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Modeling and multifractal analysis of radiation defect evolution in solids

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The results of modeling of radiation defects formation and evolution on the surface and in the volume of a crystal are presented in this paper. Statistical properties are calculated for the investigated system. It is revealed that defect structure is a multifractal and system entropy decreases, while observing self-organization of the physical system.

1 Introduction With the development of nuclear engineering and space technology the creation of radiation-resistant materials becomes a topical issue [1-4]. The investigation of processes where high-energy particles interact with some matter comes to a radiation-induced defects analysis in them. Explanation of causes and effects of formation and evolution of radiation-induced defects in solids would allow to estimate their resistance to radiation. Non-radiative decay of electronic excitations formed under the influence of ionizing radiation in crystals can lead to Frenkel pair defects [5-7]. Vast majority of experimental methods used to detect the defect formation in different types of solids enables to register structurally formed centers, but doesn't make possible to trace back step by step the electronic excitation energy transformation into primary radiation defects and their following spatial separation [8-12].

In this connection computer modeling is one of the most convenient study methods of the following physical phenomenon by using which it becomes possible to trace back the whole process of formation and evolution of studied aggregative centers [13-16].

2 Computer modeling and multifractal analysis
Real crystals in equilibrium when $T>0$ always contain a small number of defects corresponding to the minimum potential energy. Additional defects can be formed by the action of heat, deformation and radiation. These actions can be carried out specifically at certain stages of the cycle to create instruments and devices in conditions of radiation. Consequently it's of great importance to search for ways to control the properties of defects in crystals [17].

It is known that on exposure to radiation pairs of Frenkel defects form in solids, which are spatially well correlated. For most ionic crystals the main Frenkel defects are *F*- and *H*-centers [10].

At the initial stage of irradiation process their formation is random in nature and described by Poisson distribution. On long-term irradiation random distribution of combined Frenkel pairs begins to be replaced by aggregates of one-type defects [9].

The process of transition from a less ordered to a more ordered state becomes possible due to the openness of the system. In open systems under the influence of matter or energy exchange spatially correlated structures arise, i.e. process of self-organization takes place [18].

The formation of spatio-temporal structure-fractal structures in active extended media was also described in [19]. The authors researched how these reactant spatial fluctuations could be incorporated in the description of the bimolecular kinetics on a microscopic scale, and how they might manifest themselves in the experiments. The forming spatial and temporal structures can be described within the bounds of the model of a system-dynamic self-organization, which is accompanied by a decrease of entropy production. Self-organization and non-equilibrium phase transitions lead to non-classical dependences, which determine the system state as a whole at its intrinsic and extrinsic parameter variations. For example, the formation of localized structures with high concentration of reagents and accordingly high efficiency of current transfer processes leads to non-classical behavior of luminescence decay in a system of excited molecules and their nearest surrounding. At the analysis of experimental kinetic dependences it is necessary to take into account the features of the initial reagent distribution, the change of their distribution type as a result of photo-physic processes, and therefore temporal dependence of appropriate reaction rate constants, which lead to the variation of the fractal properties of the formed clusters.

Structuring process of disordered systems is characterized with nonlinearity and occurs under conditions far from equilibrium. One of the simplest possible ways of describing stochastic phenomena is the theory of fractals [20].

Used in the research multifractal analysis (MFA) in addition to the geometry of the system allows us to describe its physical and chemical properties that occur in its structure, helps to reveal the mechanism of the process, and to determine the conditions of formation , stability, decay of temporal and spatial structures. With the MFA it's practical to get the whole spectrum of Rényi generalized fractal dimensions Dq, to determine entropy S of the system, the parameters of homogeneity R^2, describing the level of self-organization of the system.

The spectrum of Dq shows the level of inhomogeneity of set of points under study and corresponds to the condition $D_{q_1} \geq D_{q_2}$ when $q_2>q_1$, where $\infty<q<\infty$. Generalized fractal dimension always monotonically decreases (or remains constant in the case of monofractal) with increasing q. When q=0, D_0 corresponds to the carrier structure of Hausdorff dimension containing multifractal. The control parameter of self-organization of a multifractal aggregate (q) is related to the density of the multifractal structure.

In case of simple regular fractal of dimension D there is the same number of points in all occupied cells, i.e. all probabilities of filling cells are equal. In this case, all the generalized dimensions Dq are equal and coincide with Hausdorff dimension. The value D_0 (q=0) is a Hausdorff fractal dimension; it determines the highest possible value of a system fractal dimension. Information entropy S is a measure of information, which a structure is obtained at the transition from one level of organization to another.

The parameter of homogeneity R^2 is defined by the probabilities (p_i) of filling geometrically identical parts of the structure under consideration. The homogeneous system is characterized by the parabolic multifractal spectrum function $f(\alpha)$, determined by the set of above mentioned probabilities. In this case, the coefficient of the quadratic approximation authenticity is R^2. The higher is the degree of non-homogeneity in the system the more is the deviation of the spectrum $f(\alpha)$ from the parabola.

The modeling was performed by using a cellular automata (CA) method [17] on a planar lattice corresponding to NaCl type size of 100×100, 500×500 and 1000×1000 units, and on a cubic one size of 50×50×50 and 100×100×100 units. Time of modeling irradiation process on a planar lattice - 10^6, the number of iterations which is proportional to the time of physical process – 5×10^4 (one iteration is equal to one cycle of modeling program). Dose rate is proportional to concentration of induced defects on a lattice at the beginning of each cycle and is equal to 0.1% out of all the nodes on the lattice.

CA is a universal model of parallel calculations. This is a discrete dynamic system, which represents a set of equal nodes or cells combined identically. Nodes form the lattice of the cellular automata. Lattices can be of any type, differing both on dimension and on the cells form. Each node state is completely defined by the nearest nodes states and its own state. All nodes develop simultaneously according to the identical rules. The extensive experimental study of CA, performed by Wolfram [21], says that all models, generated in the CA evolution process from the disordered initial state, can be divided into four classes. CA, which belongs to the IV class, are the most interesting for modeling natural phenomena. In most cases all nodes reach zero value in a finite time interval. However, in some cases propagating configurations or stable periodic structures are formed according to the local interaction rules. Those can be stable infinitely. It is possible to simulate processes of radiation defects formation in solids and defects.

The processes such as aggregation and annealing of radiation-induced defects in a crystal are nonlinear. They proceed in open systems and result in their structural recombination and spatial reorganization of configurations of their components. Owing to the spatial separation of radiation-induced defects in a crystal clusters arise from the same type of defects. Such structures are called multifractal objects or inhomogeneous fractal sets containing subsets of different fractal dimensions [22-25]. For thorough investigation of such nonlinear processes in open systems we applied fractal geometry methods [26].

Besides the geometry of studied model, the applied multifractal approach enables to consider its physical and chemical properties.

3 Results and discussion Computer-based testing results have shown that the total concentration of accumulated radiation-induced defects in a crystal increases several times, then ceases to depend on duration of radiation exposure becoming invariable (Fig. 1). The larger the dose rate, the higher is the growth rate of concentration (Fig. 2).

Concentration of accumulated defects is one of the factors measuring resistance of the material to radiation. In the case when concentration of defects reaches a certain critical value during the irradiation, collective defects can arise in a system.

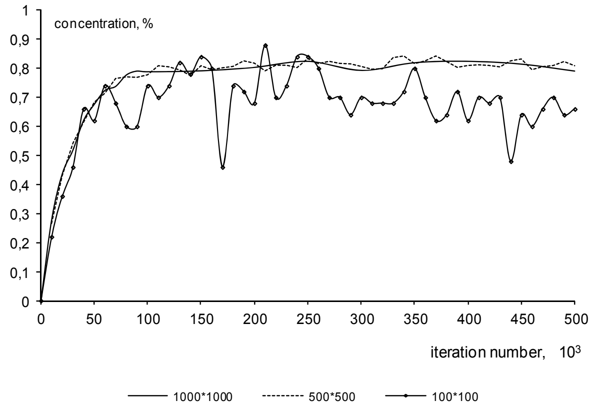

Figure 1 The relationship between concentration of residual defects and duration of irradiation of two-dimensional ionic crystals with lattices of different size. Dose radiation is equal to 0.1% all the time.

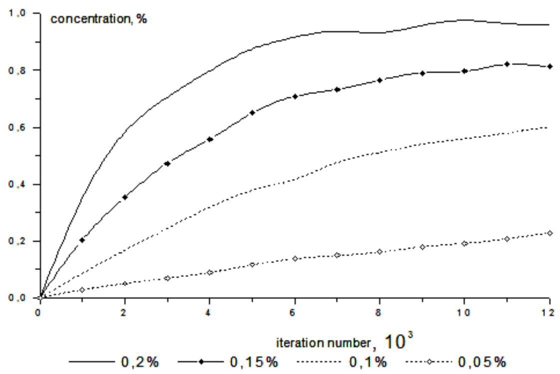

Figure 2 The relationship between concentration of residual defects and duration of irradiation of three-dimensional ionic crystal with lattices size of 100×100×100 under different dose rates.

These defects in the system lead to a process known as self-organization. Nonequilibrium state of a solid caused by the interaction of radiation with matter is the condition for the emergence of this effect.

In this case, there are abnormally big changes in the properties of the material: for example the emergence of multiple clusters.

The concentration of radiation-induced defects in clusters is higher than their average concentration throughout the crystal, so the formation of aggregative centers is more likely within the limits of accumulation of radiation-induced defects of the certain type rather than throughout the crystal as confirmed with the results of modeling.

In Fig. 3, it is shown a spatial separation of polytypic defects and nucleation of aggregates out of the defects of the same type.

Calculation of information entropy as a measure of disorder was performed for the system. Entropy of a fractal structure can be considered as a logarithm of the average number of the possible system states:

$$S = -\sum_i p_i \ln p_i, \tag{1}$$

where p_i is probability to find a certain node of the lattice in the state numbered i.

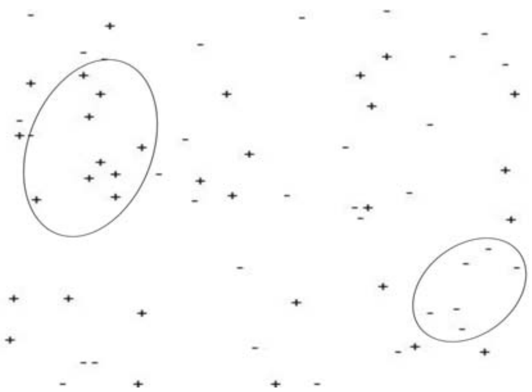

Figure 3 The picture of spatial distribution of radiation-induced defects in two-dimensional ionic crystal with lattice size of 100x100 after 50000 iterations, where «+» and «-» are two types of Frenkel defects.

The proceeding of synergetic effects in a system is proved with information entropy calculations. They cause the decrease of entropy in the system and cluster formations out of defects of the same type (Fig. 4). Lattice aggregates represent a dynamic multifractal (a sustainable formation existing only by constant rearranging of its components until the conditions of the system do not change) when the concentration of defects in a lattice reaches its saturation.

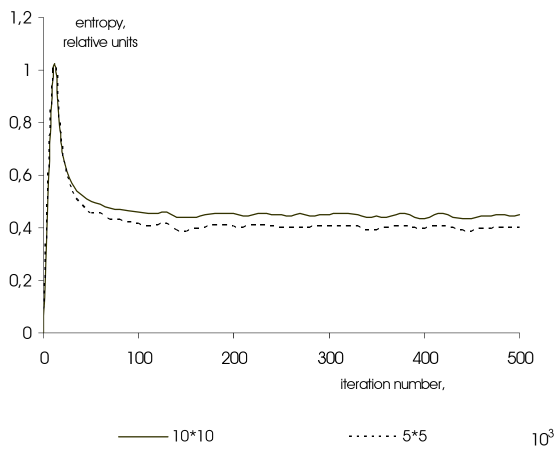

Figure 4 Relationship between the unit value of information entropy of the structure with radiation-induced defects and the duration of 0.1% dose irradiation of NaCl two-dimensional crystal with lattice size of 1000×1000.

To estimate the typical size of the formed aggregations we introduced a statistical parameter, named "conditional potential of the survived defects interaction" (CPI) [27]. This value is calculated for nonzero nodes pairs according to the formula:

$$U = \frac{1}{2}\sum_{k}\sum_{i,j}\frac{q_i q_j}{r_{ij}} \qquad (2)$$

where i, j are the indices of pair components, k is a value of interaction nodes radius, at which it is possible to split the lattice, q_i, q_j are nonzero nodes (the equally charged defects), r_{ij} is the distance between interacting nodes.

The CPI dependence on irradiation duration allows watching the evolution of the defects structure distribution in solids. The dependence of CPI on the duration of irradiation enables to trace back the evolution of distribution of defective structure of a solid (Fig. 5).

The minimums of CPI on the chart represent the state of physical chaos in a system and maximums of CPI show the process of aggregation of the same type defects. Aggregation curves change almost synchronously at different radii of partitions. This suggests the proceeding process of self-organization in a system.

The calculations show that the information entropy of the system in the crystal for a uniform random distribution of radiation-induced defects exceeds the information entropy of the system for the correlated distribution of the same defects concentration.

The decrease of the information entropy suggests the reduction of the number of thermodynamic system microstates. It can be possible only in the case when some structuring (elements recombination) of the system occurs.

This leads to the process of structure ordering having (the higher degrees of symmetry).

It is noticeable that the CPI minimum falls at the information entropy maximum, corresponding to chaotic distribution of defects on a surface and in a volume. Decrease of the system entropy and growth of CPI indicate observed effect of self-organization of defects structure.

The multifractal analysis enabled to estimate the degree of the ordering of the system and to plot the relationship among generalized Renyi dimensions $D(q)$, function of multifractal spectrum $f(\alpha)$ and the value of the self-organization control parameter $-100 \le q \le 100$. Spectrum of the values of $D(q)$ describes structural inhomogeneity and set of different values of function $f(\alpha)$ in different values of α represents the range of fractal dimensions of homogeneous subsets on which the original set can be divided. According to calculations $f(\alpha)$ changes from 0.24 to 1.96 on a plane and from 0.8 to 2.68 in volume. Fractional dimension less than spatial dimension of the system also tells about fractal nature of studied structure.

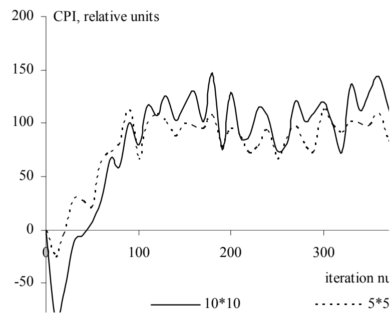

Figure 5 Relationship between CPI and the duration of 0.1% dose irradiation of NaCl two-dimensional crystal with lattice size of 1000×1000.

In Fig. 6 it is possible to follow the changing of Hausdorff dimension D_0, as well as parameter of homogeneity R^2 during irradiation.

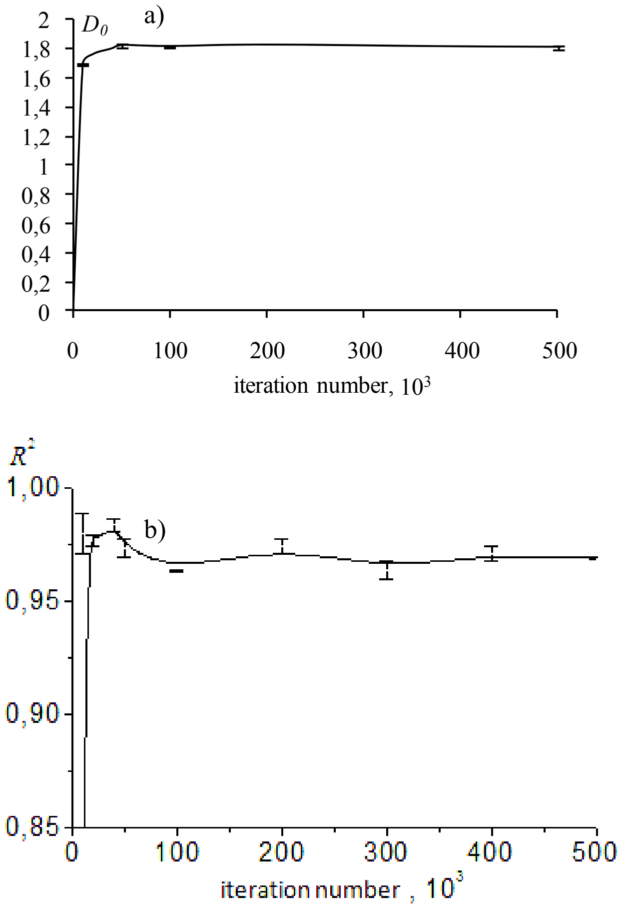

Figure 6 Change of multifractal structural properties of radiation-induced defects (two-dimensional NaCl crystal size of 1000×1000 and 0.1% dose irradiation). (a) Hausdorff fractal dimension D_0, (b) homogeneity R^2.

When the duration of irradiation roughly corresponds to 50-100 thousands iterations, the decrease of homogeneity of discontinuity in a studied system can be easily noticed on the chart (Fig. 6b). All this confirms the fact that randomly distributed clusters arise in the system in the shape of multifractal on the surface of the solid.

If the duration of irradiation exceeds 100000 iterations, the changes of multifractal parameters in the system are not noticed. Apparently, the restructuring of components in the system completely offsets the ongoing impact of external influences on it. Formed structure is a dynamic configuration, enabling the system to more fully dissipate the energy supplied from the outside. All this testifies to the stability of the resulting multifractal structure.

4 Conclusion By computer modeling it was defined that the concentration of radiation-induced defects in an ionic crystal subjected to ionizing radiation is the control parameter of the process of self-organization, resulting in the formation of clusters of the same type radiation-induced defects. The rate of accumulation of radiation-induced defects depends on the dose rate, when the concentration of defects in the crystal during irradiation reaches a critical value.

It is shown that the information entropy calculated for the system is always less than the entropy of a random uniform distribution of the same number of defects in the crystal. After reaching the value of saturation of concentration in the system a decrease to a certain value of the unit value of information entropy takes place, that is, there is a process of self-organization.

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