CHEMICAL ENGINEERING TRANSACTIONS
VOL. 81, 2020
A publication of AIDIC
The Italian Association of Chemical Engineering
Online at www.cetjournal.it
Guest Editors: Petar S. Varbanov, Qiuwang Wang, Min Zeng, Panos Seferlis, Ting Ma, Jiří J. Klemeš

ISBN 978-88-95608-79-2; **ISSN** 2283-9216
DOI: 10.3303/CET2081190

Discrete-Event Simulation of Aggregation Processes in Batch and Flow Tubular Reactors Based on the Stochastic Lattice Models

Leila Musabekova[a,*], Nurlybek Zhumatayev[b], Nurzhamal Dausheyeva[a], Madina Jamankarayeva[c]

[a]State University of South Kazakhstan, Tauke Khan, 5, Shymkent, Kazakhstan
[b]Silkway International University, Tokaev str., 27 "A", Shymkent, Kazakhstan
[c]South Kazakhstan State Pedagogical University, Baitursynov str., 13, Shymkent, Kazakhstan
mleyla@bk.ru

The novel approach to modeling clusters aggregation which is based on the paradigm of discrete-event simulation (DES) with the help of particles random walk and aggregation on the 3-D stochastic lattices has been applied for describing the aggregation in the heterogeneous poly-dispersive systems. The goal of the work is to generalize the models and results, obtained by the authors earlier for describing aggregation processes, applying to mixed kinetics in batch and flow reactors. The submitted approach allows accounting a different hierarchy of random drift times of particles and aggregation times of clusters of different orders in different areas of the apparatus working volume.
The article presents also the principal part of the developed algorithm for the numerical experiment and the main results of the numerical experiment realizing submitted algorithm for wide range of control parameters. As a result, opportunities may open up both to better understanding the nature of scaling in devices with dispersed phase aggregation and for determining the possible ways to optimize the technological regime and apparatus design.

1. Introduction

There are many areas in modern science of processes in chemical apparatuses, in which the issues of calculating the kinetics and dynamic characteristics of reactors with the formation and aggregation of polydisperse solid or liquid phases in the working volume are extremely relevant (Zhou et al., 2015). Aggregation processes play an important role in nanotechnology (Kacalak et al., 2018), chemical and pharmaceutical engineering, metallurgy and other industries (Chen et al., 2015). The term "aggregation" here applies to the processes of coagulation, coalescence, and fusion (Markus et al., 2015).
At the same time despite the long-standing interest of researchers and many outstanding works, theoretical analysis of many problems remains poorly developed (Shadrack et al., 2018).
The first visible problem is that classical models based on Smoluchowski or Becker-Doering equations accounting only binary collisions of particles are little applying to fast processes in the high-dense disperse systems (Wattis, 2006). The other open problem is a description of the influence of clusters age and current state on their internal and surface structure, what, in turn, affects the aggregation activity and aggregation kinetics (Yassen and Mansoori, 2018).
The noted problems significantly limit the ability of the engineering calculation of many technological processes and reduce the reliability of recommendations on the determination of optimal control parameters (Chowdhury et al., 2015). In several of the previous works (Brener et al., 2017), the authors submitted a novel approach to this problem, based not on the model kinetic equations, but on the paradigm of discrete-event simulation (DES) while particles random walk on the stochastic lattices with formation of aggregates (Zeigler and Sarjoughian, 2012). This approach can be applied for describing the aggregation in the heterogeneous poly-dispersive systems, and the noted limitations can be eliminated.

Paper Received: 12/05/2020; Revised: 24/06/2020; Accepted: 26/06/2020
Please cite this article as: Musabekova L., Zhumatayev N., Dausheyeva N., Jamankarayeva M., 2020, Discrete-Event Simulation of Aggregation Processes in Batch and Flow Tubular Reactors Based on the Stochastic Lattice Models, Chemical Engineering Transactions, 81, 1135-1140 DOI:10.3303/CET2081190

The direct approach that seems to be well modeling this process is in setting the probability of completion of the collision of two or more particles with a combine cluster formation (Kébaili et al., 2009). However, in fact, the calculation of this probability is a separate problem, the formulation and solution of which should be based on peculiarities of the physical mechanism of particle bonding in a single cluster (Tammet, 1995). Since the physical mechanism in different systems can be different (charged or uncharged particles, amorphous or crystalline clusters, and so on). So the control parameters of the model lose their certainty, and the simulation results become difficult to interpret (Morganti et al., 2016).
Another approach proposed in the paper (Brener et al., 2017) is in considering the competition of the characteristic times of diffusion transfer of particles and the times of particles "sticking together" during the formation of a combined cluster. The clustering mechanism does not play a principal role in such a model, since the control parameter is the characteristic time of the process.
The main goal of this work is to generalize the 3D-models and results obtained before for describing aggregation processes in bulk batch and flow reactors, applying to mixed kinetics.

2. Problem setup and results of numerical experiment

2.1 Concept and algorithm

The concept is based on the algorithm which has been submitted firstly in the work (Brener et al., 2017), and it has been adapted then to the case of flow tubular reactors in the work (Musabekova et al., 2019). The idea is that the region, in which the diffusion and aggregation processes take place become covered by a fixed spatial lattice.
The change in the local distribution of the fractional composition in the cells of the lattice caused by random drifts or captured by main stream migrations of particles as well as the aggregation processes is described according with discrete-event simulation paradigm (DES) (Zeigler et al., 2000).
In the case of diffusion-limited aggregation (DLA), the aggregation of clusters occurs immediately in the moment of the collision. In terms of the lattice model this can be interpreted as the hit of clusters in a common cell (Brener et al., 2017). In the case of mixed kinetics (Zhou et al., 2015) the scale unit of the time pace was assumed to be equal to the characteristic coagulation time of clusters falling into one cell. It means the particles that reach one cell do not necessarily have time to form a single cluster, but can continue drift separately. It is assumed that the particles are glued together only if, once they hit the common cell, they again migrate to the common cell at the next time unit (Brener et al., 2017).
In this paper, in contrast to the previous works of the authors, the algorithm for describing the process of clusters aggregation in a flow reactor for both DLA and mixed kinetics in the 3D setting has been developed. It should be noted that physically meaningful interpretation of the results of a numerical experiment, strictly speaking, makes physical sense only for 3D models.
At the same time, the algorithm used here according to its methodological content follows the one developed and described in detail in our previous works (Brener et al., 2017) and (Musabekova et al., 2019) for the 2D case. During the calculation, four 3D arrays are formed. The first array simulates the entire lattice with clusters of different orders, obtained in the process of their drifts and aggregation at each time unit. The second array simulates an analogous lattice with clusters that fall into the reactor with a fresh dispersive stream that flows into the reactor with a given average horizontal velocity. The third array simulates the situation at the end of the calculation time unit and generates the initial situation for the next unit. It is formed as the sum of the two previous arrays. The fourth array indicates the number of collisions of particles in each cell at each time unit. This algorithm used for the numerical experiment applying to 3D case.
Only the new code has been carried out. The flowchart used for the numerical experiment applying to 3D case depicts at Appendix A.

2.2 Results of simulation

In the calculation process, the distribution of clusters in orders in the reactor volume along the flow direction, depending on the flow rate, the number of collisions of various orders (i.e., the number of clusters colliding in one cell) in time and in the reactor volume were studied. All experiments were conducted in parallel for a batch reactor and a flow reactor. The order of any cluster means the number of constituent its particles-nuclei, and the order of any particle-nucleus is assumed to be 1 (Wattis, 2006).
Numerical experiments were carried out on 3D arrays simulated a flow parallelepiped reactor with a square cross section 5 X 5 and a length of 20 for flow rates W = 0 (batch reactor) and W = 2; 4; 6. The chaotic drifts of the clusters were superimposed on the rate of the main stream, and the mobility of each cluster was assumed to be dependent on its order (Zatevakhin et al., 2015). The density of the array means that at the initial moment clusters of the first-order were placed one at a time in each cell of the lattice. It was assumed that

first-order clusters are also introduced into the tubular reactor with the main stream. The aggregation of these clusters begins only in the reactor volume.
Figures 1-4 depict some numerical results for dense 3D arrays, where each calculated point is averaged over ten series of calculations.

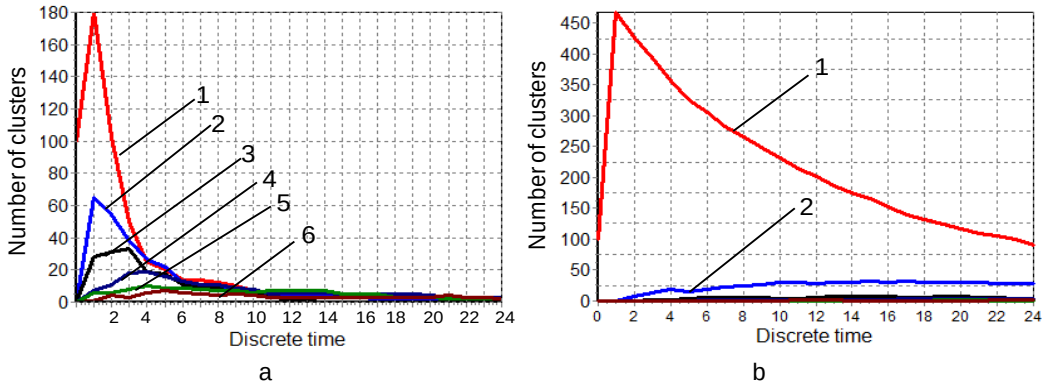


Figure 1: Time dependences of the clusters number of different orders in the batch reactor, clusters orders: 1-first, 2-second, 3-third, 4-fourth, 5-five, 6-six. A- DLA aggregation; B- mixed kinetics

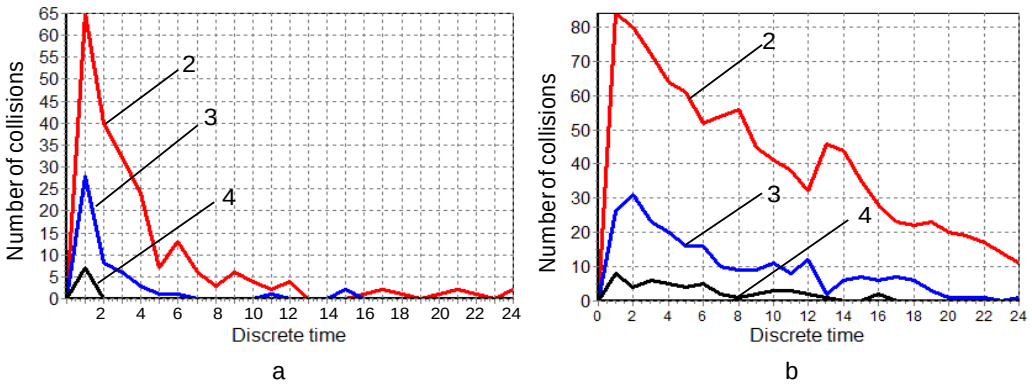


Figure 2: Time dependences of the collisions number of different orders in the batch reactor, collisions orders: 2-second, 3-third, 4-fourth. A- DLA aggregation; B- mixed kinetics

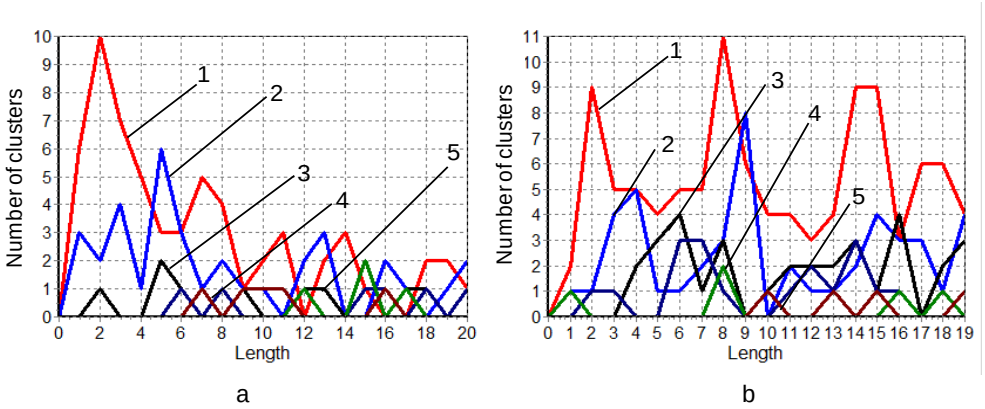


Figure 3: Dependences of the clusters number of different orders in the flow reactor (W=4) on the longitudinal coordinate L(W=2; T=24), clusters orders: 1-first, 2-second, 3-third, 4-fourth, 5-five. A- DLA aggregation; B- mixed kinetics

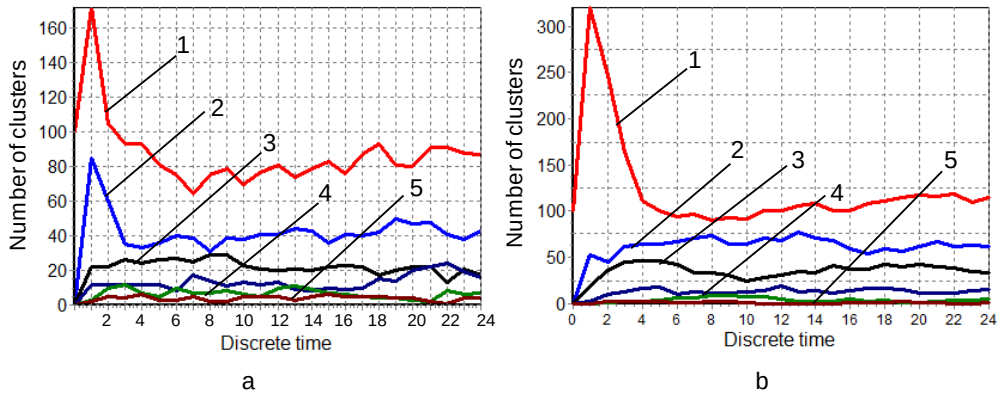


Figure 4: Time dependences of the clusters number of different orders in the flow reactor (W=4), clusters orders: 1-first, 2-second, 3-third, 4-fourth, 5-five. A- DLA aggregation; B- mixed kinetics

Figures 1, 2 depict the results of numerical experiments carried out for the batch reactor. The results obtained in 3D case for a batch reactor do not differ qualitatively from those for 2D case. These data are shown here for comparison with numerical data obtained for flow reactor in Figures 3, 4.

The results of numerical experiments show that in the case of aggregation processes occurring in the region of mixed kinetics, the formation of high-order clusters is significantly inhibited. This braking is somewhat smoothed out in flow reactors (Figures 3, 4). It can be explained by equalization of the total concentration of clusters of various orders along the reactor length.

The sharp fluctuations and peaks in the concentration of clusters of various orders in Figure 3 can be explained by the fact that the selected orders of the main flow velocity W and the amplitudes of the random particles drift are comparable and close. It should be noted that this phenomenon needs further more detail investigations.

However, the general trend is quite noticeable. The experiments showed that the number of multiparticle collisions is noticeably greater during aggregation in a batch reactor at the initial stage of the process than in a flow reactor (Let's compare Figures 2 and 4). In the case of mixed kinetics, the effect of multiparticle collisions on the aggregation intensity is also blurred along the length of the reactor (Figure 3), and does not play such an important role in a flow reactor.

This phenomenon is especially evident with increasing flow rate, because of competition of the three characteristic times: time of the random drift, kinetic time of aggregation and the particles residence time in the working area of the apparatus. It follows that the flow rate affects not only the productivity of the reactor, but also the fractional composition of the dispersion at the outlet. This confirms the conclusion that the average rate of flow through the reactor can serve as a control parameter for stabilizing the desired fractional composition. The influence of flow velocity on the kinetics of aggregation manifests itself in the 3D case much more clearly than in the 2D model (Musabekova et al., 2019).

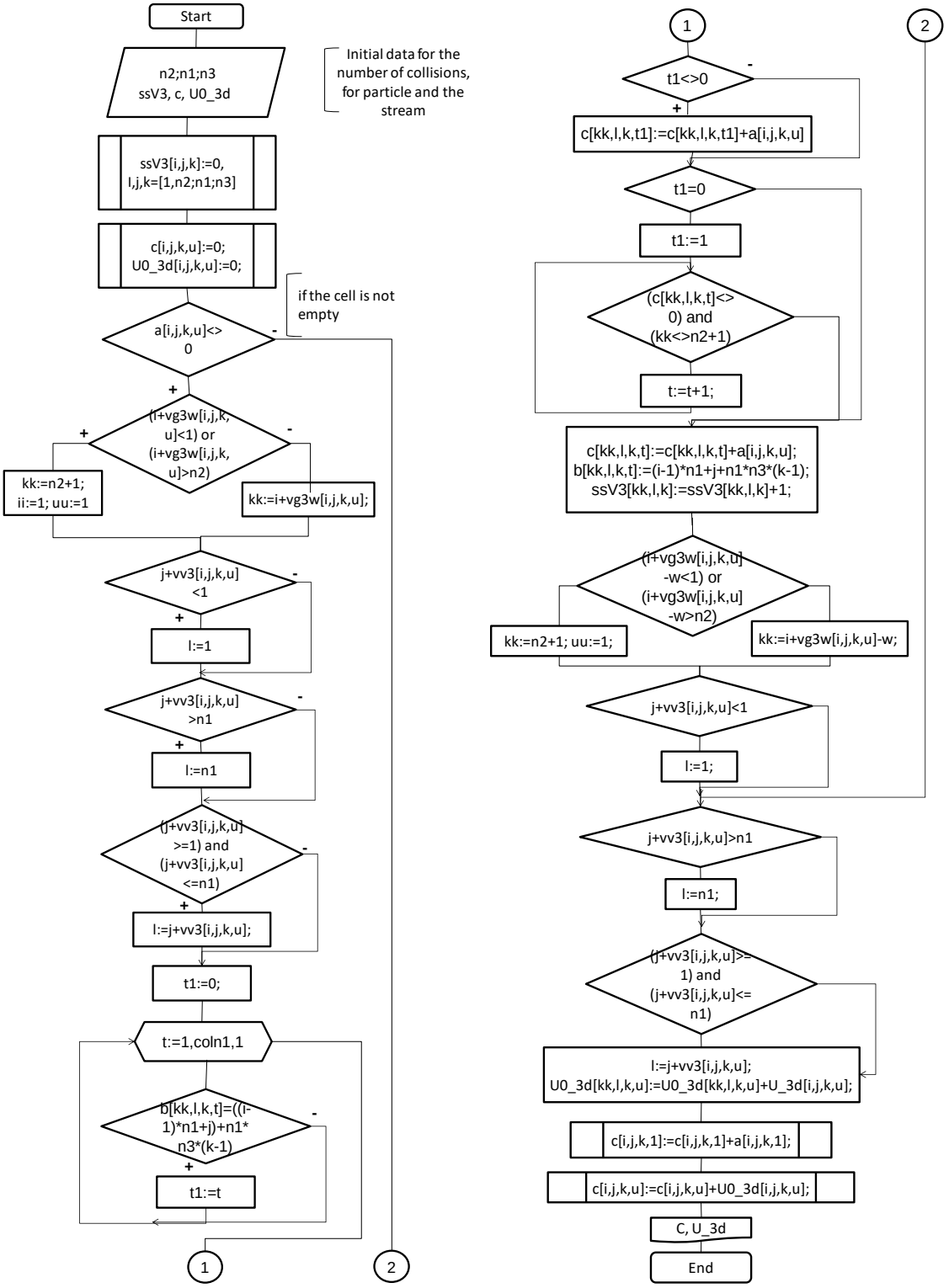
3. Conclusions

The use of the paradigm DES allows partially circumventing the difficulties arising in the numerical study of models based on systems of kinetic differential equations. Of course, this approach does not mean total criticism of the well-known kinetic models, as such, and it does not decrease their significance. The results obtained in this paper can be analyzed only by qualitative characteristics. Direct comparison with experimental data will not be correct in this case, because first it is necessary to establish the values of the control parameters as applied to specific physicochemical systems, which was not part of the objectives of the submitted study. The authors plan to carry out detailed work on checking the frames of the adequacy of the proposed model in subsequent studies. May be such work will also be undertaken by other researchers who are interested in the results presented in this article.

The results of numerical investigations manifest that as the flow rate increases, stabilization of the fractional composition along the length of the reactor occurs at shorter initial section. It was established also that the total number of collisions with a multiplicity higher than two cannot be ignored in the initial transition region. These effects are due to the greater share of multiparticle collisions in the initial sections, since the total concentration of clusters of various orders along the length of the reactor decreases due to intense aggregation.

The total number of multi-particle collisions is comparable to the number of binary collisions at the initial region, what is especially visible in the case of batch reactor. The conducted simulation allows making quantitative estimates of the ratio between the lengths of the initial and stabilized aggregation sites in flow reactors, depending on the scale of the reactor. This gives reason to conclude that the developed method for modeling aggregation processes in flow reactors may be useful in engineering practise for calculating and optimizing chemical technology apparatuses.

Appendix A. Flowchart used for the numerical experiment applying to 3D case



References

Brener A.M., Musabekova L.M., Jamankarayeva M.A., 2017. Stochastic Lattice Model of Aggregation in Heterogeneous Disperse Media. Chemical Engineering Transactions, 60, 70-84.

Chen H., Lin L., Li H., Li J., Lin J.M., 2015. Aggregation-induced structure transition of protein-stabilized zinc/copper nanoclusters for amplified chemiluminescence, ACS Nano, 9(2), 2173-2183.

Chowdhury I., Mansukhani N. D., Guiney L. M., Hersam M. C., Bouchard D., 2015. Aggregation and stability of reduced graphene oxide: complex roles of divalent cations, pH, and natural organic matter. Environmental Science & Technology, 49(18), 10886-10893.

Kacalak W., Lipiński D., Bałasz B., Rypina Ł., Tandecka K., Szafraniec F., 2018. Performance evaluation of the grinding wheel with aggregates of grains in grinding of Ti-6Al-4V titanium alloy. International Journal of Advanced Manufacturing Technology, 301-314.

Kébaili N., Benrezzak S., Cahuzac P., Masson A., Bréchignac C., 2009. Diffusion of silver nanoparticles on carbonaceous materials. Cluster mobility as a probe for surface characterization. The European Physical Journal D, 52(1-3), 115-118.

Markus A.A., Parsons J.R., Roex E.W.M., De Voogt P., Laane R.W.P.M., 2015. Modeling aggregation and sedimentation of nanoparticles in the aquatic environment. Science of the Total Environment, 506, 323-329.

Morganti P., Del Ciotto P., Stoller M., Chianese A., 2016. Antibacterial and Anti-inflammatory Green Nanocomposites. Chemical Engineering Transactions, 47, 61-66.

Musabekova L.M., Zhumataev N.S., Yunussova A,A., Dausheyeva N.N., Zhidebayeva A.N., 2019. Development of the Stochastic Lattice Model for Describing Aggregation Processes in the Polydisperse Systems (3D Case). Chemical Engineering Transactions, 76, 823-828.

Shadrack J.B., Klein R., Site L.D., 2018. Structural locality and early stage of aggregation of micelles in water: an adaptive resolution molecular dynamics study. Advanced Theory and Simulations, 1(6), 1870014

Tammet H., 1995. Size and mobility of nanometer particles, clusters and ions. Journal of Aerosol Science, 26(3), 459-475.

Wattis J.A., 2006. An introduction to mathematical models of coagulation–fragmentation processes: a discrete deterministic mean-field approach. Physica D: Nonlinear Phenomena, 222(1), 1-20.

Yassen S., Mansoori G.A., 2018. Asphaltenes aggregation due to waterflooding (A molecular dynamics simulation study), Journal of Petroleum Science and Engineering, 170, 177–183.

Zeigler B.P., Praehofer H., Kim T.G., 2000. Theory of modeling and simulation: integrating discrete event and continuous complex dynamic systems. Academic Press, San Diego, CA, USA.

Zeigler B.P., Sarjoughian H.S., 2012. Guide to Modeling and Simulation of Systems of Systems (Simulation Foundations, Methods and Applications). Springer-Verlag, London, UK.

Zatevakhin M.A., Ignatyev A.A., Govorkova V.A., 2015. Numerical simulation of Brownian coagulation under turbulent mixing conditions, Izvestiya. Atmospheric and Oceanic Physics, 51(2), 148-155.

Zhou X.H., Huang B.C., Zhou T., Liu Y.C., Shi H.C., 2015. Aggregation behavior of engineered nanoparticles and their impact on activated sludge in wastewater treatment. Chemosphere, 119, 568-576.