DOI: 10.6060/tcct.2017605.5611

УДК: 621.927

ТЕОРЕТИЧЕСКОЕ ИССЛЕДОВАНИЕ КИНЕТИКИ ГРАНУЛИРОВАНИЯ В ПЕРИОДИЧЕСКОМ КИПЯЩЕМ СЛОЕ

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Целью исследования является построение простой, но информативной модели, позволяющей качественно оценить влияние параметров процесса на кинетику гранулирования в периодическом кипящем слое. Для построения модели использован подход, основанный на теории цепей Маркова. Высота реактора кипящего слоя разбита на ячейки идеального перемешивания, и введены две параллельные цепи ячеек: одна для исходных частиц, другая для гранулированных частиц. Частицы могут стохастически двигаться вдоль своих цепей и переходить из одной цепи в другую из-за увеличения их размера вследствие гранулирования. Считается, что собственно грануляция происходит только в той ячейке цепи исходных частиц, куда подается суспензия. Объем суспензии, который оказывается в ячейке за шаг по времени, распределяется только между теми частицами, которые могут быть полностью покрыты суспензией до желаемого конечного размера. Эти частицы переходят в соседнюю ячейку цепи для уже гранулированных частиц. Затем оба сорта частиц перемещаются вдоль своих цепей в соответствие со своими матрицами переходных вероятностей. Эта «холодная» модель может быть легко скомбинирована с марковской моделью сушки в кипящем слое, разработанной в наших предыдущих работах. Численные эксперименты с моделью позволили оценить качественное влияние параметров процесса на кинетику гранулирования. Показано существование оптимальной расходной скорости газа, обеспечивающей максимальную скорость гранулообразования.

Ключевые слова: псевдоожиженный слой, грануляция, цепь Маркова, вектор состояния, связующее вещество, рост частиц, кинетика

UDC: 621.927

THEORETICAL STUDY OF GRANULATION KINETICS IN A BATCH FLUIDIZED BED

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The objective of the study is to build a simple but informative model to estimate qualitatively the influence of process parameters on granulation kinetics in a batch fluidized bed. A Markov chain approach is used to build the model. The height of fluidized bed reactor is separated into a certain number of perfectly mixed cells, and two parallel chains of such cells are introduced: one chain for original particles and another chain for already granulated particles. The particles can move stochastically along their chains and transit from one chain to another due to their size enlargement during granulation. It is supposed that the granulation itself occurs only in the cell of original particles where a binder suspension is supplied to. The volume of suspension, which enters the cell during the time step, is spread over the original particles that can be covered by the suspension up to their desired size. These particles transit to the neighboring cell of another chain for already granulated particles. Then the both sorts of particles move along their chains according to corresponding matrices of transition probabilities. This "could" model can be easily combined with the Markov chain model of drying in fluidized bed developed in our previous works. The numerical experiments with the developed model allowed qualitative estimating the influence of the process parameters on the granulation kinetics. The existence of the optimum superficial gas velocity that provides the maximum rate of granulation is shown.

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

Для цитирования:
Митрофанов А.В., Мизонов В.Е., Tannous K., Овчинников Л.Н. Теоретическое исследование кинетики гранулирования в периодическом кипящем слое. *Изв. вузов. Химия и хим. технология*. 2017. Т. 60. Вып. 5. С. 81–87.

For citation:
Mitrofanov A.V., Mizonov V.E., Tannous K., Ovchinnikov L.N. Theoretical study of granulation kinetics in a batch fluidized bed. *Izv. Vyssh. Uchebn. Zaved. Khim. Khim. Tekhnol.* 2017. V. 60. N 5. P. 81–87.

INTRODUCTION

Enlargement of size of primary particles is an ordinary procedure for particulate solids products engineering. Different types of equipment are employed for the process in many industries. There are four types of wet-agitated granulators: drum granulators, pan granulators, mixer granulators and fluidized bed granulators [1]. The advantages and disadvantages of the above mentioned granulators have been summarized in the well-known handbooks, such as [1-3].

The present study is focused on 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]. At wet fluidized bed granulation particles grow up due to spraying a binder solution onto fluidized particulate solids [5]. There are three various types of FBG according to a spray method employed. Those are top spray, bottom spray and tangential spray [6]. The particle diameter 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, the 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. Under this reason, design of fluidized bed granulators is still based on empirical data, and development of modeling approaches are relevant and timely [1-2].

The known computational models are usually based on various combinations of Lagrangian and Eulerian approaches [7]. In particular, a discrete particle model (Lagrangian-Eulerian) to predict particle motion in a pseudo-2D spout fluidized bed and its experimental validation was presented in [8]. 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 by Berthiaux et al [10], Catak et al [6], Dehling et al [11].

Dehling et al. [11] have been first to propose the cell model of particulate flow in fluidized bed. Catak et al. [6] have proposed Markov chains 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 by Dehling et al [11] the transition matrix was kept constant 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. A non-linear model with state dependent transition matrix was proposed by Mizonov et al. [12]. Later on, this approach was applied by Mitrofanov et al [13-14] to describe the drying process in fluidized bed.

The objective of the present study is to develop a simple but informative model of granulation in a fluidized bed based on the theory of Markov chains. The model is not supposed to be a predictive one. It is intended for qualitative estimation of influence of the process parameters on its technological characteristics and perhaps for separation more narrow areas for more detailed modeling.

THEORY

The present study is based on the theory of Markov chains that was successfully applied to model the drying in a fluidized bed [13-14]. According to the general strategy of the approach the height of fluidized bed reactor is separated into n perfectly mixed cells of the height $\Delta x = H/n$ where H is the height of the reactor. 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. The particulate material involved in the granulation process is presented as two fractions. The fraction A belongs to the fine particles of a monosized feed material. The fraction B is also monosized fraction of the end product. The fraction A transits into the fraction B during granulation. In order to distinguish the fractions in the model, two parallel chains of cells are separated: one for the fraction A and another one for the fraction B. The scheme of such cell model is shown in Fig. 1.

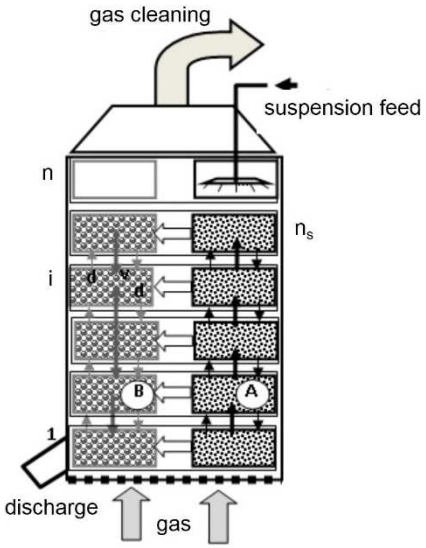

Fig. 1. Schematic presentation of the process cell model
Рис. 1. Схематичное представление ячеечной модели процесса

Thus, 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 the matrices for A and B

depend on the concentration of the both fractions in the couple of neighboring cells of the parallel chains, i.e., the model is considerably non-linear.

In order to describe the granulation itself, the following simplified model is used. The granulation as such occurs only in the cell a binder suspension is fed to. The amount of suspension that appears in the cell during Δt goes to binding only these particles of the fraction A that can reach the final size of granulation and transit to the end product B. All the rest particles of A remain as they were. After that, the both fractions are redistributing over their chains according to corresponding matrices of transition probabilities.

Evolution of fractions content distribution over the cells of the chains can be described by the recurrent matrix equalities

$$\mathbf{S}_\mathbf{A}^{k+1} = \mathbf{P}_\mathbf{A}^k(\mathbf{S}_\mathbf{A}^k - \Delta\mathbf{S}_\mathbf{A}^k) \quad (1)$$

$$\mathbf{S}_\mathbf{B}^{k+1} = \mathbf{P}_\mathbf{B}^k(\mathbf{S}_\mathbf{B}^k + \Delta\mathbf{S}_\mathbf{B}^k), \quad (2)$$

where $\mathbf{S_A}$ and $\mathbf{S_B}$ are the state column vectors of the size nx1 that describe the particle content distribution over the cells, $\mathbf{P_A}$ and $\mathbf{P_B}$ are the matrices of transition probabilities for the fractions, $\Delta\mathbf{S_A}$ and $\Delta\mathbf{S_B}$ are the exchange vectors of the A→B transitions during one time step.

The detailed description how to build the matrix of transition probabilities can be found in our previous works [12-14]. 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.

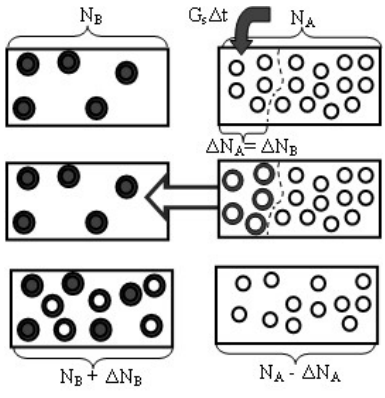

Fig. 2. Evolution of A and B fractions during one time transition
Рис. 2. Эволюция фракций A и B в течение одного временного перехода

Under the assumptions taken above the exchange vectors $\Delta\mathbf{S_A}$ and $\Delta\mathbf{S_B}$ for granulation are to be defined as follows. 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. Fig. 2 illustrates how to transit from the particle number content to their volume content. The consequential states of the cell belonging to the chain for original particles are shown on the right, and the same for the cell for granulated particles – on the left.

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 cell at a time transition. In order to enlarge the particle size from d_A (original particle) to d_B (granulated particle) the following volume of suspension is required

$$\Delta V_s = \frac{4}{3}\pi(d_B^3 - d_A^3)\cdot \quad (3)$$

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

$$\Delta N_A = \frac{G_s\Delta t}{\frac{4}{3}\pi(d_B^3 - d_A^3)} \quad (4)$$

and leave the right cell taking with them the particle volume

$$\Delta S_A = \Delta N_A \frac{4}{3}\pi d_A^3 = \frac{G_s\Delta t}{(d_B/d_A)^3 - 1}\cdot \quad (5)$$

It leads to the increase of the fraction B volume content

$$\Delta S_B = \Delta N_A \frac{4}{3}\pi d_B^3 = \frac{G_s\Delta t}{1-(d_A/d_B)^3}\cdot \quad (6)$$

Thus, the both source vectors $\Delta\mathbf{S_A}$ and $\Delta\mathbf{S_B}$ can be defined.

In order to start the recurrent computational procedure given by Eqs.(1),(2), the initial state vectors have to be given. It is obvious that $\mathbf{S_B^0}$ is the zero vector of the size mx1. The vector $\mathbf{S_A^0}$ has the non-zero elements only for the cells, which are filled with the original particles after their loading into the reactor.

The kinetics of granulation can be characterized by the current ratio of the volume of already granulated particles to the volume of all particles that can be granulated. The following formula allows calculating this characteristic

$$R^k = \frac{\sum_{i=1}^{n} S_{Bi}^k}{\sum_{i=1}^{n} S_{Ai}^0 (d_B/d_A)^3} \quad (7)$$

It is implicitly supposed that the size of granulated particles do not change during accompanying drying process when a solvent is being removed from the binding suspension. This assumption is applicable if the solid concentration in the binder suspension is high enough.

RESULTS AND DISCUSSION

Some results of numerical experiments with the above model are presented in this section. The calculations were done for the following process parameters. The diameter and height of the reactor were taken 0.1 m and 0.3 m respectively. The superficial gas velocity w was varying within the range 1.35-1.8 m/s. The properties of particles were as follows: d_A = 2 mm, d_B = 3 mm, and their densities were assumed equal $\rho_A = \rho_B$ = 900 kg/m^3. The rate of binder injection was 3 kg/h, the mass share of solid substance in the sprayer binder composition was equal to 0.4. The height of the reactor was separated into n = 20 perfectly mixed cells of the height Δx = 1.5 cm. The binder suspension was fed to the upper cell (top spray), the number of which varied due to the bed expansion.

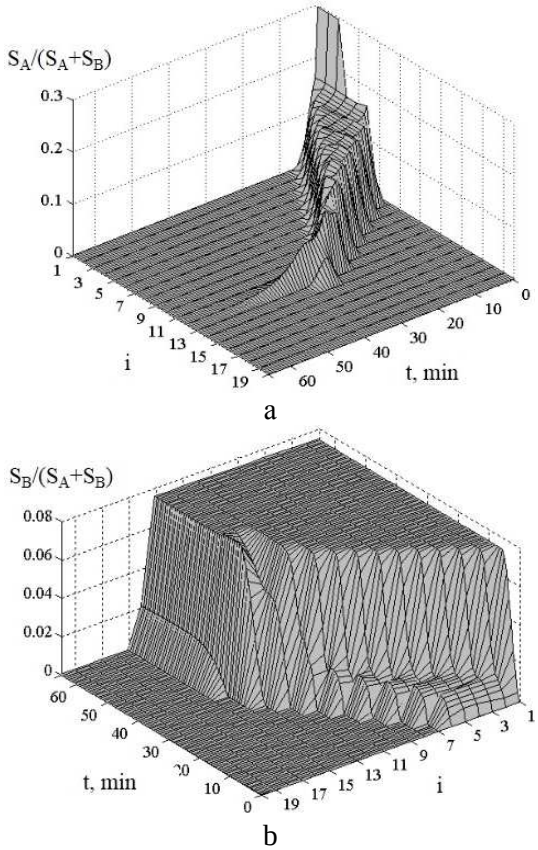

Fig. 3. Evolution of the initial fraction A distribution (a) and the target fraction B distribution (b) over the bed height (w=1.65 m/s, D (dispersion coefficient) = 0 cm^2/s)

Рис. 3. Эволюция распределения исходной фракции A (a) и целевой фракции B (b) по высоте кипящего слоя (w=1,8 м/с, D (дисперсионный коэффициент) = 0 см^2/с)

Fig. 3 shows the evolution of the initial fraction A distribution (a) and the target fraction B distribution (b) over the bed height. The feed fraction A moves up and, after reaching the top cell, interacts with the binder suspension, transforms into the target fraction B, i.e., disappears. The increase of the fraction B leads to the solid volume growth, and the bed expands. The evolution of the both distributions looks plausible that confirms the model workability and its predictability at least on the qualitative level.

Fig. 4 shows the influence of the superficial gas velocity w on the granulation kinetics calculated by Eq (7). The decrease of w first leads to faster granulation but then, after w = 1.4 m/s, its rate strongly decreases. At high w, the concentration of feed particles in the top cell is rather small, and the appearance of the fraction B is small too. However, at low w, the bed transits to almost dense state and migration of the fraction A becomes hindered, particularly at the final stage of the process when concentration of the fraction B becomes high.

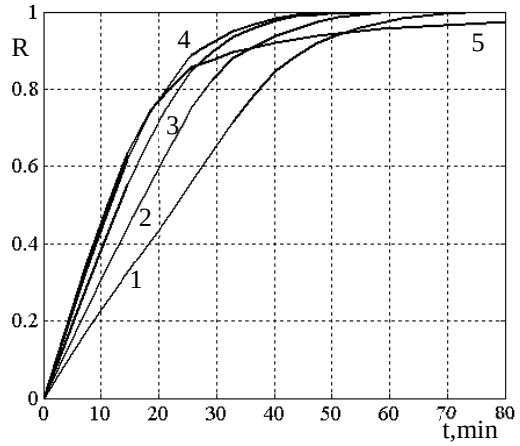

Fig. 4. Influence of the of the superficial gas velocity on the granulation kinetics (D=0.1 cm^2/s) : 1 – 1.8 m/s; 2 – 1.65 m/s; 3 – 1.5 m/s; 4 – 1.4 m/s; 5 – 1.37 m/s

Рис. 4. Влияние расходной скорости газа на кинетику гранулирования (D=0,1 см^2/с): 1 – 1,8 м/с; 2 – 1,65 м/с; 3 – 1,5 м/с; 4 – 1,4 м/с; 5 – 1,37 м/с

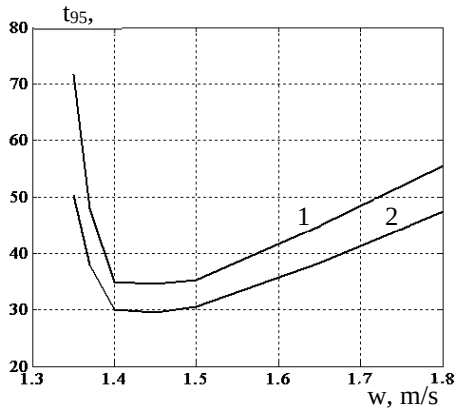

Fig. 5. Influence of the of the superficial gas velocity on the granulation rate: 1 – D=0 cm^2/s; 2 – D=0.1 cm^2/s

Рис. 5. Влияние расходной скорости газа на скорость гранулирования: 1 – D=0 см^2/с; 2 – D=0,1 см^2/с

The influence of the superficial gas velocity on the rate of granulation can be estimated more obviously if quantitative parameter of the rate is introduced. For example, it can be the time t_{95}, at which 95% of the fraction B is already obtained. Fig. 5 shows how this value varies with the superficial gas velocity at two values of the dispersion coefficient. As it was concluded above, the optimum superficial gas velocity exists within the range 1.4 …1.5 m/s that provides the minimum time of granulation, i.e., the maximum process capacity. It can be also seen that the intensity of axial mixing in the bed (the dispersion coefficient D) influences the granulation rate rather strongly. The gain for the two compared lines is about 14% within the range of optimum w.

CONCLUSIONS

A cell model based on the theory of Markov chains to describe the kinetics of granulation in a batch fluidized bed is proposed. The model allows qualitative estimation of the influence of the process parameters on the granulation kinetics. It has low computational time and can be easily combined with other cell models describing processes in fluidized beds, for instance, with a model of granules drying. Using the model, it was shown that the optimum superficial gas velocity exists that provides the maximum rate of granulation. It was also shown that the intensity of particulate solids axial mixing in the bed has the strong influence on the granulation kinetics.

Acknowledgement. *This work is supported by the Russian Foundation for Basic Research (project 15-08-01684).*

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Поступила в редакцию 10.03.2017
Принята к опубликованию 20.04.2017

Received 10.03.2017
Accepted 20.04.2017