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Unsteady and steady-state particle size distributions in batch and continuous fluidized bed granulation systems

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Abstract

A two-dimensional balance model of populations distributed by property was developed using the ordinates of particle diameter and particle density. This model makes it possible to calculate the spectrums of granulation changing over time in the fluidized bed during batch operation and in addition to calculate the time-dependent size distribution of the product granulate for continuous operation. A physics-based model was devised for the submechanism of the growth of the granulates. On the one hand, the population balance model is divided into the particle balance around the granulator and, on the other hand, when granulation is continuous, particles are constantly removed from the fluidized bed. To this end, a separator model was used that subdivides the extractor device into two areas of balancing in accordance with the geometry of the apparatus and the properties of the particle. In the case of continuous operation, the results when the product is classified using a classifier tube inside the fluidized bed are discussed. The outcome is the influence of the various particle flows on the particle diameter and the product mass withdrawn. In particular, the problematic nature of the start-up is dealt with, i.e. reaching stable points of steady operation. The simulation results are validated using results from continuous tests on a semi-industrial plant.

Keywords: Fluidized bed; Drying; Modelling; Population balance; Granulation

1. Introduction

In fluidized bed granulation and fluidized bed coating, respectively, particles of a size distribution, which are fluidized with air kept at a specified temperature, are sprayed with a solid solution, a solid suspension or a solid melt. The solvent then evaporates while the solid deposits itself on the fluidized particles. As a result of this, these granulates grow. If the particles achieve a particular property, for the most part a particular diameter, then they are removed from the apparatus. This process can be run in batches or also continuously with an extractor device. Simultaneously, extremely different mechanisms such as growth, agglomeration, abrasion and breakage occur. These mechanisms have an influence on the properties, on the number of the particles in the apparatus as well as on the mass flows occurring between the individual parts of the plant. Fig. 1 presents the active mechanisms of the real technical process with a possible variant for external classification of the discharge in oversize, product, undersize as well as the grinding of the oversize and the subsequent recirculation of the dust. A modified presentation from [1] about the growth phases in fluidized bed granulation serves as the basis.

The existing particle aggregates, both in the apparatus and in the mass flow of the product, exhibit distribution functions. Only a few studies exist for the specification of the change of these functions over time [2–10] and these usually do not deal with the complexity of the overall process. For that reason, this study presents a mathematical population balance model that specifies the parameters of influence and their effect.

2. Mathematical modelling

Disregarding the fracture and the agglomeration, the balance scheme as in Fig. 2 represents an expansion of Mörl's model presentation [11].

The essential components of the model are the areas of balancing around the granulator and the separator. An essential aspect of the modelling is that the mass flows occurring between the area of balancing have a distribution structure which can change over time, which is a function of the coordinates introduced in the system distributed by properties. These distributions are taken into account and examined in the balancing. Taking into consideration that

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Nomenclature

A	area or surface (m^2)
d	diameter (m)
D	dispersion coefficient (m^2/s)
L	length (m)
m	mass (kg)
$\dot{m}$	mass flow (kg/s)
$\dot{n}$	particle flow (s^{-1})
N	number of particles (−)
q_0	number density distribution (mm^{-1}, $1/(m\,kg/m^3)$)
t	time (s)
T	probability of discharge (−)
v	velocity (m/s)
x	water content of the liquid (mass-%)

Greek symbols

κ	$\kappa = d_{dis}^2/d_{apparatus}^2$ (%)
ρ	density (kg/m^3)

Subscripts

ax	axial
B	back from the discharge tube
bed	fluidized bed
dis	discharge of product
pr	period of revolution
R	remaining in the apparatus
sum	summary
susp	suspension

the particle systems observed exhibit a multi-dimensional property distribution and that the system described here possesses a two-dimensional property distribution, Eq. (1) with the ordinates particle diameter and particle density results from the balance around the granulator. The left term representing the changeover time of the particle in the granulator. The terms on the right side characterize the particles flowing toward or away from the areas of balancing:

$$\frac{\partial N_{sum} q_0}{\partial t} = -\frac{\partial N_{sum} q_0 v_\rho}{\partial \rho_P} - \frac{\partial N_{sum} q_0 v_d}{\partial d_P} + \dot{n}_{nuclei} q_{0,nuclei} + \dot{n}_R q_{0,R} + \dot{n}_B q_{0,B} + \dot{n}_{dust} q_{0,dust} - \dot{n}_{bed} q_{0,bed} \tag{1}$$

2.1. Particle growth

With the assumption of the uniform wetting, the following ensues for the change of mass of a particle:

$$\frac{dm_P}{dt} = \frac{A_P}{A_{sum}} \dot{m}_{susp}(1-x) \tag{2}$$

The following ensues for the growth rate of a particle:

$$v_d = \frac{dd_P}{dt} = \frac{2\dot{m}_{susp}(1-x)}{\rho_{solid} A_{sum}} \tag{3}$$

The growth rate of the particles increases by the same value for every particle diameter. The time dependency of the particle diameter can be determined for a monodisperse aggregate with a constant number of particles by integrating Eq. (3). Using the initial condition $d_P(t=0) = d_{P,0}$ stands for the particle diameter of a monodisperse aggregate

$$d_P(t) = \sqrt[3]{\frac{6\dot{m}_{susp}(1-x)}{\rho_{solid}\pi N_{sum}} t + d_{P,0}^3} \tag{4}$$

2.2. Change of density

The density of a particle consisting of a core (hold-up) and a coating (solid) results from

$$v_\rho = \frac{d\rho_P}{dt} = \frac{6 d_{P,0}^3 (\rho_{solid} - \rho_{P,0}) \dot{m}_{susp}(1-x)}{d_P^4 \rho_{solid} A_{sum}} \tag{5}$$

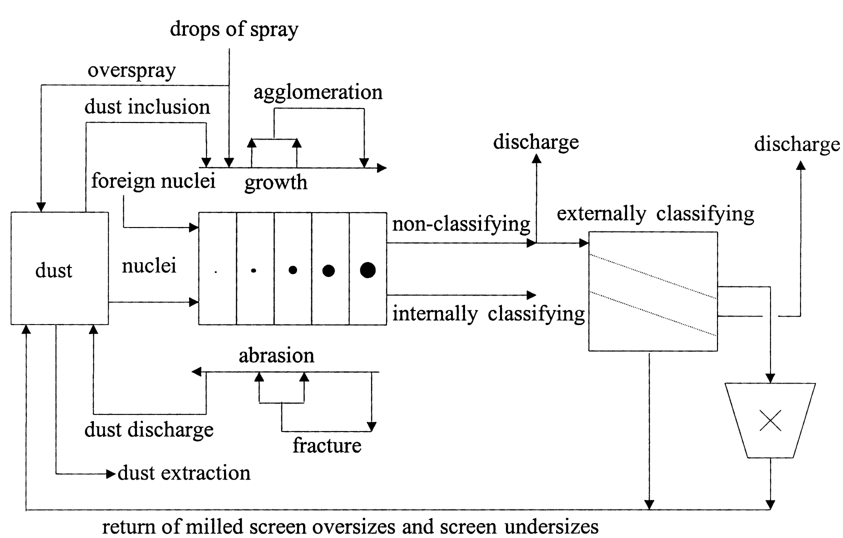

Fig. 1. Mechanisms of fluidized bed granulation and coating.

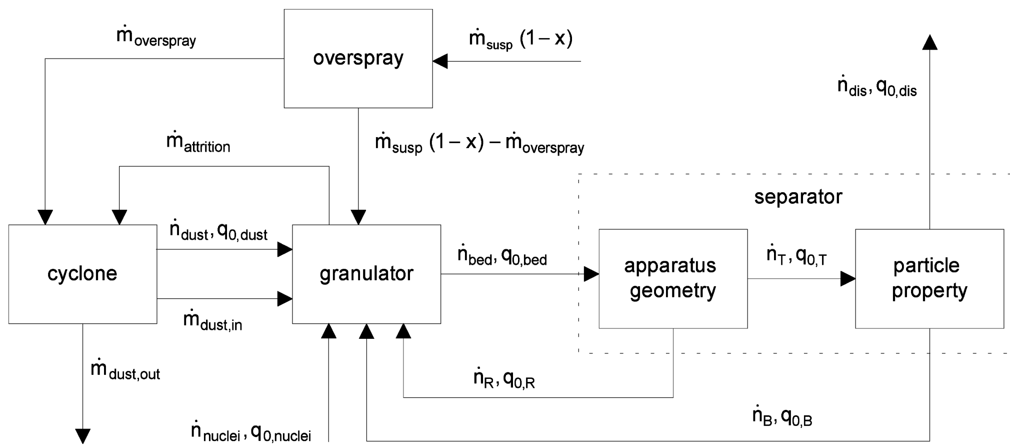

Fig. 2. Model balances of fluidized bed granulation and coating.

In comparison with Eq. (4) stands

$$\rho_{\mathrm{P}}(t)=\rho_{\mathrm{solid}}-\frac{d_0^3(\rho_{\mathrm{solid}}-\rho_0)}{d_{\mathrm{P}}(t)^3} \tag{6}$$

2.3. Modelling of the separator

The first area of balancing specifies the particle flow flowing to the extractor device as a function of the apparatus geometry and the place of residence. The second area of balancing characterizes the process of the particle discharge from the fluidized bed on the basis of the particle properties. When solving the balances, it was assumed that the particles have no residence time in these two areas of balancing. Thus a quasi-stationary analysis takes place. Various types of discharge are calculable.

2.3.1. Classification through a classifier tube

When classifying through a classifier tube (discharge tube), a separation of the particles occurs on the basis of their different sedimentation velocities. The particles are discharged through a tube in the bottom of the fluidized bed apparatus. In order to ascertain the particle flows, which reach the discharge tube conditional on the apparatus geometry, the calculation of the first areas of balancing is completed first.

The following applies to the probability of collision of a particle with the discharge tube in a period of observation Δt:

$$P(\geq 1)=1-(1-\kappa)^{\Delta t/t_{\mathrm{pr}}} \tag{7}$$

The particle flow colliding with the discharge tube can now be ascertained from this probability:

$$\dot{n}_{\mathrm{T}}q_{0,\mathrm{T}}=\dot{n}_{\mathrm{bed}}q_{0,\mathrm{bed}}P(\geq 1) \tag{8}$$

The particle flow remaining in the apparatus is therefore calculated from

$$\dot{n}_{\mathrm{R}}q_{0,\mathrm{R}}=\dot{n}_{\mathrm{bed}}q_{0,\mathrm{bed}}(1-P(\geq 1)) \tag{9}$$

In view of the different sedimentation velocities, the process of the particle discharge out of the fluidized bed is specified in accordance with Molerus and Hoffmann [12]:

$$T(d_{\mathrm{P}},\rho_{\mathrm{P}})=\frac{1}{1+(v_{\mathrm{dis}}/v_{\mathrm{sink}}(d_{\mathrm{P}},\rho_{\mathrm{P}}))\times\exp[-(v_{\mathrm{sink}}(d_{\mathrm{P}},\rho_{\mathrm{P}})-v_{\mathrm{dis}})L_{\mathrm{dis}}/D_{\mathrm{ax}}]} \tag{10}$$

In order to ascertain the particle flow discharged from the apparatus, the number of the particles flowing to the classifying area must be multiplied by the corresponding probability of discharge T:

$$\dot{n}_{\mathrm{dis}}q_{0,\mathrm{dis}}=\dot{n}_{\mathrm{T}}q_{0,\mathrm{T}}T(d_{\mathrm{P}},\rho_{\mathrm{P}}) \tag{11}$$

The particle flow flowing back from the discharge tube therefore corresponds with

$$\dot{n}_{\mathrm{B}}q_{0,\mathrm{B}}=\dot{n}_{\mathrm{T}}q_{0,\mathrm{T}}[1-T(d_{\mathrm{P}},\rho_{\mathrm{P}})] \tag{12}$$

2.3.2. Discharge through the non-classifying discharge

When modelling a granulation fluidized bed with a non-classifying discharge, which is operated so that the bed mass maintains a constant value, instead of the probability of collision, a factor must be determined, which specifies the quantity of the discharged particles as a function of the solid mass and nucleating mass brought in.

3. Numerical solution

The population balance equation (1) can be solved only with difficulty in the form presented. However, if a shift is made from the Euler analysis to the Lagrange analysis, a normal differential equation system results. Taking Eq. (1) as the starting point, the following is obtained with Lagrange coordinates:

$$\frac{\mathrm{d}N_{\mathrm{sum}}q_0}{\mathrm{d}t}=\dot{n}_{\mathrm{nuclei}}q_{0,\mathrm{nuclei}}+\dot{n}_{\mathrm{R}}q_{0,\mathrm{R}}+\dot{n}_{\mathrm{B}}q_{0,\mathrm{B}}+\dot{n}_{\mathrm{dust}}q_{0,\mathrm{dust}}-\dot{n}_{\mathrm{bed}}q_{0,\mathrm{bed}} \tag{13}$$

$$\frac{\mathrm{d}d_\mathrm{P}}{\mathrm{d}t} = v_\mathrm{d}(d_\mathrm{P}, \rho_\mathrm{P}, t), \qquad \frac{\mathrm{d}\rho_\mathrm{P}}{\mathrm{d}t} = v_\mathrm{P}(d_\mathrm{P}, \rho_\mathrm{P}, t) \tag{14}$$

This system of equations specifies the movement of a control volume in the property space in Lagrange coordinates as well as the change of the number density of this control volume. In order to specify the changeover time of the structure of the polydisperse aggregate using this equation, the bed is split up into a number of monodisperse-size fractions. Corresponding with the properties of the particles contained in them, these size fractions are assigned a control volume in the property space. This space is represented by a discrete point d_P and ρ_P. The differential equation system is now solved quasi-stationary for each of these size fractions using the explicit Euler method.

4. Simulation results

The following boundary conditions apply to all simulations: $t = 10\,\mathrm{h}$, $m_{\text{hold-up}} = m_{\mathrm{P},0} = 10\,\mathrm{kg}$, $\dot{m}_\mathrm{solid} = 3\,\mathrm{kg/h}$, $\dot{m}_\mathrm{nuclei} = 1\,\mathrm{kg/h}$, $\rho_\mathrm{solid} = 2000\,\mathrm{kg/m^3}$. The particle density of the hold-up, of the foreign nuclei and of the solid in the suspension, possesses the same value for these calculations. It is, however, also possible to vary the particle density in relation to the particle density of the base material (hold-up) in order, e.g. to calculate a coating process. In order to obtain a starting value for the numerical solution, the particle aggregates are specified by normal distributions. Both the hold-up and the added foreign nuclei exhibit an identical particle spectrum.

4.1. Batch process without addition of foreign nuclei and without discharge (simulation 1)

The hold-up in the apparatus is sprayed with a solid suspension. A particle growth then takes place. The number of the nuclei in the apparatus is constant. Fig. 3a plots the progression of the Sauter diameter against time. That a constant decrease of the growth rates can be expected in this mode of operation is visible in Fig. 3a. After 60% of the duration of granulation ($\tau = 0,6$), the growth rate amounts now only to 50% of the initial value. As is plotted in Fig. 3b, this decrease is conditional on the doubling of the bed surface. In accordance with the linear statement for growth in the granulating fluidized bed (Eq. (3)), a halving of the growth rate follows for a doubling of the surface area. This relationship is confirmed by the model. The distribution density of the number in Fig. 3c is changed in their form only to a very slight extent. The number density distribution is plotted three-dimensionally against the total

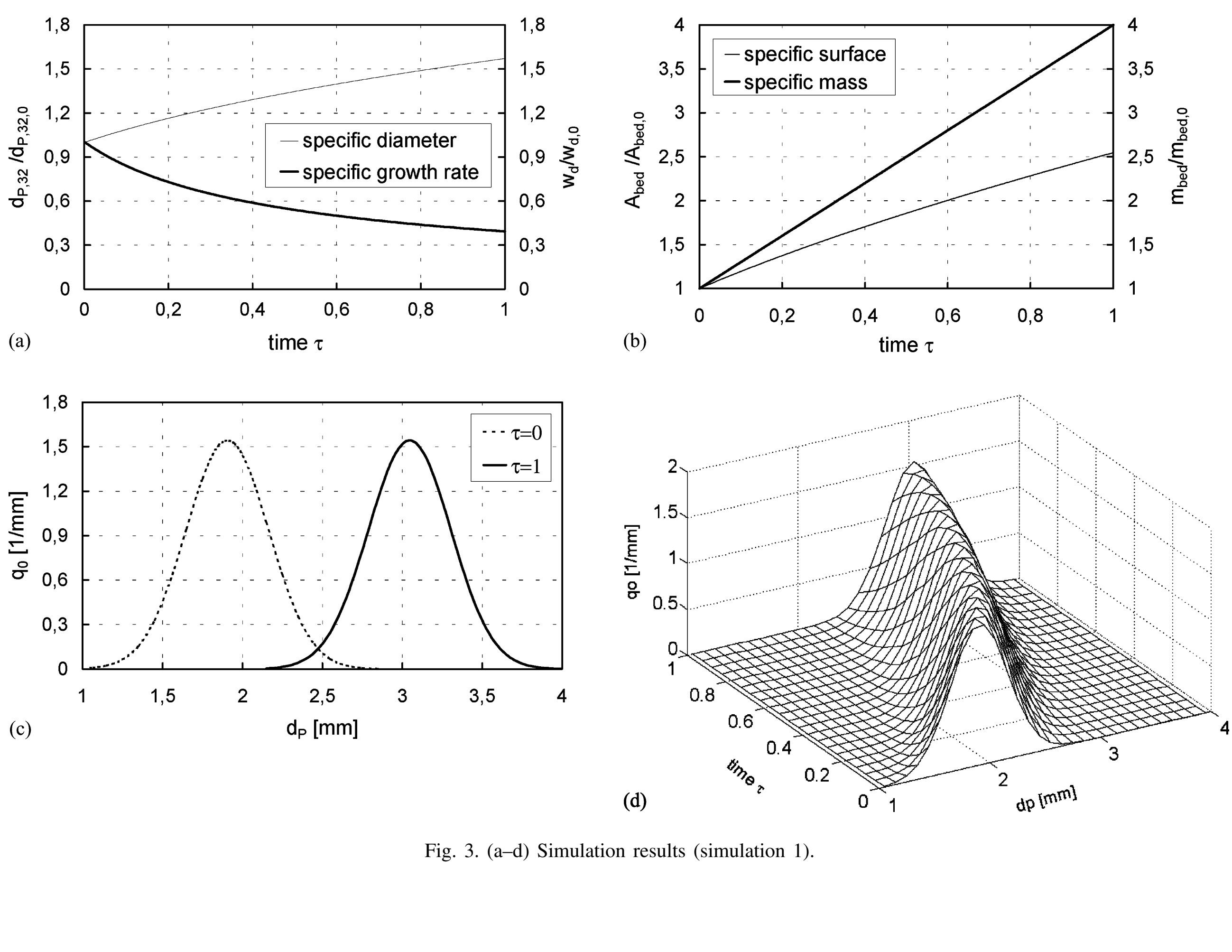

Fig. 3. (a–d) Simulation results (simulation 1).

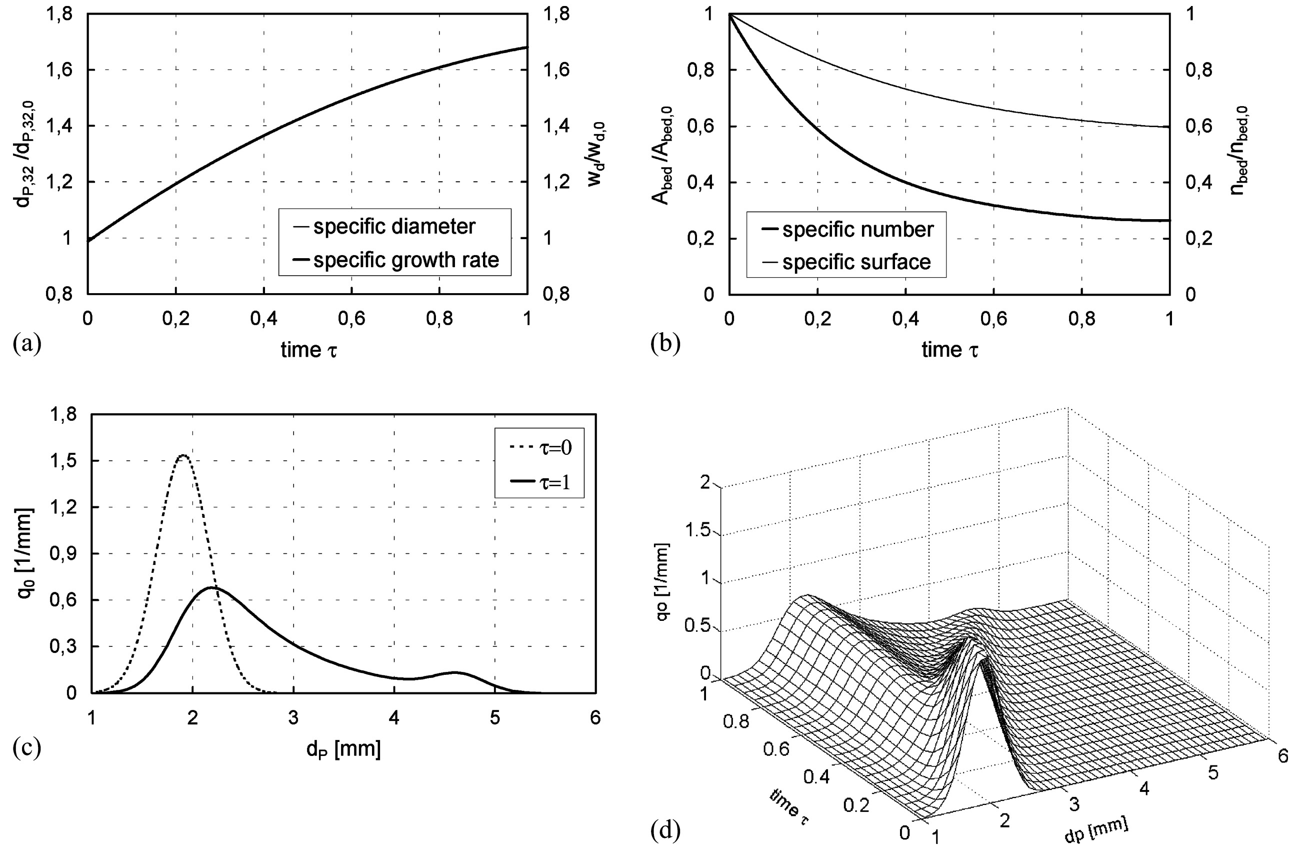

Fig. 4. (a–d) Simulation results (simulation 2).

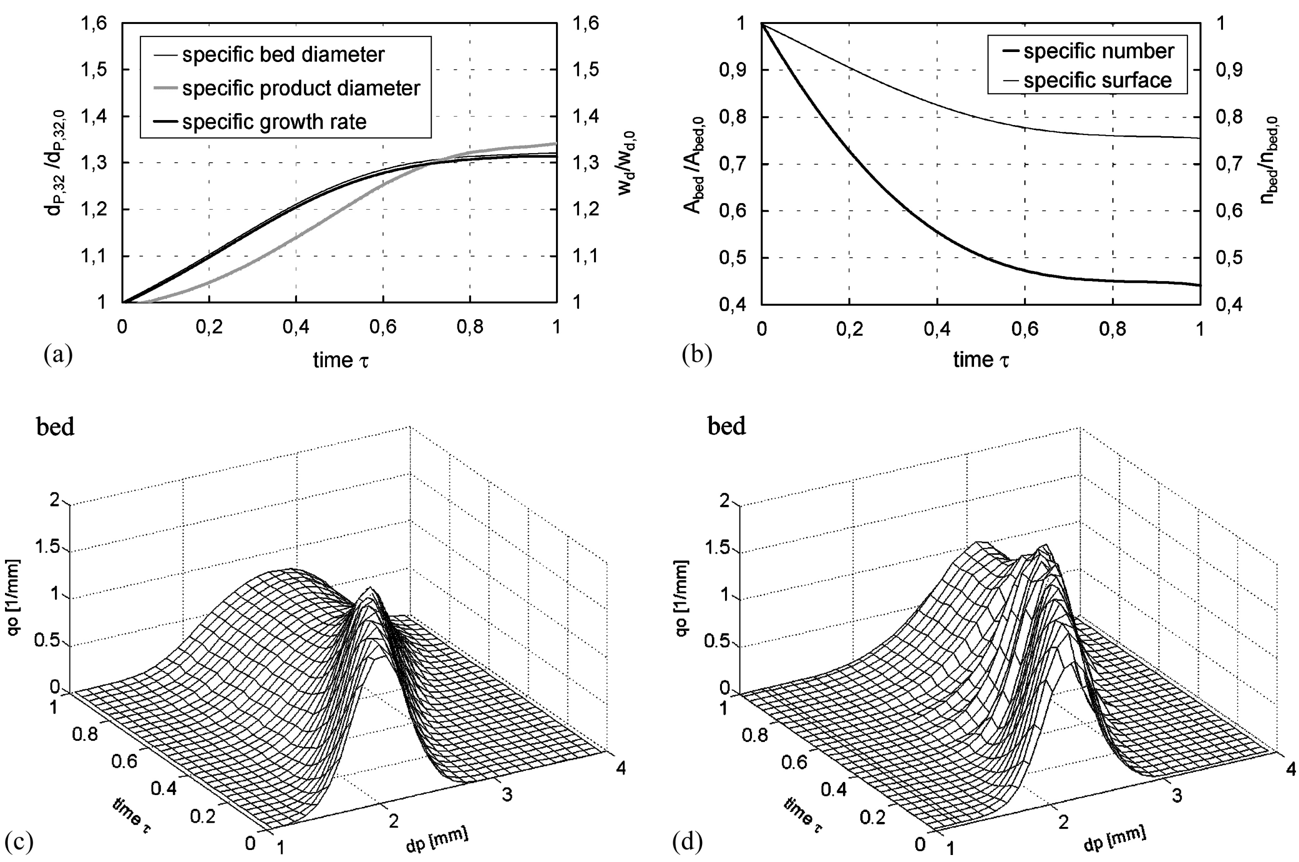

Fig. 5. (a–d) Simulation results (simulation 3).

simulation time in Fig. 3d. From this graph, very good changes over time in the structure of the distribution can be made out and facilitate discussion of the observations which follow.

4.2. Continuous process with addition of foreign nuclei and non-classifying discharge (simulation 2)

In calculation 2, a fluidized bed is simulated during the unsteady start-up phase, which is to be transformed into a steady-state by adding foreign nuclei and permanently withdrawing particles. A constant granulate spectrum is characteristic for the steady point of operation. To develop this granulate spectrum, beginning with the spraying in of the suspension, it is necessary to add foreign nuclei of specific quantity and size into the fluidized bed. The steady point of operation is only reached after the complete discharge of the granulate of the hold-up. After the simulation time of $t = 10\,\mathrm{h}$, no steady-state occurs in the particle size distribution of the fluidized bed for the selected boundary conditions. Neither the Sauter diameter (Fig. 4a) nor the bed surface and the particle number (Fig. 4b) assume a constant final value.

With the help of the particle size distributions, the relation between the steady-state and the discharge of the hold-up specified above is very recognizable. The three-dimensional plotting of the number density distribution of the bed (Fig. 4d) shows the increasing influence of the foreign nuclei brought in, while the particles of the hold-up constantly lose importance for the characteristics of the bed. This process is more clearly presented in Fig. 4c. The local maximum on the right side of the distribution represents the particles of the hold-up still remaining in the apparatus, while the left side is dominated by the nuclei alone. This moment of reaching a steady-state is clearly extended by the characteristics of the non-classifying discharge, since through this form of discharge, foreign nuclei are constantly ejected as equally as particles of the hold-up. As a result, the latter can reside longer in the fluidized bed, while newly added particles possibly leave the bed immediately.

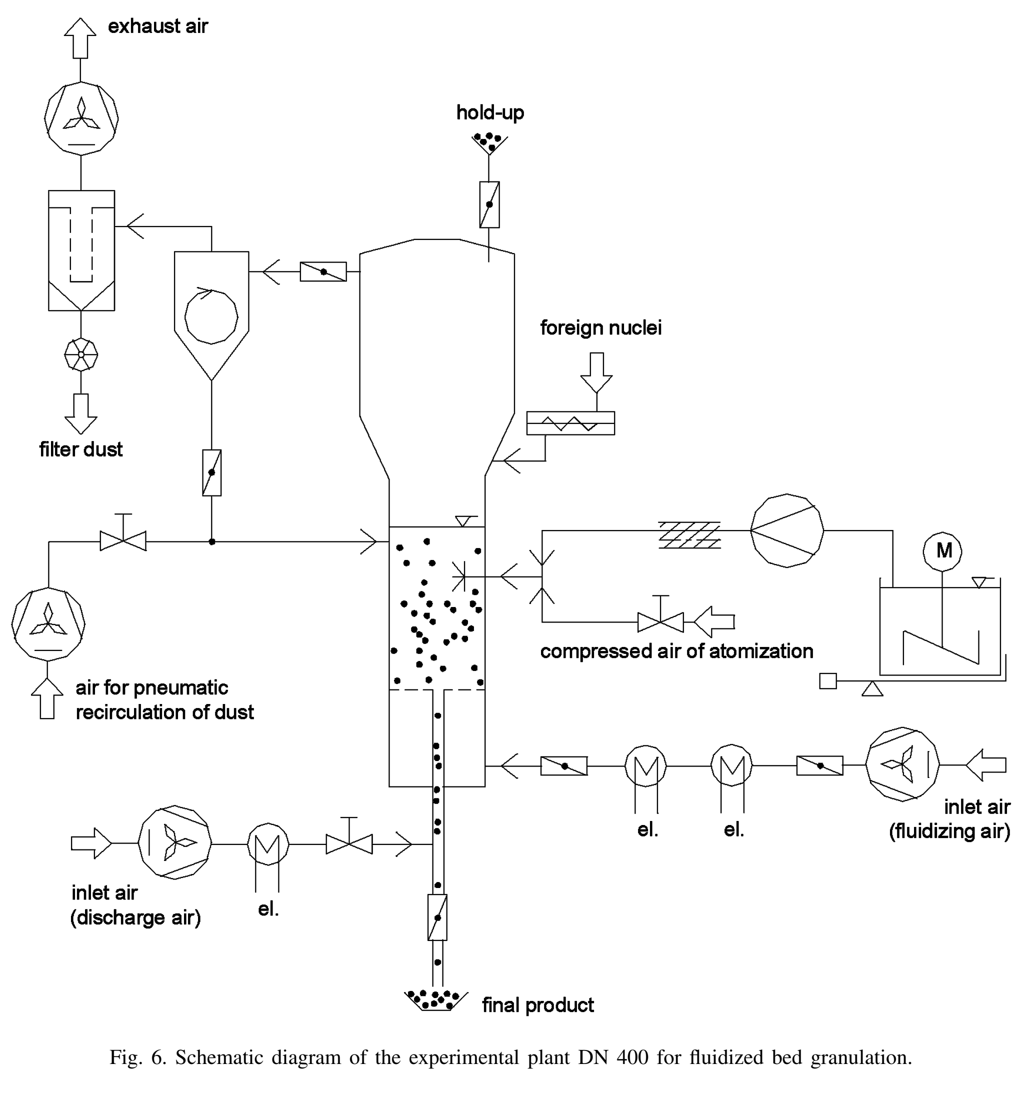

Fig. 6. Schematic diagram of the experimental plant DN 400 for fluidized bed granulation.

4.3. Continuous process with addition of foreign nuclei and classifying discharge (simulation 3)

Analogous to simulation 2, a continuous process is examined, in which the particle discharge occurs through a classifying discharge. During the start-up process, the withdrawal must be constantly changed in such a way that, by increasing the classifying air, a particle flow with a diameter spectrum growing larger and larger is removed from the apparatus, the bed mass on the other hand having to remain constant.

In contrast to simulation 2, transforming the bed into a steady-state is successful. The progression of the Sauter diameter over time is characteristic for the unsteady start-up process of a fluidized bed, in which the added foreign nuclei exhibits the diameter spectrum of the hold-up (Fig. 5a). The diameter then runs increasing monotonously, asymptomatically to the final value. After approximately 80% the breakthrough time is reached, at which the last particles of the hold-up have left the apparatus. At this moment, the Sauter diameter assumes its steady final value. In comparison with the non-classifying variant of simulation 2, it

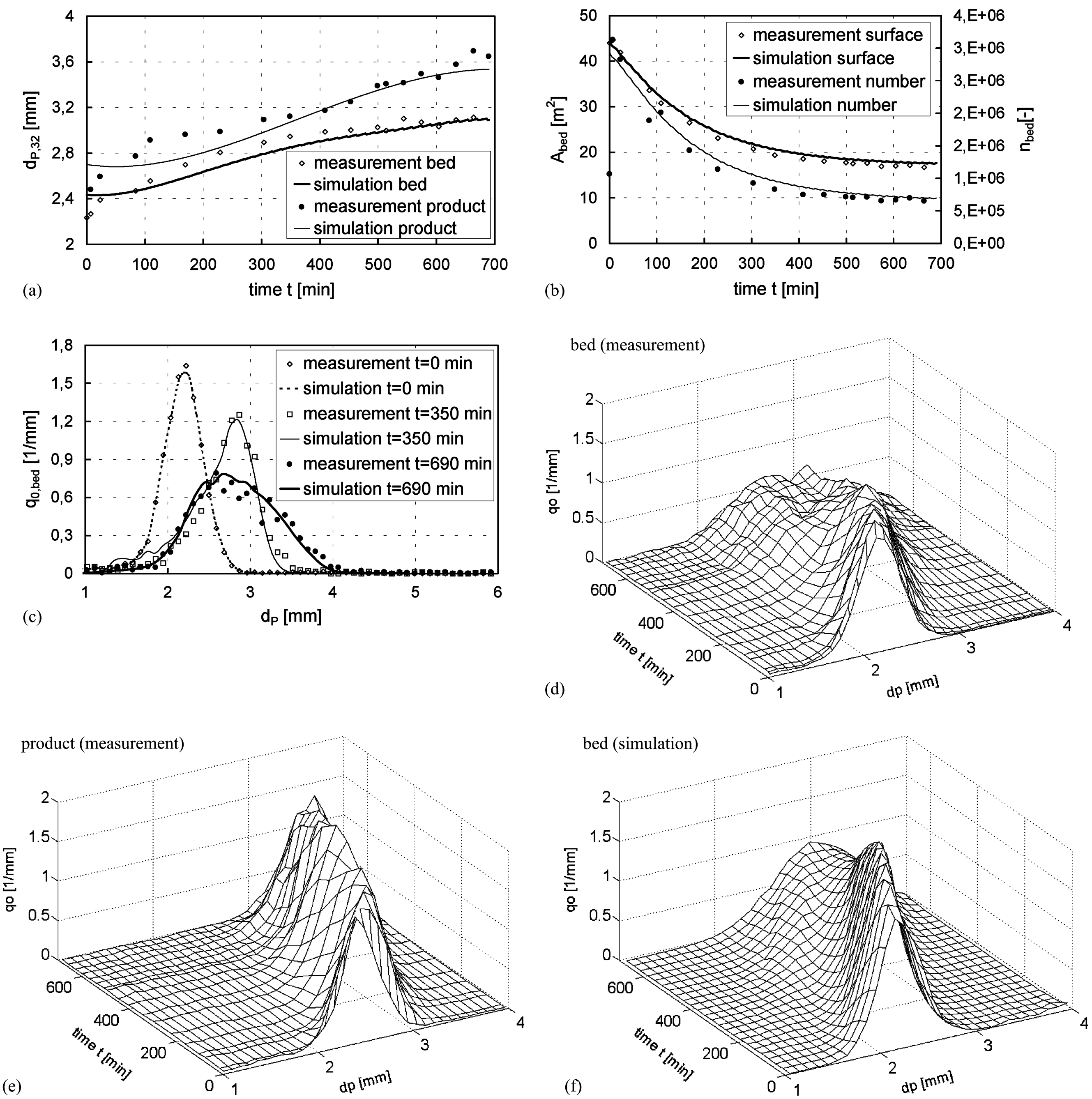

Fig. 7. (a) Measured and simulated Sauter diameter. (b–g) Measured and simulated values for the experiment in continuous mode of operation.

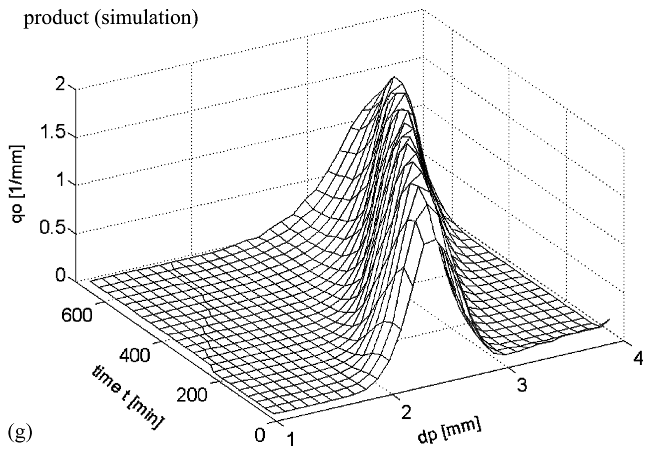

Fig. 7. (*Continued*).

becomes clear that the obtainable diameter increase turns out considerably smaller. In addition the specific product–Sauter diameter is plotted in the figure. The time progression of the bed surface as well as the number of the particles in the fluidized bed express that the bed assumes its steady final value after approximately 70% of the simulation time (Fig. 5b).

The increasing dominance of the foreign nuclei is recognizable on the distribution spectrum. Through the classifying effect of the extractor device, large particles are discharged first. These are primarily granulates of the hold-up. The condition for the steady-state, i.e. the discharge of the particles of the hold-up, is thus realized considerably faster than in the non-classifying variant. Additionally, the time progression of the product distribution is plotted in Fig. 5d. Caused by the classifier device, this distribution exhibits a considerably smaller diameter range than the bed.

5. Experimental validation

The bed mass must be kept constant during the continuous mode of operation (see simulation 3). In order to ensure this, the classifying air flow must be increase constantly, since the diameter of all particles increases and consequently larger particles must be removed constantly from the fluidized bed. The scheme of the experimental plant is shown in Fig. 6. During the granulation experiment (see Fig. 6), a lime dust suspension ($\rho_{\mathrm{lime}} = 2762\,\mathrm{kg/m^3}$) was sprayed onto mustard grains ($\rho_{\mathrm{mustard}} = 1180\,\mathrm{kg/m^3}$). To obtain a better granulating effect, carboxyl-methyl-cellulose (CMC) was added to the suspension as a binder. The suspension had the following composition: $m_{\mathrm{water}} = 7.5\,\mathrm{kg}$, $m_{\mathrm{lime}} = 2.5\,\mathrm{kg}$, $m_{\mathrm{CMC}} = 0.2\,\mathrm{kg}$. In addition, the following boundary conditions were set in the apparatus: $\vartheta_{\mathrm{air,\,in}} = 120\,^{\circ}\mathrm{C}$, $\dot{m}_{\mathrm{air}} = 0.6$–$0.5\,\mathrm{kg/s}$, $m_{\mathrm{hold\text{-}up}} = m_{\mathrm{P},0} = 20\,\mathrm{kg}$, $\dot{m}_{\mathrm{susp}} = 13.55\,\mathrm{kg/h}$, $x_{\mathrm{solid}} = 4\%$, $\dot{m}_{\mathrm{nuclei}} = 0.6\,\mathrm{kg/h}$, $t = 690\,\mathrm{min}$.

In the experiment, bringing the fluidized bed to a nearly steady-state succeeded (see Fig. 7a–g). Both the breadth and the maximum of the calculated particle size distribution correspond well with the measured values. While the left side of the bed distribution is decisively dominated by the added foreign nuclei, the flat progressions of the right side depend on the characteristics of the classifier device and the probability of collision with the discharge tube. After approximately 400 min, the particles of the hold-up increasingly lose influence over the distribution spectrum. The product exhibits a considerably narrower diameter range and, as a result, a greater maximum value than the bed.

6. Conclusions

This study presents a population balance model to calculate the granulation spectrums during fluidized bed granulation and coating, respectively. The fluidized bed is specified as a polydisperse material system, i.e. taking an initial particle size distribution as the point of departure, conclusions can be made when spraying in a solid solution, solid suspension or solid melt, taking into account the particle size distribution of the foreign nuclei introduced, the nuclei on the particle size distribution which changes over time in the fluidized bed. The number, the surface, the volume, the mass as well as the Sauter diameter of the solid particles are calculated (see Fig. 7). A physics-based particle growth model is formulated, with the help of which a diameter growth rate and a rate of density change of the particle are introduced. The model can be connected with the temperature and humidity distributions [13,14].

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