Whole-field imaging of temperature and hydrodynamics in a gas fluidized bed with liquid injection

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Abstract
This work presents results of experiments on thermal effects and flow dynamics in a flat-bed geometry gas-fluidized bed at atmospheric pressure, with and without liquid injection. Whole-field measurements have been performed using a combination of thermography, particle image velocimetry and digital image analysis. A first series of experiments demonstrates the mixing of two layers of particles at initially different temperature for a sequence of injected gas bubbles. It is shown that the pulse duration strongly affects the solids mixing and that the influence of diffusion on thermal equilibration is limited without convective mixing. Subsequently liquid has been injected into the fluidizing bed, which has been carried out with water, isopentane and hexane. Owing to the use of a combination of infrared and visual cameras, under particular conditions liquid-solid conglomerates can be observed in situ. Under the conditions applied, the flow dynamics are not clearly affected by the liquid injection. The thermal behavior of the bed is not found to be strongly dependent on which of the model liquids is injected, but more on the cooling capacity obtained by evaporating the injected amount of liquid at the selected injection rate, and the mode of operation.

1. Introduction
For application in chemical processes, gas fluidized beds have the advantage over fixed beds of operation at relatively uniform temperature (Kunii and Levenspiel, 1991) and rapid mass and heat exchange between the gas and solid phases. The mixing of the solid phase, possessing the dominant heat capacity in gas fluidized beds, is key to the beneficial thermal properties of a gas fluidized bed.
A particular and industrially important application of gas fluidized beds is polyolefin production. The production capacity per unit bottom plate surface area for this highly exothermic process...
was increased by introducing an inert hydrocarbon of relatively high boiling point, for example hexane or isopentane, into the gas phase. Due to the now induced condensation (Cai and Olson, 2005) upon cooling the recycle gas stream, the cooling capacity is enhanced. The amount of liquid that can be introduced to evaporate without disturbing fluidization properties depends on geometry (Chinh et al., 1996). Patented geometries for liquid introduction are for example bottom plate injection (Jenkins et al., 1986) and nozzle injection (Chinh et al., 1996), optionally with the use of a draft tube (Mutsers, 2002). The macroscopic reactor design, at a length scale of meters, needs to take into account factors such as sorption (Pannell et al., 2008), gas phase composition distribution in space and time, distribution of fluidization gas and the ability to discharge solid product dry from the continuously operating reactor. These are dependent on whether liquid is transported with the solid phase, or evaporates quickly, in close proximity to the point of introduction. Large amounts of liquid inside the bed may promote irreversible agglomeration (forcing reactor shutdown) and could affect product consistency (DeChellis and Griffin, 1994). At all times and positions, the particle temperature needs to be below the polymer melting temperature. For this a macroscopic solid circulation pattern without “dead zones” is needed, but also sufficiently frequent close contacting of all particles with the coolant itself or with cooled particles or gas. In efforts to increase cooling capacity and therewith production capacity even further it is thus of importance to know whether the liquid can always be relied on to evaporate quickly and how, from a fundamental understanding of the involved phenomena, this could be enhanced.

The mechanism to evaporation following the injection of liquid into dense gas fluidized beds has been subject to discussion (Bruhns and Werther, 2005). In recent years, capabilities of digital (infrared) cameras have improved and the capacity of affordable digital recording and processing of raw experimental data have dramatically increased. High speed visual cameras have been employed to observe the phenomena occurring after injection of water and ethanol onto a bed of hot FCC particles (Gehrke and Wirth, 2009) and for observation of injection of water into a bed of 0.2 mm sand (Cocco et al., 2013). The latter work achieved observation of conglomerates via the application of a fluorescent dye. Ariyapadi et al. (2004) have achieved observation of the motion dynamics of liquid injection via X-ray. Simultaneous observation of a gas fluidized bed with a visual and infrared camera was first presented by Yamada et al. (2001), studying heat transfer to stagnant particles at a window. Sakurai et al. (2005) presented a comparison between computational DEM results and observations of the time-averaged temperature profile using thermography, for a pseudo-2D bed. The work of Tsuji et al. (2010) studied the cooling of dry preheated particles in a spout fluidized bed via thermography and included particle tracking velocimetry to study particle motion. The infrared camera can also be employed with particle heating for particle tagging (Zhang et al., 2011, 2013). Recently, Patil et al. (2015) presented a combination of particle image velocimetry and infrared thermography to study the cooling of preheated particles in a non-spouting fluidized bed. Further work by Surkar et al. (2015) studied a spouting pseudo-2D fluidized bed with internal baffles. It employed injection of water and showed snapshots of the temperature distribution in time, to demonstrate the transport of particles through the bed.

![Fig. 1. The experimental setup employed in this work.](image)

<table>
<thead>
<tr>
<th>Designation</th>
<th>Material</th>
<th>Glass</th>
<th>Polymer</th>
</tr>
</thead>
<tbody>
<tr>
<td>Size range (mm)</td>
<td>Soda lime</td>
<td>LLDPE</td>
<td></td>
</tr>
<tr>
<td>Settled bulk density (kg/L)</td>
<td>0.4–0.6</td>
<td>1.0–1.3</td>
<td></td>
</tr>
<tr>
<td>Manufacturer</td>
<td>Sigmund Linder (Type S)</td>
<td>Lyondell-Basell</td>
<td></td>
</tr>
</tbody>
</table>

Table 1
Particle material employed in this work.
This work focusses on the thermal effects in conjunction with hydrodynamics, when introducing LLDPE particles of reduced temperature into a gas fluidized bed. The experimental work is done via a combination of visual and infrared cameras, providing a non-invasive whole-field observation with high spatial and temporal resolution.

As an introduction to the technique and to demonstrate the importance of convection in the thermal equilibration of gas fluidized beds, first a demonstration for two layers of particles at initially different temperature is given. As a controlled analog to the seemingly random passage of bubbles under fluidizing conditions, a sequence of bubbles is injected. Simultaneous to the mixing dynamics that would be observed for two layers of differently colored particles (Bokkers, 2005), now also time-dependent thermal effects are observed.

Subsequently, liquid injection is performed, with and without atomization gas being fed to the liquid injection nozzle. The LLDPE material used in the experiments is actual particle material taken directly from a polyolefin production gas fluidized bed, sieved to reduce the particle diameter range. Two of the liquids separately employed are used in polyolefin production as the induced condensing agent and are thus injected industrially (with condensed dissolved components) into a bed of closely similar particle material to evaporate. In this way the experiment mimics, albeit on a very small scale, the conditions of condensed mode polyolefin production, with the heat source due to reaction now replaced by heating by the inlet gas and back- and side wall contact. Experiments with water as liquid and with glass beads as solid material are also performed, to provide a reference for materials that are more readily available in a lab environment. The effects of the liquid injection on the temperature distribution and solids flux dynamics are demonstrated.

<table>
<thead>
<tr>
<th>Camera</th>
<th>Visual</th>
<th>Infrared</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wavelength range</td>
<td>Visual (CCD)</td>
<td>2.5–5.0 μm</td>
</tr>
<tr>
<td>Integration time (µs)</td>
<td>370</td>
<td>800</td>
</tr>
<tr>
<td>Pair interval (µs)</td>
<td>2</td>
<td>Not appl.</td>
</tr>
<tr>
<td>Maximum vertical resolution</td>
<td>1024</td>
<td>512</td>
</tr>
<tr>
<td>Maximum horizontal resolution</td>
<td>1280</td>
<td>640</td>
</tr>
<tr>
<td>Bits per pixel</td>
<td>16</td>
<td>14</td>
</tr>
<tr>
<td>Memory (maximum)</td>
<td>2 GB</td>
<td>PC RAM</td>
</tr>
<tr>
<td>Brand</td>
<td>Lavision</td>
<td>FLIR</td>
</tr>
<tr>
<td>Type</td>
<td>ImagerPro SC7000</td>
<td></td>
</tr>
</tbody>
</table>

![Fig. 2. Piping and instrumentation for the setup employed in this work.](image)
Digital image analysis is applied to ensure proper application of infrared calibration, using a method via detection of the dense emulsion phase in the simultaneously captured visual images. This digital image analysis also provides solids holdup information. Statistics are reported for temperature dynamics and solids fluxes, including higher order statistics. These results provide further insights into condensed mode operation and heat transfer limitations in gas fluidized beds. In addition the obtained results can be used for verification of computer models.

3. Experimental

A flat-bed geometry gas fluidized bed was constructed with a width of 80 mm and depth of 15 mm (see Fig. 1). The window was equally wide as the bed and 180 mm tall. To avoid static charging, the particle material (Table 1) as well as the setup front window were pre-treated with antistatic. Heater rods in the alumina back wall kept the back wall at constant pre-set temperature. Nitrogen gas preheated to the temperature of the back wall was fed to the windbox. Nitrogen at room temperature was used for gas bubble injection and as atomization gas in the liquid injection nozzle (Fig. 2). The nitrogen flow rate was metered and controlled via mass flow controllers.

A pair of cameras (see Table 2) was used to simultaneously observe the fluidized bed in the visual and infrared (MWIR) wavelengths. The selected integration times were found empirically to be sufficiently low to give observation of individual particles in the emulsion phase for this work. In order to ensure that both cameras can observe the bed through the same window, the window material was selected to be 3 mm thick sapphire, based on the operating wavelength ranges and resistance to the operating conditions. For the postprocessing it is essential that images are taken simultaneously by both cameras, at preset intervals. This was achieved by means of a pulse generator.

Sections 3.1 and 3.2 give detailed descriptions of the arrangements for pulsed gas bubble injection and liquid injection. More details on the experimental setup can be found in Kolkman et al. (2016).

Any desired assumption for pseudo-2D behavior in the chosen flat-bed geometry breaks down especially when the gas flow to the liquid injection nozzle is enabled. There occurs most certainly a temperature profile in the depth direction and the possibility cannot be excluded that undetected clusters of wet particles move behind a front layer of relatively hot particles. The setup with flat-bed geometry however does enable whole-field observation that can be used for validation of models to be used for detailed studies of the process. Above all, it brings observation of phenomena that thus far were not directly observable.

3.1. Arrangement for gas bubble injection

The bottom section of the experimental setup contained a special arrangement to achieve timed injection of pulses of gas, schematically shown in Fig. 3. The pulses were injected via a tube with 6 mm outer diameter, inserted through the center of the bottom plate. The tube had a wire mesh to prevent particles from falling through. The first mass flow controller provided a local gas flux...
over the tube consistent with that passing through the bottom plate. The second was at a pre-set flow rate, which could be directed into the bed via a three-way valve. In preliminary measurements it was verified with the pressure sensor sampling at 100 Hz, that indeed injection times of 0.1 s or 0.2 s could thus be achieved as set.

The exhaust system shown in Fig. 1 included a holder connected to a slide. This was used to suddenly drop particles at room temperature into the setup.

The gas bubble injection experiments proceeded as sketched in Fig. 4. With the back plate and inlet gas at a preset temperature of 80 °C, a pre-metered amount of particles was fluidized to equilibrate their temperature with the wall temperature. Next, the background gas velocity was reduced to below the minimum fluidization velocity (0.24 m/s at room temperature (Laverman et al., 2008)) and a pre-metered amount of particles was added. As soon as all the particles had dropped in, the experimenter started recording on the cameras and started the pre-programmed injection of a series of gas pulses. Settings common to all bubble injection measurements are given in Table 3.

### 3.2. Arrangement for liquid injection

Liquid injection was achieved via an installed airbrush nozzle with internal gas-liquid mixing (Sogolee AB200, 0.2 mm tip diameter, tip pressure drop 1.5–2.5 bar). It was installed at a height 35 mm above the bottom plate and injected horizontally and parallel to the back plate. This position at the side of the column, instead of via the bottom plate, was chosen for ease of (re)placement of the injection nozzle. Liquid feed was pressurized as shown in Fig. 2, by the controlled pressure of the gas phase in the supply vessel as well as via a liquid pump. The latter also served to circulate liquid through the pressurized liquid circuit to remove gas bubbles from the lines. The liquid flowmeter was a coriolis meter.

In the exhaust there was an arrangement to add additional nitrogen (Fig. 2), so that flammable components evaporated into the gas phase could be diluted to below their lower explosion limits. When doing experiments with such organic solvents, for safety also the top exhaust part in Fig. 1 was replaced by a leak-tight component.

Experiments with all three fluids given in Table 4 were performed. As seen from Table 4, the selected hydrocarbon fluids have a volumetric enthalpy of evaporation that is of an order of magnitude higher than that of water, a fluid that is more readily available and poses less risk in a laboratory environment. To account for this significantly higher enthalpy of evaporation by weight (Table 4) of water versus isopentane or hexane, the mass flow rate for water was chosen lower than for the other two fluids to avoid too significant cooling of the setup. Flow rates are given in Table 5.

In all cases the heat source for the heat of evaporation was the heat supplied to the solid phase by the temperature controlled back and side walls and bottom gas supply. In the case of injection of water, the temperature was set at the maximum the setup was considered to be able to handle, 80 °C, so below the boiling point of water. In the case of hexane injection, this temperature was set at 70 °C, so equal to the atmospheric boiling point of the fluid. Since the temperature of the solids phase will drop below this point, this hydrocarbon was injected into a solid phase at a temperature below its boiling point. For isopentane the bed wall temperature was set to 60 °C, below the expected boiling point at the pressure inside the nozzle (Stull, 1947). Upon nozzle exit, this liquid contrary to the others encounters a solid phase above its boiling point. These two extremes were chosen because under typical operating conditions in polyolefin reactors (e.g. 98 °C and 20.7 barg (DeChellis and Griffin, 1994)) the fluids in Table 5 are below their boiling points (Stull, 1947; Weast, 1977), although co-condensed components may boil off at initial solid-liquid contact.

The solid material in all presented cases is LLDPE particles (Table 1), except one. For reference, also one case with water injection into a bed of 0.5 mm diameter glass particles is presented, to Table 4

<table>
<thead>
<tr>
<th>Liquid</th>
<th>Isopentane</th>
<th>Hexane</th>
<th>Water</th>
</tr>
</thead>
<tbody>
<tr>
<td>b.p. (°C)</td>
<td>27.85</td>
<td>68.95</td>
<td>100.0</td>
</tr>
<tr>
<td>Density (kg/L)</td>
<td>0.6201</td>
<td>0.6603</td>
<td>1.000</td>
</tr>
<tr>
<td>Mol. wt. (g/mol)</td>
<td>72.15</td>
<td>86.18</td>
<td>180.16</td>
</tr>
<tr>
<td>$H_v$ (kJ/mole)</td>
<td>27.07</td>
<td>31.91</td>
<td>44</td>
</tr>
<tr>
<td>$H_v$ (MJ/L)</td>
<td>0.233</td>
<td>0.244</td>
<td>2.44</td>
</tr>
<tr>
<td>$H_v$ (MJ/kg)</td>
<td>0.38</td>
<td>0.37</td>
<td>2.44</td>
</tr>
</tbody>
</table>

**Bold values indicate those flows for which the quantity differs intentionally from the default settings.**

**Table 5**

<table>
<thead>
<tr>
<th>Inj. liquid</th>
<th>$u_{inj}$ (m/s, STP)</th>
<th>$\Phi_{inj}$ (L/min, STP)</th>
<th>$\Phi_{inj}$ (g/min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hexane</td>
<td>0.68</td>
<td>0</td>
<td>6.3</td>
</tr>
<tr>
<td>Hexane</td>
<td>0.68</td>
<td>7</td>
<td>6.3</td>
</tr>
<tr>
<td>Hexane</td>
<td><strong>0.34</strong></td>
<td>7</td>
<td>6.3</td>
</tr>
<tr>
<td>Isopentane</td>
<td>0.68</td>
<td>7</td>
<td>6.3</td>
</tr>
<tr>
<td>Isopentane</td>
<td><strong>0.34</strong></td>
<td>7</td>
<td>6.3</td>
</tr>
<tr>
<td>Water</td>
<td>0.68</td>
<td>7</td>
<td><strong>1.3</strong></td>
</tr>
</tbody>
</table>

*Fig. 5. Simplified distributions of temperature in particle temperature, and resulting histograms. The statistics standard deviation $\sigma$ and modified third moment $\tau$ are used to provide information on the shape of the distribution.*

---

**Table 3**

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_{wall}$</td>
<td>80 °C</td>
</tr>
<tr>
<td>$u_{bg}$ (m/s) at 80 °C</td>
<td>0.16</td>
</tr>
<tr>
<td>Bottom gas flux (kg/m²/s)</td>
<td>0.18</td>
</tr>
<tr>
<td>$n_{inj}$</td>
<td>5</td>
</tr>
<tr>
<td>$\Phi_{inj}$ (L/min, STP)</td>
<td>12</td>
</tr>
<tr>
<td>$f_{sample}$ (Hz)</td>
<td>50</td>
</tr>
<tr>
<td>$n_{samples}$</td>
<td>525</td>
</tr>
</tbody>
</table>
show a case in which capillary forces are expected to be of greater importance than for the LLDPE particles. The difference also demonstrates the importance of the selection of proper model materials.

Before recording was started, the liquid injection was already operating steadily for several minutes (at least five) until quasi-steady state was observed in the infrared signal monitored real-time on the computer screen, after which the recording was started. Then, data was recorded at a frequency of 5 Hz for at least two minutes, typically three. Afterward the attainment of quasi-steady state was also verified on the recorded data from a plot of the temperature distribution versus time.

4. Digital image processing

The digital postprocessing used in this work is explained in detail in Kolkman et al. (2016). The first step is to align the simultaneously taken raw images from both cameras to the same grid. In the lab, the infrared camera was always taking images at maximum resolution. It was typically aligned such that it captured the lower square of the bed (the zone of highest interest) and a bit of the wall around it. This was done to provide a reference and to ensure that the entire bed width was recorded. For the final resolution of the infrared images a square of 256 × 256 pixels was chosen, so that in post-processing for the alignment digital interpolation and not extrapolation is done. This is the resolution of the infrared images finally presented in this work. With the bed width of 80 mm this thus gives a final resolution of 3.2 pixels per millimeter, the latter being the typical length scale of the polymer particles used. Thus individual particles can be observed in the final resulting infrared images.

The images obtained from the visual camera are used to distinguish zones containing dense and dilute emulsion phase. Dilute zones are here defined as zones in which observation into the bed depth for multiple particle diameters is possible, typically showing the back wall. Dense zone should show only the front layer of particles and that directly behind it. Contours around dilute zones found via the empirically tuned image processing are illustrated e.g. in Fig. 6. This is applied as a pre-processing step for particle image velocimetry (which is also based on the visual images), to remove any particles “raining” through relatively large bubbles (Laverman et al., 2008) and to account for the solids holdup in the determination of the local solids flux. The

<table>
<thead>
<tr>
<th>Table 6</th>
<th>Variation of configuration of gas bubble injection experiments.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pulse interval (s)</td>
<td>2.0</td>
</tr>
<tr>
<td>Pulse duration (s)</td>
<td>0.1</td>
</tr>
<tr>
<td>( V_{bh} + V_{ba} ) (mL)</td>
<td>( 30 + 30 ) Case L1</td>
</tr>
</tbody>
</table>

Fig. 6. Snapshots for bubble location for cases L1 and H1 (Table 6). The time indicated is the time since the first frame with observed influence of the gas pulse injection. The two types of contours shown correspond with two complementary methods of distinguishing dense and dilute zones, explained in more detail in Kolkman et al. (2016). Both these complementary methods are applied simultaneously.
information on the location of the dense emulsion phase obtained from visual images is also applied in the processing of infrared images, to identify zones in which the emulsion phase is in a condition for which calibration is available.

The alignment of images with the standardized grid for the setup is done manually. The alignment of the images of the visual camera, always observing the bed head-on, was based on the interface between wall and bed. The subsequent alignment of the infrared image, on the contrary, was always based on the alignment of bubbles as observed inside the bed at the front window. This is because the window and front wall plate protrude in front of the front bed surface (Fig. 3). Since the infrared camera observes the bed at an angle from the right (varying between experiments), it thus can occur that part of the right side of the bed is obscured by the wall, showing as a zone of low temperature due to the low infrared emissivity of the wall. Results of the digital image analysis were inspected for both the infrared camera and the visual camera. In all cases bubble detection worked satisfactorily, despite the simplicity of the principles applied. A general advice for improvement in future work can be to align the infrared camera close to perpendicular to the setup, to reduce the distortion required for the alignment. A trade-off is to be chosen between reducing the angle and reducing camera distance, while keeping the infrared camera out of view of the visual camera. A potential source of error in the manual handling of images is the manual selection of the first image in which the effect of gas pulse injection is visible. The deviation is at most one frame, so 0.02 s.

The principle behind infrared thermography is that the digital signal obtained from an infrared camera can be related to the temperature of the object from which radiation was received by the camera. There are some complications. In particular, there is a distribution in the camera signal received for a fluidizing bed even if there is no intentional cooling introduced in the system. The width of this distribution increases with increasing difference between bed temperature and room temperature. The distribution is in part due to reflections and it is suspected that this is in part due to a particle temperature distribution existing in the particle bed. The method of the calibration procedure applied is described in detail in Kolkman et al. (2016). Sample data used in calibration was taken on a fluidizing bed for various pre-selected temperatures, to obtain the signal distributions including the effect of reflections. It was endeavoured to account for the distribution in particle temperature. A linearization was introduced to suppress any skewing effect that the occurrence of the distribution of signals (instead of just a single value) corresponding to a single bed particle temperature might have on the calculated average bed temperature. Two methods of averaging are employed in this work. The first is local time averaging. If, however, for each moment in time statistics are to be obtained, the average of a row of pixels in a single snapshot can be used to average out variation. The latter, second method will be referred to as the horizontal average. In order to filter out effects of minor errors in the detection of the inclined bed surface position, values for the horizontal average are only reported for a particular vertical position if for that data points are available for a threshold chosen as at least \( \frac{1}{16} \)th of the width of the bed.

**Fig. 7.** Snapshots for bubble location for cases L2 and H2 (Table 6). The main difference with the cases shown in Fig. 6 is that the duration of the gas pulse has been doubled.
In the infrared thermography, there is also an influence of the temperature of the window at the front of the setup. For experiments with injection of gas pulses, it is assumed that the window temperature is uniform and equal to the imposed temperature of the back wall. For experiments involving liquid injection, it is assumed that the local window temperature is equal to the observed local time-averaged temperature. An option for further future improvement could be determining the local temperature of the window via an infrared camera that operates in a wavelength range in which the window is not transparent.

5. Statistics representing distributions

In this work it is expected to be observing various particle temperatures for any given position in the gas fluidized bed. Presenting only the local average \( \mu \) (Eq. (1)) would lead to loss of information. Presenting additionally the standard deviation \( \sigma \) (Eq. (2)) provides a means to demonstrate whether the average is a good representation for all members in the collection (Case I in Fig. 5), or whether there is a distribution of values around this average (Cases II and III in Fig. 5).

A modification of the third moment is given by Eq. (3) and designated with \( \tau \). This parameter serves to detect a skew in the distribution. If the distribution is symmetrical (Cases I and III in Fig. 5), the value of \( \tau \) is zero. If the distribution is skewed to the lower values (Case II in Fig. 5), it is negative. If it is skewed to higher values the value is positive.

\[
\mu = \frac{1}{n} \sum x_i \tag{1}
\]

\[
\sigma = \sqrt{\frac{1}{n-1} \left( -n\mu^2 + \sum x_i^2 \right)} \tag{2}
\]

\[
M_3 = \frac{1}{n-1} \sum (x_i - \mu)^3
\]

\[
\tau = \text{sgn}(M_3) \sqrt{|\text{sgn}(M_3)M_3|} \tag{3}
\]

Scalar data, such as the average temperature, is represented primarily in color scales. The statistics defined by Eqs. (1) and (2) are also applied for results of particle image velocimetry. The standard deviation is represented by rectangles, with the dimension in a particular direction proportional to the magnitude of the standard deviation of the distribution in that direction.

For the bubble injection experiments, for the infrared as well as for the solids holdup, integration was done over all frames taken at 50 Hz over the total experiment time of 10.5 s. For the liquid injection experiments integration was done for all frames taken at 5 Hz over an interval of at least 2 min.

6. Results for injection of gas bubbles in a dry bed

This section presents the results for the bubble experiments as introduced in Section 3.1. Results are presented for four selected combinations of pulse duration (single (“1”) or double (“2”) length) and amount of solids (low (“L”) or high (“H”)), as given in Table 6.

6.1. Bubble trajectory

The primary phenomenon visually observed when applying the conditions and gas pulses, as defined in Tables 3 and 6, is the passage of gas bubbles through the emulsion phase. Figs. 6 (pulse duration 0.1 s) and 7 (pulse duration 0.2 s) each show snapshots of the injected bubbles taken at 0.1 respectively 0.2 s after the start of pulse injection. Up to 0.1 s the injection of the gas pulse is identical for the pair L1 and L2 (each 60 mL of particles) and for the pair H1 and H2 (each 90 mL of particles). For these pairs indeed the bubbles observed after 0.1 s closely correspond. Beyond this point in time, for cases L1 and H1 the injection of the gas pulse ceases. As the bubble approaches the bed surface, it collapses. For cases L2 and H2 injection of the gas pulse proceeds, and as the wake becomes inclined the bubble obtains a kidney shape. The effect of the protruding wake is also found from Fig. 8, showing that for the case with a relatively long gas pulse (case H2) in a relatively large fraction of samples near the center of the bubble trajectory, dense emulsion phase is detected by digital image analysis. For the short pulse (case H1) this effect is absent.

After the bubble collapses, the top bed surface at rest typically is inclined towards the center of the bed (see e.g. Fig. 10(a) and (b)).

6.2. Temperature equilibration

In this section the temperature distribution in the particle bed is tracked as a function of time. This is done for the four pulse injection experiments defined in Table 6.

Fig. 8. Fraction of samples from the visual images, per position, detected to contain dense emulsion phase for cases H1 (a) and H2 (b) (Table 6). The color light blue at a given position indicates that no valid samples were detected for this position at all. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)
Fig. 9 shows side by side the images taken simultaneously by the infrared camera (left) and the visual camera. The visual image corresponds with that in Fig. 7, for case L2 at $t = 0.2$ s. By comparison with the initial condition illustrated in Fig. 4 the infrared image demonstrates that the injection of the first pulse has caused solids at relatively high temperature to have been transported upward through the center.

Fig. 10 demonstrates snapshots obtained from the infrared camera, uncorrected for intrinsic variation, for case H2 (Table 6). The passage of bubbles is seen to cause solids to be convected upward in the center of the setup, and down along the side. Fig. 10(c) shows the local time average temperature after the injection of five pulses. Here the intrinsic variation has locally averaged out, but information on the time sequence of events was lost.
**Fig. 11** shows the time sequence of pulse injection by presenting for each vertical position the horizontal average of the temperature (vertical axis), as a function of time (horizontal axis). The injection of the gas pulses is indicated by the rectangles on top of the diagram. Their width represents the duration of the pulse. The passage of the bubbles is seen as relatively warm particles being pushed up, and then again dropping down, into the bed. The stepwise convection of particles at relatively low temperature downward into the bed is also observed in **Fig. 11(a)**. It is seen that between the pulses, the temperature distribution in the bed is fairly constant and heat transfer by diffusion plays only a minor role. A representative sample for the horizontal temperature distribution therefore can be taken at the times indicated by the arrows below **Fig. 11(a)** along with another at the initial condition \( t = 0 \) s. The result is shown in **Fig. 11(b)**. At the top of the bed (\( y = 70 \) mm) the horizontally averaged temperature increases as relatively warm particles are pushed upward with the passage of the bubble. After the final pulse, the horizontally averaged temperature has become higher at \( y = 70 \) mm than at \( y = 50 \) mm. At this point a significant layer of particles at relatively high temperature covers the initial top layer of particles at relatively low temperature, as also shown in the snapshot of **Fig. 10(b)**. This inversion is not observed in the sequence H1, with shorter pulses, for which results are shown in **Fig. 11(c)**.

In both **Fig. 11(b)** and (c) the wall temperature is given by the curve \( t = \infty \). In absence of heat losses to the environment the bed should equilibrate to this temperature with time. It is seen that in the bottom region of the bed a temperature higher than the imposed wall temperature is found. This is most likely due to an overcorrection for heat losses in setting the bed wall temperature. Also, since the initial layers exceed the expected height in the setup of 50 mm for 60 mL particles, from the bed height observed in the results it can be observed that the bulk volume of particles inside the setup is larger than given in Table 6. This may be due to a bias in reading the scale on the beaker, increase of particle size with increasing temperature or due to the increase of the fraction of particles in contact with the wall.

**Fig. 11** shows the application of higher order statistics to the local temperature observations for case H2. Due to the intrinsic variation of the temperature as determined from the bed signal, these can be used only as qualitative indicators (Kolkman et al., 2016). In the lower corners, the standard deviation is zero. Due to the relation between the intrinsic infrared signal variation and particle geometry (the cavities between the particles radiate more), this means that in the corners the particles have not moved at all. In between the corners there is a minor standard deviation, indicating the particles have moved position but have not been exchanged for particles of significantly different temperature. Similarly, such zones at the top left and right indicate that relatively cold particles have been present throughout the observation. In the top center there is a strong standard deviation, since there relatively cold particles have been replaced by particles of different temperature. Two additional zones of high deviation, directed from the top corners to the bottom center, show the zone into which the relatively cold particles have been transported.

Additional information on the stepwise transportation of the solids is provided by the modified third moment. In the average (**Fig. 10(c)**), it is seen that in all zones with significantly non-zero
(\(|\tau| > 3{\,}^\circ\mathrm{C}\) modified third moment at some point in time there have been particles at relatively low temperature. A strongly negative modified third moment indicates that relatively cold particles have been present less than half of the time for the top middle. A negative value is also found in the zones into which colder particles have only been transported in a late stage. Zones which initially contained relatively warm particles, but in which relatively cold particles are found more than half of the time, have a strongly positive value for the modified third moment. The modified third moment \(\tau\) therefore serves as an indicator of the fraction of time particles of a certain temperature offset, relative to the mean, are found in a zone.

The small asymmetry of the results relative to the center axis is the result of a slight initial asymmetry in the distribution of the added particles. As a result, the first injected bubble followed a leftward trajectory, while at the same time the initial height of the layer of relatively cold particles was relatively thick on the right. This demonstrates the distribution found for this configuration is very sensitive to the initial conditions.

For brevity for cases L1 and L2 (Table 6), so cases for which in total 60 mL particle material was loaded into the setup, only the time sequence is shown in Fig. 13. The omitted statistics on the time-average solids holdup, per position, are very similar to those for cases H1 and H2 in the lower part of the bed. In case L2 the relatively cold particles reach the bottom zone into which the gas pulse is injected after the third gas pulse. As a result, they get ejected by the next pulse into the freeboard, and subsequently onto the top of the bed, then already covered by relatively warm particles. For case L1, with the shorter pulse duration, vertical particle displacement is slower.

7. Results for injection of liquid

In this section the results for experiments under fluidizing conditions, with liquid injection as defined in Section 3.2, are presented.

7.1. Fluidization without liquid injection

Fluidization without liquid injection was presented before (Kolkman et al., 2016). The flow dynamics show the typical upflow of solids around the center axis and downflow along the walls, with a slight asymmetry ascribed to imperfection of the bottom plate. For temperature statistics it is shown that there is an intrinsic fluctuation in the observed signal even without the introduction of intentional cooling (\(\sigma \approx 1.8{\,}^\circ\mathrm{C},\ \tau \approx 1\{^\circ\mathrm{C}\)\). The injection nozzle protrudes the right wall at a height of 35 mm and is fed with gas at room temperature. The injection of cold gas via the injection nozzle only has a minor effect on the bed temperature, as seen from Fig. 14. The temperature being slightly higher than the intended 80\(\,^\circ\mathrm{C}\) is most probably due to an overcorrection for heat losses in setting the back wall temperature.

7.2. Fluidization with hexane injection without nozzle gas

Hexane was injected via the liquid injection nozzle, without atomization gas being fed to the nozzle. In this case the liquid exits the nozzle as a thin jet of liquid. The statistics on the solids fluxes are very similar to those in the case without liquid injection (given by Kolkman et al. (2016)) and therefore not shown here. When observing raw visual images of the fluidized bed (Fig. 15(a)), there is no clear sign of change due to the liquid injection. The image taken at the same moment with the infrared camera (Fig. 15(b)), however, reveals clusters of relatively cold particles. These appear to form a trace that originates from the position \(x = 50\,\mathrm{mm}\) on the bottom plate. Fig. 15(c) shows the infrared image that was taken 0.2 s after that of Fig. 15(b). Again a relatively large cluster of colder particles is seen. This relatively large cluster is seen at practically the same position for six consecutive frames, so one second, suggesting it is the same conglomerate. The conglomerate disappears from view and gets replaced by, supposedly, another. This sequence repeats itself.

Fig. 16(a) shows the time-averaged temperature, which has dropped clearly below the wall temperature at 70\(\,^\circ\mathrm{C}\). However, the time average does not reflect the unsteady observations described for Fig. 15. On the contrary, statistics on the variation in the observed temperature reveal that the strongest skew in temperature distribution is found near the center of the bottom plate. The modified third moment \(\tau\) (Fig. 16(c)) now has a negative value, contrary to dry fluidization (Section 7.1). This is in line with the occasional observation of a relatively cold particle in a bed of otherwise uniform temperature (see Fig. 5).

Given the injection of liquid from the right hand side, the approximation of symmetry around the vertical center axis of the bed in Fig. 16 is unexpected. The observations are in line with a mechanism in which particles are initially wetted and cooled by
the injection of liquid at the injection nozzle, and transported with the emulsion phase towards the lower center of the bed, at which they appear in view in the relatively stagnant zone around the center of the bottom plate, from $x = 20$ mm to $x = 60$ mm. The fact that the conglomerate stays put on the bottom plate suggests the occurrence of (supposedly capillary) cohesive forces. A relatively cold conglomerate of particles dropping in one piece through a bubble, as shown in Fig. 17, proves the occurrence of such forces. To counter the formation of liquid-solid conglomerates, Section 7.3 presents an enhancement to the operation of the liquid injection nozzle.

7.3. Injection of hexane with nozzle gas

The liquid injection nozzle is designed to be operated with a supply of a gas flow, in addition to liquid. In the application for liquid injection into a gas-fluidized bed, this has twofold action. First, it atomizes the liquid, so that it is converted into small droplets. Second, it introduces an additional gas jet, carrying liquid droplets into the bed. Fig. 18 presents statistics for solids fluxes with both liquid injection and gas injection via the injection nozzle. The gas flow rate via the bottom plate is the same as before. The gas added via the injection nozzle has a strongly shaping effect on the solids flow pattern. However, fluctuations (Fig. 18(c)) are still in the order of magnitude of the time average solids flux (Fig. 18(b)), with the notable exception of the lower right, showing a steady mixing cell. The time averaged solids flux is the solids flux observed at the front window. It shows solids flowing towards the nozzle injection position at 35 mm above the bottom plate on the right. The acceleration of particles by the injected gas is obscured from view, but solids re-appear on average at the window at the position 25 mm from the left, 50 mm from the bottom approximately. The zone primarily occupied by bubbles in the top of the bed has shifted somewhat to the right, the side at which the nozzle injects gas. Fig. 19 shows the key statistics for the temperature distribution.

The temperature drop of the particle bed, compared to the wall temperature is very similar to the case without atomization gas.
Fig. 15. Instantaneous images, taken at the same moment, from visual camera (a) and infrared camera (b). Image (c) was taken 0.2 s after image (b). Hexane injection at 6.3 g/min with the wall temperature at 70 °C and a background gas velocity of 0.68 m/s (STP). The small arrow on the right (figures (b) and (c)) indicates the locus and direction of liquid injection.

Fig. 16. Temperature statistics for hexane injection with wall temperature 70 °C and a background gas velocity of 0.68 m/s (STP), without atomization gas. The small arrow on the right indicates the locus and direction of liquid injection.
(Fig. 16), but it is more localized. The average temperature shows a clearly cut-off triangular zone above the bottom plate from \(x = 50\) mm to \(x = 80\) mm. At the interface of this zone, the standard deviation for the observed temperature is high and the modified third moment shows a zone of remarkably high values, compared with the rest of the bed. The statistics for the solids fluxes (Fig. 18) assist in interpreting the origin by showing that this is the aforementioned localized mixing cell. There is no trace of relatively large conglomerates of colder particles in the snapshots, nor in the statistics. There are conglomerates of a few particle diameters wide (not shown). By comparing Figs. 18(b) and 19(c) it is seen that the zone in which these conglomerates show up, as indicated by the negative value of the modified third moment, coincides with the zone towards which solids are transported from the point at which solids appear after passing the nozzle zone \((x = 25\) mm, \(y = 50\) mm).

### 7.4. Injection of hexane with reduced background gas velocity

Injection of hexane was performed with atomization gas fed at 7 L/min (STP) to the injection nozzle. To observe what happens under badly fluidizing conditions, the background superficial velocity was set at a relatively low value. Bubble activity in general is low, but very pronounced above the liquid injection nozzle (Fig. 20(a)). At bed heights below the nozzle injection position, the average solids flux is low (Fig. 20(b)). The reduced mobility of the solids phase leads to a strong time-averaged temperature gradient (Fig. 20(c)).

![Fig. 17. Observation of a conglomerate of particles at relatively low temperature inside a bubble for hexane injection at 6.3 g/min with a background gas velocity of 0.68 m/s (STP) and without atomization gas.](image1)

![Fig. 18. Hydrodynamics for hexane injection with 7 L/min (STP) atomization gas fed to the nozzle. Wall temperature at 70 °C and a background gas velocity of 0.68 m/s (STP).](image2)
7.5. Injection of isopentane

Isopentane is injected into a particle bed that is at a temperature well above its atmospheric boiling point, so that there are no mass transfer limitations in the evaporation of the liquid. The results are, however, very similar to the injection of hexane with atomization gas and therefore only a small selection is shown in figures, for brevity. The atomization gas has a strong influence in shaping the time-average solids flow and the flow profiles are very

Fig. 19. Temperature statistics for hexane injection with 7 L/min (STP) atomization gas fed to the nozzle. Wall temperature at 70 °C and a background gas velocity of 0.68 m/s (STP).

Fig. 20. Statistics for flux and holdup of solids, as well as temperature statistics, for hexane injection with 7 L/min (STP) atomization gas fed to the nozzle. Wall temperature at 70 °C and a background gas velocity of 0.34 m/s (STP).
similar to those for the injection of hexane. At a background superficial gas velocity of 0.68 m/s, in the modified third moment for the temperature, spikes due to significantly colder-than-average particles are seen throughout the bed, indicating cold particles are transported throughout the bed. At this background gas velocity the time-average temperature is also rather uniform (Fig. 21(a)), but the relatively high temperature near the walls and in the mixing cell on the lower right stand out.

When the background fluidization gas velocity is reduced significantly, convection of the solid phase is reduced as well. Indicators (a negative value of $\tau$) of relatively cold particles being transported are not longer seen in the lower right of the bed. A strong gradient in temperature, on average (Fig. 21(b)), results.

The data obtained for the particle temperature shows, in a small fraction of the data in particular for isopentane injection, single particle temperatures between 40°C and 50°C. Due to the closely linear operation observed for the infrared camera (after precorrection) and general experience with the camera, the linear trend is expected to extend into this region below the lowest temperature (50°C) for which bed temperature was calibrated, so that there is no major effect on the results obtained. Future experiments should however employ a wider calibration range for bed temperatures.

7.6. Injection of water

Fig. 22 shows that the introduction of water leads to the observation of relatively cold single particles being transported throughout the bed. The statistics for solids holdup, solid fluxes and temperature again are very similar to the case of hexane and are omitted for brevity.

7.7. Agglomeration for glass beads

Injection of water into a bed of soda lime glass beads (Table 1) of 0.5 mm diameter under the conditions given in Table 7 led to the formation of conglomerates of solids, that can be observed in Fig. 22. Snapshot from visual camera (a) and infrared camera (b) for injection of water into a bed of LLDPE particles with the wall temperature at 80°C, a background gas velocity of 0.68 m/s and a nozzle gas flow rate 7 L/min (STP).

<table>
<thead>
<tr>
<th>Settings for case with water injection with glass beads.</th>
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<tbody>
<tr>
<td>Case</td>
</tr>
<tr>
<td>$u_{bg}$ (m/s, STP)</td>
</tr>
<tr>
<td>$\Phi_{m,bg}$ (kg/m²/s)</td>
</tr>
<tr>
<td>$\Phi_{b}$ (L/min, STP)</td>
</tr>
<tr>
<td>$V_{bulk}$ (mL)</td>
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<tr>
<td>$\Phi_{m1}$ (g/min)</td>
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<tr>
<td>$T_{wall}$</td>
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Fig. 21. Statistics for injection of isopentane at 6.3 g/min with atomization gas at 7 L/min (STP). Background gas velocities are 0.68 m/s (a) and 0.34 m/s (b), respectively, the wall temperature was 60°C.
dropping through bubbles. This can happen despite the supply of atomization gas to the injection nozzle. In the infrared, these can additionally also be detected when submerged in the emulsion phase adjacent to the window and have a lower temperature than the surrounding emulsion phase, presumably due to the evaporating liquid they contain. This is illustrated in Fig. 23. The liquid cannot be separately discerned. The agglomerates, however, can move out of view by moving deeper into the emulsion phase and therefore cannot be tracked from their formation to their destruction. It would be possible to obtain statistics on the size and location of such conglomerates throughout the front face, as a function of the conditions applied. This is, however, outside the scope of the current work.

The application of the glass beads in this work was not straightforward, since the antistatic strongly affects the interparticle forces. There was a fine line between the bed becoming static (too little antistatic) and the particles cluttering (too much antistatic). From the observation that the same goes for some glassware, it is suspected that the glass beads are weakly transparent in the waveband of the infrared camera (MWIR) applied in this work. This should be considered in further work applying the glass beads, although this observation might not occur for infrared cameras operating in a different wavelength range such as LWIR (8–14 μm). The relatively strong tendency to conglomerate, compared to the hexane/LLDPE experiments, stresses the importance of choosing the proper particle material and liquid to obtain relevant results.

7.8. Relation to industrial application

The small flat-bed geometry setup at atmospheric pressure certainly does not lend for straightforward scale-up of the results to assist in the design of an industrial reactor. As stated in Section 3.2, the geometrical position of the nozzle was chosen for practical reasons and does not convey an advise for industrial design. The experiment does however demonstrate a few phenomena that should be taken into account in design:

- Heat transfer limitations can occur in a gas fluidized bed. Solids mix-up should be ensured (and promoted).
- Liquid-solid agglomerates can be formed under fluidization for injection of hydrocarbon fluids with polymer particles.
- Formation of agglomerates is discouraged by co-injecting the liquid with gas, but might not be prevented depending (at least) on the particle size distribution.

Furthermore, results from the presented experiments may be used in the verification of computational models used to assist in the design of industrial reactors.

8. Conclusions

A combination of a high speed visual and an infrared camera and digital image analysis has been employed to obtain whole field data on hydrodynamics and temperature distribution for a flat-bed geometry gas fluidized bed. This was done for experiments with timed gas pulse injection and for experiments with liquid injection.

Injection of gas bubbles into a stagnant bed with particles with strongly different initial temperatures demonstrated that the pulse duration strongly affects the resulting solids mixing patterns and demonstrated that statistics other than time averages can provide valuable insights.

Injection of water, isopentane and hexane into a bed of 1.0–1.3 mm LLDPE particles gave similar results for hydrodynamics and temperature statistics in case the liquid was injected together with atomization gas. Injection of hexane without atomization gas resulted in clusters of colder particles reaching the bottom plate of the setup. Injection of water in a bed of 0.4–0.6 mm glass beads resulted in the formation of rigid agglomerates, that can be directly observed using the infrared camera. This happened despite the supply of atomization gas.

The introduction of higher order statistics in the description of solid fluxes has shown beneficial in improving understanding of the development of the time-averaged temperature profile, by pinpointing a steady local mixing cell. The modified third moment is a valuable indicator of the presence of individual significantly colder (or hotter) particles, the size of which was confirmed by evaluating snapshots of raw data.

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References


