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Development of the Stochastic Lattice Model for Describing Aggregation Processes in the Polydisperse Systems (3D Case)

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This paper is devoted to the problem of developing the newly efficient method for modeling the aggregation processes in polydisperse systems, without limitation of considering only binary collisions. The submitted method is an extension of the method previously developed by the authors for the case of a three-dimensional stochastic lattice. Such an extension increases the practical significance and reliability of the simulation results. The deals with the results of a numerical experiment conducted to study the kinetics of the aggregation process occurring in a dispersed system flowing through a 3D tubular reactor based on the discrete-event simulation paradigm (DES). The model is based on the mathematical apparatus of a random walk on mathematical lattices. As shown in this work, this approach allows removing the problem linked to accounting the effect of many-particles collisions, and it allows reducing the problem of calculating the aggregation kernels in kinetic equations to taking into account the hierarchy of characteristic times of the various stages of the aggregation process.

1. Introduction

Aggregation processes play an exceptionally important role in nano-technologies (Chowdhury et al., 2015), chemical and pharmaceutical engineering (Chen et al., 2015), metallurgy (Golubev and Brener, 2002) and other industries (Yaseen and Mansoori, 2018). In general, it can be singled out a number of areas of contemporary science on the processes and apparatuses of chemical technologies, in which the issues of calculating the kinetics and dynamic characteristics of reactors with the formation of a polydisperse solid phase in the working volume are extremely relevant (Gholizadeh and Wang, 2018). The areas of applicability of aggregation models cover the range from fine chemicals production and pharmacy to environmental cleaning (Markus et al., 2015) and industrial waste (Zhou et al., 2015). However, despite of long-standing interest of researchers and exist of many outstanding works (Wattis, 2006), theoretical analysis of many issues remains poorly developed (Zeigler and Sarjoughian, 2012). The first obvious problem is that a known model for accounting multi-particles collisions is, so far, little effective in practical calculations (Conway et al., 2015), and only binary collisions are considered in most models (Gambinossi et al., 2015). However, it was also theoretically shown that multi-particles collisions can make a significant contribution to the kinetics of aggregation (Krapivsky, 1991), especially at a high concentration of the dispersed phase and high intensity of a random fine particle drift (Kacalak et al., 2018). The situation becomes much more complicated when the system contains sources of the dispersed phase in the form of chemical reactions (Waite et al., 2001) or phase transitions (Wang et al., 2012), as well as in the case of simulating the flow reactors, when a multi-phase polydisperse mixture flows continuously into the working volume (Zatevakhin et al., 2015). The other open problem is a description of the influence of clusters age on their internal and surface structure, and this, in turn, affects the aggregation activity and aggregation kinetics (Brener, 2011). This factor can also essentially change regimes of aggregation processes (Andreassen, 2005). The noted problems significantly limit the ability of the engineering calculation of many technological processes and reduce the recommendations reliability on the determination of optimal parameters (Kébaili et al., 2009). In

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previous works, the authors proposed a stochastic lattice model of aggregation in the heterogeneous poly-dispersive systems, for which the noted limitations can be eliminated (Brener et al., 2017). The special algorithm allowing for simulating the aggregation processes in flow reactors and flow systems has also been submitted (Musabekova et al., 2018). The problem of accounting the clusters ages under describing aggregation processes was partially considered too (Brener and Dil'man, 2016). However, in the previous works of the authors and the well-known works of other researchers (Wattis, 2006), the similar approach was used only for describing the random drift of particles and their aggregation on 2D lattices.
In the present paper, the novel approach based on the DES paradigm of processes simulation extends to the case of 3D lattices. The novelty of this approach is that it allows investigating the distribution of different orders clusters throughout the volume of the reactor at different points in time.
The contribution of the results to practices is that the ones open up the possibility of actually applying the developed model for describing the processes of aggregation in the volume of industrial devices, taking into account the peculiarities of the aggregation kinetics in the volume and near the walls of the devices. The cases of batch and flow tubular apparatus have been considered.

2. Algorithm details

In according to DES (Zeigler and Sarjoughian, 2012), the region, in which the diffusion and aggregation processes take place become covered by a fixed spatial lattice (Brener et al., 2017). Moreover, unlike the previous works, in this case 3D array is considered for describing the diffusion-limited aggregation (DLA), and 4D array approaches should be introduced for describing the aggregation in the case of mixed kinetics, when the competition between characteristic times of diffusion and aggregation processes should be described (Brener et al., 2017). Such an algorithm allows taking into account the different time scales of diffusion processes and aggregation processes. In this paper, the DLA case only has been described. The case of mixed kinetics will be submitted in forthcoming papers. Thus, the characteristic aggregation time was completely determined by the characteristic diffusion time of the particles, which was supposed to be equal to the time of drift of particles from one cell of the lattice to another. Further, the term "cluster order" will mean the number of particles-monomers in a given cluster.

2.1 Batch reactor

The algorithm is described below following the previously discussed work (Brener et al., 2017). Namely, at each time beat, a random selection of the drift components of the particle in the horizontal and vertical directions was made. The choice was made from a given set of characteristic displacements. This set was formed taking into account the order of the cluster being moved, namely: the mobility of the cluster was assumed to depend on its order. For clusters of a higher order, the mobility decreased, which corresponds to the known physics of the random drift of particles both in the volume (Tammet, 1995). If a random choice of one time displacements along a given direction for clusters of orders 1 or 2 can be made from the sequence (-3; -2; -1; 0; 1; 2; 3), then for clusters having orders higher than two and less than four, the choice should be made from the sequence (-3; -2; -2; -1; -1; 0; 0; 1; 1; 2; 2; 3). Thus, an increase in the probability of moving higher order clusters to closer cells during a time step is simulated. The behavior of clusters near the walls can also be taken into account in the general case by modifying the sequence of a random selection of displacements. In the numerical experiment applying to the batch reactor, it was assumed for particles trapped in the boundary cells to zero the displacements in the case of random selection, which leads outside the boundaries of the lattice under consideration (Brener et al., 2017).
A cluster of order 1 was initially placed in each cell. Then the process of particles random walk from cell-to-cell was repeated at each time step. Aggregation of particles occurs immediately (DLA case) in the moment of their collision, i.e. without any delay, after they hit the common cell. In contrast to the planar case (Brener et al., 2017), two 3D arrays were formed. The first array modeled the entire lattice with particles of different orders, obtained in the process of displacements and aggregation at each time step. The second array indicated the number of collisions of particles in each cell at each time unit.

2.2 Flow tubular reactor

The algorithm is briefly described below in accordance with the previously discussed work (Musabekova et al., 2018). 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 displacements and aggregation at each time unit. The second array simulates an analogous lattice with clusters that fall into the reactor with a fresh flow 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.

The principal part of the developed code is shown below (Figure 1):

```
procedure Tform1.calcAM;//Main procedure for calculating particles in space
var k,l,ii,al,v,k1,v1,al1:Integer;
begin
 for l:=1 to n2+1 do for i:=1 to n1 do for j:=1 to n3 do
  begin
   ss[l,i,j]:=0; rez[l,i,j]:=0;//initial data on the number of collisions and particles
   if l<=w then U[l,i,j]:=1 else U[l,i,j]:=0;//flow at the initial time
  end;
   for l:=1 to n2+1 do for i:=1 to n1 do for j:=1 to n3 do
   U0[l,i,j]:=0; //initial data on the stream
   for l:=1 to n2+1 do for i:=1 to n1 do for j:=1 to n3 do
   begin
     if (ch[l,i,j]<>0) then //If the number of particles in the cell is not zero, then calculated
        begin
          if (l+vg[l,i,j]<1) or (l+vg[l,i,j]>n2) then //if the value of moving the particle
          //horizontally is outside the length range, then the particle moves to the end of the pipe
          begin k:=n2+1; ii:=1;  end  else k:=l+vg[l,i,j]; //otherwise, the particle
           //motion is calculated: coordinate l + horizontal velocity
          if i+vv[l,i,j]<1 then al:=1; //if the vertically movement of the particle is < 1, the
           //particle will move to cell 1
          if i+vv[l,i,j]>n1 then al:=n1; // if the particle moving vertically is larger than the
           //border, the particle will move to the last cell
          if (i+vv[l,i,j]>=1) and (i+vv[l,i,j]<=n1) then al:=i+vv[l,i,j];
          //for component depth
          if j+vf[l,i,j]<1 then v:=1; // if the movement of the particle in depth is < than the first cell, the
          //particle will move to the first cell
          if j+vf[l,i,j]>n3 then v:=n3; // if the particle moving in depth is greater than the boundary cell,
          //then the particle will move to the last cell
          if (j+vf[l,i,j]>=1) and (j+vf[l,i,j]<=n3) then v:=j+vf[l,i,j];
          rez[k,al,v]:=rez[k,al,v]+ch[l,i,j]; // calculation of the number of particles
          ss[k,al,v]:=ss[k,al,v]+1;//collision number calculation
         end;
    //displacement calculations for flow particles
    if (l+vg[l,i,j]-w<1) or (l+vg[l,i,j]-w>n2) then k1:=n2+1; //if the value of horizontally moving
    //the flow particle is outside the length of  //the pipe, then the particle moves to the end of the pipe to n2the +1 cell
    if (l+vg[l,i,j]-w>=1) and (l+vg[l,i,j]-w<=n2) then k1:=l+vg[l,i,j]-w;
    if i+vv[l,i,j]<1 then al1:=1; //if the vertical movement of the flow particle is less than the  first cell, the
    //particle will move to the first cell
    if i+vv[l,i,j]>n1 then al1:=n1; //if the vertical movement of the flow particle is larger than the boundary
    // cell, the particle will move to the last cell
    if (i+vv[l,i,j]>=1) and (i+vv[l,i,j]<=n1) then al1:=i+vv[l,i,j];//particle flow through the
    //pipe depth
    if j+vf[l,i,j]<1 then v1:=1;
    if j+vf[l,i,j]>n3 then v1:=n3;
    if (j+vf[l,i,j]>=1) and (j+vf[l,i,j]<=n3) then v1:=j+vf[l,i,j];
    U0[k1,al1,v1]:=U0[k1,al1,v1]+U[l,i,j];//calculation of the movement and the number of particles flow
    //in the cells
  end;
 for l:=1 to n2+1 do for i:=1 to n1 do for j:=1 to n3 do
 begin
   rez[l,i,j]:=rez[l,i,j]+U0[l,i,j];//calculation of displacement and medium particles
   ch[l,i,j]:=rez[l,i,j]; U[l,i,j]:=U0[l,i,j];
 end;
end;
```

Figure 1: Main code used for occurring the numerical experiment

3. Results of simulation

Figure 2 depicts the scheme of calculated volume and the example of calculation. Figures 3, 4, 5, 6, 7 and 8 depict some numerical results, where each point is averaged over ten series of calculations for several dimensionless flow rates w=0, 2, 4. As a conventional unit of time in the simulation process, the calculation step is used. The number of clusters of the orders above 5 is essentially less than clusters with the lowest orders in the volume of the reactor. So these clusters are not depicted everywhere on the Figures 3, 4, 5, 6, 7 and 8.

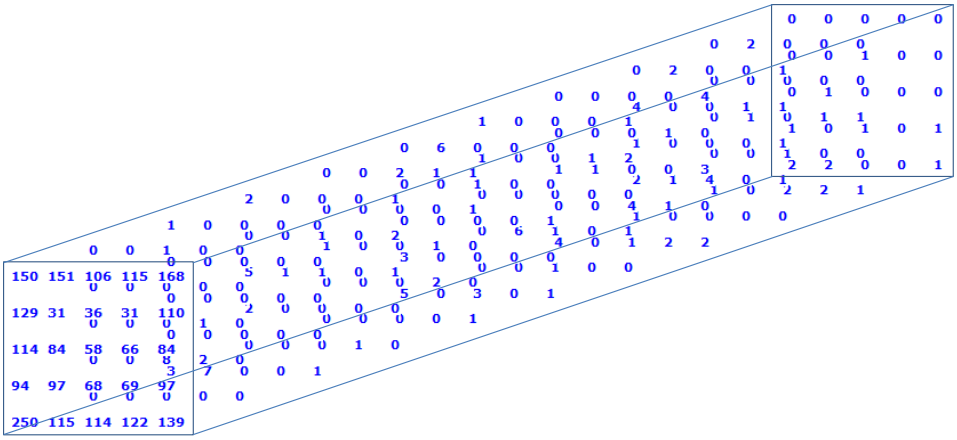

Figure 2: Scheme of calculated volume (5X5X10)

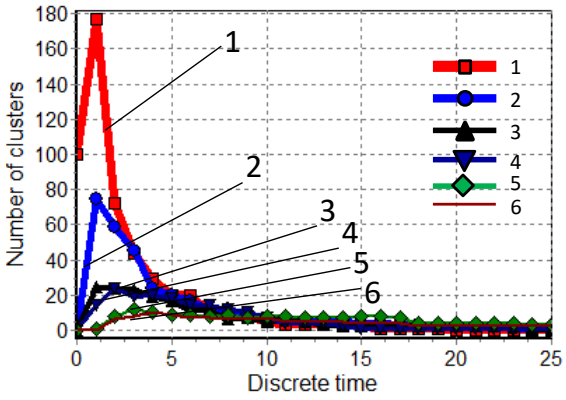

Figure 3: Time history of the numbers of clusters of different orders for w=0 (batch reactor). Experiment of the DLA case. Cluster orders: 1-first, 2-second, 3-third, 4-fourth, 5-five, 6-six

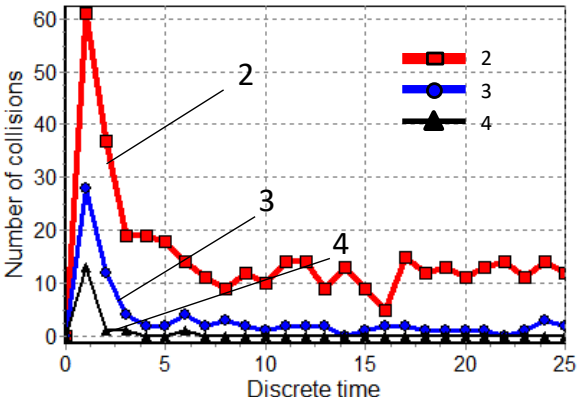

Figure 4: Time history of the numbers of collisions of different orders for w=0 (batch reactor). Experiment of the DLA case. Collision orders: 2-second, 3-third, 4-fourth

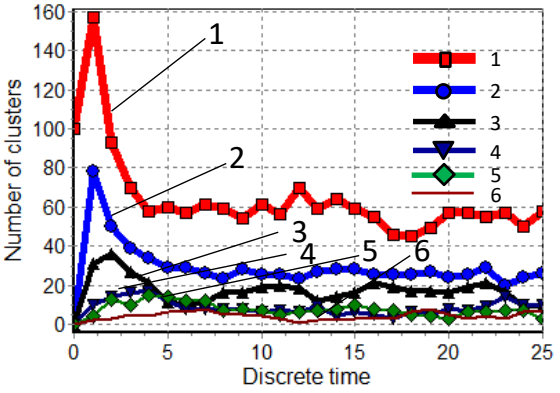

Figure 5: Time history of the numbers of clusters of different orders for w=2 (flow reactor). Experiment of the DLA case. Cluster orders: 1-first, 2-second, 3-third, 4-fourth, 5-five, 6-six

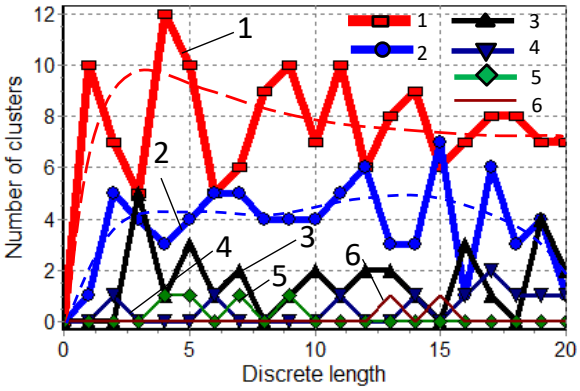

Figure 6: The change of the numbers of clusters of different orders along the longitudinal coordinate in the direction of flow for w=2 (flow reactor). Experiment of the DLA case. Cluster orders: 1-first, 2-second, 3-third, 4-fourth, 5-five, 6-six

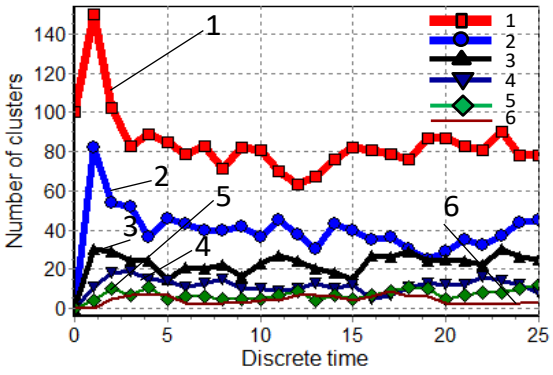

Figure 7: The change of the numbers of clusters of different orders along the longitudinal coordinate in the direction of flow for w=4 (flow reactor). Experiment of the DLA case. Cluster orders: 1-first, 2-second, 3-third, 4-fourth, 5-five, 6-six

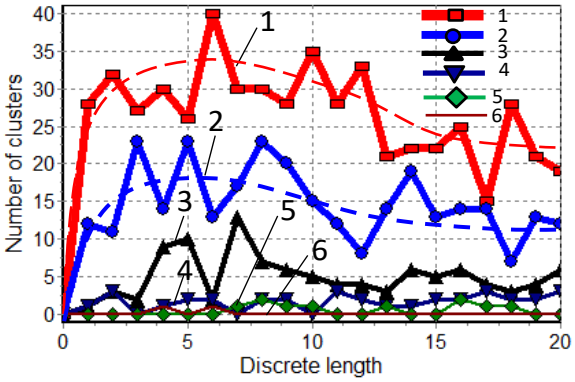

Figure 8: The change of the numbers of clusters of different orders along the longitudinal coordinate in the direction of flow for w=4 (flow reactor). Experiment of the DLA case. Cluster orders: 1-first, 2-second, 3-third, 4-fourth, 5-five, 6-six

In Figures 6, 8 the dashed lines show some ironed by the minimum squares method curves. The results of numerical experiments showed that the change in the number of clusters in a bulk batch reactor occurs at the initial stage of the aggregation process at a much higher rate than this phenomenon was described by the numerical experiment on a planar lattice. The completion of the process with the formation of a single cluster (the so-called gelation) is also observed much faster. Such a result should not be interpreted unequivocally, since the dependence of the clusters mobility on their orders may be different for different real physicochemical systems. In a flow reactor, the cluster size distribution also changes in the initial section near the inlet to the reactor, but the length of the transition section is longer than the experiment on the planar grid shows. As the flow rate increases, stabilization of the fractional composition along the length of the reactor occurs at shorter initial section. This observation is consistent with known data (Shadrack et al., 2018). Just as it was observed in the numerical experiments on a planar lattice, the modeling of aggregation on a three-dimensional array shows that the total number of collisions with a multiplicity greater than two cannot be ignored in the initial transition region. However, the multiplicity of collisions rapidly decreases with distance from the initial part due to a decrease in the volume concentration of clusters.

4. Conclusions

The main result of the work can be considered as the confirmation of the efficiency of the method of modeling aggregation on stochastic lattices on the basis of DES simulation without using the model aggregation kinetic equation. The results of numerical experiments showed that the change in the number of clusters in a bulk batch reactor as well as in the flow reactor occurs at the initial stage of the aggregation process at a much higher rate than this phenomenon was described by the numerical experiment on a planar lattice. In the conducted numerical experiment, this happened approximately 1.4 times faster. In the case of the batch reactor, the formation of a single cluster (the so-called gelation) is also observed much faster. In the flow tubular reactor, the cluster size distribution also changes in the first section near the inlet to the reactor, but the length of the transition section is approximately 1.2 times longer than in the experiment on the planar grid shows.

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. The total number of multiparticle collisions (about 47 collisions) was comparable to the number of binary collisions (60 collisions) at the initial region that is approximately 25 % of the total calculation time for the batch reactor. The main subject of the further investigations should be scaling problems as the results of the conducted simulation do not allow making quantitative estimates of the ratio of the lengths of the initial and stabilized aggregation sites in flow reactors, depending on the scale of the reactor. The submitted method can be developed without the considerable complication of the algorithm for describing the processes of stochastic aggregation on three-dimensional arrays in the case of DLA. The transition to the description of more complex mechanisms of aggregation kinetics will require an increase in the dimension of the problem. In this case, there should be no significant mathematical problems, but the volume and duration of the calculation may noticeably increase.

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