Particle transport in fluidized beds
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Chapter 4

Freely Bubbling Batch Fluidized Bed: Model Validation

4.1 Description of a Stochastic Model for Freely Bubbling Batch Fluidized Bed

The model that we propose for the freely bubbling batch fluidized bed of this chapter, is based on Dehling and Hoffmann’s (1999) modeling concepts for particle transport in fluidized beds. That model, which was outlined in Chapter 3, was designed for a continuously operated fluidized bed. As our experiments were carried out with a batch fluidized bed, where there is no inflow and outflow of particles, the model of Dehling and Hoffmann (1999) required appropriate modification.

In our model, the motion of a single particle in the reactor is defined as a stochastic process in which only the vertical position of the particle is modeled and the radial motion is ignored. The model reflects the physical processes governing the vertical particle transport that have been proposed by Rowe and Partridge (1962), including:

- Upward transport in bubble wakes and deposition on top of the bed,
- Downward transport in the bulk due to the removal of material low in the bed in bubble wakes
- Dispersion due to the disturbance of the bulk material by fluidization bubbles.

Our model is a Markov process $(X_t)_{t \geq 0}$ where $X_t$ gives the vertical position of the particle in the reactor, measured from the top. We first formulate a discrete model, where the bed is divided into $N$ horizontal segments of equal width $\Delta$. In this case $X_t$ gives the index of the cell, where we start counting at the top.

For the stochastic model we can compute the probability distribution of a single particle at time $t \geq 0$. This distribution can be described by a probability vector $(p(t,1),\ldots, p(t,N))$ in a discrete model and by a probability density $p(t,x)$ in a continuous model. When studying the dispersion of a pulse of marked particles,
the model can be applied to predict the evolution of the density of marked particles in the reactor. This is a consequence of the law of large numbers, which states that for a large number of independently moving particles, the fraction of particles in any part of the reactor equals the probability that a single particle is found in that part. Thus \( p(t,i) \) in the discrete model gives the fraction of particles from the pulse that is found in cell \( i \) and \( p(t,x) \) in the continuous model gives the concentration of marked particles, i.e. fraction per length and, since we base our analysis on unit cross-sectional area, therefore per volume.

![Figure 4.1 A discretized fluidized bed.](image)

When applying a single particle model to predict the dispersion of a pulse of marked particles, the independence assumption is crucial. In the experiments of this chapter, independence is a reasonable assumption, because the marked particles form a small percentage of the particles in the reactor and thus have little or no interaction.

Our modeling approach starts from a model with discrete state space and in discrete time. Space discretization is achieved by subdividing the reactor into \( N \) horizontal segments of equal width \( \Delta = \frac{h}{N} \), where \( h \) denotes the height of the reactor, see Figure 4.1. The state space for the discrete model is \( \{1, \ldots, N\} \), and by \( X_t \) we denote the index of the cell containing the particle at time \( t \). Time is discretized by considering the process at times \( t = n\varepsilon, n = 0,1,\ldots \) only.

During one time step, between \( t = (n-1)\varepsilon \) and \( t = n\varepsilon \), four transitions are possible. The particle can stay in the same cell, move either one cell upward or downward or
it can be caught in the wake of a rising fluidization bubble and be deposited at the
top of the bed, i.e. move to cell 1. The corresponding transition probabilities
\[ p_{ij} = P(X_{ne} = j \mid X_{(n-1)e} = i) \]
are parameters of the model. Let \( \alpha_i, \beta_i, \delta_i \geq 0 \) with \( \alpha_i + \beta_i + \delta_i = 1 \) and \( \lambda_i \geq 0 \) be
given, \( i = 1, \ldots, N \). We then assume the transition probabilities
\begin{align*}
    p_{i,i+1} &= \beta_i (1 - \lambda_i) \\
    p_{i,i} &= \alpha_i (1 - \lambda_i) \\
    p_{i,i-1} &= \delta_i (1 - \lambda_i) \\
    p_{i,1} &= \lambda_i \\
\end{align*}
(4.1)
Thus \( \lambda_i \) is the probability that a particle that is presently in cell \( i \) gets caught in the
wake of a rising fluidization bubble and is subsequently dropped in cell 1. Given
that this does not happen, the particle either moves one cell ahead, stays in the
same cell or moves on cell back, with probabilities \( \beta_i, \alpha_i, \delta_i \) respectively. This part
of the process, for particles in the bulk phase, specifies a birth and death process.
The transition probabilities specified in Equation (4.1) hold for \( i = 2, \ldots, N-1 \).
For \( i = 1 \) and \( i = N \) we have to specify boundary conditions, which we give as follows
\begin{align*}
    p_{1,1} &= 1 - \beta_1 (1 - \lambda_1), q_{1,2} = \beta_1 (1 - \lambda_1) \\
    p_{N,N-1} &= \delta_N (1 - \lambda_N), q_{N,N} = 1 - \delta_N (1 - \lambda_N) - \lambda_N, q_{N,1} = \lambda_N.
\end{align*}
These are the reflecting boundary conditions.
In Figure 4.2 we have plotted a sample path of the Markov chain with transition
probabilities. A useful picture is that in the first instance we perform a birth- and
death-process with transition probabilities specified by \( \beta_i, \alpha_i, \delta_i \) and that after each
transition a biased coin is tossed, which with probability \( \lambda_i \) shows Tail indicating
a move to the top of the bed, and with the remaining probability \( 1 - \lambda_i \), for no
further action.
Chapter 4: Freely Bubbling Batch Fluidized Beds: Model Validation

The goal of one type of experiments that we performed using the PET-technique was the visualization of the path of a single marked particle, in order to allow comparison with the paths predicted by our model. The results are presented in section 4.4.

For the comparison of experimental results and model predictions for the dispersion of a pulse of marked particles, we have to compute the probability distribution of $X_t$. If we define

$$ p(n\varepsilon, j) := P(X_{n\varepsilon} = j), $$

we get the recursion formula

$$ p(n\varepsilon, j) = \sum_{i=1}^{N} p((n-1)\varepsilon, i) \cdot p_{ij}. $$

For the probability vector $p(n\varepsilon) := (p(n\varepsilon, j), 1 \leq j \leq N)$ this yields the recursion formula $p(n\varepsilon) = p((n-1)\varepsilon) \cdot P$, which we can iterate to obtain

$$ p(n\varepsilon) = p(0) \cdot P^n. $$

Figure 4.2 A sample path of the Markov chain with transition probabilities.
Here $p(0)$ gives the initial probability distribution of the particle: i.e. for a particle starting in the top or in the bottom cell, $p(0) = (1, 0, \ldots, 0)$ or $p(0) = (0, \ldots, 0, 1)$, respectively. In Figure 4.3 we have plotted the particle distribution for $t = 1, 2, 3, 4$ and 10 for $p(0) = (1, 0, \ldots, 0)$.

We derive the model parameters $\alpha_i$, $\beta_i$, $\delta_i$ and $\lambda_i$ from the continuous quantities $v(x)$, $D(x)$ and $\lambda(x)$, for $0 \leq x \leq h$, which give the mean velocity, the mean square displacement per second and the rate of return to the top of the reactor, respectively, for a particle located at $x$. The specification is as follows,

$$\beta_i = \frac{\varepsilon}{2\Delta^2}D(i\Delta) + \frac{\varepsilon}{2\Delta}v(i\Delta) \quad (4.2)$$

$$\delta_i = \frac{\varepsilon}{2\Delta^2}D(i\Delta) - \frac{\varepsilon}{2\Delta}v(i\Delta) \quad (4.3)$$

$$\lambda_i = \varepsilon\lambda(i\Delta) \quad (4.4)$$
and $\alpha_i = 1 - \delta_i - \beta_i$. The argument for this choice of parameters is the same as in section 3.7. We also impose a relationship between $\epsilon$ and $\Delta$ by requiring $\epsilon = \frac{\Delta^2}{2D_0}$, with $D_0 = \sup_{0 \leq x \leq 1} D(x)$. If $\nu(x)$, $D(x)$ and $\lambda(x)$ are all strictly positive, continuous functions, all the above parameters fall into the interval $[0, 1]$ and are thus probabilities, if $\Delta$ is small enough.

In Chapter 2 we have shown how the excess gas flow determines bubble size, wake fraction and finally wake flow in the reactor. The wake flow $Q(x)$, $0 \leq x \leq h$, again determines both the mean particle velocity $\nu(x)$ as well as the rate of return to the top. The wake flow is an upward flow of particles, which for reasons of balance of particles must be compensated by a downward flow of particles in the bulk phase. When relating our model to a physical bed, the model neglects the volume occupied by the bubble/wake regions, and the whole model description is based on unit cross-sectional area, so that we get $\nu(x) = Q(x)$.

In order to compute the rate $\lambda(x)$, we consider a small horizontal segment between height $x$ and $x + \Delta x$.

Per unit area, there is an inflow of $Q(x + \Delta x)$ and an outflow of $Q(x)$ in the wake phase. Per second, the difference is picked up in the wake of rising fluidization bubbles and this constitutes the fraction

$$\lambda(x) = \frac{Q(x + \Delta x) - Q(x)}{\Delta x}$$

(4.5)

Letting $\Delta x \to 0$, we obtain the equation

$$\lambda(x) = -\frac{d}{dx}Q(x),$$

i.e. the return rate is the derivative of the wake flow.
When computing the probabilities $\lambda_i$, we actually approximate the derivative $\frac{d}{dx} Q(x)$ by the difference quotient (4.5), for $\Delta x = \Delta$. In this way, we only need information about $Q(x)$ at the points $x = i \cdot \Delta, i = 1, \ldots, N$.

### 4.2 Background of the Experiments

The motion of individual particles and the dispersion of a pulse of particles in a freely bubbling batch fluidized bed reactor were studied in a series of experiments performed in the Academic Hospital Groningen (AZG) and analyzed using Positron Emission Tomography (PET). This highly advanced experimental technique, which was used here in connection with fluidization, allows direct non-invasive monitoring of physical processes at high time resolution. The main goal of our experiments was to gain insight into particle transport processes in fluidized bed reactors and to validate the concepts behind our stochastic models by direct observation. Previous experiments had only given indirect validations, for example, by comparing observed residence time distributions with model predictions. The experimental results confirm our basic modeling assumptions and they also give rise to modifications.

The nature of fluidized beds and the particle transport in such beds has long been the subject of intense investigation. Due to the nature of the process, determining particle and gas dynamics is not an easy task. During the last decade a number of techniques such as X-rays, Positron Emission Particle Tracking (PEPT), Electrical Capacitance Tomography (ECT) and direct visualization by endoscopes have been pioneered and used to monitor the dynamics of fluidized beds. All these methods throw light on different aspects of the gas and particle dynamics.

Recently the method of “positron emission particle tracking” (PEPT) was introduced at the University of Birmingham to study particle dynamics in process apparatus (Parker et al., 1997) and fluidized beds (e.g. Stein et al., 1998, Snieders et al., 2000). Also at the Technical University of Delft, PEPT was used for studying fluidized beds (Abellon et al., 1997) with the object of investigating the particle residence time in interconnected fluidized beds. One limitation in this technique has, until now, been the size of the tracer particle, which had to be relatively large to be given sufficient activity for tracking. Therefore, it was not possible to use a bed particle as tracer particle. It was, however, possible to use a pellet of the same bulk density as the bed material, and this was shown to behave similarly to the bed particles themselves (e.g. Snieder et al., 1999). The motion of a
small pellet of the same density as the bulk density of the fluidized particles provided information about the particle movement in 3-D for the first time.

Advanced techniques for better results are being pioneered continually. In this chapter, a new technique for visualizing the particle movement in process equipment, which makes direct tracking of particle pulses or individual particles possible and provide 3-D information, was applied to better understandings on gas and particle dynamics in fluidized beds.

The most advanced cameras for Positron Emission Tomography (PET) are found in the medical sector. Cooperation with the University Hospital in Groningen (AZG) made it possible to make use of a state-of-the-art ECAT EXACT HR+ PET camera. To do this, a fluidized bed was constructed to fit in the cylindrical measuring region of the camera. Using this technique made it possible to use an actual bed particle as tracer rather than a pellet. Further details of the PET camera and data analysis are presented in Appendix 4A.

The experiments can be categorized into:

- Pulse experiments: following the dispersion of a pulse of marked particles and
- Single particle experiments: tracking a single tracer particle typical of the bed material with high temporal and spatial resolution.

Tracking an individual particle gives information about the nature of fluidized beds and the particle motion in them. If such tracking is done for a sufficiently long period, inferences can also be made about the time-mean particle motion, which is also the assembly-mean if the process is steady state.

A more direct way of determining the assembly-mean particle behavior, however, is to study the behavior of pulses of marked particles in the bed. The objective of the pulse experiments was to explore the possibilities and limitations of imaging pulses of radioactive particles in a PET camera for studying particle dynamics in fluidized beds, in this case a freely bubbling batch fluidized bed, and to compare the results with our stochastic model for the particle dynamics.
4.3 Experiments and Data Analysis

4.3.1 Experimental Set-Up

The experiment was, as mentioned above, designed to study the dynamics of both tracer pulses and of individual particles. Descriptions of the apparatus used and of experimental method are presented below.

Vessel and rig

Sketches of the fluidized bed and the rig used for the experiments are shown in Figure 4.4. Two cylindrically shaped glass tubes of diameters 15 and 10 cm, respectively, and both of height 35 cm were used as fluidized bed reactors. These sizes were chosen to be compatible with the reactor in the PET scanner.

Sintered plates were connected with a metal ring both on the top and on the bottom of the reactor. The sintered plate at the top of the bed was mounted to prevent the particles from exiting the bed. It had a high porosity and a low-pressure drop. The distributor plate at the bottom, a grade 0.3-sintered plate (made from copper) had a low porosity, so that the pressure drop across it was formally sufficient to ensure cross-sectionally uniform gas distribution. To attain a better distribution of air passing through the particles the windbox under the distributor was filled with small plastic cylinders. A particle outlet was made at the bottom of the vessel for later experiments with continuous beds.
Powder

The powder used for these experiments needed to fulfill the following requirements:

- It should be fluidizable, preferably falling in the Geldart group B category.
- The particles should absorb the radioactive labeler well using a simple procedure, which is fast relative to the labeler’s half-life.
- The particles should be tough enough to withstand the fluidization.

Two types of particles were found to meet these requirements. A macroporous anion exchange resin and a catalyst powder for fluidized catalytic cracking (FCC). The physical properties of these two particles are shown in Table 4.1.

<table>
<thead>
<tr>
<th>Table 4.1 Physical properties of the powders used</th>
</tr>
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<tbody>
<tr>
<td>Lewatit MP500</td>
</tr>
<tr>
<td>Average particle size [µm]</td>
</tr>
<tr>
<td>Density [kg/m³]</td>
</tr>
<tr>
<td>Sphericity [-]</td>
</tr>
<tr>
<td>( \varepsilon_{mf} [-] )</td>
</tr>
<tr>
<td>Geldart group</td>
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The Lewatit MP500 was a macroporous anion-exchange resin, which could easily be labeled with the radioactive tracer. Enough labeler could be attached even to perform experiments with a single particle as a tracer.

Ion exchange resins work by ions residing on the resin being replaced by ions in solution. In these experiments chloride ions were replaced by fluoride ions:

\[ R^+Cl^- (s) + F^- (aq) \rightarrow R^+F^- (s) + Cl^- (aq) \]

where \( R^+ \) is the exchanging material, of which the resins consist.

Macroporous anion exchange resins are tough and spherical (see Figure 4.5). When these particles are dried, they are sandlike. They can be chosen to be selective for Fluoride ions, and the process of marking these resins is very simple. The particles only have to be soaked in a fluoride containing solution for a short time, so the ions can exchange and the particles are radioactive. After this marking process the particles need to be dried. The strongly basic Lewatit MP500 resin was chosen, after initial tests showed it to be sufficiently effective in adsorbing fluoride ions.

Using the data in Table 4.1 and the physical properties of ambient air, the minimum fluidization velocity \( (U_{mf}) \) of the powder was predicted to be 0.75 m/s by the Wen and Yu equation (Wen and Yu, 1966) and 0.102 m/s using the full Ergun equation (Ergun, 1952). From several measurements of \( U_{mf} \), the value is 0.116 m/s. The value is slightly less than the value given in Hoffmann et al. (2003), and agrees well with model predictions.
The FCC catalyst powder was supplied by Shell Global Solutions. It was porous, providing enough surface area to label a pulse of particles. The measured $U_{mf}$ was 0.004 m/s which again agrees well with model predictions.

The particle densities given in Table 4.1 are envelope densities (including the internal pores in the particle volume) not skeletal densities.

### 4.3.2 Experimental Procedure and Plan

Experiments were done inside the PET (ECAT EXACT HR+) camera’s cylindrical detection zone of 83 cm diameter and 15 cm depth as shown in Figure 4.4B.

**Single Particle Experiments**

Only Lewetit MP500 powder was used in the single particle experiments. To mark a particle of the resin powder, a few wet particles were placed in 2 ml demi-water containing 4000 MBq of $^{18}$F-ions, and allowed to soak for about 1 hr. The particles were then dried. Handling of the single particles was done with extreme care to avoid damage.

Three experiments were performed, all in the larger reactor of 15 cm diameter. In two experiments, the tracer particle was initially placed low in the bed, while in the third it was placed in the middle.

In all single particle experiments PET measurements were done for about 10 s before fluidizing the bed, to get an image of the still condition. After this, the airflow was started, taking care that the desired flowrate of about 0.13 m/s was reached as quickly as possible. The PET measurements were continued for about 5 minutes at a constant flowrate.

**Pulse Experiments**

Both Lewatit MP500 and FCC catalyst particles were used in the pulse experiments. The procedure for labeling the particles for pulse experiments was as follows: 3 g of particles were soaked in a solution containing 1500 MBq $^{18}$F in 150 ml demi-water. After stirring for 15 minutes, the material was separated from the water using a Büchner funnel with sintered plate. The marked particles were dried in the small reactor by fluidizing for a half to a whole hour together with dry particles (900 ml in case of the Lewatit MP500, 1400 ml in the case of the FCC powder). The resulting dry powder mixture constitutes the “tracer”. In the case of the FCC catalyst, the initial activity was measured to be about 400 MBq.
The pulse of tracer was arranged in a bed of unmarked particles according to a pre-planned scheme (see Figure 4.6), and the bed placed in the PET camera. The camera was started up a couple of seconds before the gas was charged to the bed. The fluidizing gas pump was activated, and initially the gas was discharged to the atmosphere. At \( t = 0 \), the gas was suddenly diverted to the bed. The fluidization was initiated in this way to minimize start-up effects and to get as true as possible a picture of the dispersion of the tracer pulse in the bed at steady operating conditions.

The height of the powder bed was 20 cm in all cases. For the coarse Lewatit MP500, this remained almost the same when fluidizing at the low excess gas velocities used for these experiments, while the bed height rose to about 25 cm when fluidizing the much finer FCC powder. The point at which the gas was diverted to the bed could always be recognized by the pulse rising slightly.

The experiments were designed with well-defined pulses of marked particles arranged in the bed initially. When the bed was fluidized the dispersion of the pulse through the bed was monitored. A number of different types of pulses were used, as illustrated in Figure 4.6. The three first pulses, Figure 4.6(a) – (c), comprised layers at the bottom, top and middle of the bed respectively, and were intended to show the axial particle dispersion in the bed. The central and annular columns illustrated in Figure 4.6(d) and (e) were used to investigate the radial particle dispersion. The final pulse configuration shown in Figure 4.6 (f), a disk in the middle of the column, was intended to give an over-all impression of the particle mixing in the bed.

![Figure 4.6 The types of pulses used in the experiments.](image)

As mentioned, the superficial velocity required to just fluidize the powder or ‘minimum fluidization velocity’, \( U_{mf} \), of the Lewatit MP500 powder was found to be 0.116 m/s, and the experiments were carried out at 0.130 m/s, while for the FCC
powder the measured $U_{mf}$ was 0.004 m/s, and the experiments were carried out at 0.01 m/s.

4.3.3 Data Output and Analysis

The PET technique is based on the radioactive decay of a radioactive labeler, here $^{18}$F, which leads to the emission of a positron and neutrino. The positron travels a few mm through the system before annihilating with an electron. This annihilation normally results in the emission of two photons, in this case of 511 keV. For conservation of energy and momentum, the two photons are normally emitted back-to-back, and thus define a straight line going through the annihilation site called a "line of response" (LOR), see Figure 4.7.

The emitted photons were detected by cylindrically arranged sensors as illustrated in Figure 4.7A. To analyze data obtained from these sensors, different tools for collecting data were used. For the single particle experiments, the data used were collected in so-called list mode files. A sinogram, on the other hand, was used for the pulse experiments. This is a tool for storing the detector pairs in a way suitable for future image reconstruction and display. More details on the PET camera and data analysis are included in Appendix 4A.
4.4 Experimental Results and Discussion

As discussed in Section 4.3, the distributor plate used in all experiments was a copper sintered plate. For the Lewatit MP500 powder, this sintered plate distributed the gas as intended. However, in the case of the FCC catalyst powder, the plate did not always distribute the gas sufficiently well. It could be seen in some of the experiments that defluidization occurred at one side of the bed. Nevertheless, we were able to obtain initial results that clearly demonstrated the usefulness of the PET technique. A new distributor plate, which significantly improved the air distribution, was designed and placed in the test vessel. This work is described in Appendix B.

For the pulse experiments images of the particle distribution profiles were obtained using in-house developed software that can show horizontal and vertical planes within the reactor for different time frames, calculate the sum of the emission intensity over each plane and make bitmap pictures of the results. The images presented using this software show the mean intensity of the radioactive emission during the ‘exposure period’. The darker the shade of a specific voxel, the higher the mean intensity, so that the marked particles show up as dark-gray colors. In addition to the images shown below, more images and data can be found in van der Zwan (2002) and van der Wiel (2003). Van der Zwan (2002) also contains the source code for the mentioned software.

For the single particle experiments the data were retrieved from so-called "list-mode files", containing information about the LOR's on an event-by-event basis. These data were used for determining the particle's position in the bed once per millisecond. Also for this, software was developed in-house.

Details of the data retrieval and the analysis procedure are given in Appendix 4A.

4.4.1 Pulse Experiments

The results of the pulse experiments are presented as bitmaps showing the marked particle concentration in the bed in a succession of 1 s time frames, and in the form of 3-D plots and contour plots of the marked particle concentration as a function of position in the bed and time.

These experiments were performed by preparing some amount of marked (radioactive) particles and by laying them in different positions, as shown in Figure 4.6. Experimental results are presented below, firstly for the axial particle mixing
by showing the dispersion of horizontal layers, and subsequently for the radial particle mixing by showing centrally and annularly placed particle pulses.

**Axial Particle Mixing**

Figure 4.8 shows the dispersion of a horizontal layer initially placed at the top of a bed consisting of FCC catalyst particles.

![Figure 4.8](image)

The bitmaps and the 3-D plot show the layer first moving upwards at the onset of fluidization and then descending through the bed while being dispersed. Once reaching the bottom of the bed, the layer is rapidly brought to the top, where it is still vaguely recognizable, whereafter it again descends and disperses completely.

Figure 4.9 shows similar results for a layer initially positioned in the middle of the bed. Additionally this figure shows a contour plot of the relative radiation intensity as a function of height and time. Here also we see the layer initially ascending at the onset of fluidization and then descending while being dispersed, being brought...
quickly from the bottom to the top of the bed, and descending again while being dispersed further. The 3-D plot shows the descent of the layer as a series of successive ridges with the same slope in the height-time plane. In the contour plot this is visible as light streaks with the same slope in the height-time plane.

Figure 4.9 The dispersion of a horizontal layer from the middle of a bed of FCC catalyst powder shown by a series of x-z bitmaps, and a 3-D plot and a z-time contour plot of x-y averaged tracer concentration
The initial ascent of the layer is due to the fluidizing gas entering the bed at the onset of fluidization, forming fluidization bubbles and expanding the bed. The layer descent sets in once the fluidization bubbles have reached the layer. All the features can be related qualitatively to the particle transport associated with the fluidization bubbles as described in Chapter 2.

For comparison with the results using the FCC powder, the dispersion of a middle layer in a bed of Lewatit MP500 powder is shown in Figure 4.10. As mentioned, this powder is much coarser than the catalyst powder, and the superficial fluidization velocity in excess of the minimal fluidization velocity, \((U-U_{mf})\), which determines the bubble activity (see below), is much larger. This explains the relatively rapid dispersion of the layer.

![Figure 4.10 The dispersion of a horizontal layer from the middle of a bed of Lewatit MP500 shown by a series of x-z bitmaps.](image)

The dispersion of a layer initially in the bottom of a bed of FCC catalyst is shown in Figure 4.11.

![Figure 4.11 The dispersion of a horizontal layer from the bottom of a bed of FCC catalyst powder shown by a series of x-z bitmaps.](image)

A degree of asymmetry between the left and right hand sides of the bed of FCC powder is evident in all three series of bitmaps, but most clear in this last series.

We will discuss these results, their relation to the physical events in the bed and their comparison to a stochastic model for particle transport in fluidized beds in Section 4.5.
Radial Particle Mixing

Figures 4.12 and 4.13 show the radial dispersion through a bed of FCC catalyst particles of pulses initially shaped as a column in the middle and an annular cylinder, respectively.

Comparing the two figures it is clear that the central column remains recognizable for a couple of seconds more than the annular pulse. This, however, was not always the case. It is, in any case, clear that radial dispersion is fast even at these low excess fluidization velocities, the pulses being dispersed within a few seconds of operation.

Figure 4.12 The radial dispersion of a vertical center column spanning the length of a bed of FCC catalyst powder, shown by a series of \(x-y\) bitmaps and an \(x\)-time contour plot of \(y-z\) averaged tracer concentration in a central \(y\)-slice of the bed.
Figure 4.13 The radial dispersion of an annular column spanning the length of a bed of FCC catalyst powder, shown by a series of x-y bitmaps and an x-time contour plot of y-z averaged tracer concentration in a central y-slice of the bed.

Figure 4.14 shows the axial and radial dispersion of the disc-shaped pulse in the middle of the bed. The axial dispersion (left contour plot) shows the same characteristics as the middle layer, with descent and dispersion interrupted by fast movement to the top of the bed, while the radial dispersion shows a similar pattern to that of the central column, except that left-right asymmetry in the x-direction is clearer in this case, consistent also with the bitmaps.
4.4.2 Single Particle Experiments

In this section we show different aspects of the results from the single particle experiments. We start with the short time-scale motion, i.e. the individual ms points, study the standard deviation of the points in the static and fluidized conditions, and try to detect any fast, short-range motion of the fluidized particles. Thereafter, we consider the particle motion on a longer time scale and relate this to the particle transport mechanisms envisaged in fluidized beds.

Precision of the Data and Short Time-Scale Particle Motion

The particle tracking was initiated about 10 seconds before the bed was fluidized, making it possible to ascertain the spread in the millisecond (ms) positions due to
experimental error only. Figure 4.15A and B show ms positions during periods where the bed was unfluidized and fluidized, respectively.

To quantify the precision with which the particle's position can be determined per ms, we determined the standard deviation in the data corresponding to the unfluidized state while using different numbers of LORs per ms. Since the number of cutpoints between LORs increase with the square of the number of LORs we would expect the standard deviation of ms points to decrease linearly with the number of LORs. In fact, it decreased less so, leading us to seek some source of scatter in addition to that incurred by the sources from the analysis alone, see Appendix 4A (4A.3).

The standard deviation in the data shown in the Figure 4.15A is $485 \pm 6 \mu m$, indicating that when using 100 LORs the particle position can be determined to within two particle diameters once per ms. The standard deviation in the $x$-coordinates was about the same, while that of the $z$-coordinate was three times as large. These standard deviations varied slightly between the experiments, and varied with the position in the field of view (see Appendix 4A).

The motion of the particle in the fluidized bed is evident in Figure 4.15B, and we shall return to this large-scale, long-term motion. One question that arises is

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**Figure 4.15** A and B are plots of time (ms) vs. vertical positions ($y$-coordinate) in unfluidized and the fluidized state, respectively.
whether we can detect any short-range, fast particle movement, which may be obscured by the scatter in the ms points. We can attempt to answer this question by comparing the standard deviation in the unfluidized state with that in the fluidized state. To do this, the particle motion in the fluidized bed was subdivided in small time segments of 10 ms, in which the path could be well fitted with a straight line, and the residual standard deviation determined. This was done during a period where the particle occupied a similar position to that in the static bed. The resulting estimate of the scatter in the particle position in the fluidized state was $487 \pm 9 \mu m$, which is the same as in the unfluidized bed. Therefore, we cannot detect any fast, short range motion of a single particle in a fluidized bed.

**Long-Term, Large-Scale Particle Motion**

The plots of Figure 4.15 indicate that we can smooth the data over a number of ms reducing the scatter without losing information about the real motion of the particle. A preliminary study of the data indicates that the fastest motion of the particle will be faithfully reproduced if some running average spanning between 10 and 50 ms is calculated.

Kendall (1985) discusses the issue of smoothing data in an optimal way and also suggests other useful methods for characterizing time series. Figure 4.16 shows plots of the $y$-, $x$- and $z$-coordinates (height and lateral position in the bed) as a function of time. The data are smoothed by calculating simple averages every 20 ms.

Several features can be noted in Figure 4.16. The bed was 19 cm high, and in the fluidized state about 25 cm. The top graph ($y$- coordinate) shows that the particle almost always traversed the entire length of the bed on its upward and downward motion, giving a regular, cyclic motion of the particle. The upward motion is faster in general than the downward one. Furthermore the velocity of the downward motion appears in many cases to decrease in the bottom section of the bed. The motion in the $x$- and $z$-directions is less regular. As mentioned, the field of view was 15 cm deep, and the internal bed diameter was also 15 cm. Occasionally, the particle would therefore move to the extreme of the field of view in the $z$-direction, resulting in short time segments with very high scatter. An example of this phenomenon is also included in the time interval in the figure, and indicated by an arrow.

Snieders *et al.* (1999a) related the upward and downward velocities of their pellet to the bubble rise and bulk descent velocities, respectively, calculated using empirical relations presented in Equations (4.6) to (4.13) below. They found that the observed velocities corroborated the explanation of Rowe and Partridge (1962)
mentioned in section 4.1 for vertical particle transport, i.e. their upward particle velocity was about equal to the bubble rise velocity and their downward velocity about equal to the descent velocity of the bulk material in the bed, although the descent velocity seemed to be slightly high, which was attributed to segregation of the pellets. Below, we perform similar calculations of the expected vertical particle velocity.

The bubble diameter, \( D_B \), is defined here as twice the radius of curvature of the front of the spherical-cap shaped fluidization bubble. The mean value of \( D_B \) can be estimated by empirical relationships put forward in the literature (see the review by Baron et al., 1990). One such relation that is widely used is, as we have mentioned in Chapter 2:

\[
D_B = \frac{1.3}{g^{0.2}} \left( \frac{U - U_{mf}}{1000} \right)^{0.4} \left( \frac{1}{1 - f_w} \right)^{0.33} + 2.05h \left( U - U_{mf} \right)^{0.94}. \tag{4.6}
\]

The first term describes the bubble size upon formation at the porous plate distributor (Kato and Wen, 1969), and the second the bubble growth in the bed due to coalescence (Geldart, 1972). Experiments show that the bubble and associated wake together approximately form a sphere, and \( f_w \) is the volume fraction of the bubble-wake sphere taken up by the wake material. \( f_w \) can be estimated from:

\[
f_w = 0.45 \left( 1 - e^{-60D_B} \right)^{2.75} \tag{4.7}
\]

(Bosma and Hoffmann, 2003), this is an approximation of Equation (2.4).

It turns out, however, that for our system Equation (4.6) estimates rather a low value for \( D_B \) compared to the bulk of the proposed relationships, and we therefore prefer here to use the relationship of Mori and Wen (1975), presented in Equation (4.8), which has been recommended recently by a number of workers, and gives a more typical value.

\[
D_B = D_{B,m} - \left( D_{B,m} - D_{B,o} \right) \exp \left( -0.3 \frac{h}{D_{bed}} \right) \tag{4.8}
\]

where the bubble diameter upon formation at the porous plate distributor is given by:

\[
D_{B,o} = \frac{1.38}{g^{0.2}} \left( \frac{1}{1000} \left( U - U_{mf} \right) \right)^{0.4} \tag{4.9}
\]
and the largest bubble diameter that will form by coalescence in the given bed of diameter $D_{\text{bed}}$ is given by:

$$D_{B_{\text{m}}} = 1.49 \left(D_{\text{bed}}^2 \left(U - U_{mf}\right)\right)^{0.4}$$  \hspace{1cm} (4.10)

Once the size of a fluidization bubble is known, its rise velocity, $U_b$, can be estimated from a relation due to Davis and Taylor (1950) originally for gas bubbles in liquids:

$$U_b = 0.711 \sqrt{gD_{eq}}$$ \hspace{1cm} (4.11)

where $D_{eq}$ is the diameter of a volume equivalent sphere for the spherical-cap shaped fluidization bubble. Rowe and Partridge (1965) and Hoffmann and Yates (1986) have reviewed the issue of bubble rise velocities in light of experiments using X-rays.

To finally estimate the total upward material flow in the wakes of fluidization bubbles, we estimate the total flow of empty bubble volume from the simple “two-phase theory”, in which all gas exceeding that required to just fluidize the particles travels in the form of fluidization bubbles:

$$\frac{Q_b}{A_{\text{bed}}} = U - U_{mf}$$ \hspace{1cm} (4.12)

where $Q_b$ is the volumetric gas flow in the bubble phase, and $A_{\text{bed}}$ the bed cross-sectional area. Equation (4.12) in combination with Equations (4.8) and (4.7) gives the desired volumetric material flow in the wake phase, which is also the downward material flow in the bulk, as:

$$Q_w = A_{\text{bed}} \left(U - U_{mf}\right) \frac{f_w}{1 - f_w}$$ \hspace{1cm} (4.13)

In our case, the voidage in the bed due to the bubbles is low, so that the downward particle velocity in the bulk can be estimated as the superficial downward velocity obtained by dividing the downward flow by $A_{\text{bed}}$. 
Figure 4.16 Time-series of the tracer particle’s y-position (height in bed), x-position and z-position. Simple averages of groups of 20 ms particle positions are plotted.
The result of these calculations for the velocity, $U_b$, of a bubble with the mean size, is indicated in Figure 4.16 (top) by the dash-dot line. It seems that the upward velocity of the tracer particle agrees with the bubble rise velocity.

On the other hand, the descent velocity in the bulk material is calculated to be 0.003 m/s, while the downward velocity of the tracer particle is about 0.08 m/s, an order of magnitude higher. We are thus looking for a mechanism causing the circulation in the bed to be much higher than that caused by the transport in fluidization bubble wakes.

Studying the movement in Figure 4.16 more closely reveals a correlation between the vertical and the horizontal particle motion (made clearer by the broken lines in the figure). A high vertical velocity corresponds to a low horizontal one and vice versa. The general impression is of a circulating particle motion. This impression is reinforced by the 3-D plots of the particle motion shown in Figure 4.17.

![Figure 4.17 3-D tracks of the tracer particle seen from two different angles, the dimensions of the bed of particles are also shown.](image)

Seen from one angle, the particle motion seems to be straight up and down, but when turning the bed, the motion is seen to be circulatory in nature.

To study the nature of the particle motion further, the starting points for significant upward and downward paths were charted using an algorithm. In order for the
algorithm to give results consistent with visual observations, it had to be run on a file containing simple averages of 50 ms particle positions, which is consistent with the result that averaging over 10-50 ms positions reduces the scatter in the results without loss of information about the particle motion.

The results are shown in Figures 4.18 and 4.19. Consistent with the observations of regular vertical movement of the tracer spanning the entire bed height, the starting points for upward and downward motion are concentrated in the bottom and top of the bed, respectively.

Figure 4.18 Starting points for significant upward paths of the tracer particle.
Figure 4.19 Starting points for significant downward paths of the tracer particle

It is clear, however, that these points are distributed axially to some extent.

When looking at the radial distribution, the starting points for upward motion are not distributed evenly over the bed cross-section, while those for downward motion are better distributed. The visual observation was that the bubbling activity was well distributed over the bed.

These results are therefore all showing the same picture. The bubbling activity is to some extent concentrated one region of the bed, and the particle is rising when in this part of the bed, while descending when in the less active region.

In the region of high bubble activity, the bulk material is likely to be brought upwards by the rising fluidization bubbles. The bubbles are therefore bringing much more material up than accounted for when calculating the flow in their individual wake regions. This phenomenon is called “gulf streaming” and is explained further in the following section.
4.5 Comparison with the Model and Further Discussion

The parameters needed for the freely bubbling batch fluidized bed were obtained as described in the above section. We then compare the model characteristics with the pulse experimental results.

The previous chapter shows that all that is needed for calculating the descent velocity for the stochastic model is $Q_w$, the material flow in the wake phase, and $D$, the dispersion coefficient. The relations in the previous section are sufficient to find $Q_w$.

A value for the dispersion coefficient $D$ can be found in the manner explained in Chapter 2 from a literature study of the vertical particle displacement caused by one rising fluidization bubble (Tanimoto et al., 1981) and calculating $D$ as the mean square particle displacement caused by fluidization bubbles per second (this is twice the Fickian diffusion coefficient, as mentioned). Again, as will be seen below, the exact value of $D$ is not an issue here, since other factors cause a large deviation between model and experiment.

Since the bubble flow is low, we can take the superficial downward velocity of the bulk phase to be about $Q_w/A_{bed}$. The above equations then predict bulk descent velocities of about 0.001 m/s, while the downward velocity of the pulse in the bulk phase, see for instance Figure 4.8, seems to be of the order of 0.05 m/s, since the pulse travels from the top to the bottom of the bed in 4-5 seconds. We will return to the reason for this discrepancy in the following section. Below we show that upward transport of the particles associated with the bubble flow and downward flow with dispersion in the bulk can actually explain the features of the experimental results qualitatively.

Figures 4.20 and 4.21 show comparisons of model predictions using empirically optimized values for $Q_w$ and $D$ with experimental results for the dispersion of the middle layer in the FCC catalyst bed. As indicated by the numbers given above for the expected and actual descent velocity of the layer, the circulation rate $\nu$ had to be multiplied by a factor of 50. The dispersion $D$ had to be multiplied by a factor of 20.

The model predictions clearly account for all the features of the experimental results. More detailed analysis shows that agreement would be even better if the fast material transport to the top of the bed with the fluidization bubbles was associated with some dispersion as well. This, of course, is quite reasonable in light
of the wide spread in the sizes and velocities of fluidization bubbles and the fact, also shown by the single particle experiments, that particles at the end of their upward trajectory are deposited not exactly at the bed surface, but somewhat axially dispersed over the top region of the bed.

When performing this comparison between model and experiment, it must be realized that the model only accounts for the system’s behavior after fluidization has been commenced and the bubble stream has reached the layer.

![Image of dispersion comparison](image)

**Figure 4.20** Dispersion of a horizontal layer from the middle of the bed, comparison of experimental results (top figures) and model predictions (bottom figures).
Chapter 4: Freely Bubbling Batch Fluidized Beds: Model Validation

Figure 4.21 Dispersion of a horizontal layer from the top of the bed, comparison of experimental results (top figures) and model predictions (bottom figures).

We attribute the quantitative discrepancy between experiment and model to “gulf streaming” of the solids in the bed. The principle is illustrated in the sketch in Figure 4.22. Due to a non-uniform bubble distribution over the bed cross-section, regions exist where, in addition to the wake material, also bulk material interstitial between the bubbles is dragged up with the bubble stream. This flow of bulk material can be considerably more than the wake flow, causing also a considerably faster downward bulk flow in the rest of the bed. Merry and Davidson (1973) were among the first to discuss this phenomenon. Maldistribution of bubble activity and the associated solids movement in fluidized beds have been discussed by a number of workers (e.g. Werther, 1975 and Farrokhalae and Clift, 1980). Matsen (1996) states that gulf-streaming of solids in industrial fluidized beds is unavoidable and that this constitutes a powerful axial mixing mechanism that is often absent in small, laboratory fluidized beds with good gas distribution. Werther (1975) states that, even if the gas distribution is perfect at the distributor plate, regions of high and low bubble activity will still develop; higher in the bed the bubbles will have concentrated in the middle due to coalescence, and this will give rise to gulf-streaming effects.
Region of high bubble activity, bulk material is transported upwards

Downward bulk velocity higher than expected from only wake flow

Figure 4.22 Sketch illustrating the principle of “gulf streaming” in a fluidized bed.

We have shown that in the beds of Lewatit MP500 powder the motion of a single particle indicates that gulf-streaming is taking place, even though the beds visually appeared to have a reasonably uniform cross-sectional gas distribution. Studying also the figures of pulse experiments more closely indicates that gulf-streaming is taking place: for instance in the 7th bitmap in Figure 4.8 and the 4th bitmap in Figure 4.11 it can be seen that a section of the layer is separated from the main part, and moving upward relative to the main part.

The phenomenon of gulf-streaming in fluidized beds is obviously an important one, which should be included in a model for particle dynamics in fluidized beds. Gulf-streaming is mathematically fairly easy to incorporate in a stochastic model for vertical particle transport, but it is not easy to model physically. Going from extremely bad to ideal cross-sectional bubble distribution we might expect:

Very bad bubble distribution gives a very localized region of high bubble activity and bubble velocity, the bulk material flows rapidly upwards in this region, and relatively slowly down in the rest of the bed.
Better bubble distribution gives a larger region of lower, but still relatively high, bubble activity; the upwards flow of bulk material in this region is larger in terms of volume but smaller in terms of velocity than in 1). Downwards bulk velocity in the rest of the bed is higher than in 1). This is what we see in our experiments.

Ideal bubble distribution, material is only brought upwards in bubble wakes; the downward velocity in the bulk is cross-sectionally uniform and low. This is probably the case in the experiments of Snieders et al. (1999).

The transition from state 2) to state 3) is not easy to visualize, or to predict. Moreover, any cross-sectional non-uniformity of fluidization bubble flow is likely to be self-augmenting in that the downwards flow of bulk material in regions of low activity probably diverts fluidization bubbles away from that region. Also the bed aspect ratio and the absolute gas flow through the bed in excess of the minimum fluidization velocity are likely to play a role in determining the extent of gulf-streaming of the solids.

These experiments show that visual observation of the bed is not enough to determine the extent of gulf-streaming, and thus the degree of axial particle mixing in the bed. The degree of axial particle mixing will determine, for instance, the particle residence time in continuous beds and the progress of such processes and fluidized bed granulation and coating. Also the axial heat and mass transfer in fluidized beds, and the degree of particle segregation, depends on the axial particle mixing.

Gulf-streaming is, as mentioned, likely to be unavoidable in industrial fluidized bed processes. It may in many cases be beneficial in decreasing temperature and concentration gradients in fluidized bed processes. In any case, it is important to model, and to control.

### 4.6 Concluding Remarks

Using a PET scanner to obtain the results have shown that the stochastic model captures qualitatively the dynamic movement of particles in freely bubbling batch fluidized beds and confirm the basic concept of the model. However, the results have also shown that in many important cases the upward particle transport associated with the fluidization bubbles is not, even approximately, accounted for by the flow in bubble wakes, and that a fundamental investigation of the phenomenon of gulf streaming in fluidized beds is needed. Gulf streaming can be incorporated in the model, and the model is also relatively easy to apply to different types of fluidized beds with only a few modifications as also shown later in Chapters 5 and 6.
4.7 Notation

\[ A_{\text{bed}} = \text{bed vessel cross-sectional area} \]
\[ D_B = \text{diameter of fluidization bubble} \]
\[ D_{B,m} = \text{maximal bubble diameter in the bed} \]
\[ D_{B,o} = \text{bubble diameter at the gas distributor} \]
\[ D_{\text{bed}} = \text{diameter of the bed vessel} \]
\[ D = \text{dispersion coefficient (twice the Fickian diffusion coefficient)} \]
\[ d = \text{distance of LOR from center of vision} \]
\[ D_{eq} = \text{diameter of a sphere with the volume of the fluidization bubble} \]
\[ f_w = \text{wake fraction} \]
\[ g = \text{gravitational acceleration} \]
\[ h = \text{height in the bed} \]
\[ i,j = \text{indices} \]
\[ N = \text{number of detectors in the ring} \]
\[ p(n) = \text{probability vector for the particle’s position at time step } n \]
\[ p(n,i,j) = \text{elements of } p(n) \]
\[ Q_B = \text{volume flow in the bubble phase} \]
\[ Q_w = \text{volume flow in the wake phase} \]
\[ P = \text{matrix of transfer probabilities} \]
\[ p_{i,j} = \text{transfer probability from cell } i \text{ to cell } j, \text{ elements of } P \]
\[ t = \text{time} \]
\[ U = \text{superficial gas velocity} \]
\[ U_{mf} = \text{superficial gas velocity required to just fluidize the powder} \]
\[ U_b = \text{fluidization bubble velocity} \]
\[ v = \text{velocity due to circulation} \]
\[ x = \text{x-coordinate} \]
\[ y = \text{y-coordinate, along the bed axis} \]
\[ z = \text{z-coordinate} \]
\[ z' = \text{distance from top of bed divided by the total bed height} \]

Greek symbols:

\[ \alpha, \beta = \text{transfer parameters for a particle in the model} \]
\[ \delta, \lambda = \text{transfer parameters for a particle in the model} \]
\[ \lambda(z') = \text{probability rate (s}^{-1}\text{) of the particle being caught in a bubble wake} \]
\[ \Delta = \text{height of a cell in the discretized bed} \]
\[ \varepsilon = \text{length of a time step} \]
\[ \varepsilon = \text{void fraction or ‘voidage’} \]
\[ \epsilon_{mf} = \text{voidage at minimum fluidization conditions} \]
\[ \theta = \text{angle between LOR and the vertical} \]
4.8 References


Ergun, S., “Fluid flow through packed columns”, Chemical Engineering Progress, 48(1952), 89-94.


Appendix 4A

The PET Technique

4A.1 Introduction

This appendix describes both analytical and physical aspects around the PET camera used in this research.

Positron Emission Tomography (PET) is a technique used in clinical medicine and biomedical research to create 3-D images that show functional structure and physiological processes. Through co-operation between the staff at the PET center of the Groningen University Hospital (AZG) and university-based process technologists, it was possible to use modern PET facilities to study particle dynamics in fluidized beds.

A miniature-fluidized bed was used to conduct this research, placing the bed in the ECAT EXACT HR+ scanner, as shown in Figure 4A.1. Tracer particles were made radioactive by doping them with a proton-rich 18-F isotope using the AZG cyclotron (see Section 4.3 for a description of the experimental set-up and procedure). The tracer has the same properties as the bed material itself, and therefore does not disturb the dynamics of the system.

Figure 4A.1 A: The ECAT EXACT HR+ PET scanner. B: Sketch of a fluidized bed in the ring of the PET scanner.
4A.2 Principles and Construction

To monitor physiological processes, radioactive nuclei are injected into the body as a labeled substance. These labeled substances contain positron-emitting isotopes, produced in a cyclotron. The most useful positron emitters are shown in Table 4A.1 with their half-lives. Atoms from these positron-emitting isotopes are then used to “tag” molecules of a compound of interest, which is subsequently introduced into the human body, usually by intravenous injection (Toga et al., 1996).

At any given time, some of the atoms of the positron emitting isotope will decay, emitting a positron and a neutrino. The positrons will annihilate with electrons. The annihilation converts the mass of the electron and positron into energy which is liberated in the form of gamma rays (see Figure 4A.2).

<table>
<thead>
<tr>
<th>Isotope</th>
<th>$^{11}$C</th>
<th>$^{13}$N</th>
<th>$^{15}$O</th>
<th>$^{18}$F</th>
</tr>
</thead>
<tbody>
<tr>
<td>Half-life (min)</td>
<td>20.4</td>
<td>9.96</td>
<td>2.07</td>
<td>109.8</td>
</tr>
</tbody>
</table>

For energy conservation this annihilation normally results in back-to-back emission of two high energy gamma photons (gamma rays), in this case of 511 keV. These two gamma rays are detected by detectors in a ring surrounding as shown in Figure 4A.3. The detectors used in PET scanners are scintillation detectors, which look for “coincidence” events, where two gamma rays impact almost simultaneously on opposite sides.

Figure 4A.2 Positron emission and annihilation.
Two such detected gamma rays represent a straight line, the line of response (LOR), along which the event took place. An assembly of such lines gathered and processed finally produces an image. Spatial resolution (the ability to accurately locate events) and sensitivity (the number of events registered per unit dose of activity initially present) are the two most important parameters for a PET scanner. The PET scanner, the ECAT EXACT HR+ model, used for this work is state-of-the-art in both respects.

4A.2.1 Basic Construction of a PET Scanner

![Figure 4A.3 Blocks in detectors inside an ECAT EXACT HR+ PET scanner.](image)

Figure 4A.3 shows the construction inside the scanner. The important characteristics of a scanner are the numbers of detector rings and blocks. The volume where the object is scanned is called the field of view (FOV). The blocks are laid out in a circular form around the FOV as shown in Figure 4A.3. This volume has a cylindrical shape having a diameter of 83 cm and a depth of 15 cm. The ECAT EXACT HR+ camera has 4 rings. Each ring has 72 blocks surrounding the FOV (Figure A.3). Each block has 64 crystals in an 8x8 array although in terms of a detector ring, each block contains a partial ring of 8 crystals. The total number of ECAT EXACT HR+ detectors is $4 \times 726 \times 4$ (or 18,432).

The detectors used in PET scanners are scintillation detectors. A typical scintillation detector consists of a scintillating crystal coupled to a photomultiplier tube, with some simple electronics housed in an aluminum shield. Scintillator materials used include Sodium Iodide, Gadolinium Oxyorthosilicate, Lutetium Oxyorthosilicate, Barium Fluoride and Bismuth Germanate (BGO). The ECAT
EXACT HR+ scintillator material is BGO. The features of this scanner are summarized in Table 4A.2.

Table 4A.2 Summary of technical features of ECAT EXACT HR+ PET scanner.

<table>
<thead>
<tr>
<th>Crystal material</th>
<th>BGO (Bismuth Germanate crystal)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Number of crystal rings</td>
<td>32</td>
</tr>
<tr>
<td>Number of crystals/ring</td>
<td>576</td>
</tr>
<tr>
<td>Total Number of crystals</td>
<td>18,432</td>
</tr>
</tbody>
</table>

The principle of a scintillation detector is that when the radiation enters the detector, it will ionize and excite the atoms in the crystal, causing the crystal to scintillate and emit a small flash of light. The photomultiplier tube will respond to this light and convert the light into an amplified electronic pulse. There are two very important properties of the scintillation signal; 1) the approximately linear response to the deposited energy and 2) the fast time response. The latter is the time needed for the scintillation to die out and the detector to be ready for a new event, also called dead time. The scintillation detector used in the ECAT EXACT HR+ has an especially short time compared to other types of detectors. So the measurements of time differences between events, the time resolution, can be made with higher accuracy with this quality of scintillator crystal.

4A.2.2 Limitations of the PET Scanner

Statistical Limitations

The statistical noise in PET images is determined by:

- The number of image counts (the scan time and scanner efficiency)
- The statistical properties of the method of reconstruction from projections.

Geometric Limitations

The spatial resolution of the images predominantly depends on the radius of the ring and the size of the detectors. It is preferable to have a ring diameter of at least twice the field of view because the resolution improves with the decrease in parallax afforded by a larger ring diameter. The angle of incidence should be close to perpendicular, although the solid angle decreases with enlargement of the ring. These considerations lead to the choice of diameter for the ring. Since the radius of the scanner’s ring is limited (83 cm), miniature beds were made of height 80 cm,
and 10 or 15 cm in diameter, in order to fit our fluidized beds inside the field of view.

**Data Storage**

Data acquired by the PET detectors and electronics are processed in ASC II (Advanced Computational Systems) format. Gamma rays that hit the detectors are recorded and the signals produced by the photomultiplier tubes (PMTs) are passed on to a coincidence processor, where events are classified as true, random or multiple.

The standard procedure is that the data are then sent to a real-time sorter (RTS) and distributed into a storage mode called sinograms, tools for storing the collection of detector pairs in a specific way. The sinograms are transferred to ASC II memory boards, where they are stored temporarily.

Eventually, filtered back-projection algorithms are applied to the sinograms to produce images. Following this procedure, images are obtained with a minimum time frame of 1 second.

While this type of data storage and analysis with the associated limitations in time- and spatial resolution were adequate for the experiments wherein a pulse of marked particle was followed, the experiments following a single particle demanded a different type of data record and data analysis. For this, another manner of storage was invoked: binary event-be-event records of the LORs in so-called list-mode files.

The details of sinogram and list mode file are discussed in the following section.

**4A.3 Data Analysis**

A line of response (LOR) is drawn between the two detectors if two detections occur within a narrow time window, in which case they are adjudged to have emanated from the same annihilation. The position of the LOR is stored as:

- Its distance to the camera center.
- Its angle with the vertical.
- The numbers of the two detectors in the direction normal to the paper.
Figure 4A.4 A sketch of an LOR through a given point \((x,z)\), and its defining parameters, also a circular pulse of radioactive material is sketched. B sketch showing “traces” of the point and the pulse in a plot of \(\theta\) vs. \(d\).

We first focus on detections in one particular detector ring, i.e. in the \(y\)-plane. If the angle \(\theta\) was given in radians and the distance \(d\) in some unit of length (see Figure A.4), \(d\) would depend on \(\theta\) according to:

\[
d = \sqrt{x^2 + z^2} \left\| \cos \left( \theta + \arctan \frac{z}{x} \right) \right\|
\]

We can choose a sign convention for \(d\), which is consistent with the way the single LOR data are stored in list-mode files by including the factor \((\pm x/|x|)\) on the right-hand-side, the positive sign applies when \(\theta\) is less than \(\pi/2\). Thus, a plot of \(d\) vs. \(\theta\) would give a sinusoidal shape, the phase and amplitude depending on the position of the point. The range of \(\theta\) need only be from 0 to \(\pi\), due to the symmetry of the system.

**Sinogram**

A *sinogram* (detailed information about the construction of sinograms can also be found in Nichols, 2001) is a two-dimensional array containing values of \(\theta\) in the
rows and values of $d$ in the columns, and is thus equivalent to a plot with $\theta$ on the vertical axis and $d$ on the horizontal one, as the one indicated Figure A.4B. In a sinogram, however, both $d$ and $\theta$ are given in terms of number of detectors, as described below, rather than a length unit and radians, giving a distortion compared to the plot in Figure 4A.4B.

Figure 4A.5 shows the construction of the sinogram by a simplified example. “Whole angles” (rotating one detector at both ends of the LOR each time, starting from the vertical as indicated in the figure) and “half angles” (rotating one detector only in one end as indicated, the LORs going through the center of the sensing zone are of this type) are “interleaved” in the sinogram, meaning that they occupy the same row as indicated in the figure. The distances for whole and half angle LORs occupy separate columns as indicated. For a given angle, only the central half of the LORs are considered. The resulting sinogram matrix is square and has dimension $N/2$, where $N$ is the number of detectors in a ring; in the example in Figure 4A.5 $N = 48$, while in the actual camera $N = 576$.

As mentioned above, if the sinogram cells corresponding to the LORs going through one point are marked, a sort of sinusoidal trace, somewhat distorted compared to the traces in Figure 4A.4B, would result. The trace in the sinogram resulting from some pulse of active material, such as that indicated in gray in Figure 4A.4A will result from combining the traces generated by the differential areas making up the pulse. The original spatial shape of any pulse can be reconstructed from the sinogram.

![Figure 4A.5 Simplified diagrams showing the construction of a sinogram from the detected LORs.](image)
The above explains the organization of the LOR data in one ring, i.e. at one particular y-position; the y-coordinate is given by the number of the ring.

However, the two ends of LORs will also impact on detectors in different rings, and if we designate one half-side of the rings as A and the other as B as indicated in Figure 4A.5, we obtain separate sinograms corresponding to each possible ring pair. Moreover, these are no longer symmetrical between halves A and B. LORs impacting on half A in ring $i$ and half B in ring $j$ are organized in a separate sinogram from those for which the converse applies. Thus there are $(4 \times 8)^2 = 1024$ possible sinograms. To limit storage space the number of sinograms is reduced by grouping them together according to the principle shown in Figure 4A.6.

To further limit storage space, (two) angular positions can be grouped together without serious loss of image quality, which is equivalent to grouping rows two by two in Figure 4A.5B.

When reconstructing the 3-dimensional image from the sinograms, attenuation of the gamma rays traveling through the medium is corrected for by measuring the attenuation of gamma rays sent through the subject by the scanner itself.

![Figure 4A. 6 Detector rings seen from above (not to scale). Sinograms cutting in the same z-position in the center are grouped together as long as the corresponding rings are not further apart than a pre-set limit. To the left, four sinograms are grouped with a sinogram from a physical ring, to the right six sinograms are grouped around z-position in-between rings.](image)

The reconstructed images are stored in so-called image files. In the case of the ECAT EXACT HR+ scanner an image is stored as radiation intensity in 128 by
128 by 63 voxels (three-dimensional pixels of 5.148 by 5.148 by 2.381 mm). For this research these images were constructed once per second and converted into files of one-byte numbers representing one voxel. Software was developed in-house to further analyze these files (van der Zwan, 2001).

The software developed makes it possible to analyze the intensity data in the three coordinate planes as indicated in Figure 4A.7.

The data were suitably averaged and normalized. Figure 4A.8 illustrates how. In Figure 4A.8 data for $y$-planes are tabulated. To eliminate differences in absolute intensity between time frames, due to radioactive decay during the experiment, and due to different levels of intensity at $t = 0$ between experiments, all the intensity values were summed at each time frame (A to B in the figure), and normalized to all sum to unity (B to C). To minimize noise, the minimal value for all the time frames was then subtracted from all the values (C to D), and finally the data were again normalized to sum to unity (D to E).

The software has three functions:

- Analyzing the raw data as mentioned above, writing the results to ASCII files for further analysis and visualization
- Creating bitmaps giving the intensity distribution in $x$-, $y$- or $z$ planes.
- Tracking a single particle tracer by searching for the voxel with maximal intensity in each time frame for single particle experiments.
Some blurring of the images takes place due to unavoidable scatter in the LORs obtained. Scatter is due to a number of effects:

- The finite size of the detectors limits spatial resolution, and therefore the LOR precision.
- The positron travels a few mm through the medium before annihilating with an electron.
- The emission of photons may not be exactly collinear, this depends on the momentum of the electron-positron pair before the annihilation.

Moreover, completely false LORs, which give rise to noise in the images, are obtained due to ‘random coincidences’. In ‘random coincidences’ two different events both give rise to single detections within the same time window, the other photons from both events either penetrating the sensors undetected or falling outside the detection zone, giving a bad LOR. Inbuilt in the camera software is an estimation of the frequency of random coincidences, but it is impossible to identify and eliminate the actual false LORs.

![Table and Figure](image-url)
List Mode Files

A drawback of the standard use of sinograms for data reconstruction is that it is limited to predefined time frames with a minimum of 1 second. Smaller time frames require another way of handling the data. The second way of data storage is event-by-event storage of the distance and angle data for the individual LORs. This is useful for tracking a single tracer particle. The data are stored in so-called “list-mode files” in binary form, in buffers of 32 words each of 32 bits, which had to be read and deciphered.

Two types of words are stored in list-mode files: event words giving the data for an LOR and timing words, of which one is stored every ms. The format is the big-endian format with the most significant bit to the left (with the highest address). The binary numbers are thus converted to integers by:

\[ b_{n-1}b_{n-2}......b_0 = \sum_{i=0}^{i=n-1} 2^i \cdot b_i \]  

(2)

For example, if the angle segment of an event word is the binary number ‘011001010’, the angle value is 202:

\[ 011001010 = 2^8 \cdot 0 + 2^7 \cdot 1 + 2^6 \cdot 0 + 2^5 \cdot 1 + 2^4 \cdot 0 + 2^3 \cdot 1 + 2^2 \cdot 0 + 2^1 \cdot 0 + 2^0 \cdot 0 = 202 \]  

The structure of the event words is given in Figure 4A.9.

![Figure 4A.9 The structure of an event word. Bit 31 is to the left (the Tag), and bit 0 to the right.](image-url)
The tag zero indicates an event word. The ‘Event type’ section gives the detector rings in which the event (A and B are the two ‘ends’ of the LOR) was registered. The number of the detector ring registering end A of the LOR, counting from the back, is given by:

$$8 \cdot \left( 2 \cdot BR_{A,i} + BR_{A,0} \right) + D$$

where $BR_{A,i}$ is bit $i$ of ‘Block ring A’ in Figure A.9 similarly with end B. When looking from the front, the left half of the scanner is denoted A, the right half B. The last three flags in the ‘Event type’ section are set to zero if the event is considered a valid one (see below).

Use of the data is illustrated in Figure 4A.10, the numbers given in the text below refer to the actual camera, and those in parenthesis to the simple example in the figure.

One distinguishes between ‘whole angles’, where both ends of the LOR is moved one detector in the rotation, and ‘half angles’ where only one end is moved, as indicated in the figure. To find the LOR corresponding to given $N_\theta$ and $E_{ld}$, one rotates $N_\theta$ whole angles in the clock-wise direction starting from the vertical centre-line as indicated. For this operation whole and half angles are thus lumped, and $N_\theta$ takes on values from 0 (0) to 287 (23).

Of all the possible LORs parallel with the resulting line, only the central half are considered, thus, including the half-angles, there are 289 (25) almost parallel LORs. An element number, $E$, is calculated from the ‘Element ID’, $E_{ld}$, by the formula:

$$E = E_{ld} - \begin{pmatrix} 1 \\ 0 \end{pmatrix} \cdot 512 + 144$$

where the number between brackets is 1 if $E_{ld} > 256$ and 0 if $E_{ld} < 256$. The LOR is then identified by counting $E$ from the left (rotating with the ‘Angle’) as indicated in the figure.

In the listmode files the ‘Element ID’ takes on values in the ranges [0,144] and [368,511], and Equation (4) converts these values to the range [0,288], the range for $E$. 
Figure 4A.10 Simple example illustrating the use of the list-mode data for identifying the LOR in the x-y plane

The structure of the timing words is very simple, the tag is set to 1 rather than zero, and the time in ms is given by bits 26-0.

On basis of these data, the Cartesian coordinates of the detectors A and B were calculated. The equation for a line between two points \((X_A, Y_A, Z_A)\) and \((X_B, Y_B, Z_B)\) in 3-D Cartesian space is:

\[
\begin{align*}
\frac{x - X_A}{X_B - X_A} &= \frac{y - Y_A}{Y_B - Y_A} = \frac{z - Z_A}{Z_B - Z_A}
\end{align*}
\]  

(5)

About 500 LORs were registered per ms in the experiments where a single tracer particle was used. Figure 2B in section 4.3.3 shows 150 such LORs from a particular ms from such an experiment.

Apart from scatter in the ‘good’ LORs, there are also some ‘bad’ ones. Coincidence in time (detector signals within the same narrow time window) does not guarantee that the two detector signals form a good LOR. In ‘random coincidences’ two different events both give rise to single detections within the same time window, the other photons from both events either penetrating the sensors undetected or falling outside the detection zone, giving a bad LOR. The camera estimates the number of these by ‘overlaying’ delayed time windows and determining the number of random ‘coincidences’: a single detection in both windows. This is the ‘Delayed’ flag. More than two detections within one window...
are indicated by the ‘Multiple’ flag. Another possibility is that one or both photons are scattered on the way to the detector, giving rise to deflection and decrease in energy. The camera attempts to identify these by a threshold in photon energy. This is indicated by the ‘Scatter’ flag.

When only a single tracer particle is used in the experiment, two LORs detected close in time can be expected to emanate from the same position, and this position can in principle be found by cross-triangulation. However, scatter is inherent in both the position of the radiation event and in its detection, and the following procedure for finding the position of the particle was therefore adopted.

It was decided to determine one particle position per ms. Bad LORs were first eliminated, and the particle position in the \( x-y \) plane was then determined by averaging the cutpoints between the rest. Computing power was a limiting factor for the analysis; the number of operations for computing particle positions increase with the square of the number of LORs included in the analysis. Writing a program in a compiled language (PASCAL), made it possible to include 100 LORs per ms. The most efficient algorithm was one of ‘zeroing in’ on the average of the good cutpoints by successive elimination of bad cutpoints, i.e. narrowing the window containing the cut-points used for the averaging. The optimal final window size was around 10 mm. Like the crystal coordinates, the position data were generated in a Cartesian coordinate system, which is convenient for viewing projections on the coordinate planes.

Once the \( x-y \) position was found, the \( z \)-position was determined by a similar procedure in the \( y-z \) plane, using only the cutpoints contributing to the \( x-y \) average. The scatter in the millisecond (ms) data and the possibility for fast particle motion in the bed is investigated and shown that for this process the data may be smoothed without loss of information about the particle motion by calculating simple averages of 10-50 ms particle positions (Hoffmann et al., 2003). The investigation also showed that this method allows the particle position to be determined to within about 1 mm\(^3\) (based on ± one standard deviation) once every 0.2 ms. This is high enough spatial and temporal resolution for studying most industrial processes, even those where the particle velocity is high. In single particle experiments simple averages of the ms positions are taken every 20 ms, and the particle position is therefore determined to within about 0.01 mm\(^3\) 50 times per second.
Appendix 4B

Distributor Plate Investigation

Due to the bad distribution of gas that occurred at the first distributor plates for our fluidized beds we carried out a separate investigation of gas distribution and its consequences for the movement of the particles. This investigation was performed in collaboration with the fluidization research group at the Harriot-Watt University by applying different imaging techniques; Particle Emission Tomography technique (PET) by our group and Electrical Capacitance Tomography (ECT) reported by Makkawi et al. (2002) from Harriot-Watt University.

Our initial experimental data showed an uneven axial dispersion of solids indicated by a shift in the profile of tracer solids towards one side of the column (i.e. poor cross-sectional distribution of the drag force exerted on the solids by the fluidizing gas), and even the final data exhibited this to some extent as evidenced by some of the bitmap images in this chapter. This problem probably exists in many industrially operating fluidized beds, but due to the limitation in monitoring techniques and difficulties in visual observation, this subject has not received enough attention. This problem is usually associated with three main defects: (1) low pressure drop across the distributor (2) poor distribution of holes or of porosity over the cross-section of the distributor plate (3) inclination of the fluidization column, or mis-alignment of the distributor inside the column. Since the distributor used in our work was a sintered plate, with a sufficiently high pressure drop to ensure even distribution of the gas flow through it, we are left with the latter two possibilities.

Porous plates like those we used are supposed to give a uniform distribution of gas if the pressure drop over the plate is sufficient, but it is known that non-uniform porosity is a perennial problem with this type of distributor. The work reported from Makkawi et al. (2002) with ECT was undertaken to better understand the role of bed tilt on gas mal-distribution, and to compare the appearance of mal-distribution between two completely different tomographic methods: ECT and PET.

In our PET experiments, we leveled our bed with a spirit level, and we observed that our bed had to be tilted somewhat off the vertical to compensate for a slight cross-sectional mal-distribution. Although this resulted in the appearance of uniform fluidization, the PET results showed this not to be the case.
The ECT method measures the cross-sectional bulk solids concentration. The setup for these experiments comprised a column equipped with a precise tilting mechanism, and experiments were conducted under very similar operating conditions with the same particles and in the same bed geometry as for the PET experiments. The results from ECT studies were compared with PET measurements.

The ECT study demonstrated that a very minor inclination as low as 2° may lead to a dramatic change in the bed dynamics with serious defluidization spots, especially at low excess gas velocity. However, our studies showed that the distributor has to take most of the blame for the gas mal-distribution in our system, although the results of the gas mal-distribution appear very similar whatever the cause. More details about this study can be found in Makkawi et al. (2002).

After this investigation, we changed the distributor plate to a new one and paid careful attention to level the fluidized bed inside the PET’s field of view. The experimental results shown in this chapter are the results of this change.