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РАСЧЕТНО-ЭКСПЕРИМЕНТАЛЬНОЕ ИССЛЕДОВАНИЕ ГРАНУЛИРОВАНИЯ В РЕАКТОРЕ С КИПЯЩИМ СЛОЕМ

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Целью работы является построение простой, но информативной модели для описания кинетики гранулирования в периодическом реакторе кипящего слоя. Для описания этой кинетики предложена ячеечная модель, базирующаяся на теории цепей Маркова. В соответствии с числом наблюдаемых фракций по размеру частиц введено несколько параллельных цепей, составленных из ячеек идеального перемешивания. Состояние процесса описывается векторами объемного содержания частиц в ячейках. Эволюция состояния обусловлена переходом частиц из ячеек одной цепи в другую из-за увеличения их размера при грануляции и миграцией частиц вдоль цепей из-за взаимодействия с восходящим потоком ожижающего газа. Состояние процесса наблюдается в дискретные моменты времени. Считается, что объем раствора, поступающий в ячейку в течение одного временного перехода, взаимодействует только с теми частицами, размер которых может быть увеличен до значения, переводящего их в соседнюю более крупную фракцию. Миграция частиц вдоль цепей для свой фракции контролируется матрицей переходных вероятностей, которая различна для каждой фракции и зависит от полной концентрации частиц в ячейках. Модель позволяет производить количественную оценку влияния параметров процесса на кинетику гранулирования. Для проверки адекватности модели были выполнены экспериментальные исследования гранулирования сульфата аммония в лабораторном реакторе кипящего слоя. Сравнение расчетных и опытных данных выполнено на примере влияния расхода раствора на кинетику гранулирования. Получено хорошее соответствие расчетных и опытных данных как по росту среднего размера частиц, так и по изменению во времени их фракционного состава.

Ключевые слова: псевдоожиженный слой, грануляция, цепь Маркова, вектор состояния, связующий раствор, рост частиц, кинетика

COMPUTATIONAL AND EXPERIMENTAL STUDY OF GRANULATION IN FLUIDIZED BED REACTOR

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The objective of the study is to build a simple but informative model to describe the kinetics of layering granulation in a batch fluidized bed reactor. A cell model based on the theory of Markov chains to describe this kinetics is proposed. Several parallel chains of perfectly mixed cell according to the number of size fractions, which are under observation, were introduced. The vectors of particles volume content in the cells describe the state of the process. Evolution of the state is conditioned by particles transition from the cells of one chain to another due to their size enlargement during granulation and by particles migration along the chains due to their interaction with fluidizing gas upstream flow. The process is observed in a discrete moments of time. It is supposed that the volume of binding solution coming into a cell of a chain during one time step interacts only with the particles that can enlarge their size to transit to the cell of the next larger size fraction. The migration of the particles of a size fraction along its chain is controlled by the matrix of transition probabilities, which is different for each size fraction and depends on the total particles concentration. The model allows qualitative estimating of influence of the process parameters on the granulation kinetics. In order to validate the model, the experimental study of ammonium sulphate granulation in the lab scale fluidized bed reactor was carried out. The comparison of theoretical and experimental results was done for the example of particle size enlargement at different flow rate of the binding solution feed. A good correlation between theoretical and experimental data was found for both the mean particle size growth and the fraction size distribution at different moments of time.

Key words: fluidized bed, granulation, Markov chain, state vector, binder solution, particles enlargement, kinetics

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INTRODUCTION

The processes of particle size enlargement are widely used in many industries. Such processes can be realized in different types of equipment. One of them is the wet-agitated granulation. Drum granulators, pan granulators, mixer granulators and fluidized bed granulators are commonly used for its realization [1]. Comparison of advantages and disadvantages of these granulators can be found in the well-known handbooks, such as [1-3].

The present study is devoted to the wet fluidized bed granulation (FBG). Fluidization is an operation in which particulate solids is transformed in a fluid-like suspended state by upward gas flow [4].

Spraying a binder solution onto fluidized particulate solids leads to the growth of particle size [5]. The top spray, bottom spray and tangential spray can be used to cover these particles by a binding solution [6]. The particle size growth can occur due to adhesion of sticky particles (agglomeration mechanism) and due to deposition of films of binder solution onto particle surface and its following dehydration (layering mechanism). Thus, we deal with a fluidized bed with particles of time-varying properties that must be taken into account in modeling of fluidized bed hydrodynamics. FBG is a complex process that involves multiple sub-processes (layering and growth, wetting and drying, breakage and attrition), which occur simultaneously and affect each other. The papers [10-13] are devoted namely to the particle growth kinetics during coating and layering granulation, i.e., to their properties variation. However, this kinetics is not connected with the particulate flow hydrodynamics in a fluidized bed, which is strongly stochastic.

Usually various combinations of Lagrangian and Eulerian approaches [7] are used to model the fluidized bed hydrodynamics. For example, a discrete particle model (Lagrangian-Eulerian) is used in [8] to predict particle motion in a pseudo-2D spout fluidized bed. The Eulerian-Eulerian approach was successfully used in [9] to describe dynamics of spouted beds with conical-cylindrical and conical geometry. However, according to the authors' viewpoint, the approach based on the theory of Markov chains has some undoubted advantages. The comprehensive reviews on its application in powder technology can be found in the works [6,14-16].

Dehling et al. [16] have been first to propose the cell model of particulate flow in fluidized bed. Catak et al. [6] have proposed Markov chain model of particle size enlargement in fluidized bed due to aggregation of solids simultaneously with their breakage process. An essential part of any Markov chain model is the transition matrix, which represents the set of transition probabilities between small but finite cells during a small time duration. In the paper [16], the transition matrix was kept and independent on the current state of fluidized bed. It was a linear model that could not take into account some important specific features of the process, in particular influence of particles growth on hydrodynamics. A non-linear model with state dependent transition matrix was proposed in [17]. Later on, this approach was developed to the case of particles with time-varying properties [18]. Its application to the layering granulation is described in [19]. However, only a binary mixture of particles was examined in this work: the primary particles and completely granulated particles. The gradual growth of them was not taken into account. Besides that, no experimental validation of the model was demonstrated.

The objective of the present study is to generalize the approach described in [19] to the case of gradual growth of particles and to perform experiments to compare the computational and experimental results.

THEORY

Fig. 1 shows the scheme of a granulator the proposed model is based on. The operating volume is separated into n perfectly mixed cell of the height $\Delta x = H/n$ where H is the height of the reactor. In turn, these cells are separated into z vertical chains of cells according to the number z of size fractions which are under observation. The process is observed in the discrete moments of time $t_k = (k-1)\Delta t$ where Δt is the time step, or transition duration, and k is the transition number (a discrete analogue of time).

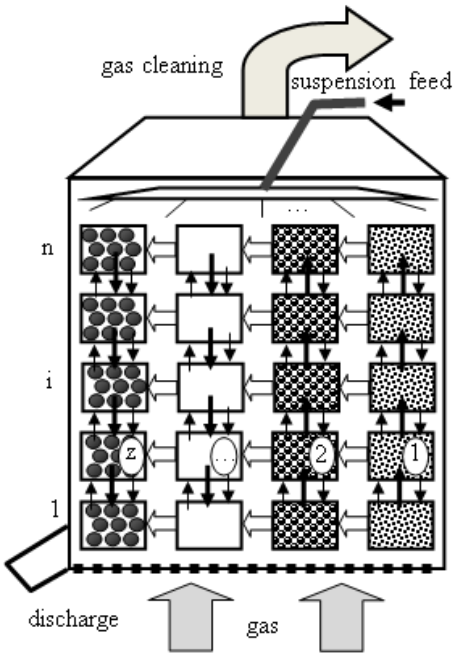

Fig. 1. Schematic presentation of granulator and its cell model
Рис. 1. Схема гранулятора и его ячеечной модели

It is supposed that granulation goes only by the layering mechanism, i.e., the total number of particles is conserved. As the particle size grows due to binding solution deposition it can transit from a cell of one vertical chain to the neighboring cell of another vertical chain.

Let us assume that two virtual stages occur separately during one time step Δt. The first stage is the transition of particles from one vertical chain to another due to their size growth. The second stage is the spatial transitions of the particles along the vertical chains due to their stochastic motion in the fluidized

bed. It is necessary to emphasis that, in fact, the both stages go simultaneously.

The state of the entire process at the moment of time t_k can be presented as the sequence of z state vectors $\mathbf{S_j^k}$ (j = 1, 2, …z) of the size nx1 where S_{ij}^k is the volume content of the j-th fraction in the i-th cell of the vertical chain. The evolution of the state can be described by the set of z recurrent matrix equalities

$$\mathbf{S_j^{k+1}} = \mathbf{P_j^k}(\mathbf{S_j^k} + \Delta\mathbf{S_{j-1,j}^k} - \Delta\mathbf{S_{j,j+1}^k}) \quad (1)$$

where $\mathbf{S_j^k}$ is the current state vector, $\mathbf{S_j^{k+1}}$ is the state vector after the k-th transition, $\Delta\mathbf{S_{j-1,j}^k}$ is the part of particles that comes into the fraction j from a smaller fraction due to granulation during Δt, $\Delta\mathbf{S_{j,j+1}^k}$ is the part of particles that leaves the fraction j to the larger fraction due to granulation during Δt, $\mathbf{P_j^k}$ is the matrix of transition probabilities that describes the migration of particles over the cells of the j-th chain.

The imitation model to describe the flows $\Delta\mathbf{S_{j-1,j}^k}$ and $\Delta\mathbf{S_{j,j+1}^k}$ is presented in Fig. 2. In order to simplify the description, only 3 size fractions are under observation. They are numbered as A, B and C where A is assigned to the finest fraction (original particles). The granulation as such occurs only in the cells a binder solution is fed to. The consequential states of the cell belonging to the chains for different size fractions (the first virtual stage) are shown in the figure.

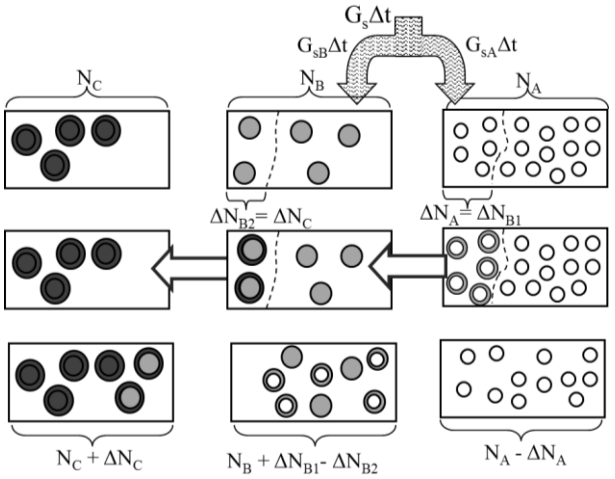

Fig. 2. To the model on granulation mechanism
Рис. 2. К модели механизма гранулирования

Let the binder suspension is fed to a cell with the volume flow rate G_s. It means that the volume $G_s\Delta t$ of it appears in the cells at each time transition. It is supposed that this volume is distributed over the size fractions proportionally to their surface. It is necessary to remind that the total volume content of solid particles is not subjected to the law of conservation because the volume of binding suspension participates in the process. However, the number of particles is conserved. Thus, the fraction A gets the volume of binder solution $G_{sA}\Delta t$. It is enough to enlarge a certain number of A particles up to the size that allows their transition to the neighboring fraction B of the larger size. These particles are separated in the figure by the dashed line. They will transit to the corresponding cell of the neighboring fraction B. Now it is necessary to calculate what volume of particles will appear in B and will be lost in A [19].

In order to enlarge the particle size from d_A (original particle) to d_B (the neighboring fraction size) the following volume of binding solution is required

$$\Delta V_s = \frac{\pi}{6}(d_B^3 - d_A^3) \quad (2)$$

It means that ΔN_A of original particles will be fully covered by the binder solution

$$\Delta N_A = \frac{G_{sA}\Delta t}{\frac{\pi}{6}(d_B^3 - d_A^3)} \quad (3)$$

and will leave the fraction A taking with them the particle volume

$$\Delta S_A = \Delta N_A \frac{\pi}{6} d_A^3 = \frac{G_{sA}\Delta t}{\left(d_B / d_A\right)^3 - 1} \quad (4)$$

It leads to the increase of the fraction B volume content

$$\Delta S_{B1} = \Delta N_A \frac{\pi}{6} d_B^3 = \frac{G_{sA}\Delta t}{1 - \left(d_A / d_B\right)^3} \quad (5)$$

Eqs. (2)-(5) describe the exchange with the volume fractions content between the fractions A and B. The exchange between other fractions can be described by completely analogous formulae.

The next virtual stage is the longitudinal migration of the fractions. The particles can travel along their chains due to their interaction with the upstream gas flow (convection transitions) and surrounding particles (diffusion transitions) and transit from one chain to another due to granulation. The particles travel along the chain is controlled by its matrix of transition probabilities, which is the basic operator of any Markov chain model. It is necessary to note that each matrix $\mathbf{P_j^k}$ depends on the total volume concentration of all fractions in the neighboring cells of the parallel chains, i.e., the model is considerably non-linear.

The detailed description how to build the matrix of transition probabilities can be found in our previous works [15, 17, 18]. It is supposed that transitions only to the neighboring cells of a chain are allowed during Δt. Thus, the transition matrix becomes a tridiagonal one. Each column of it belongs to a cell. The element on the main diagonal is the probability of a particle to stay within the cell, the above element is the probability to transit into the upper cell, and the below element is the probability to transit into the lower cell.

The described above theoretical model allows predicting the evolution of particulate solids during layering granulation in a batch fluidized bed reactor.

MATERIALS AND METHODS

The lab-scale granulator used for experiments is shown schematically in Fig. 3.

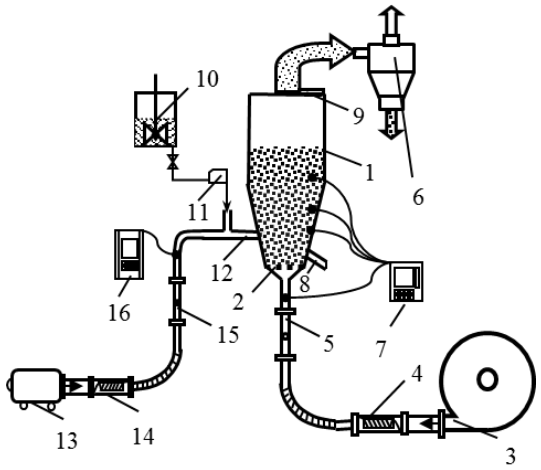

Fig. 3. Scheme of laboratory experimental set-up for granulation of ammonium sulphate particles: 1 – granulator body; 2 – gas distributor; 3 – air blower; 4 – electrical heater; 5 – rotameter; 6 – cyclone; 7 – 8-channel temperature detector; 8 – discharge branch pipe; 9 – inspection window; 10 – pressure tank; 11 – precision feeder; 12 – pneumatic sprayer; 13 – compressor; 14 – electrical heater; 15 – rotameter; 16 – thermometer

Рис. 3. Схема лабораторной экспериментальной установки для гранулирования частиц сульфата ам-мония: 1– корпус гранулятора; 2 – газораспределительная решетка; 3 – воздуходувка; 4 – электро-калорифер; 5 – ротаметр; 6 – циклон; 7 – восьмиканальный термометр; 8 – выгрузочный патрубок; 9 – инспекционное окошко; 10 – напорный бак; 11 – микродозатор; 12 – пневматическая форсунка; 13– компрессор; 14 – электрокалорифер; 15 – ротаметр; 16 – термометр

The fluidized bed reactor circuit consists of the cylinder-conic granulator body 1 with the gas distributor 2, the air blower 3, electric air heater 4, the rotameter 5, and the cyclone 6. The granulator body is equipped with the 8-channel thermometer 7, the discharge branch pipe 8, and the inspection window 9.

The line of binding solution supply contains the pressure tank 10, the precision feeder 11, and the pneumatic sprayer 12. The air for the pneumatic sprayer is supplied by the compressor 13 through the electric heater 14 and the rotameter 15. The air temperature is measured by the thermometer 16.

The feed material was the fraction 1.6…2 mm of ammonium sulphate particles. The mean fraction size was defined as the arithmetical mean of the fraction border sizes, i.e., it was 1.8 mm. Five fractions of particles in the discharged material were under control: <1.6 mm, 1.6…2.0 mm, 2.0…2.5 mm, 2.5…3.0 mm and >3 mm. The fraction size distribution was defined by sieving.

At certain moments of time the samples of about 100g were taken from the granulator to define the fraction mass content and the mean particle size. The total mass of the feed material loaded into the granulator was 2 kg.

The fluidizing air was heated up to 200 °C. Its flow rate was selected to guarantee the stable fluidization during the entire process.

The pneumatic sprayer was placed at 3.5cm above the gas distributor, i.e., it was inside the material. The 40% water solution of ammonium sulphate was used as the binding suspension. The air supplying the pneumatic sprayer was heated up to 90 °C.

Thus, the obtained experimental data allowed determining the fraction size distribution and mean fraction size at different moments of time, i.e., defining the granulation kinetics.

RESULTS AND DISCUSSION

The theoretical and experimental results are compared in this section. Fig.4 shows the growth of the mean particle size with time at three different flow rates of the suspension injection. It is obvious that the rate of the particle size growth is higher at higher this flow rate. Besides that, the reasonable correlation of calculated and experimental data can be seen.

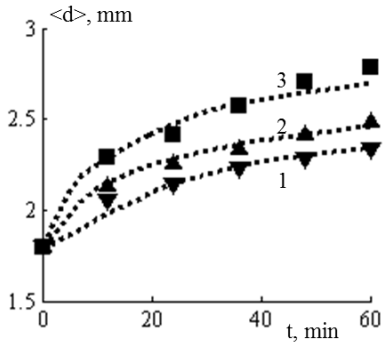

Fig. 4. Evolution of particles average size at different flow rate of binding suspension: 1 – 2,5 kg/h; 2 –3,5 kg/h; 3 –5,5 kg/h (lines – theory, markers – experiment)

Рис. 4. Изменение во времени среднего размера частиц при различном расходе суспензии: 1 –2,5 кг/ч; 2 –3,5 кг/ч; 3 –5,5 кг/ч (линии – теория, маркеры – эксперимент)

However, the fraction size distribution at different moments of time is more informative characteristics of the process than the mean fraction size. These distributions are shown in Fig. 5.

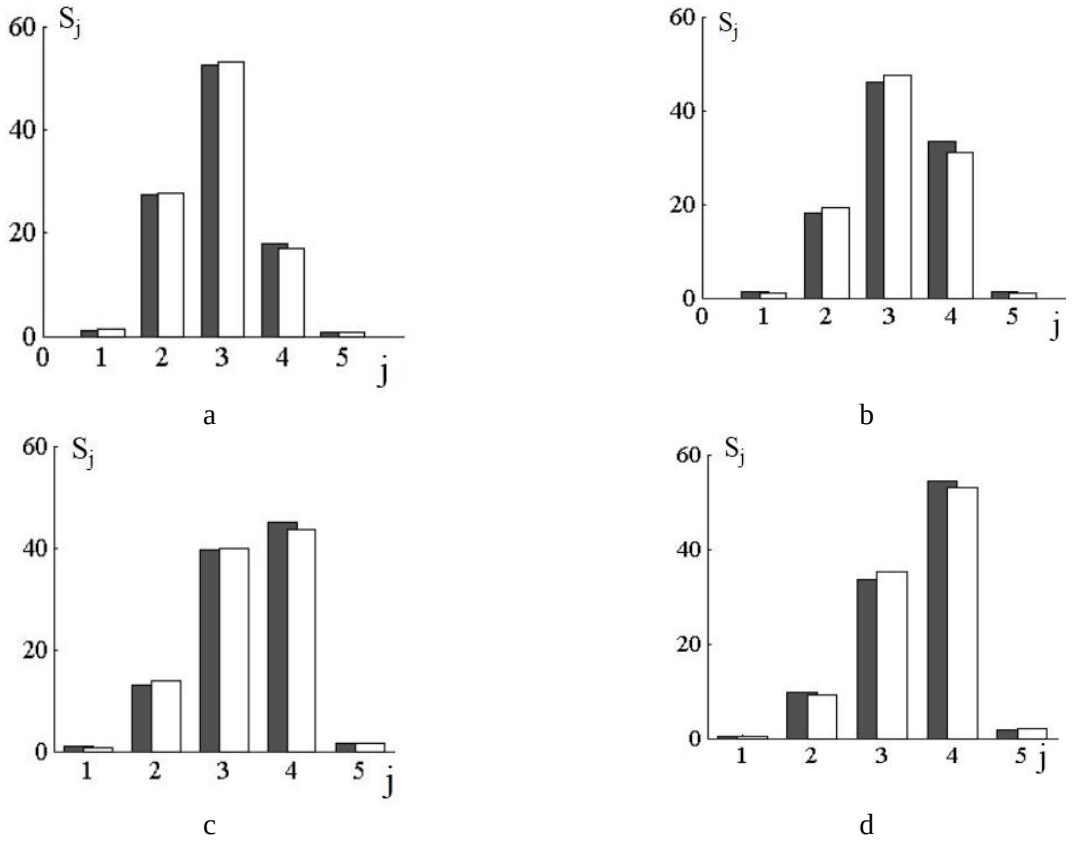

Fig. 5. Fraction size distribution at different time of granulation: a – 12 min; b – 24 min; c – 48 min; d – 60 min (light bars – theory, dark bars – experiment)

Рис. 5. Фракционный состав частиц при различной продолжительности грануляции: a – 12 мин; b – 24 мин; c – 48 мин; d – 60 мин (светлые столбики – теория, темные столбики – эксперимент)

It can be seen from the graphs that the original fraction 1 transits to the larger fraction very fast because practically all binding solution lays on its particles. However, when a considerable part of its particles leaves the fraction it gets less and less of binding solution, and growth of its particles becomes very slow. It practically disappears only after 60 min of granulation. The same can be said about the largest fraction 5 (>3 mm). Until 24 min of granulation it is present as traces, and only after 48 min its more, or less visible growth can be seen. As far as the fraction 4 is supposed to be the objective fraction, it follows from the graphs that the process of granulation is not completed yet during 60 min.

CONCLUSIONS

A cell model based on the theory of Markov chains to describe the kinetics of granulation in a batch fluidized bed reactor is proposed. The model allows qualitative estimation of influence of the process parameters on the granulation kinetics. In order to validate the model, the experimental study of ammonium sulphate granulation in the lab scale fluidized bed reactor was carried out. The comparison of theoretical and experimental results was done for the example of particle size enlargement at different flow rate of the binding solution feed. A good correlation between theoretical and experimental data was found for both the mean particle size growth and the fraction size distribution at different moments of time.

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