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# DESIGN OF NEW FLUIDIZED BED REACTORS FOR CVD - PROCESSES

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# DESIGN OF NEW FLUIDIZED BED REACTORS FOR CVD - PROCESSES

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## 1. ABSTRACT

A new concept of fluidized bed will be shown which can be used for various gas-to-solids reactions (e.g. chemical vapour deposition (CVD) reactions) with high throughput like the production of new materials. For the design and scale-up of the new bubbling fluidized bed with vertically aligned vertical nozzles the fluid dynamics of the fluidized beds have to be determined and analysed, especially the flow around the gas nozzles. A jet region around a single centrally arranged injector lance in a bubbling fluidized bed reactor is characterized by different parameters like solids concentration and jet gas distribution. It can be shown that – depending on the related parameter – different jet regions are obtained.

## 2. INTRODUCTION

Bubbling fluidized beds, in which particles are kept in abeyance by a vertically upwards orientated gas stream, have different advantages, which make them interesting for the chemical industry: easy handling of solids due to the liquid like behavior, fast solids mixing leading to nearly isothermal behavior and an intensive particle-fluid-contact, resulting in high material- and heat transfer coefficients (Werther (16)). Surveys of the hydrodynamics of gas-solid-fluidization are given by, e.g. Lim et al. (8) and Yang (18). A possible application of fluidized beds is the use as reactor for CVD-processes. A good overview over such processes gives e.g. Choy (17). In the case of CVD-reactions, viz the deposition of solid material on the surface of particles, the reactant gas is injected into the fluidized bed reactor through nozzles in different number and position.

Many results of investigations from different authors are published up to now, but most of them deal with gas jets in two-dimensional fluidized beds or if the investigations were made in three-dimensional fluidized beds, the diameters of the reactors were very small (Yan (19), Massimilla (4), Merry (2, 3), Wu et al. (5), Copan et al. (13), Kimura et al. (7), Markhevka et al. (1), Hong et al. (9), Musmarra (12), Guo et al. (14), Cleaver et al. (6), Vaccaro (10,11)).

The most important parameters to describe a jet region are the jet penetration depth and the jet angle. Several correlations are published to calculate these two values. The most famous are the correlations of Merry for the penetration height $L_{Jet}$ and the half jet opening angle $\theta$ of a vertical gas jet in a fluidized bed (Merry (3)).

Penetration depth of vertical gas jet:

$$\frac{L_{Jet}}{d_0} = 5.2 \cdot \left(\frac{\rho_f \cdot d_0}{\rho_s \cdot d_p}\right)^{0.3} \cdot \left(1.3 \cdot \left(\frac{U_0^2}{g \cdot d_0}\right)^{0.2} - 1\right) \qquad (1.1)$$

Half jet angle of vertical gas jet:

$$\theta = \arctan \cdot \left(\frac{1}{10.4} \cdot \left(\frac{\rho_s \cdot d_p}{\rho_f \cdot d_0}\right)^{0.3}\right) \qquad (1.2)$$

The aim of the present paper is to investigate the flow around the nozzles in bubbling fluidized beds of scale which is typical for scale-up experiments. The investigations are focused on the fluid dynamics in a bubbling fluidized bed with centrally aligned vertical injection nozzle.

## 2. EXPERIMENTAL

The objective of this work was the characterization of a jet region in a bubbling fluidized bed with additional vertical gas injection through a centrally arranged nozzle (see Fig. 1).

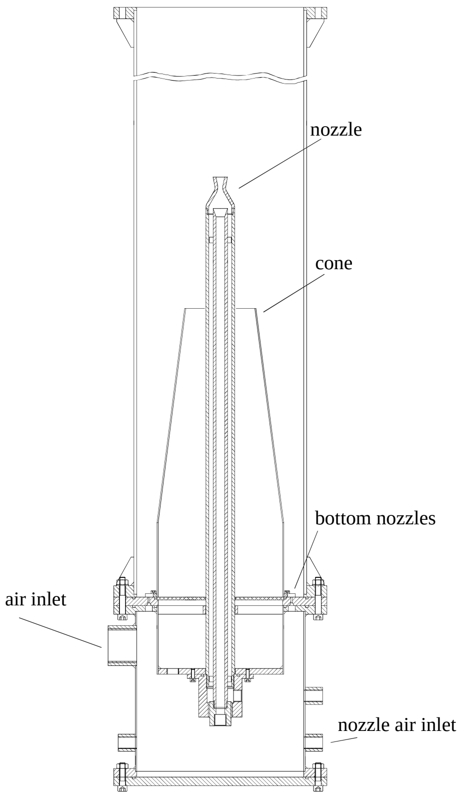



Fig. 1: Fluidized bed reactor model

The fluidized bed reactor model had an inner diameter of 190 mm and a height of 2000 mm. It was made out of plexiglass and in the centre an injector lance with a length of 482 mm was installed. The nozzle on top of the lance had an outlet diameter of 10 mm. To intense the solids movement in the fluidized bed a conical tube was concentrically installed around the lance (see Fig. 1). The air for fluidizing the solids was applied through 12 bottom nozzles with a diameter of 1.9 mm in the bottom of the reactor outside the cone. The experiments were carried out at ambient conditions. The used solid (fused quartz) had a mean diameter of $d_p$ = 732 µm and a density of $\rho_S$ = 2480 kg/m$^3$. The reactor was filled with particles up to a static bed height of 700 mm, thus 220 mm above the

nozzle mouth. All presented results were obtained at a superficial gas velocity of $U_G$ = 0.46 m/s and a jet gas velocity of $U_{Jet}$ = 60.13 m/s.
In order to measure the secondary gas distribution a mass spectrometer in conjunction with a tracer gas (helium) was used. The helium was added to the air applied through the injector lance and its concentration was measured by the mass spectrometer at different radial and axial positions.

## 3. RESULTS AND DISCUSSION

In the following, the jet region in a fluidized bed reactor with additional gas injection through a centrally arranged injector lance is characterized by different parameters (solids concentration and jet gas distribution). Contrary to most publications up to now the nozzle mouth is in a relatively large distance from the bottom gas distributor.

### 3.1 Solid concentration distribution

Radial solids concentration profiles were recorded by using capacitance probes (Richtberg (15))Fig. 2 shows some measured concentration profiles for a superficial gas velocity of $U_G$ = 0.46 m/s and a jet gas velocity of $U_{Jet}$ = 60.13 m/s.

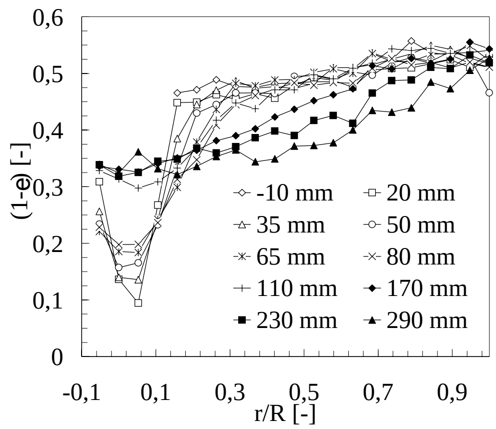
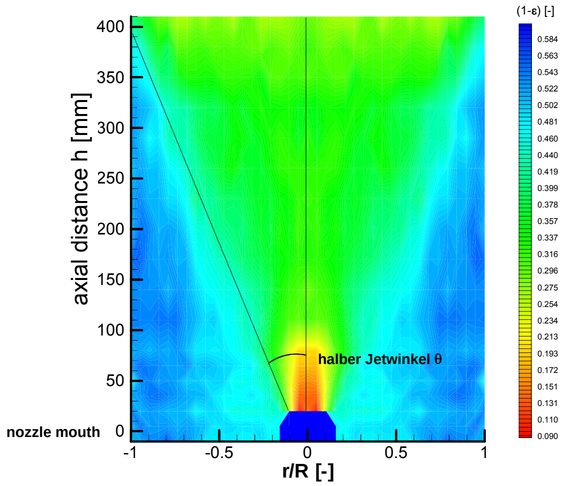



Fig. 2: Radial solids concentration profiles for different axial distances h from the nozzle mouth; $U_G$ = 0.46 m/s, $U_{Jet}$ = 60.13 m/s, $d_p$ = 732 μm, $\rho_S$ = 2480 kg/m$^3$, $h_{bed}$ = 700 mm

Above the injector lance at the radial position r/R = 0, a jet region with low solids concentration down to (1-ε) < 0.1 is formed. With increasing the axial distance h from the nozzle mouth, the solids concentration in the centre of the fluidized bed increases and an increasing radial extension of the jet region can be detected. A value of 80 % of the solids concentration in the suspension close to the reactor wall is assumed to be the boundary of the jet region, a half jet angle of θ = 13.7° can be calculated (see Fig. 3). This angle is clearly smaller than the calculated angle by means of Merry´s correlation (see equation 1.2). The correlation provides a half jet angle of θ = 22.8°.

Merry´s correlation for the penetration depth of vertical gas jets in fluidized beds gives a value of $L_{Jet}$ = 113 mm (see equation 1.1). Regarding the solids concentration (1-ε) at the radial position r/R = 0 the jet penetration depth can be defined to $L_{Jet}$ = 110 mm (see Fig. 4).



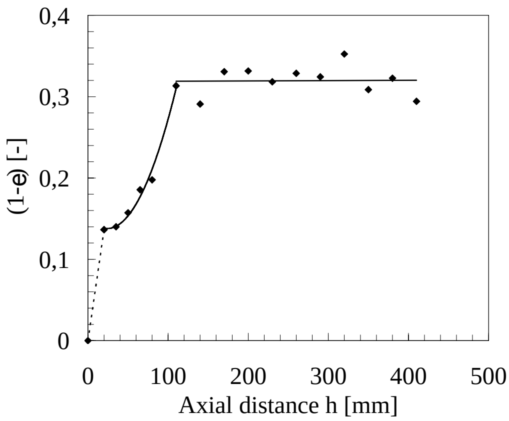




Fig. 3: Solids concentration distribution; $U_G$ = 0.46 m/s, $U_{Jet}$ = 60.13 m/s, $d_p$ = 732 µm, $\rho_S$ = 2480 kg/m$^3$, $h_{bed}$ = 700 mm

Fig. 4: Solids concentration at the radial position r/R = 0 against the axial distance h; $U_G$ = 0.46 m/s, $U_{Jet}$ = 60.13 m/s, $d_p$ = 732 µm, $\rho_S$ = 2480 kg/m$^3$, $h_{bed}$ = 700 mm

Between h = 20 mm and h = 110 mm a distinct increase in the solids concentration (1-ε) can be detected. Above h = 110 mm, the values for the concentration show fluctuations around a value of about (1-ε) = 0.32 and no clear tendency is observable. Not only the solids concentration directly in the centre of the bed, but also the mean cross-sectional solids concentration within the jet region, mentioned above (θ = 13.7°), shows the same jet length $L_{Jet}$ (see Fig. 5).
The solids concentration outside the jet region is nearly constant over the total investigated axial distance from the nozzle mouth.

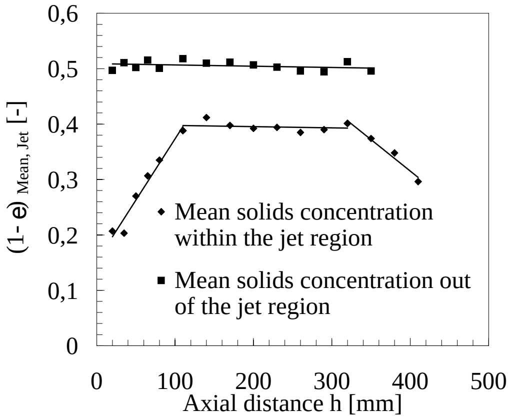



Fig. 5: Dependency of mean solids concentration within and outside the jet region on axial distance h; $U_G$ = 0.46 m/s, $U_{Jet}$ = 60.13 m/s, $d_p$ = 732 µm, $\rho_S$ = 2480 kg/m$^3$, $h_{bed}$ = 700 mm

The solids concentration distribution shows a half jet angle of θ = 13.7° and a jet penetration depth of $L_{Jet}$ = 110 mm. The jet length agrees well with the value calculated by using Merry´s correlation but the jet angle is significantly smaller than calculated by means of Merry´s correlation. The reasons for that might be the definition of the jet angle by Merry (2), using the diameter of separating bubbles from the jet and the investigation of gas jets in a two-dimensional fluidized bed.

### 3.2 Jet gas distribution

Beside the solids concentration, the injected gas (e.g. reactant gas) is of particular interest. To estimate the distribution of the jet gas, the applied air was tagged with helium and its concentration was measured with the use of a mass spectrometer. Radial profiles were recorded and the local concentration of helium C was related to the cross-sectional average value $C_0$. The dimensionless radial concentration profiles are mirrored at the vertical reactor axis at r/R = 0. Profiles are diagrammed for different axial distances h from the nozzle mouth in Fig. 6. For smaller distances clearly higher values of $C/C_0$ in the centre of the fluidized bed (r/R = 0) are obtained (e.g. $C/C_0 > 50$ for h = 5 mm).

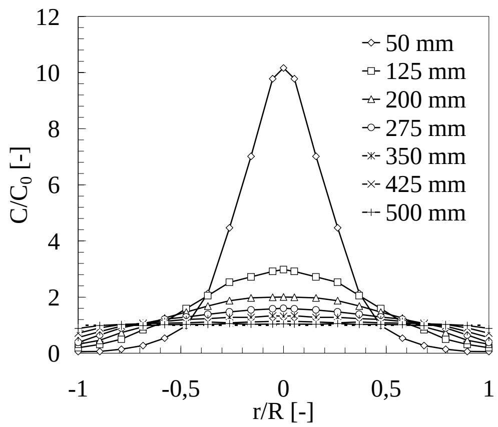



Fig. 6: Radial dimensionless tracer gas concentration profiles $C/C_0$; $U_G$ = 0.46 m/s, $U_{Jet}$ = 60.13 m/s, $d_p$ = 732 μm, $\rho_S$ = 2480 kg/m$^3$, $h_{bed}$ = 700 mm

With increasing the axial distance h, the dimensionless concentration decreases in the centre of the reactor and next to the wall an increase can be detected. A dimensionless concentration of $C/C_0$ = 1 is assumed to be the limiting concentration of the jet region. In Fig. 7 the radial positions at which the concentration profiles cross $C/C_0$ = 1 are plotted against the dimensionless radius r/R. The graph shows no linear dependency over the total investigated distance from the nozzle mouth, but it can be divided into two parts: from h = 0 mm to h = 50 mm and from h = 50 mm to the highest regarded distance of h = 500 mm. The following values are obtained to calculate the jet radius $r_{Jet}$ using $r_{Jet} = r_0 + h \cdot \tan\theta$:

$r_0$ = 43.0 mm and θ = 4.9° (50 mm < h < 500 mm)

$r_0$ = 10.3 mm and θ = 38.7° (0 mm < h < 50 mm)

$r_0$ = 8.3 mm and θ = 11.4° (0 mm < h < 500 mm)

For h = 0 mm the nozzle radius is set to be the jet radius.

Beside the jet angle θ, the jet length $L_{Jet}$ is of interest regarding a potential reaction region. Fig. 8 shows the axial characteristics of the dimensionless helium concentration $C/C_0$ at the radial position r/R = 0 directly above the nozzle mouth.
Up to a vertical distance of h ≈ 120 mm, a clear decrease of the concentration can be detected. For larger distances, the decrease in concentration takes place much weaker. Thus a jet penetration depth of about $L_{Jet}$ = 120 mm – comparable with the jet length estimated by the solids concentration – can be determined.

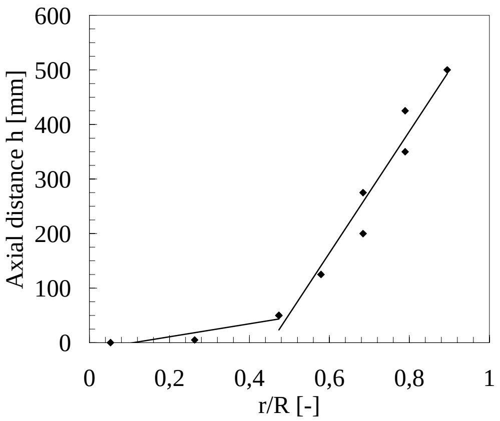



Fig. 7: Boundary of jet region on the basis of jet gas distribution; $U_G$ = 0.46 m/s, $U_{Jet}$ = 60.13 m/s, $d_p$ = 732 µm, $\rho_S$ = 2480 kg/m$^3$, $h_{bed}$ = 700 mm

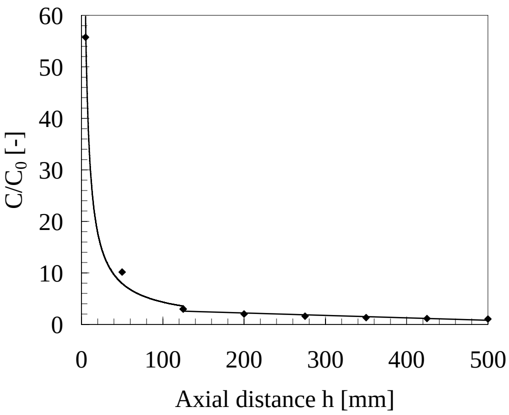



Fig. 8: Dimensionless jet gas concentration $C/C_0$ at the radial position r/R = 0 depending on the axial distance h; $U_G$ = 0.46 m/s, $U_{Jet}$ = 60.13 m/s, $d_p$ = 732 µm, $\rho_S$ = 2480 kg/m$^3$, $h_{bed}$ = 700 mm

The determination of the jet length using gas mixing experiments with the chemically inert gas helium inert results in the largest extension of the jet length. With a chemical reaction the relevant jet length is much shorter.
For changing parameters like temperature or different fluidizing medium standard scale-up criteria can be used.

## 4. CONCLUSIONS

By using different parameters like solids concentration and jet gas distribution different jet region boundaries can be estimated which is shown in Fig. 9.

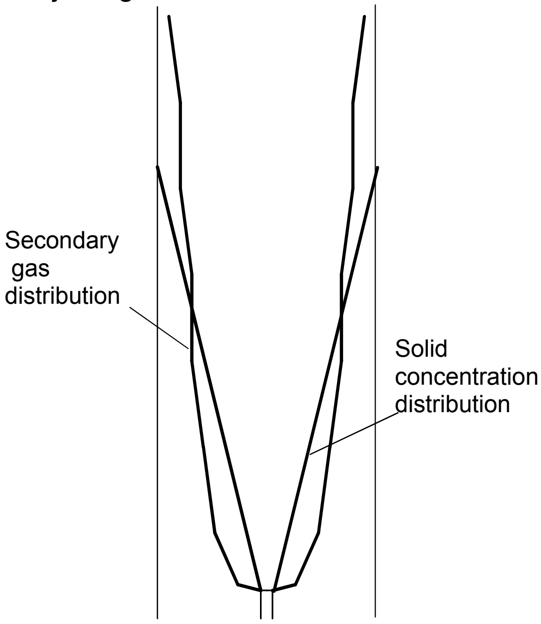



Fig. 9: Boundaries of jet region on the basis of different parameters; $U_G$ = 0.47 m/s; $U_{Jet}$ = 60.13 m/s; $d_p$ = 732 µm; $\rho_S$ = 2480 kg/m$^3$; $h_{bed}$ = 700 mm

If a CVD-reaction should be implemented in a fluidized bed reactor, some terms must be fulfilled: a solids concentration as high as possible near the nozzle to provide the injected reactant gas enough surface area for deposition, avoiding e.g. homogenous gas phase reactions. The jet gas should be well distributed over a wide range of the cross sectional area to use the possible reactor volume for the reaction and guarantee a homogenous coating of the particles. But a problem can be the deposition of solid material on the reactor wall. Thus the contact between the reactant gas and the normally hot reactor wall should be in large distances h from the

nozzle. Fig. 9 shows regions next to the nozzle mouth with high solids concentration and high jet gas concentration, thus adequate conditions for chemical reactions at the particle surface exist.
The definition of a jet region depends on the related parameter like solids and jet gas distribution which are relevant for the reaction considered and may influence them in a different way.

## 5. LIST OF SYMBOLS

| | |
|---|---|
| $C$ | Jet gas concentration |
| $C_0$ | Mean cross sectional jet gas concentration |
| $c_S$ | Solids concentration |
| $d_p$ | Mean particle diameter |
| $d_0$ | Nozzle diameter |
| $h$ | Axial distance from the nozzle mouth |
| $h_{bed}$ | Bed height |
| $L_{Jet}$ | Jet length |
| $L_I$ | Injector lance length |
| $r$ | Radius |
| $R$ | Radius of fluidized bed |
| $U_0$ | Nozzle gas velocity |
| $U_G$ | Superficial gas velocity |
| $U_{Jet}$ | Jet gas velocity |
| $U_S$ | Solids velocity |
| | |
| $(1-\varepsilon)$ | Solids concentration |
| $(1-\varepsilon)_{Mean, Jet}$ | Mean solids concentration within the jet |
| $\rho_S$ | Solids density |

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## 7. KEYWORDS

fluidized bed, gas jet, jet region, nozzle, secondary gas injection, bubbling bed