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ANALYSIS OF CHANGES IN FRACTAL, STATISTICAL AND KINETIC PARAMETERS DURING THE HETEROANNIHILATION OF PARTICLES ON THE SIMULATED STRUCTURALLY INHOMOGENEOUS SURFACES

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To investigate the mechanisms of interaction of particles on structurally inhomogeneous surfaces, the influence of surface morphology, distribution of interacting particles and their mobility on the mechanisms of ongoing processes, various modeling approaches are widely used. The paper presents the results of comparison of kinetic, statistical and fractal parameters changing in the process of heteroannihilation of particles distributed chaotically and multifractally on a simulated structurally inhomogeneous surface. To simulate the complex behavior of particles in time and space during heteroannihilation with changes in their mobility across the surface and various interaction probabilities, a class IV probabilistic cellular automaton method was applied. Based on the $_{simulation}$ results, it is shown that the interaction probability, initial distribution, and mobility of interacting particles influence the formation of kinetic modes at different iterations of the heteroannihilation process. It was found that a decrease in the mobility of interacting particles leads to a longer preservation of spatial heterogeneity in the system. An increase in the mobility of interacting particles and the probability of their interaction increases the rate of change in the fractal dimensionality and the rate coefficients of heteroannihilation kinetics. Decrease of fractal dimensionality during heteroannihilation is accompanied by decrease of asymmetry and increase of excess characterizing shape and degree of symmetry of distribution of interacting particles. At the same time, large values of asymmetry and excess indicate a significant deviation from the normal distribution of interacting particles on the surface.

Keywords:*heteroannihilation, fractal kinetics, fractal dimensionality, structurally inhomogeneous surface, multifractal analysis, interaction probability.*

1. Introduction

To investigate the interaction mechanisms of particles distributed on structurally inhomogeneous surfaces, various modeling approaches can be used to take into account the distribution of molecules, the interaction radius, the rates of running processes, the surface morphology, thermodynamic and statistical characteristics of the system. Such approaches include methods of fractal and multifractal analysis, which can be used to describe numerically the evolution of complex systems, the mechanisms of processes occurring in them, to identify the factors affecting the behavior of structurally inhomogeneous systems, to establish empirical relationships between fractal parameters and experimentally determined microstructural characteristics, to describe the inhomogeneous fractal object by means of the spectrum of fractal dimensions, to establish connection between geometrical, statistical and kinetic parameters of the system, to identify the type and number of interacting particles in the considered system [1-3]. In addition, fractal and multifractal analysis are widely used in the study of luminescence kinetics and electron excitation energy transfer processes in various media and on surfaces with different topologies [2,4-8], fermentative processes [9], the dynamics of intracellular reactions [10-12], and others.

2. Theory and research methods

Various experimental studies have shown that the kinetics of processes in structurally inhomogeneous media cannot be explained by the law of acting masses because of the anomalous, time-dependent behavior of the rate constant [7]. The key factor in this special behavior is the reaction probability.

For example, according to the approach proposed by Kopelman to describe the kinetics of all diffusion-controlled reactions (reaction order ≥ 2) in inhomogeneous media, the dependence of the rate coefficient on time is described by the equation [7]:

$$k(t) = k_0 \cdot t^{-h} \quad (1)$$

where k_0 is the rate constant in classical kinetics, t is time $(t > 0)$, h is a fractal parameter or measure of system dimensionality $(0 \leq h \leq 1)$. The fractal parameter h in the Kopelman equation increases with increasing concentration of obstacles and particles that undergo transformations in the interaction. In this case, the individual influence of the concentrations of obstacles and interacting particles is cumulative with time. Schnell and Turner, who consider fractal kinetics, the rate coefficient of which follows the Zipf-Mandelbrot distribution, propose another dependence of the rate coefficient on time:

$$k(t) = \frac{k_0}{(r+t)^h}, 0 \leq h \leq 1 \quad (2)$$

In equation (2), the parameters k_0 and h have the same value as in equation (1), and r is a positive constant that represents the time after which the reaction is affected by inhomogeneities in the distribution of molecules, such as clusters of molecules [13]. In addition, a number of works have found that the diffusion anomaly and fractality of kinetics increase with increasing medium density, decreasing the mobility and size of interacting particles or obstacles [14-17]. In accordance with this, in this paper, we compare and contrast the kinetic, statistical and fractal parameters of the processes occurring on the simulated structurally inhomogeneous surface with chaotically and multifractally distributed interacting particles on it. The method used in this work allows to standardize the description of the loss kinetics of interacting particles on surfaces of different structure and morphology.

In this work we consider a model of a surface with inhomogeneously distributed interacting particles of two varieties (*A* и *B*) on it, which form disordered structures of different topology at the initial moment of time and as a result of heteroannihilation. The interest in this kind of systems is related to the need to understand the most general regularities in the behavior of complex nonlinear dynamical systems that reveal a self-consistent tendency towards critical regimes [4-10,18].

Disordered structures on the surface were modeled by introducing the probability of filling the surface nodes with molecules of each variety. The modeled surface and the particles of the two varieties distributed on it were used to simulate the heteroannihilation process described by the equation of the form $A+B \rightarrow 0$. The particles considered in the simulation are analogs of donors and acceptors of electron excitation energy.In modeling the heteroannihilation process, it was assumed that all particles are mobile. Particle mobility was modeled as diffusion motion in a randomly chosen direction. To change the mobility of the particles, the temperature of the simulated surface was varied in the range 193-281K. When distributing the particles over the surface and modeling their mobility, it was taken into account that two particles cannot occupy the same position on the surface at the same time. To simulate the complex behavior of interacting particles on a simulated surface in time and space at a given probability of interaction in the considered temperature interval and to reproduce the processes of heteroannihilation, the method of probabilistic cellular automata of class IV was used [5, 6]. A cellular automaton is a discrete dynamic system whose behavior is completely determined by a set of local rules. The dynamics of cellular automata of class IV depends on the peculiarities of the initial distribution of interacting particles. Some initial distributions may lead to degeneration of the automaton, others to the emergence of a cyclic sequence of states, others to continuously changing activity patterns of interacting particles. In the process, various spatiotemporal structures may emerge, change shape, and die. In the same environment qualitatively different processes with different direction of evolution may arise. Application of cellular automata allows to describe nonlinear processes observed in the system under consideration in discrete language, considering that the measured quantity itself can take a finite set of values. The using of IV class probabilistic cellular automaton method for modeling heteroannihilation processes between particles of different varieties at any iterative step results in simultaneous changes in the state of the whole system. This allowed in the present paper to take into account the connection between the state of the system at the microlevel and changes at the macrolevel during computer modeling. The lattice model was used to simulate random wandering of particles involved in the process of heteroannihilation, energy migration along the donor subsystem and its annihilation in the donor-

acceptor pair when two particles of different varieties approach to a distance equal to the interaction radius. The structure of the modeled surface was specified by the number of nodes and lattice dimension. The dynamics of heteroannihilation processes on the modeled surface depended on the temperature of the system under consideration, the activation energy, the initial concentration of particles of each variety, and the nature of their initial distribution on the surface.

An iteration time (*N*), was chosen as the time step for modeling the processes of annihilation occurring on the surface, representing the act of moving a particle to a neighboring free node on the surface and interacting with a particle of a different sort located in the neighboring node of the simulated surface (interaction radius) with a given probability. The position of interacting particles at each step of iteration was varied synchronously with the number of free nodes in the nearest neighboring cells. After that the number of remaining particles of each kind was counted.The analysis of the modeled system at different iterations was performed using the multifractal analysis (MFA) method, which allows estimating the fractal parameters of the matrix, such as the generalized Renyi fractal dimensions D_q, the ordering parameter Δ, the heterogeneity parameter *h*, and the entropy of the system*S*. The heteroannihilation process was modeled at different interaction probabilities (from 20% to 100%). An interaction probability equal to 100% corresponds to the instantaneous annicc yjb6hilation of a pair of particles of different species located at a distance of the interaction radius, while a decrease in the interaction probability allows the modeling of longer-lived pairs. The interaction probability between particles at a distance greater than the interaction radius was assumed to be 0%. All modeling results presented in this work included 3000 iterations on a 500x500 knot lattice. The degree of surface coverage by particles of the two varieties was 40%. The ratio of particles of different types was varied in the ratios 1:1, 1:2, and 1:5. No additional kinetic modes were observed at iterations greater than 3000 for the selected lattice size and degree of surface coverage by the particles under consideration. The program used for simulation was written in C++.

3. Results and discussion

A system representing inhomogeneously distributed particles on a structurally inhomogeneous surface can be considered as a multifractal set consisting of subsets with different fractal dimensionality characterizing the degree of spatial filling of the multifractal set. A change in the filling probability of fractal subsets leads to a change in the kinetic dependences and characteristics of the processes occurring in such systems. Taking this into account, when considering the kinetics of heteroannihilation on a structurally inhomogeneous surface, several fractal subsets with different distribution patterns can be distinguished.The total number of such subsets will be considered equal to*K*, and the fraction of particles of sort *A*, distributed over such subsets will be considered equal toA_1, A_2, A_3,..,A_K. For different fractal subsets the following distributions of particles *A* and B relative to each other are possible: 1) particles of kind *A* have no particles of kind *B* in the nearest coordination spheres.This case will correspond to the longest decay time (τ_1), 2) the number of particles of kind *A* ≤ the number of particles of kind *B* that are at a distance of the interaction radius (the first coordination sphere). This case corresponds to the shortest decay time (τ_K). All other types of distributions in which the interaction zone of particle *A* contains several particles of kind *B* will be intermediate, the decay time for which (τ_i) corresponds to the condition$\tau_1 < \tau_i < \tau_K$.This approach allows us to approximate the kinetic dependence by the sum of exponents in the form:

$$I(t) = \sum_{i=1}^{K} A_i \, e^{-t/\tau_i}. \quad (3)$$

In this case, if the distance between *A* and *B* is greater than the interaction radius, the exponent in formula (3) must contain a value that depends on the medium dimensionality, the concentration of the interacting molecules, and the interaction radius. This approach can also be used to study the processes of remote radiation-free energy transfer, contact quenching of luminescence, reversible capture of charge carriers by traps, and others [2].

Taking into account the presence of different kinetic modes in the fractal subsets of the system being modeled, the fractal, statistical and kinetic parameters at different iterations that characterize the change in the distribution of interacting particles during heteroannihilation were calculated. The fractal dimension d_f, was chosen as a fractal parameter, asymmetry and excess were used as statistical parameters, and the velocity coefficient *k*was used as a kinetic parameter. Fractal dimension, or Hausdorff dimension, is a

quantitative characteristic of a set of points in n-dimensional space, showing how densely points fill a subspace when their number becomes very large. Excess and asymmetry have been used to describe the shape and degree of symmetry of the distribution of interacting particles. Calculations of fractal dimensionality, excess and asymmetry were performed using the Gwyddion software [19].

In determining the fractal dimensionality we considered the set of points *F*embedded in the Euclidean space $E_n$of dimension $n \geq 1$. $n=1,2,3,...$,an arbitrary arbitrarily small number $\varepsilon > 0$, defining an n-dimensionalε-cube in the space E_n. The minimum number ofn-dimensional ε-cubes covering the set of points *F* with the accuracy ε denote $N_n(\varepsilon)$.Then, the expression for the fractal dimension of the set *F* will be [18]:

$$d_f = d_f[F] = -\lim_{\varepsilon \to 0} \frac{\ln(N_n(\varepsilon))}{\ln(\varepsilon)}. \quad (4)$$

From formula (4) we see that for Euclidean space E_n the fractal dimension $d_f = n$. In the general case $d_f \neq n$. Negative values ofd_f characterize the degree of emptiness of the set. The limit in formula (4) exists if the set *F* has the property of self-similarity or scale invariance.If the number of points in an n-dimensional ε-cube with number *i*is denoted by $n_i(\varepsilon)$, then we can determine the probability that a randomly taken point from the set is in an n-dimensional ε-cube with number*i* by the formula:

$$p_i(\varepsilon) = \lim_{N_n(\varepsilon) \to \infty} \frac{n_i(\varepsilon)}{N_n(\varepsilon)} \quad (5)$$

Accordingly, $p_i(\varepsilon)$in formula (5) characterizes the relative population of n-dimensional ε-cubes in the spaceE_n. If the distribution of points in the cells is unequal, then the fractal is inhomogeneous, i.e. it is a multifractal. In general case, multifractal is characterized by some nonlinear function [20]:

$$\tau(q) = \lim_{\varepsilon \to 0} \frac{\ln Z(q,\varepsilon)}{\ln \varepsilon}, \text{where } -\infty < q < +\infty,, \quad (6)$$

determining the behavior of the statistical sum $Z(q, \varepsilon)$for $\varepsilon \to 0$:

$$Z(q,\varepsilon) = \sum_{i=1}^{N(z)} p_i^q(\varepsilon) \approx \varepsilon^{\tau(q)} \quad (7)$$

Taking this into account, surfaces with multifractal and chaotic initial distributions of interacting particles were modeled and the process of heteroannihilation was simulated using the probabilistic cellular automaton of class IV. When modeling a surface with an initial chaotic distribution of interacting particles, the Randomize random number generator was used to select the coordinates of particles on a planar lattice. The change in the entropy of the system was chosen as a criterion for multifractal formation, the decrease of which indicated the formation of surface structures differing from each other by statistical parameters such as homogeneity and ordering. The change in the statistical characteristics of the particle distribution over the modeled surface as a result of heteroannihilation can be estimated using the function $f(\alpha)$, which describes the Hausdorff dimension spectrum of homogeneous subsets of the original set, which is a multifractal: $f(\alpha) = q\alpha - \tau(q)$, where $\alpha = \frac{d\tau(q)}{dq}$. The homogeneous subsets that give the largest contribution to the statistical sum (7) for a given q are taken into account. The spectrum of generalized fractal dimensions of Renyi D_q, $-\infty < q < +\infty$, whose calculation methodology is described in detail in [5, 21, 22, 23], can be used to analyze changes in the fractal parameters of the particle distribution during heteroannihilation of particles on the modeled structurally inhomogeneous surface. The values of the parameter q determine the contribution to the statistical sum (7) of cells with different particle densities: from cells with the lowest particle density and low probability of their filling ($q \to -\infty$) to cells with the highest particle density and high probability of detection of such cells ($q \to \infty$). The function $D_q(q)$ is monotonically increasing with increasing q. Thus, for a homogeneous fractal all generalized fractal Renyi dimensions will coincide. The parameters D_0, D_1 were the most informative for the purposes of this paper. D_0 is calculated at q=0 and represents a constant value, which can be considered as a rough parameter for a multifractal. The calculated value of the fractal dimension D_1 at q=1 characterizes the rate of increase in the amount of information with

decreasing scale of the investigated structure. The numerical values of the parameters D_0, D_1 did not exceed 2.36.

The examples of simulated surfaces with initial chaotic and multifractal distributions of interacting particles at temperature T=273K and interaction probabilities of 100% and 20% in different iterations (N=100 and N=2000) are presented in figure 1.

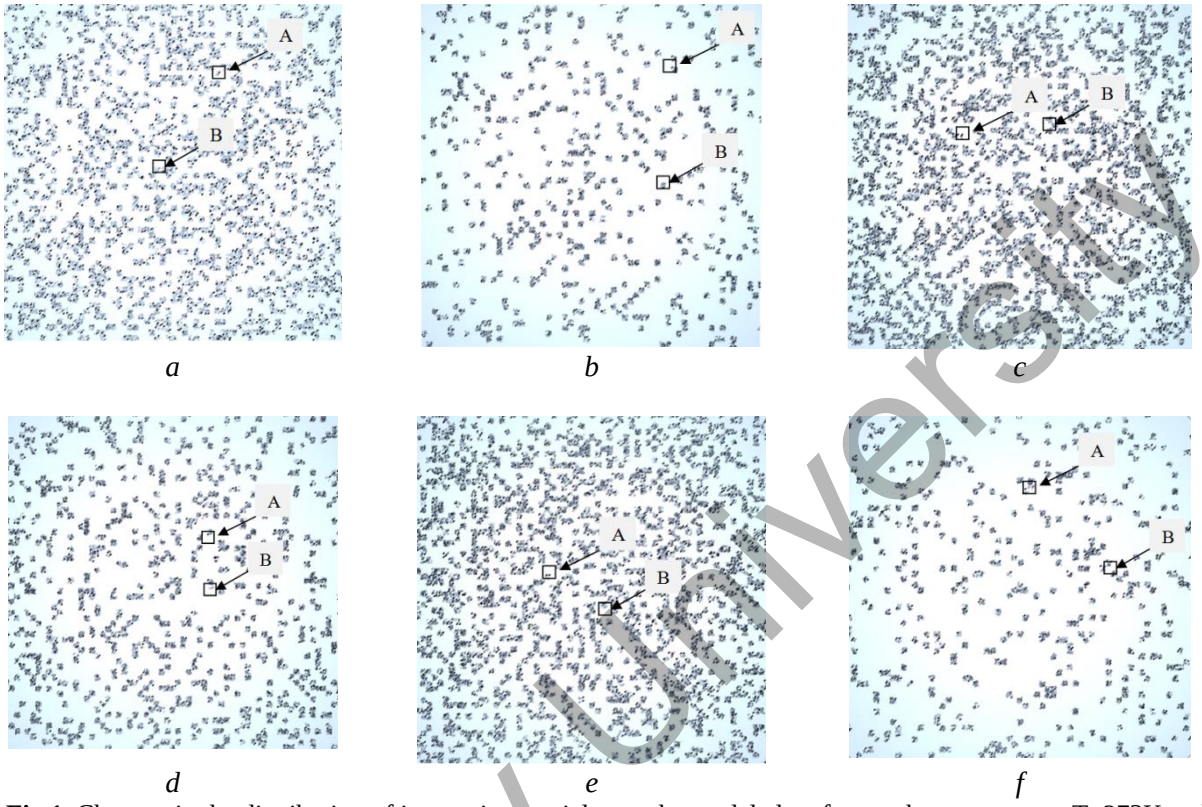


Fig.1. Changes in the distribution of interacting particles on the modeled surface at the temperature T=273K and interaction probabilities of 100% (a, b, e, f) and 20% (c, d): a), c), e) - number of iterations N=100, b), d), f) - number of iterations N=2000. Initial distribution of interacting particles is chaotic (a, b, c, d) and multifractal (e, f)

The normalized kinetic curves for the simulated heteroannihilation process at temperatures T=193K, 237K and 273K and interaction probabilities equal to 20% and 100% are shown in Figure 2 for the cases with initial chaotic and initial multifractal distributions of interacting particles.

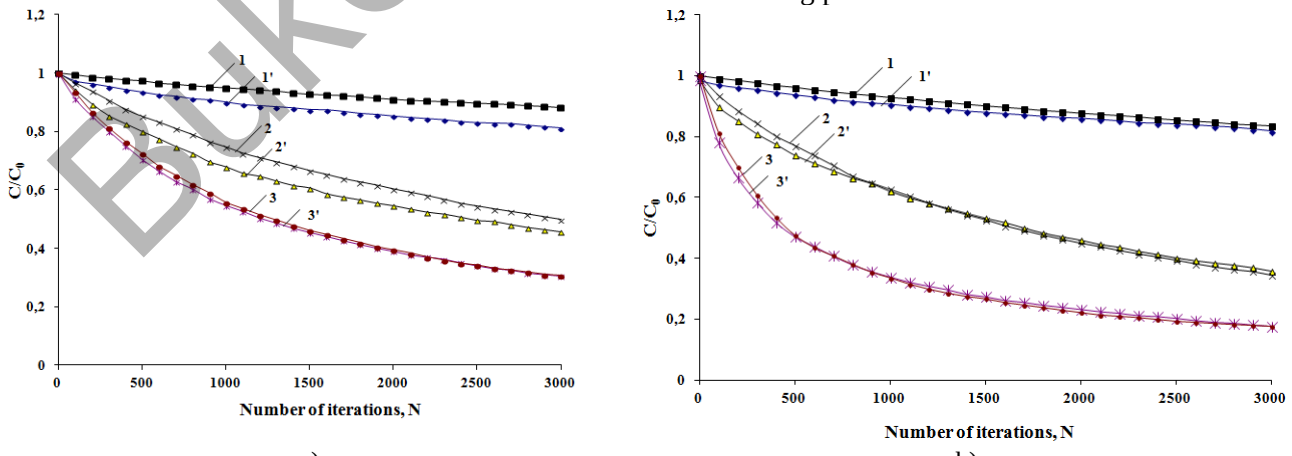


Fig.2. Normalized kinetic curves for the simulated heteroannihilation process with initial chaotic (curves 1',2',3') and initial multifractal (curves 1,2,3) distributions of interacting particles at temperatures 193K (curves 1 and 1'), 237K (curves 2 and 2'), 273K (curves 3 and 3') and interaction probabilities 20% (a) and 100% (b)

In Figure 2 the initial number of particles of kind A on the modeled surface is C_0, C=C(N) s the number of particles of kind *A* for each *N*-th iteration of the simulated heteroannihilation process As can be seen from Figures 1 and 2, the interaction probability, the initial distribution of interacting particles, and the temperature-dependent mobility of interacting particles affect the formation of kinetic modes at different iterations of the heteroannihilation process.

Figure 2 shows that the influence of the initial distribution of interacting particles is most pronounced at lower temperatures (193K and 237K) and lower values of interaction probability (20%). This may be due to the anomalous nature of diffusion of interacting particles over the simulated surface and dependence of the "diffusion coefficient" *K(r)* of a particle on the distance r between its current and starting positions: $K(r) \approx r^{-\theta}$, where θ is the index of anomalous straying on the modeled surface [24].This is confirmed by the observed dependence of fractal dimensions on the number of iterations (Figure 3). The decrease in $d_f$in Figure 3 is associated with an effective decrease in the number of space points available to the diffunding particle.

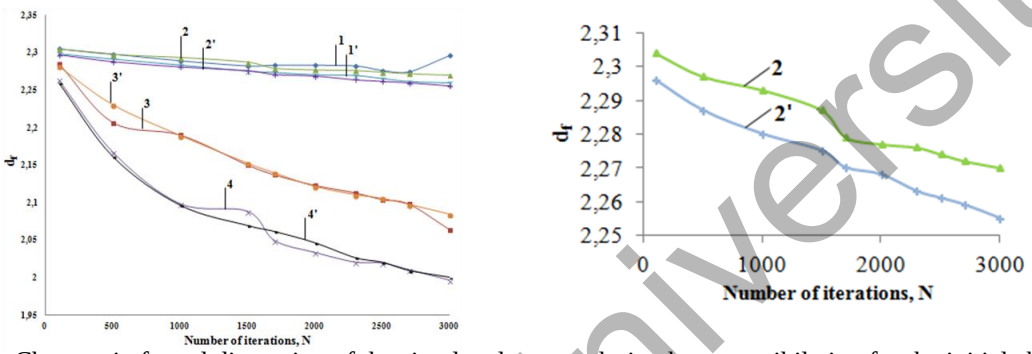
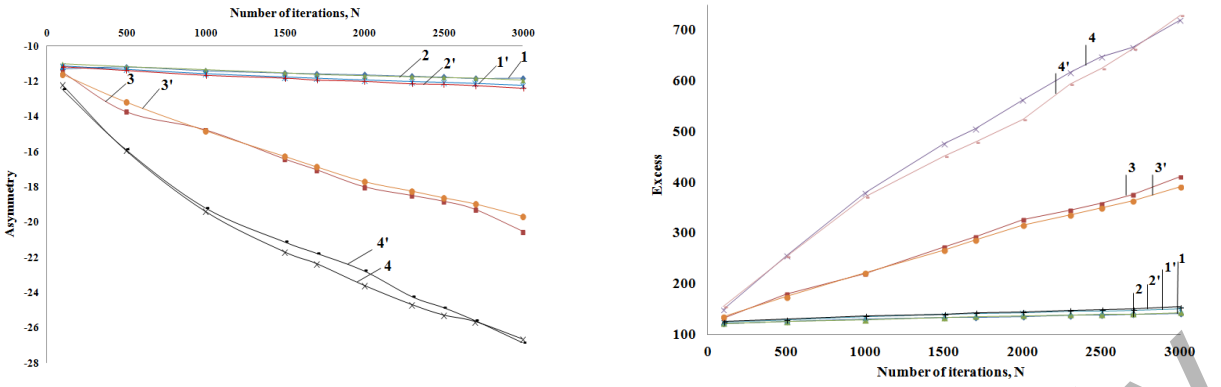


Fig.3. Changes in fractal dimension of the simulated system during heteroannihilation for the initial chaotic (curves 1',2',3,4') and multifractal (curves 1,2,3,4') distributions at 20% (curves 1,1',3,3') and 100% (curves 2,2',4,4') interaction probabilities and 193K (curves 1,1',2,2') and 273K (curves 3,3',4,4') temperatures. Inset: change in fractal dimensionality during heteroannihilation for the initial chaotic (curve 2') and multifractal (curve 2) distributions at 100% interaction probability and 193K temperature

The values of fractal dimensions at 193K remain higher at 20% and 100% probabilities compared with the same data for 273K. This indicates that in the distribution of interacting particles at low temperatures the spatial heterogeneity persists longer. At the same time, as seen in the inset to Figure 3, the values of fractal dimensions at low temperatures for the initial chaotic distribution remain lower than for the initial multifractal distribution over the whole considered time interval. Thus, an increase in the mobility of interacting particles, interaction probability, and changes in the free volume available to interacting particles leads to a sharper change in the fractal dimensionality, and, consequently, to a change in the fractality of heteroannihilation kinetics. This conclusion is consistent with the results of other works, e.g. [9, 25].

Similar trends are observed in the changes in asymmetry and excess during heteroannihilation (Figure 4). Large values of asymmetry and excess indicate a significant deviation from the normal distribution of interacting particles over the surface. A positive excess means that the outliers in the data are more intense than for the normal distribution. A distribution with significant negative asymmetry has a long tail to the left of the mathematical expectation. At the same time, comparison of the dependencies presented in Figures 3 and 4 shows that decrease of fractal dimension as a result of heteroannihilation leads to decrease of asymmetry and increase of excess. Analysis of the kinetic dependences of the modeled heteroannihilation process showed the existence of two different velocity regimes at different time intervals: the short-term interval ($N \leq 1500$) and the long-term interval ($N \geq 1500$). The calculations showed that the values of the velocity coefficients at both the short- and long-time intervals depend on the initial distribution of interacting particles, their mobility, and the interaction probability.As the mobility of particles increases, the velocity coefficient for the short-range interval increases for both multifractal and chaotic initial distributions. The only exception is the case with the multifractal initial distribution at the interaction probability of 20%, for which the decrease of the rate coefficient with the increase of mobility of interacting particles was observed. This can be due to anomalous diffusion of interacting particles, caused by formation of local stable structures on the surface, limiting rapprochement of particles.



a) b)

Fig.4. Changes in asymmetry (a) and excess (b) of the simulated system during heteroannihilation for the initial chaotic (curves1',2',3',4') and multifractal (curves 1, 2,3,4') distributions at interaction probabilities of 20% (curves 1,1',3,3') and 100% (curves 2,2',4,4') and temperatures of 193K (curves 1,1',2,2') and 273K (curves 3,3',4,4')

For the long-time interval for both the chaotic and multifractal distributions, regardless of the interaction probability, the velocity coefficient decreased as the particle mobility increased. The ratio of velocity coefficients for the short- and long-time intervals increased as the mobility of particles increased, irrespective of the interaction probability. The observed trends in simulations of interaction processes of particles on a structurally heterogeneous surface indicate the fractal nature of the kinetics with different degrees of fractality depending on the particle mobility, interaction probability and initial distribution of interacting molecules, which is in agreement with the results of other works [9, 26, 27].

4. Conclusion

In this paper the process of heteroannihilation in a system representing a structurally inhomogeneous surface with chaotically and multifractally distributed on it particles of two kinds at the initial moment of time is modeled by the method of probabilistic cellular automata of class IV. Comparison of kinetic, statistical and fractal parameters calculated for the considered system has shown their dependence on interaction probability, mobility of interacting particles and character of initial distribution of interacting particles throughout the considered iteration interval. The analysis of fractal dimensions at different iterations of the heteroannihilation process allowed to establish that the distribution of interacting particles at low temperatures retains spatial inhomogeneity longer, which is associated with a change in the fractality of the kinetics of this process.

The effect of the initial distribution of interacting particles is most pronounced at lower temperatures (193K and 237K) and lower values of the interaction probability (20%). This may be due to the anomalous diffusion character of the interacting particles, caused by the formation of local stable structures on the surface, limiting the convergence of the particles. In this case, the values of fractal dimensions at low temperatures for the initial chaotic distribution remain lower than for the initial multifractal distribution throughout the considered iteration interval. An increase in the mobility of interacting particles, interaction probability, and changes in the free volume available for interacting particles leads to a sharper change in the fractal dimensionality and, consequently, to a change in the fractality of the heteroannihilation kinetics. Analysis of the kinetics of heteroannihilation showed the existence of two different rate regimes at different iteration intervals. Thus, the method used in this work allows us to standardize the description of the loss kinetics of interacting particles on surfaces of different structure and morphology.

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