q_L = 9.6 \text{ mL/min} \]

\[ H_{sb} = 17.5 \text{ cm} \]
Figure 16. Comparison of the experimental and simulated results for different nozzle configurations at $3u_{mf}$, where $T_1$ and $T_2$ were at 4.5 cm and 14 cm above the gas distributor, respectively.

It is worth noticing that the stable temperature for different nozzle configurations depends on the heat transfer efficiency between segments, and the time needed for reaching the stable temperatures is also a reflection of localized mixing. Thus, in Figure 17 and Figure 18, time-averaged lateral particle velocities and vorticity near nozzle are compared, where each bubble cycle were about 1s, including bubble generation and eruption process.

Near nozzle position, the particle vorticity can reflect the entrainment effect and particle mixing and the vertical velocity reflected the mass interchange between adjacent segments. Above the nozzle position for the DS case, the particles were wetted and the bubble surface force reduced. Unstable agglomerates were easily formed at the exit of the nozzle in the shallow insert case and the particle fluctuation was decreased. Therefore, it can be seen from Figure 17(b) and Figure 17(d) that increasing temperature largely promotes particle velocities because of more sufficient alcohol evaporation, resulting in lower temperature in Figure 16(b).
It is observed that above 120 mm, particle motions were not as violent because bubbles were still developing for $H_{sb}=17.5$ cm. As analyzed above, bubble numbers and bubble diameters were both increased for SS cases, and therefore, the increase of the fluidized bed material height also leads to the sufficient dispersion of the liquid and the evaporation, leading to lower stable temperature in Figure 18(c). Similar results were observed for NS and TS case. However, the inter-particle force constrained local vortices and resulted in the decrease of the particle velocities. Variation of the bubble number and bubble diameter showed the competitive effect, mainly reflecting the change of local mixing and the heat transfer behaviors.

5. Summary and conclusion

The effect of the nozzle configuration and particle circulation path on bubble dynamics and particle temperature distributions have been characterized and visualized.
by novel RTM coated particles. The methods of image processing and the bubble statistical analysis provide the possibility of visualization and quantitative analysis of low temperature RTM coated particles and the influence of the liquid spray on the particle temperature. The deep-insert (DS), shallow-insert (SS) side-wall spray and top spray (TS) nozzle configurations were considered to have significant influence on hydrodynamics and heat transfer behaviors in the fluidized bed with continuous liquid feed. Furthermore, the material level and background temperature also affected the liquid dispersion pathway.

DS and SS nozzle configuration largely promoted the bubble generation in low static material level cases. The bubble number and bubble density increased due to the liquid evaporation. In these cases, the agglomerates were formed instantaneously at the exit of nozzle and the agglomerates were with high liquid content and low temperature. However, in TP cases, because the liquid droplets were collided with upward moving particles, the liquid penetrated to lower height and the bubble formation at the bottom of the material level was decreased.

Higher static material level mainly affected the liquid dispersion patterns and the bubble generation was promoted for all the cases because of the increase of the liquid evaporation path. It should be noticed that the trend of the change of bubble diameters was not necessarily the same as that of the bubble density. The aggregation of liquid-containing particles increased the bubble diameter and thus the possibility of the generation of large bubbles raised.

Due to the presence of the liquid bridge force, the liquid evaporation and different
collision pattern for different nozzle configuration, the complex flow dynamics involving the liquid spray was then analyzed by PIV. DS nozzle configuration had less influence on particle motions, whereas the SS and TP reduced the particle fluctuation obviously. It can be concluded there existed the competition mechanism of bubble motion with liquid bridge force, which lead to the change of the particle fluctuation. From the snapshots of the color change process of RTM coated particles, details of the liquid dispersion path were obtained. A stable low temperature zone was observed for TS case with high static material level and the liquid was found to evaporate rapidly with the particle circulation. For SS configuration, the liquid was dispersed relatively uniformly compared to DS configuration.

A multi-zone heat transfer model (MHTM) was established to predict the heat transfer behavior based on the two-phase theory. The evaporation of liquid droplets and liquid film were modelled based on the general four- and fifth-order Runge-Kutta method. The results showed that the liquid evaporation for SS cases was largely promoted and the inter-segment exchange of particles was also enhanced. Moreover, under proper operating conditions, the heat transfer efficiency could be enhanced significant. The simulated values compared well experimental results, and therefore can provide instruction for the design and optimization for fluidized beds with liquid spray.

**Notation**

- $B_M$ mass transfer number
- $A_t$ cross-section area of the fluidized bed
- $C$ concentration of alcohol
- $C_{p,s}$ specific heat of polypropylene
- $C_{p,l}$ specific heat of alcohol
- $C_{p,g}$ specific heat of gas
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
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</thead>
<tbody>
<tr>
<td>$D$</td>
<td>diffusion coefficient</td>
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<tr>
<td>$D_b$</td>
<td>bubble diameter</td>
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<tr>
<td>$D_0$</td>
<td>bubble diameter at orifice</td>
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<tr>
<td>$d_p$</td>
<td>particle diameter</td>
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<td>$\Delta H_b$</td>
<td>heat power of background</td>
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<tr>
<td>$\Delta H_{vap}$</td>
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<td>$H_{sb}$</td>
<td>static bed height</td>
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<tr>
<td>$H_{be}$</td>
<td>bubble phase to emulsion phase heat transfer coefficient</td>
</tr>
<tr>
<td>$K_{be}$</td>
<td>bubble phase to emulsion phase mass transfer coefficient</td>
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<td>$L$</td>
<td>liquid in $N$th segment</td>
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<tr>
<td>$m_d$</td>
<td>droplet weight</td>
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<td>$P$</td>
<td>reactor pressure</td>
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<tr>
<td>$P_{sat}$</td>
<td>saturated vapor pressure of component</td>
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<td>$Q_{L}$</td>
<td>liquid volumetric flow rate</td>
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<tr>
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<tr>
<td>$Q_e$</td>
<td>gas entering emulsion phase</td>
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<tr>
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<td>downward particle interchange between adjacent segments</td>
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<td>Schmidt number</td>
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<td>gas viscosity</td>
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<tr>
<td>$\tau_d$</td>
<td>particle response time</td>
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</table>

**Abbreviations**

- B-G-F: brown to green to fluorescent yellow
- DIA: digital image analysis
- D-R-T: dark violet to red to transparent
- MHTM: multi-zone heat transfer model
- PIV: particle image velocimetry
RTM reversible thermochromic material
TSCC thermosensitive color-changing coating
DS deep-inset
NS no spray
SS shallow-insert
TS top spray

Acknowledgement

Reference


