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# CONTROL OF THE REACTION ZONE IN BUBBLING FLUIDIZED BEDS BY MEANS OF SECONDARY GAS INJECTION

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# ABSTRACT

Fluidized bed reactors with secondary gas injection enjoy great popularity in process industry, e.g. as a reactor system for gas-solid reactions but also as an apparatus for coating of solid particles. Investigations presented in this article deal with the characterization of the solids distribution as well as the distribution of the injected secondary gas within the fluidized bed. In addition to that the residence time behavior of single particles is analyzed by means of positron emission particle tracking. The obtained results provide valuable information for design of fluidized bed reactors with a well-defined injection zone.

## INTRODUCTION

For the first time fluidized bed technology was mentioned in 1922 in a patent by Winkler (<u>1</u>), in which a process for generation of synthesis gas by coal gasification has been described. Owing to their characteristic properties such as intense mixing of solids, excellent mass and heat transfer conditions as well as easy handling of solids, fluidized beds have become a frequently applied type of apparatus in various fields of process engineering nowadays. Amongst other applications they are found in granulation and mixing technology, in chemical reaction engineering, e.g. as a reactor for heterogeneously catalyzed fluid-solid reactions (<u>2</u>), but also in power plant technology (<u>3</u>).

The investigations presented in this article refer to the field of fluidized bed applications with secondary gas injection, i.e. fluidized bed apparatuses which are equipped with injector nozzles that allow for feeding a secondary gas into the fluidized bed in addition to the primary gas fed through the bottom gas distributor. This configuration is the preferred setup for fluidized bed reactors in which contact of the involved reactants with the reactor wall or with the bottom gas distributor is to be avoided in order to prevent undesired reactions. This is the case, for instance, in chemical vapor deposition (CVD) reactions.

Characterization of the jet region around the nozzle orifice has been a focal topic of research concerning fluidization technology since the 1970s (4, 5, 6). Correlations to determine the penetration depth of the jet  $I_{jet}$  and the half jet opening angle  $\theta_{jet}$  have been published by Merry (7). The objective of this article consists in the fluid dynamic characterization of the jet region surrounding a centrally arranged nozzle in a bubbling fluidized bed. Investigation of solids distribution, jet gas distribution as well as residence time behavior of single particles are performed in order to establish understanding of mass transport phenomena at the boundaries of the jet region.

# **EXPERIMENTAL**

The experimental setup for conduction of the above mentioned investigations is displayed in Fig. 1. The zone comprising the fluidized bed consists of a hollow 1.4301 steel cylinder with an inner diameter of 0.19 m, its length amounts to 1.9 m. Particles within the rig are fluidized by pressurized air, which is fed through a sintered metal base plate that is located at the bottom end of the cylindrical section. In the center of the rig a cylindrical nozzle with an outer diameter of 0.035 m is installed. The upper part of the nozzle consists of a truncated conical section with an orifice diameter of 0.010 m and injects the applied secondary gas

into the fluidized bed in vertical upward direction. The orifice of the nozzle is located 0.145 m above the distributor plate.

The fluidized bulk material consists of glass beads with a Sauter diameter of  $732 \,\mu\text{m}$ , a density of 2480 kg m<sup>-3</sup> and a minimum fluidization velocity of 0.31 m s<sup>-1</sup>. Prior to switching on the bottom gas supply the rig is filled with solids up to a height of 0.50 m above the distributor plate.

All series of measurements presented in this article were performed at ambient temperature and pressure. The superficial gas velocity  $v_g$  was 0.5 m s<sup>-1</sup> and the jet gas velocity  $v_{jet}$  at the orifice of the nozzle was 60 m s<sup>-1</sup>. Air was used as secondary gas. In order to track the motion of the injected gas, helium was added as a tracer to the process air before feeding the nozzle. The concentration of the injected gas is measured at several axial positions above the nozzle orifice by means of tracer gas probes, connected to a mass spectrometer. By means of traversing units the probes could be directed to various radial positions. The distribution of solids was measured in a similar approach





using capacitance probes, by means of which the concentration of solids at a certain position in the bed is determined as a function of the permittivity within a cylindrical capacitor. Besides the two above mentioned, invasive measurement techniques positron emission particle tracking (PEPT) was performed to gain understanding of the residence time behavior of single particles. For this purpose a single particle was labeled radioactively in a cyclotron up to an activity of 20 MBq. Due to emission of radiation the position of the particle within the bed could be tracked from outside by means of two oppositely arranged ADAC Forte y-ray cameras, each with an active area of 590 mm by 470 mm.

#### **RESULTS AND DISCUSSION**

In the following, the jet region in a fluidized bed with vertical secondary gas injection through a centrally arranged nozzle is characterized with regard to solids and tracer gas distribution. In addition to that the residence time behavior of a single particle is investigated in different radial and axial sections of the bed.

#### Solids distribution

The solids concentrations measured at different axial positions are presented in Fig. 2. Due to axial symmetry of the bed the displayed data have been measured on positions between the center and the inner wall of the bed.





Fig. 3: Solids concentration in the center of the bed at different axial distances above the nozzle orifice;  $v_g = 0.5 \text{ ms}^{-1}$ ;  $v_{jet} = 60 \text{ ms}^{-1}$ 

Immediately above the nozzle orifice, where the secondary gas enters the fluidized bed, low solids concentrations below  $(1-\epsilon) < 0.05$  are observed. As

measurements at a height of  $\Delta h = 15$  mm above the nozzle opening show, the concentration of solids increases sharply when moving from the center of the bed towards the reactors wall. At a dimensionless radial position of r/R = 0.2 solids concentration reaches the value of  $(1-\epsilon) = 0.63$ , which is close to concentration at minimum fluidization. With increasing height  $\Delta h$  above the nozzle orifice the increase of the solids concentration stretches out over a larger radial distance, i.e. broadening of the jet region is found. Along with radial expansion of the jet region comes an increase of the solids concentration in the center of the fluidized bed. At a height of  $\Delta h = 155$  mm above the nozzle orifice the solids concentration equals that in the suspended phase. Hence no more influence of the jet can be observed.

The solids concentrations measured in the center of the bed, and thus at the same radial position as the nozzle orifice, are depicted in Fig. 3 as function of axial distance from the injection point. The solids concentration first shows progressive increase with increasing axial distance from the nozzle orifice. At distances larger  $\Delta h = 155$  mm no further increase is observed. Thus the jet length can be determined as  $I_{jet} = 155$  mm.

#### Tracer gas distribution

The measurement positions for analysis of the tracer gas concentration were equivalent to those during the solids concentration measurements. The tracer gas concentration c was recorded and subsequently related to the mean cross sectional tracer gas concentration  $c_0$ .





Fig. 4: Radial tracer gas distribution profiles at different axial distances above the nozzle orifice;  $v_g = 0.5 \text{ ms}^{-1}$ ;  $v_{jet} = 60 \text{ ms}^{-1}$ 



As depicted in Fig. 4, tracer gas concentrations exhibit a maximum in the center of the fluidized bed. The highest tracer gas concentration occurs in close vicinity of the nozzle orifice. With increasing distance from the nozzle the concentration

in the center reaches smaller values. However, along with decreasing tracer gas concentrations in the center comes broadening of the concentration profiles with increasing axial distance from the point of injection. In proximity to the inner wall of the bed the tracer gas concentration reaches very low values on all axial levels, which is desired if contact of gaseous reactants with the reactor wall is to be avoided. This is the case e.g. in reactors for CVD reactions.

On the basis of the central tracer gas concentration the penetration depth of the jet  $I_{jet}$  can be determined. Fig. 5 plots the central concentrations versus the axial distance from the nozzle orifice. A degressive trend is observed, at which the concentration of secondary gas decreases quickly upon entering the fluidized bed through the nozzle orifice and levels out at an axial distance of  $\Delta h = 155$  mm. Hence the jet length amounts to  $I_{jet} = 155$  mm, which is in good agreement with that based on the solids concentration measurements (Fig. 3). The influence of superficial gas velocity as well as jet gas entrance velocity on the jet dimensions have been treated in a previous publication by Schober and Wirth (<u>8</u>).

#### Positron emission particle tracking (PEPT)

Measurement techniques treated above allowed for characterization of the jet gas distribution as well as the behavior of the solid phase but not of single particles. In order to investigate the motion of single particles, a particle is selected from the bulk and labeled radioactively. Thus, by means of PEPT the location of the labeled particle within the fluidized bed can be tracked in real-time (9, 10).

In the following the residence time behavior of a single particle within a fluidized bed with secondary gas injection is investigated. For this purpose, the bed has been subdivided into several theoretical sections: Firstly into five radial sections arranged concentrically around the center of the bed (Fig. 6), secondly into six vertical sections stacked on top of each other (Fig. 7). Residence time behavior of the labeled particle after k intervals of residence in a certain section i is determined by referring the residence time  $\sum_k \Delta t_{i,k}$  of the labeled particle within that section to the total duration of measurement  $t_{tot}$ . Furthermore the influence of differently sized zones on the residence time to the volume fraction of the regarded section in the entire bed  $\Delta V_i/V_{tot}$ . Thus the following relative residence time  $E_i$ ' is defined.

$$E'_{i} = \frac{\sum_{k} \Delta t_{i,k}}{t_{tot}} \cdot \frac{V_{tot}}{\Delta V_{i}}$$
(Eq. 1)

The radial distribution of the relative residence time of the labeled particle is shown in Fig. 8. Data reflect the mean value of the relative residence time in the respective annular volume elements and are plotted in the middle of the corresponding interval. Besides that the calculated average value in radial direction is plotted, which serves as a reference to the expected relative residence time in an ideally mixed system, within which E' equals one.



 $V_{i}$ ; i = 1-6

Fig. 6: Radial zoning of the fluidized bed

Fig. 7: Axial zoning of the fluidized bed



Fig. 8: Relative radial residence time for a solid particle in a fluidized bed with secondary gas injection;  $v_g = 0.5 \text{ ms}^{-1}$ ;  $v_{jet} = 60 \text{ ms}^{-1}$ 

Fig. 9: Relative axial residence time for a solid particle in a fluidized bed with secondary gas injection;  $v_g = 0.5 \text{ ms}^{-1}$ ;  $v_{jet} = 60 \text{ ms}^{-1}$ 

In the center of the bed short residence time is observed. Moving towards larger radial positions the residence time of the tracked particle increases and reaches its maximum at 0.6 < r/R < 0.8. In close proximity to the wall a slight decrease of the residence time is found.

Short residence time in the center of the bed originates from low solids concentrations in the jet region and is consistent with the measurements shown in Fig. 2. Besides that, between the jet and its surrounding regions a large fluid velocity gradient occurs ( $\underline{8}$ ), which causes radial acceleration of the particles and thus supports the observed behavior. When reentering the suspended phase of the bed, particles decelerate and remain nearly statically. Due to the lower solids

velocity ( $\underline{8}$ ) and the larger solids concentration (Fig. 2) residence time in the suspended phase is higher.

In Fig. 9 relative residence time is shown as a function of the axial distance h from the bottom gas distributor. As seen in the diagram, the residence time of the labeled particle is evenly distributed over the entire height of the bed. However, at the position of the jet region, between h = 100 mm and h = 255 mm, residence time is slightly reduced. This effect might be attributed to vertical acceleration of the labeled particle by the injected secondary gas.

Combining PEPT data with solids concentration measurements, information on the residence time of a single particle in the region of the jet can be derived. In consideration of the jet length that has been determined according to Fig. 3 the central section of the bed (0 < r/R < 0.2) can be divided into a lower section, that includes the jet, and an upper section, in which suspended phase conditions prevail. Applying PEPT data the relative residence time in the lower section is determined to  $E_{jet}$  = 0.54. This states that the residence time of a single particle within a cylindrical section above the nozzle orifice with length equal to that of the jet is 54 % compared to the residence time in an equally sized volume element in the suspended phase – i.e. the residence time in the nozzle region is approximately half of that in a well-mixed fluidized bed reactor.

The derived understanding of radial and axial residence time behavior of a single particle in a fluidized bed with secondary gas injection provides a powerful tool for the design of fluidized bed reactors for gas-solid reactions with a well-defined reaction zone.

## CONCLUSION

Fluidized beds with secondary gas injection enjoy great popularity as a reactor system for gas-solid reactions. In the present article fluid dynamic investigations covering the solids distribution as well as the distribution of the injected secondary gas have been carried out. Moreover PEPT experiments have been performed in order to analyze the residence time behavior of a single particle.

On the basis of the measured data the jet region could be characterized and information on the penetration depth of the jet into the fluidized bed could be obtained. In addition to that PEPT data provide valuable information on the residence time behavior of a single particle in the jet region and suspended phase.

It can be concluded that the obtained data provide a powerful tool for design and optimization of the reaction zone in fluidized bed reactors with secondary gas injection.

# NOTATION

C E' h i k I <sub>Jet</sub> r	tracer gas concentration relative residence time density height above bottom gas distributor index - volume element number of residence time intervals jet length radial position	- - mm - mm			
			R	inner radius of the fluidized bed	mm
			t	time	S
			v	gas velocity	m s⁻¹
			V	volume	mm³
			Δ	difference operator	-
			3	void fraction	-
$\theta_{Jet}$	half jet opening angle	o			

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