



The 7th World Congress on Particle Technology (WCPT7)

**Particle Tracking In Fluidized Beds With Secondary Gas Injection**

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**Abstract**

Fluidized beds with secondary gas injection enjoy great popularity in process industry. Owing to their characteristic properties such as intense mixing of solids, excellent mass and heat transfer conditions as well as easy handling of solids, this type of apparatus is applied in various fields of process engineering nowadays. In the past decades research concerning fluidized beds with secondary gas injection has focused on understanding how solid particles and the injected gas are distributed within the apparatus. With the aid of invasive measurement techniques the region surrounding the injector nozzle was investigated with respect to the penetration depth of the gas jet above the nozzle orifice as well as the jet opening angle. A major drawback of the previously used measurement techniques consists in their invasive nature. Penetration of the injection zone by a probe can severely influence the local flow pattern and consequently has a detrimental effect on the reliability of the measured data. Therefore in the presented work for the first time the solids distribution as well as the motion of a single particle in a fluidized bed with secondary gas injection has been investigated by positron emission particle tracking (PEPT). This non-invasive technique is based on labeling one single particle, randomly selected from the bulk, radioactively, which allows for tracking its motion with high temporal and spatial resolution. The obtained data are compared with results derived from invasive measurements. Moreover PEPT-data have been used to perform investigations on the residence time behavior of particles within the jet region and the suspended phase. It could be found that the combination of invasive measurements and PEPT provide valuable information for the design and optimization of fluidized bed reactors with a well-defined injection zone.

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Selection and peer-review under responsibility of Chinese Society of Particology, Institute of Process Engineering, Chinese Academy of Sciences (CAS)

*Keywords:* Fluidized bed; secondary gas injection; positron emission particle tracking; residence time behavior; solids distribution

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## 1. Introduction

In various fields of process engineering ideally mixed systems enjoy great popularity owing to the fact that they provide gradient free conditions with regard to temperature and material distribution. In contrast to that there are also processes requiring oriented flow fields with precisely adjusted local velocity and temperature distribution. Due to their contrary properties it is challenging to realize both of these systems within one single vessel. However, an example for a type of apparatus, which meets this challenging requirement, is the fluidized bed with secondary gas injection. Amongst other applications fluidized bed technology with secondary gas injection is well established in granulation and mixing processes, in chemical reaction engineering, e.g. as a reactor or regenerator system in heterogeneous catalysis, but also in power plant technology.

On closer examination of the fundamental setup of a fluidized bed with secondary gas injection one can distinguish two different zones: The first zone is composed of the actual fluidized bed, in which solid particles are fluidized at moderate superficial gas velocities by feeding the primary process fluid through a distributor plate at the bottom of the plant. Apart from bubble formation, which is commonly observed in fluidized beds, the solids are evenly distributed within the bed. The second zone is induced within the fluidized bed by the injection of a secondary gas, which is fed through the orifice of a nozzle penetrating the fluidized bed. Depending on the velocity of the injected gas a region with reduced solids concentration is formed above the nozzle orifice. According to Merry [1] this region has been termed the jet region.

Within the past years several studies have been conducted to analyze the distribution of the solids and the injected gas within such two-zone reactor systems. Hence the physical dimensions of the jet region [2-5], such as the penetration depth as well as the jet opening angle, could be described. Based on experimental data correlations were derived, by the help of which the jet dimensions can be estimated in dependence of the velocity of the injected secondary gas at the nozzle orifice and the superficial velocity of the primary process fluid. A drawback of previous measurements, however, consists in the fact that experimental data have been obtained using invasive measurement techniques. Penetration of the injection zone by a probe can severely influence the local flow pattern and consequently has a detrimental effect on the reliability of the measured data. Moreover the established measurement techniques only provide information relating to the behavior of the bulk material but do not provide insight into the behavior of individual particles. To overcome these inconveniences in the presented work positron emission particle tracking (PEPT) is used to investigate the solids distribution as well as the motion of a single particle in a fluidized bed with secondary gas injection. This non-invasive technique is based on labeling one single particle, randomly selected from the bulk, radioactively, which allows tracking its motion with high temporal and spatial resolution.

In order to investigate the reliability of data, solids concentrations derived from PEPT measurements are compared to those obtained from invasive solids concentration measurements by means of capacitance probes. Moreover the obtained solids concentration profiles are used together with particle tracking data in order to analyze the residence time behavior of a single particle within the jet region.

## 2. Experimental setup and procedures

The above named investigations concerning the solids distribution and motion of a single particle in a fluidized bed with secondary gas injection are performed at ambient temperature and pressure. The experimental setup is displayed in Fig. 1. The section which comprises the fluidized bed is located in a hollow 1.4301 steel cylinder. The inner diameter of this steel tube is 0.19 m and its length amounts to 1.90 m. At the base of the plant the primary process fluid is fed via a sintered metal base plate. In the course of the experiments reported in this article pressurized air was used as the primary process fluid. On top of the sintered metal gas distributor the fluidized bulk material is located. The fluidized bed is penetrated by a vertically arranged nozzle, which is installed in the center of the gas distributor plate. The structure of the nozzle consists of a cylindrical section with an outer diameter of 0.035 m at the bottom and a truncated conical section at the top. The orifice of the nozzle has a diameter of 0.010 m and is located 0.145 m above the sintered metal distributor plate. Within the scope of the fluid-dynamic investigations performed in this study pressurized air is used as secondary gas.

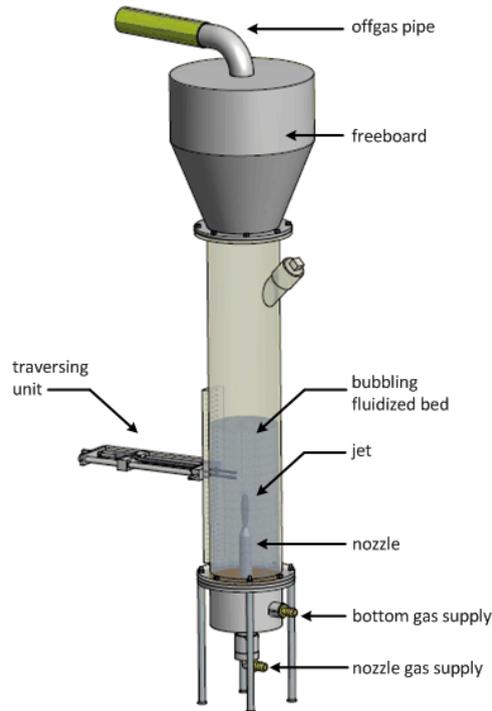


Fig. 1: Fluidized bed with central injector nozzle

Upon passing through the fluidized bed the process gas is directed into a freeboard, which serves to reduce entrainment of single particles that have been swept along with the gas before leaving the plant through a filter.

As fluidized bulk material glass beads with a density of  $2480 \text{ kg m}^{-3}$  and a Sauter-diameter of  $732 \mu\text{m}$  were used. For the mentioned system the point of minimum fluidization was determined by defluidization experiments at ambient conditions, from which the minimum fluidization velocity was inferred as  $0.31 \text{ m s}^{-1}$ . The height of the provided bulk material in the plant amounts to  $0.50 \text{ m}$  prior to switching on the bottom gas supply.

At all series of experiments the superficial gas velocity of the primary process gas was kept constant at  $0.5 \text{ m s}^{-1}$ . The secondary gas was injected with a velocity of  $60 \text{ m s}^{-1}$  at the nozzle orifice. In order to investigate the solids distribution within the bed capacitance probes are used, by the help of which the solids concentration can be inferred as a function of the permittivity in a cylindrical capacitor [6]. Mounted on traversing units those probes can be inserted into the bed at several axial positions above the distributor plate and directed to various radial positions. At each measuring position 125,000 data points are recorded with a frequency of  $5.0 \text{ kHz}$ . Subsequently the mean solids concentration is derived.

In addition to the above named invasive measurement technique the behavior of the bed is analyzed non-invasively by means of positron emission particle tracking (PEPT) [7]. For this purpose a single particle is randomly selected from the bulk and labeled radioactively up to an activity of  $20 \text{ MBq}$  in a cyclotron. Herein oxygen  $^{18}\text{O}$ -isotopes contained in the particle are converted to the radioactive fluorine  $^{18}\text{F}$ -isotope in a nuclear reaction. The  $\beta^+$ -decay of this isotope leads to the emission of positrons that instantaneously annihilate with electrons in the close surroundings of the particle and thus generate  $\gamma$ -radiation. With the aid of two oppositely arranged ADAC Forte  $\gamma$ -ray cameras the position of the particle can be tracked from outside the bed. The active area of the cameras is  $590 \text{ mm}$  by  $470 \text{ mm}$ .

Continuous PEPT-measurement over a time period of 40 minutes yields a data set containing the coordinates of the particle with high temporal and spatial resolution. As described by Goldhirsch [8] discrete particle positions are used to derive continuous fields of different properties by coarse graining. In the present study the bulk density of particles was derived on the basis of the time-dependent particle positions in combination with the coarse graining

function, which in this case is represented by a Gaussian distribution. The bulk density was then used to calculate local solids concentrations.

For the investigation of the residence time behavior of a single particle the bed is divided into annular sections with a height and width of 2.5 mm. As the labeled particle can enter a regarded section  $i$  several times in the course of the measurement, the residence time behavior of the particle is determined by referring the cumulative residence time  $\sum_k \Delta t_{k,i}$  in that section after  $k$  intervals of residence to the total duration of measurement  $t_{tot}$ . As the size of the individual volume elements differs depending on their radial position, the volume fraction of the regarded section  $\Delta V_i/V_{tot}$  needs to be considered. Thus the relative residence time density  $E'_i$  is defined as shown in Eq. 1.

$$E'_i = \frac{\sum_k \Delta t_{k,i} \cdot V_{tot}}{t_{tot} \cdot \Delta V_i} \quad (1)$$

For an ideally mixed system the relative residence time density equals  $E'_{i,ideal} = 1.0$  at every location. This is for example the case for a particle in an ideally fluidized bed with negligible bubble formation, in which the probability to detect that particle at a certain moment  $t_i$  is equal for every location in the bed due to statistical motion of particles. In case that there is a section, in which it is unlikely to detect that particle the relative residence time is smaller than one. In analogy to that  $E'_i$  is larger than one in sections, in which the residence time of the regarded particle is larger than in others.

### 3. Results and discussion

In the following the jet region in a fluidized bed with secondary gas injection will be analyzed with regard to the solids distribution in the surroundings of the injector nozzle orifice. Firstly, measurement results obtained from invasive capacitance probes are presented. Secondly, solids concentrations as determined by the non-invasive measurement technique PEPT are presented and their conformity with invasive measurement data is assessed. Subsequently the residence time behavior of a single particle within the bed is evaluated.

#### 3.1. Solids distribution and penetration depth of the jet

Solids concentrations that were determined invasively by means of capacitance probes are depicted in Fig. 2 a). The figure shows the radial distribution of the solids concentration measured at different axial positions  $\Delta h$  above the nozzle orifice.

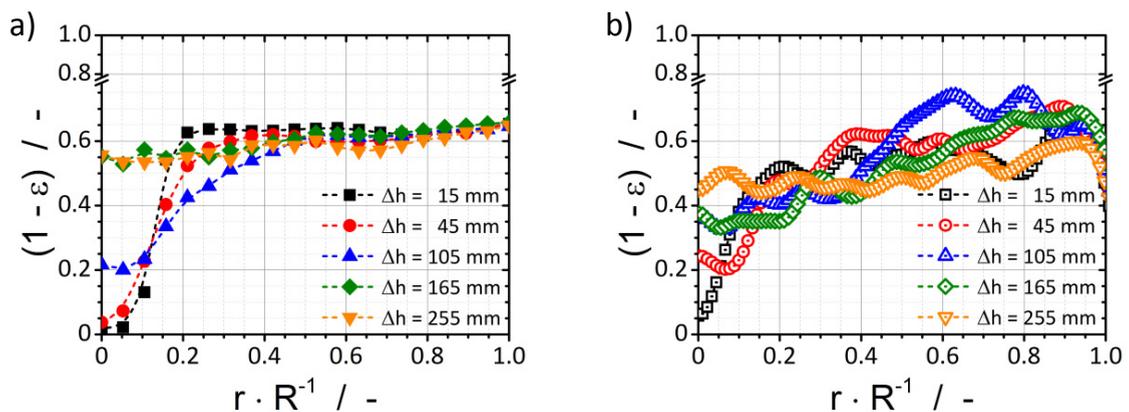


Fig. 2: Radial solids distribution at different axial distances above the nozzle orifice;  $u_G = 0.50 \text{ m s}^{-1}$ ,  $u_{jet} = 60 \text{ m s}^{-1}$ : (a) invasive measurement by capacitance probes; (b) non-invasive measurement by PEPT

Assuming axial symmetry of the bed the displayed data sets were measured between the center and the inner wall of the plant. Data measured on a height of  $\Delta h = 15$  mm above the nozzle orifice show that in the center of the bed above the orifice of the injector nozzle very low solids concentrations prevail. However, when moving in the direction of the wall the solids concentration increases sharply. At a radial position of  $r/R = 0.2$  the concentration levels off at a value of  $(1-\varepsilon) = 0.63$ , which corresponds to the solids concentration in the suspended phase and is close to that at incipient fluidization.

With increasing distance  $\Delta h$  from the nozzle orifice an increase of the solids concentration in the center of the bed is observed. Besides that, flattening of the slope between the center of the bed and the suspension phase indicates broadening of the jet with increasing distance from the nozzle orifice. At an axial distance of  $\Delta h = 165$  mm above the nozzle orifice the influence of the gas injection on the concentration profile can hardly be detected.

As a general trend a slight decrease of the solids concentration in the suspension phase is observed with increasing distance from the point of secondary gas injection. This effect is attributed to the intrusion of the injected gas into the suspension phase, which goes along with broadening of the jet.

Comparison of Fig. 2 a and Fig. 2 b shows that the evaluation of solids concentrations derived from non-invasive PEPT-measurements yields similar trends as obtained from invasive measurements with capacitance probes. At the axial position of  $\Delta h = 15$  mm also PEPT-data show low solids concentrations in the center of the bed, followed by a sharp increase. Levelling of the solids concentration is again observed at  $r/R = 0.2$ . Regarding concentrations derived at larger axial distances above the nozzle orifice broadening of the jet is observed. A major difference, however, exists in the spatial resolution, which is much higher in the case of PEPT-derived data compared to invasive measurements. This adds a high value to PEPT-measurements. However, PEPT-derived solids concentrations exhibit strong fluctuations, which have not been observed during invasive measurement. This is particularly pronounced in the suspension phase.

As shown in Fig. 2 a and Fig. 2 b the lowest solids concentrations prevail in the center of the bed at  $r/R = 0$ . In order to determine the penetration depth of the jet, central solids concentrations are plotted as a function of their axial distance from the point of injection. For both invasive and non-invasive measurements this is depicted in Fig. 3. Regarding data obtained from the capacitance probes the low solids concentration directly above the nozzle orifice is followed by a progressive increase in concentration. At an axial distance of  $\Delta h = 165$  mm above the nozzle orifice the solids concentration reaches that of the suspension phase and remains constant thereafter. Based on invasive measurements, the penetration depth of the jet region thus amounts to  $l_{\text{jet}} = 165$  mm.

Considering particle tracking data, strong fluctuations in the solids concentration are observed. Also here low solids concentrations above the point of injection are followed by an increase of the concentration. Generally the trend of the curve appears to be degressive. Levelling occurs in the range of  $\Delta h = 150$  mm to 200 mm. However, due the strong fluctuation of data the penetration depth of the jet cannot be determined clearly.

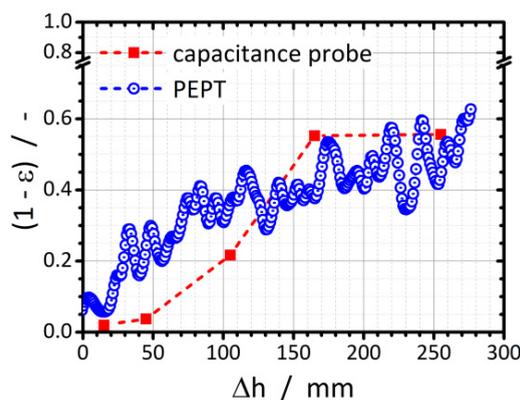


Fig. 3: Axial solids distribution in the center of the bed;  $u_G = 0.50 \text{ m s}^{-1}$ ,  $u_{\text{jet}} = 60 \text{ m s}^{-1}$

### 3.2. Residence time behavior of a single particle

Owing to the high spatial resolution that comes along with particle tracking, PEPT-data were used to determine the boundary between the jet region and the suspended phase and to derive the residence time behavior of a single particle in the jet region subsequently. As shown in Fig. 4 a the outline of the jet region has a conical shape and is determined as the transition point between low concentrations in the jet region and high concentrations of  $(1-\varepsilon) \approx 0,6$  prevailing in the suspended phase. On the basis of the concentration contour plot the half jet opening angle of  $\theta_{\text{jet}} = 16.1^\circ$  is derived.

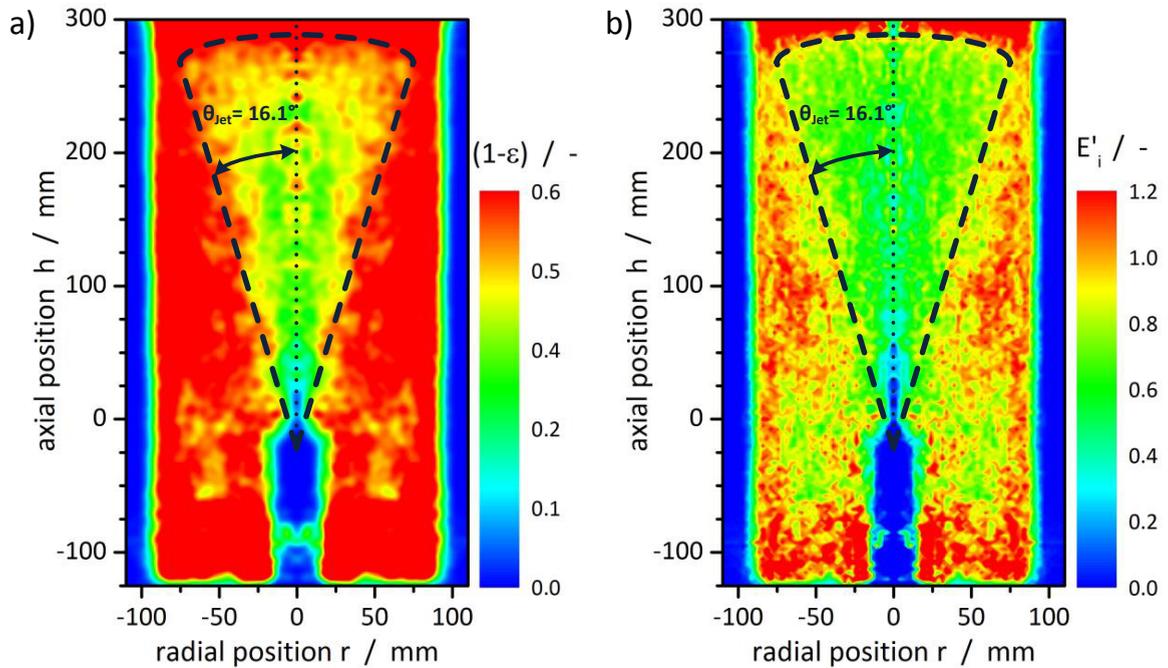


Fig. 4: (a) Solids concentration profile determined by PEPT; (b) relative residence time distribution in the bed;  $u_G = 0.50 \text{ m s}^{-1}$ ,  $u_{\text{jet}} = 60 \text{ m s}^{-1}$

Applying Eq. 1 the residence time behaviour of the labelled particle is inferred. Fig. 4 b shows the relative residence time density  $E'_i$  of the labelled particle in the different sections of the bed. The position of the nozzle is clearly distinguished from the rest of the fluidized bed by very low relative residence time. Furthermore, directly above the nozzle orifice a section with equally low residence time is observed. It may be concluded that the probability of entering this zone is very low for the labelled particle. According to Merry [1] this section is defined as the jet. Above the jet the conically shaped jet region is observed. Considering the boundary line of the jet region as depicted in Fig. 4 a the relative residence time density of the labelled particle within the jet region is obtained as  $E'_{i,\text{jet region}} = 0.699$ . This shows that the residence time of the labelled particle in the observed jet region is 69.9 % of that in the ideally mixed phase with even solids distribution. If the total volume of the bed is known this information provides valuable information for optimization of continuous processes with regard to the solids feed flow rate.

In the zone surrounding the jet region the relative residence time density is in the range between  $E_i = 0.8$  and  $E_i = 1.2$  and fluctuates around  $E_i = 1.0$ , which is typical for ideally mixed beds. Generally fluctuations may be

attributed to bubble formation. However, certain zones are found, in which the relative residence time deviates strongly from that observed in ideally mixed beds. This is for example the case above the gas distributor plate in the bottom section of the bed. Here relative residence time densities larger than  $E_i = 1.0$  are found. A possible explanation for this is demixing of the bed due to the width of the particle size distribution. The volume flow rate of the primary process fluid might not be sufficient to fluidize larger particles, which consequently settle and form a bed with reduced particle motion above the gas distributor. Upon entering this section it is difficult for the labelled particle to leave this section, leading to increased residence time of the particle in that section of the bed.

In contrast to that relative residence time densities smaller than  $E_i = 1.0$  are found in the radial surroundings of the nozzle orifice. This effect might be attributed to vortex flow induced by the high velocity of the injected gas.

#### 4. Conclusion

The focal topic this article consists in the investigation of a fluidized bed with one centrally arranged gas injector nozzle by means of positron emission particle tracking. This novel non-invasive technique allows for tracking the motion of a single particle in the bed with high temporal and spatial resolution. In order to investigate the reliability the measured data, solids concentrations obtained from particle tracking were compared to those obtained from invasive measurements by means of capacitance probes. It could be found that both measurement techniques yield the same trends regarding the distribution of solids, however, particle tracking data provide insight into disparities of the solids concentration throughout the bed. On the basis of invasive measurements the penetration depth of the jet region was derived for the given operating conditions.

Moreover, particle tracking data were used to investigate the residence time behavior of the labelled particle in various sections of the bed. Due to high spatial resolution of the applied technique a map of the residence time density could be established. Based on this map the relative residence time density of a single particle within the jet region was determined. In addition to that the map gave further insight into the residence time distribution throughout the suspended phase. In this way sections with induced flow pattern could be revealed as well as sections with reduced particle motion. With regard to fluidized bed technology in industrial applications the derived understanding of solids distribution and residence time behavior of single particles provides valuable information for the design of new plants as well as optimization for existing plants.

#### Nomenclature

$E_i$	relative residence time density, -
$h$	height above nozzle orifice, mm
$k$	number of residence time intervals, -
$l_{\text{Jet}}$	jet length, mm
PEPT	positron emission particle tracking, -
$r$	radial position, mm
$R$	inner radius of the fluidized bed, mm
$t$	time, s
$t_{\text{tot}}$	total duration of measurement, s
$u_G$	superficial gas velocity, $\text{m s}^{-1}$
$u_{\text{jet}}$	secondary gas velocity at the nozzle orifice, $\text{m s}^{-1}$
$V_i$	volume of section $i$ , $\text{m}^3$
$V_{\text{tot}}$	total bed volume, $\text{m}^3$
$\varepsilon$	void fraction, -
$\theta_{\text{Jet}}$	half jet opening angle

## Acknowledgements

The authors would like to thank Prof. D. Parker for his kind support in conducting PEPT measurements at the Positron Imaging Centre of the University of Birmingham. The support by the German Research Foundation (DFG) within the framework of the Collaborative Research Center SFB-814 and the cluster of excellence Engineering of Advanced Materials is gratefully acknowledged.

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