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Computational study of the percolation phenomena in inhomogeneous two- and three-dimensional systems

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Abstract

In this paper, the results of computer investigation of the percolation processes in inhomogeneous lattices are discussed. The inhomogeneity is simulated by a random distribution of obstacles differing in size and number. The influence of obstacles on the parameters (critical concentration, average number of sites in finite clusters, percolation probability, critical exponents, and fractal and spectral dimensions of a percolation cluster) characterizing the percolation in the system is analysed. It is demonstrated that all these parameters essentially depend on the linear size and relative area of the obstacles.

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1. Introduction

Percolation processes were first considered by Broadbent and Hammersley [1]. These processes and related phenomena can occur in different physical systems. Among them are rock fracture, fragmentation [2] and gelation [3, 4], conduction in a random resistance grating [5] and strongly inhomogeneous media [6], and propagation of forest fires [7, 8] and epidemics [9, 10], the electronic properties of doped semiconductors [11].

Relying on the percolation theory, Kopelman *et al* [12, 13] developed cluster formalism for describing the electronic excitation in inhomogeneous systems. This model deals with such mathematical functions as the percolation probability P_∞ and the average number I_{av} of sites in a cluster. The dependence of these quantities on the concentration C of sites through which the energy migrates is determined by the scaling relationships [14]

$$I_{\mathrm{av}} \propto |C/C_{\mathrm{c}} - 1|^{-\gamma} \tag{1}$$

$$P_\infty \propto |C/C_{\mathrm{c}} - 1|^{\beta} \tag{2}$$

where C_{c} is the critical concentration of sites and β and γ are the critical exponents, which depend on only the space dimension for the homogeneous (habitual) case. Investigations into

Table 1. Values of critical exponents.

Critical exponents	Percolation theory 2D	Percolation theory 3D	Isotopically mixed molecular crystals (habitual cases)	Solid solution of benzaldehyde in ethanol (habitual cases)	Solid solution of benzaldehyde in ethanol (habitual cases)	Ethanol solution of benzaldehyde in porous glass (unhabitual case)
β	0.14	0.41	0.13	0.13	0.41	0.25
γ	2.1	1.6	2.1	2.09	1.7	1.95

the transfer of electronic excitation energy in mixed molecular crystals [15, 16] and solid solutions of organic compounds in low-molecular vitrifying solvents [17] have demonstrated that the critical exponents obtained experimentally coincide with those given by the percolation theory for two- and three-dimensional spaces (see table 1, habitual cases). However, recent studies [18–20] of similar processes in porous matrices revealed a discrepancy between the experimental and theoretical critical exponents (see table 1, unhabitual case). Saha *et al* [21] also note that the matrix affects the topology of the energy transfer. In [18–28] this effect was explained in terms of the inhomogeneous properties of porous glasses used as matrices. A microscopic inhomogeneity of porous glass brings about a change in the effective topology of the space in which percolation processes occur. In turn, this can affect the formation and growth of a cluster from incorporated molecules.

In this work, we performed the computer simulation of the percolation process on a square and cubic lattices with introduced obstacles differing in size and relative area (volume) in order to elucidate the possible effect of these obstacles on the critical concentration C_c, the average number I_{av} of sites in a cluster, the percolation probability P_∞, the fractal dimension of an infinite cluster and the critical exponents.

2. Computational technique

We will solve the site percolation problem, because it is the most important from the viewpoint of energy migration in heterogeneous systems. Let us consider a lattice formed by a set of sites and bonds. It is assumed that C is the part of sites painted black in a random manner. Any two nearest-neighbour black sites are considered to be connected to each other. An aggregate of black sites connected to one another either directly or through chains of connected black sites is referred to as a cluster. Within the cluster formalism, the dynamics of the arising percolation with an increase in C is as follows. At $C = 0$, black clusters are absent in the system. At $C \ll 1$, black clusters consist of a small number of sites: single sites, pairs, triads, etc. However, as the percolation threshold is approached, particular clusters merge together and their average size increases. The average number of sites in finite clusters is defined by the expression

$$I_{av} = \frac{\sum_m i_m m^2}{\sum_m i_m m} \tag{3}$$

where i_m is the number of clusters containing m sites. The analytical dependence of I_{av} on the fraction C is unknown. Numerical calculations showed that, at $C \to C_c - 0$, the quantity I_{av} goes to infinity (see relationship (1)). At $C = C_c$, an infinite cluster extending over the whole space arises for the first time. The concentration C_c at which an infinite cluster of black sites is formed corresponds to the percolation threshold. According to [23, 24], the percolation probability is defined as the ratio between the number of sites forming an infinite cluster and the total number of sites in the lattice. In practice, we deal with systems with finite size and the

value $P_{\max}$ (4) coincides with P_∞ (2) after the percolation threshold, because the percolation cluster does not exist before and coincides with the maximum cluster after. In the numerical simulation, the number of sites contained in the maximum cluster ($m_{\max}$) is calculated and the percolation probability is estimated from the formula

$$P_{\max} = \frac{m_{\max}}{L \times L} \tag{4}$$

where L is the linear size of the lattice. Extensive simulation and theoretical considerations show that, near $C \to C_c + 0$, the percolation probability decreases as the power law (2).

All the results presented in this work were obtained from simulations of the percolation process on 200×200 and $100 \times 100 \times 100$ lattices for two- and three-dimensional cases, respectively. According to the currently available methods of computer reconstruction of inhomogeneous condition of internal structure (see, for example, Vycor porous glasses [25]), the inhomogeneous system was initially simulated by randomly introducing square obstacles with a specified size into the lattice, as was done by Bujan-Nunez *et al* [26]. For this purpose, a new lattice with a size of cell equal to the linear size of obstacles was constructed on the primary lattice. For example, if we have a system with linear size equal to 200 elementary sites and linear size of the obstacles 5, then the new lattice has the length of 40 subsystems, and the subsystems' linear size is 5 elementary sites. Some of the cells (subsystems) of the new lattice were chosen as obstacles and all sites lying within the chosen cell were eliminated (removed) from consideration. In addition, the parts which are isolated by the obstacles, were considered as obstacles too (it leads to an increase of average linear size and concentration of the obstacles, but due to the special checking algorithm not more than 5%). Note that the computer experiments were performed with such inhomogeneous lattice configurations, for which the percolation could occur in two (three for 3D space) mutually perpendicular directions simultaneously. Figure 1 displays variants of the heterogeneous matrices with obstacles differing in linear size and relative area, which were used in our research.

3. Results and discussion

First and foremost, we analysed how the inhomogeneity of the matrix affects the critical concentration. In each case, the value of C_c was determined by two methods. According to Hoshen *et al* [27], the critical concentration can be determined from the position of the maximum in the dependence of the reduced average number I'_{av} of sites in clusters on the site concentration C:

$$I'_{\text{av}} = \frac{\sum_m i_m m^2}{\sum_m i_m m} - \frac{m^2_{\max}}{\sum_m i_m m}. \tag{5}$$

A similar dependence for a clear lattice is depicted in figure 2. Note that, in this case, the accuracy of determining the critical concentration is not very high. As can be seen from figure 2, the dependence obtained by averaging over 200 realizations (2D space) exhibits a rather smeared maximum, even though the concentration in the course of the experiment was changed with the step $\Delta C = 0.001$. For this reason, the critical concentration for each realization was taken as the concentration corresponding to the onset of the percolation between opposite sides of the lattice. This approach made it possible to determine the average critical concentration in the system. Figure 3 displays a histogram that allows one to judge the probability of an infinite cluster forming at a given concentration of black sites. The critical concentration determined from these data agrees closely with the value obtained using other methods for a square lattice [7, 11, 14, 24]. The introduction of obstacles into the lattice

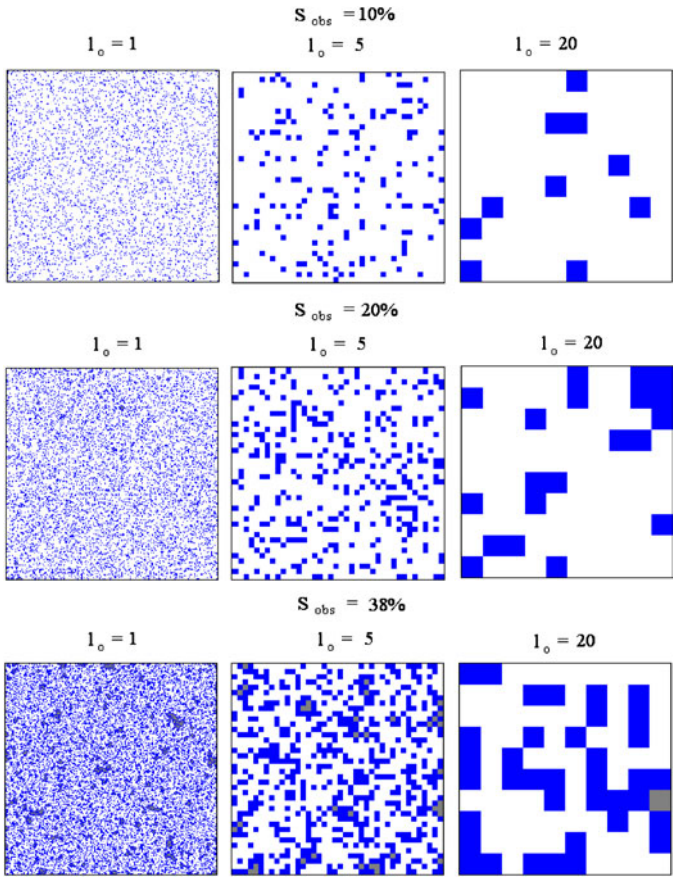

Figure 1. Variants of matrices with obstacles differing in linear size and relative area.

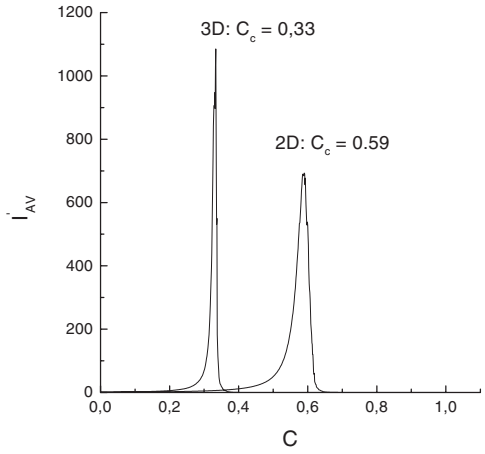

Figure 2. Dependence of I'_{av} on the concentration of occupied sites C for a 200 × 200 square and for a 100 × 100 × 100 cubic lattice without obstacles.

considerably affects the critical concentration: its value increases substantially (figure 4). The strongest effect is observed for the smallest obstacles. An increase in the critical concentration

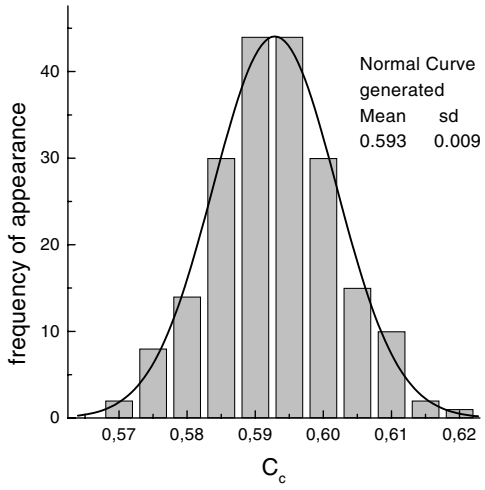

Figure 3. Probability of percolation cluster forming as a function of the concentration of occupied sites C_c for the percolation process on the square and cubic lattices.

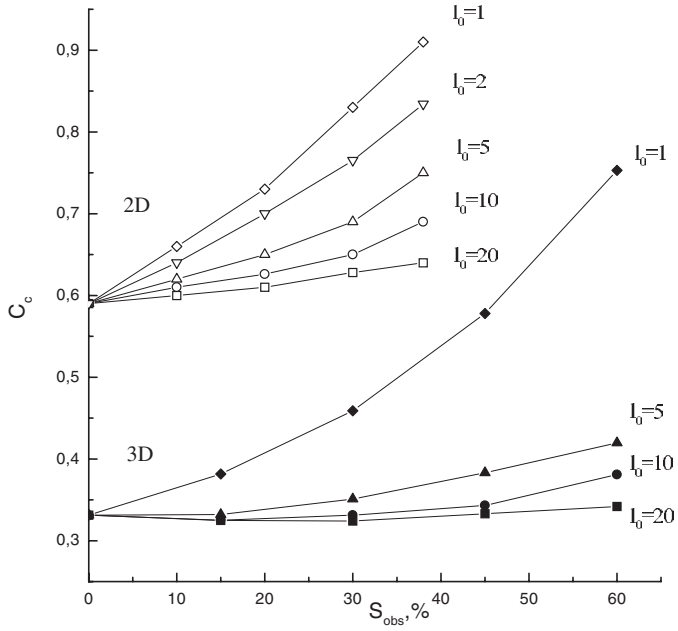

Figure 4. Dependence of the critical concentration C_c on the homogeneity fraction S_{obs} in the matrix at different linear size l_0 of obstacles.

in the inhomogeneous matrix can be explained in the following way. In a homogeneous lattice, the shortest path between any two points is a straight line (without regard for the lattice structure on minimum scales). In an inhomogeneous matrix, the shortest path can substantially deviate from a straight line. It is clear that the larger the number of obstacles (or the larger the relative area of obstacles) in the matrix the longer the shortest path between any two points. Upon introduction of obstacles into the lattice, the total number of accessible sites decreases, whereas the number of black sites required for connecting any two points in

the system increases. Consequently, the critical concentration in the inhomogeneous matrix should increase. Recall that, in this case, the critical concentration is equal to the ratio of the number of black sites (this number corresponds to the onset of percolation) and the total number of sites in the system. This effect becomes less pronounced with an increase in the linear size of obstacles, because, at the same relative area, the larger-sized obstacles turn out to be located in a certain lattice region. As a result, part of the matrix behaves as a homogeneous lattice. The larger the linear size of obstacles, the greater the fraction of the homogeneous part.

As is known, the behaviour of different quantities in the vicinity of the percolation threshold is adequately described by the critical exponents. For the homogeneous case the critical exponents depend only on the space dimension [14]. However, for each space dimension, there exists a great number of different problems. According to modern concepts, the critical exponents for all problems in a homogeneous space of the same dimension are identical to one another. It is possible that the physical reasons for the universality of critical exponents lie in the fact that the exponents are determined by the structure of clusters in the vicinity of the percolation threshold. In this case, the geometric properties of clusters play the dominant role, because they manifest themselves at large distances (of the order of the correlation radius). These distances in the vicinity of the percolation threshold are considerably larger than the lattice spacing (in the case of lattice problems). Therefore, the cluster geometry does not depend on the type of lattice used in solving a particular problem. Moreover, a particular problem can be specified not a periodic lattice but on sites randomly arranged in space: this circumstance will not affect the structure of large-sized clusters. However, the cluster geometry is substantially affected by the space dimension. For these reasons, the critical exponents depend on the dimension of a particular problem rather than on its type.

Therefore, unlike the percolation thresholds, which depend on the type of problem involved, the critical exponents exhibit a certain universality. This leads us to the important conclusion: if the results of physical experiment are treated within the percolation theory and the microscopic structure is not quite clear, it is necessary, first of all, to compare the critical exponents with the theory. Because (for the homogeneous case) they depend only on the space dimension, and do not depend on the type of the lattice, nevertheless, the introduction of the obstacles leads to more complex behaviour, and the exponents are no more the constants.

The dependence of the average number of sites in a cluster on the reduced concentration C/C_c of occupied sites is plotted on a log–log scale in figure 5. As is clearly seen, this dependence over a wide range of concentrations is well described by the power law predicted by formula (1). The deviation from the power dependence near the critical concentration is caused by the finite sizes of the lattice. In fact, as follows from formula (1), this quantity should increase to infinity at the critical point, which, in principle, is impossible in systems of finite size. The critical exponent γ determined from this dependence coincides with the value obtained by the same method in [27], and is slightly less than the exponents derived from other techniques [14]. Figure 6 displays the dependence of the critical exponent γ (determined in a similar manner) on the linear size of obstacles at different values of their relative areas. It can be seen that the critical exponent for obstacles with $l_0 = 1$ coincides with the exponent for the homogeneous lattice. As the linear size increases, the critical exponent first increases (to $l_0 = 10$) and then decreases. An increase in the relative area of obstacles into the lattice brings about separation of sites belonging to the same cluster. The critical exponent γ characterizes the cluster growth with an increase in the concentration. The larger the size and the larger the relative area of obstacles, the higher the concentration at which sites begin to coalesce into clusters and small-sized clusters merge into large-sized clusters. To state this

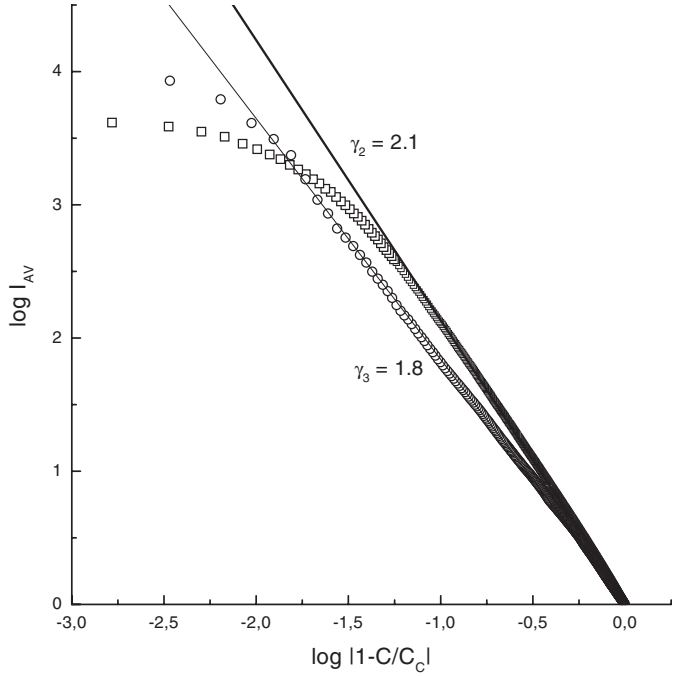

Figure 5. Dependence of log I_{av} on log $|1 - C/C_{\mathrm{c}}|$ for square and cubic lattices.

differently, an increase in the average cluster size with an increase in the concentration is more pronounced than that in the system with a homogeneous matrix; in fact, this corresponds to an increase in the critical exponent γ. It is worth noting that all the lattices studied are characterized by a linear dependence similar to that depicted in figure 5. The observed decrease in the exponent γ for matrices with obstacles of size $l_0 = 20$ can be explained by the finite sizes of the lattices. As can be seen from figure 1, an increase in the size of the obstacles brings about the transformation of the inhomogeneous matrix into a matrix with a specially confined structure. This is essentially pronounced for lattices with the maximum relative area of the obstacles used in the computer experiment. Numerical simulation revealed that, for 2D matrices at $l_0 = 20$ and $S_{\mathrm{obs}} = 38\%$, an increase in the lattice size to 400×400 is accompanied by an increase in the critical exponent γ to 2.94 (for 200×200 lattice, $\gamma = 2.50$). Note that an increase in the size of the obstacles to 40 results in a further decrease in exponent γ (figure 7).

A more intricate situation arises with the dependence of the percolation probability on the concentration of occupied sites. According to formula (2), this dependence on log–log scale should be represented by a straight line whose slope corresponds to the critical exponent β . Unfortunately, the treatment of our results for the square lattice demonstrated that this dependence does not exhibit a linear behaviour with the appropriate slope over the entire range of concentrations ($C > C_{\mathrm{c}}$). Furthermore, an analysis of the available data on this problem also showed some disagreement regarding the range of applicability of relationship (2). In particular, Hoshen *et al* [27] observed the scaling dependence (2) for a 4000×4000 triangular lattice only in the $(C - C_{\mathrm{c}})$ concentration range from 10^{-4} to 2×10^{-2}. At higher concentrations, the dependence deviated from linear behaviour. In our simulation, the results obtained in this concentration range strongly depend on the finite size of lattice, as is the case with the average number of sites in clusters. Moreover, Hoshen *et al* [27] observed a linear dependence for a 400×400 square lattice in the concentration range from 2×10^{-3} to

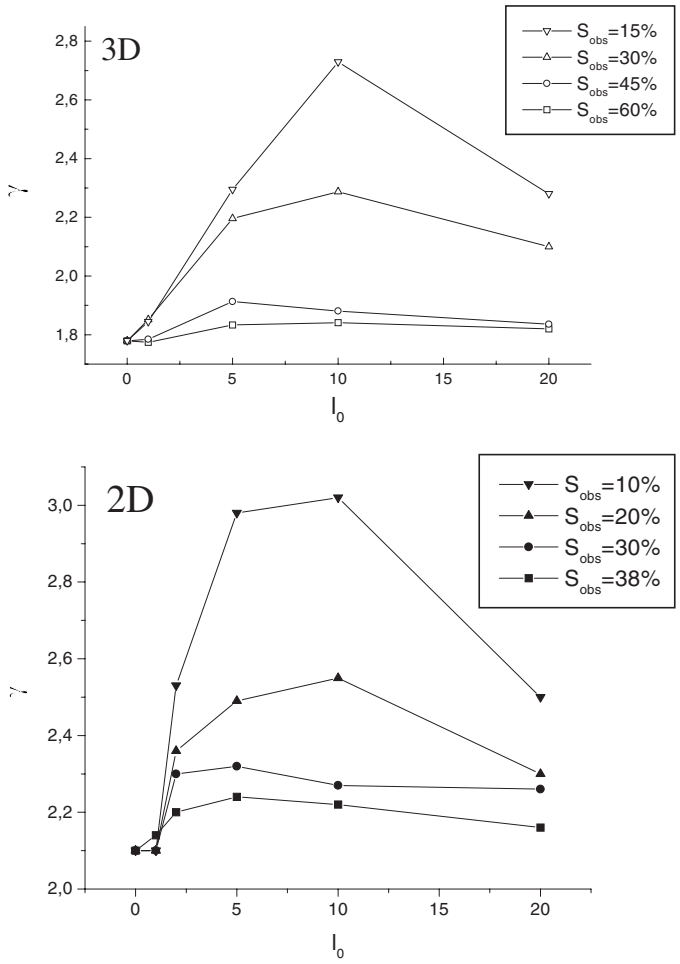

Figure 6. Dependence of the critical exponent γ on the linear obstacle size l_0 in lattices at different relative areas of the obstacles S_{obs}.

7×10^{-2}, even though the slope corresponded to $\beta = 0.19$. Therefore, relationship (2) is valid only in a very narrow concentration range in the vicinity of the percolation threshold. However, reasoning from the results of investigation into the transfer of electronic excitation energy in mixed molecular crystals, Kopelman [13] made the inference that the concentration range of applicability of the critical exponents for the energy migration is considerably wider than that for any other critical phenomenon in physics. From the viewpoint of the energy transfer, it is important that the dependence of the probability of trapping an exciton (which is governed by the percolation probability in the range of concentrations higher than the critical concentration [12]) on the reduced concentration C/C_c of activator molecules in the inhomogeneous matrix is steeper than that in the homogeneous matrix [20]. In terms of the critical exponents, this corresponds to a decrease in the exponent β.

Figure 8 depicts the dependence of the percolation probability on the reduced concentration for the lattices with different fractions of the obstacles. It is clearly seen that the introduction of obstacles into the lattice is attended by a more rapid increase in the percolation probability with an increase in the concentration. The observed effect becomes

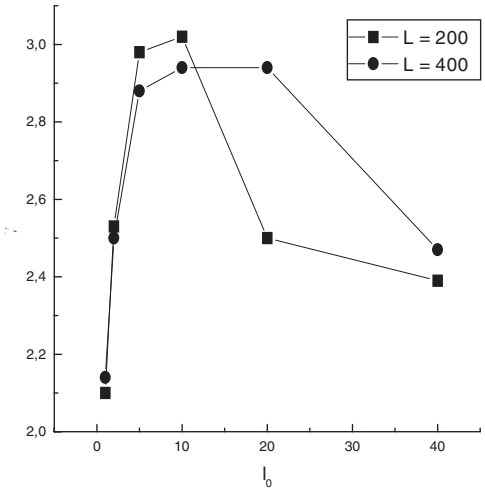

Figure 7. Dependence of the critical exponent γ on the linear size of the matrix for the 2D case at $l_0 = 20$ and $S_{\text{obs}} = 38\%$.

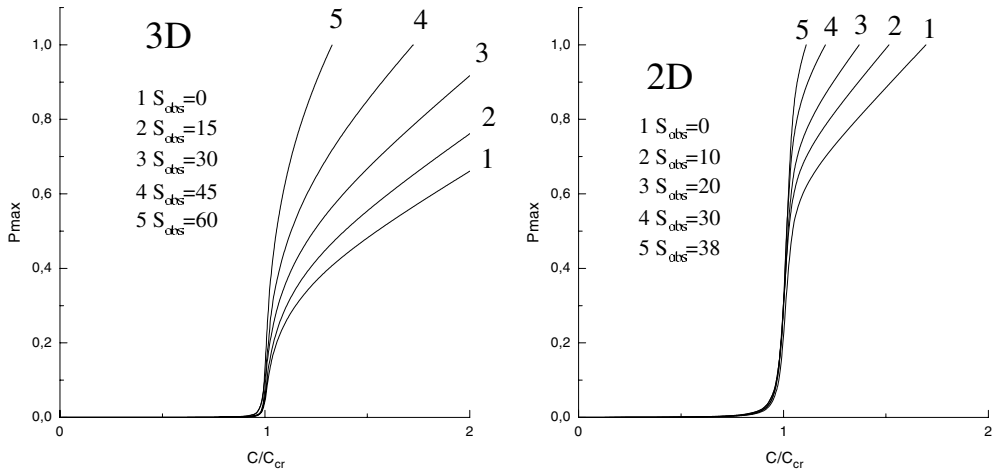

Figure 8. Dependence of $P_{\max}$ on the reduced concentration C/C_{cr} for lattices with obstacles of the linear size $l_0 = 1$ at different fractions of the obstacles in the matrix.

less pronounced with an increase in the linear size of obstacles. However, our investigations showed that, in any case, the presence of obstacles in the matrix leads to a change in this dependence.

The fractal dimension d_f is a principal characteristic of the infinite cluster at the critical point. Mandelbrot [29, 30] was the first to introduce the notion of fractal. Subsequently, he specified the tentative concept [31] and defined the fractal as a structure consisting of parts that, in some sense, are similar to unity [32]. However, until presently, there has been no rigorous and complete definition of fractals. An infinite cluster at the critical point exhibits a statistical self-similarity [24]. The fractal geometry of the infinite cluster and its statistical self-similarity are interrelated. This interrelation leads to the following relationship between the mass and the linear size of the cluster:

$$M = l^{d_f}. \tag{6}$$

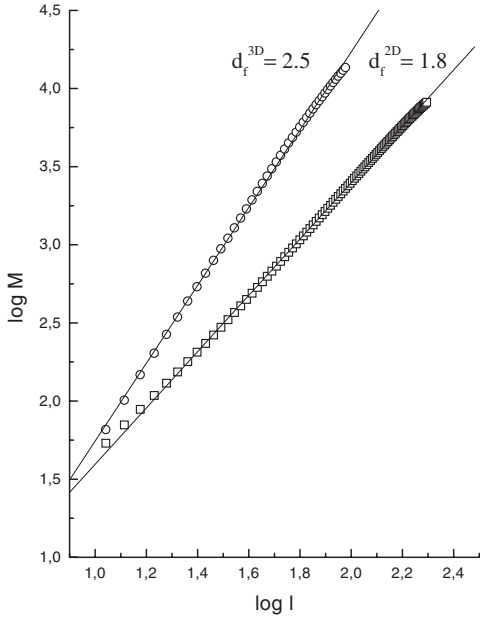

Figure 9. Dependence of log M on log l for the square and cubic lattices free of obstacles.

Stanley [33] and Sokolov [34] showed that the fractal dimension in virtually all physical problems is defined as the exponent in relationship (6). At present, the fractal dimension is determined using different methods [35]. One of them is the embedded square method proposed in [24]. In essence, this method is as follows. In the object under study, a central point is chosen in a random manner and several embedded squares are arranged around this point. The number of sites in each square is counted, and the dependence of the object mass (the number of sites) on the linear square size is constructed. This dependence is used for calculating the fractal dimension. Forrest and Witten [36] proposed to bring the central point into coincidence with the centre of gyration of the studied object in order to improve the reproducibility of the results. In our work, we also determined the fractal dimension of the infinite cluster by using the embedded square method. To accomplish this, among all the possible realizations, we chose clusters whose centres of gyration were close to $L/2$. It should be noted that the introduction of this critical concentration did not affect the statistics obtained for the critical concentration (figure 3). The results presented below were obtained by averaging over 300 different clusters. For lattices with large-sized obstacles, additional averaging was performed over 20 configurations of distribution of the obstacles in these lattices.

The dependence of the cluster mass (the number of sites) on the square size is plotted on a log–log scale in figure 9. It is easy to see that the dependence exhibits a linear behaviour beginning with square sizes of the order of 20×20. The fractal dimension determined from the slope of this dependence is equal to 1.8. This value is slightly less than the exact dimensional $d_f = 91/48$, which was calculated in [7, 14] in terms of the scaling theory. This difference can arise for two reasons. First, as was shown in [35], the fractal dimension determined by the embedded square method is underestimated compared to that obtained by other methods. Second, the underestimated value of d_f can be dictated by the finite size of the lattice. It is obvious that the percolation cluster on a finite lattice is only a part of an infinite cluster

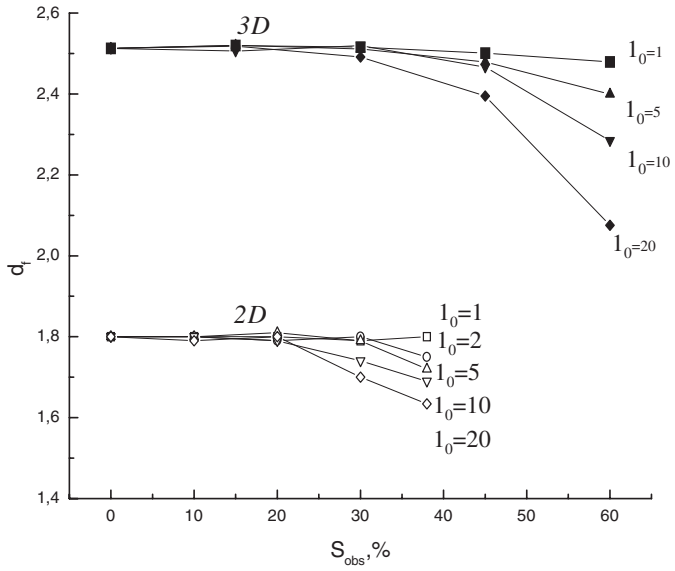

Figure 10. Dependence of the fractal dimension of a percolation cluster on the fraction of the obstacles S_{obs} in the matrix at different linear size l_0 of obstacles.

on the infinite lattice for which the exact dimension was deduced. Consequently, particular sites that are not involved in the percolation cluster on the lattice of size L, in actual fact, belong to the infinite cluster, because they are connected to it through bonds lying outside the fragment under consideration. In any case, our prime concern is with the influence of the inhomogeneous properties of the lattice on the fractal dimension rather than in its absolute value. This influence is illustrated by the data shown in figure 10. As can be seen, no change in the fractal dimension is observed for lattices with the linear size of the obstacles $l_0 = 1$. The value of d_f decreases for lattices with larger-sized obstacles only in the case when their fraction in the system is sufficiently high. The effect is enhanced with an increase in the obstacle size. This behaviour can easily be explained with due regard by the fact that the percolation cluster is a strongly porous object. Therefore, when the size of obstacles and their relative area are small, the probability that obstacles occupy these pores is high. As the size of obstacles and their fraction in the matrix increase, they begin to affect the geometry of the percolation cluster and this effect manifests itself in a decrease in its fractal dimension.

Another dimension associated with random walks on a cluster is a spectral dimension d_{s}. In [37], it has been shown that at the percolation threshold the number S_N of distinct sites visited during an N-step random walk on an infinite cluster varies asymptotically as

$$S_N \sim N^{d_{\text{s}}/2} \qquad d_{\text{s}} = 4/3 \tag{7}$$

in all Euclidean dimensions (Alexander–Orbach hypothesis) [38].

Here we are reporting our simulation investigations of random walks on a percolating cluster for a two-dimensional lattice. To obtain this result for every lattice 2×10^2 different realizations of percolation cluster were used. For every percolation cluster 10^2 statistics of random walks were investigated. Initial position and direction of jump of the walk have been chosen in a random manner. Random walks that never need to cross the boundary sites were

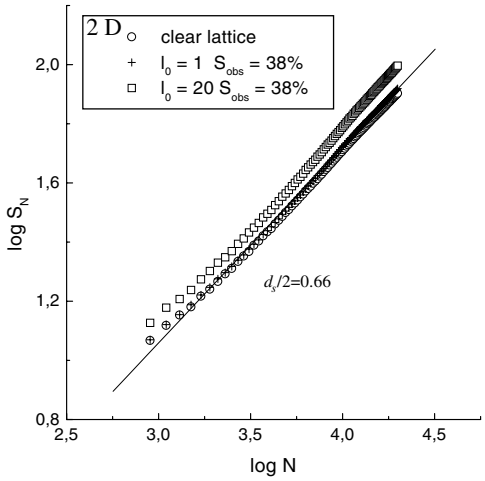

Figure 11. Dependence of log S_N on log N for square lattices.

considered. The realizations of the walk with initial site attended to the boundary of the lattice were not taken into account.

In our investigation we obtained the values of spectral dimension close to $4/3$ for lattices differing in relative area and linear size of the obstacles (figure 11). It is seen from figure 11 that the introduction of the obstacles to the lattices does not strongly affect d_s. This result is in a good agreement with the Alexander–Orbach hypothesis.

4. Conclusion

Thus, the results obtained in this work confirm the assumption made earlier (on the basis of the available data on the energy transfer in disordered system, specifically, in matrices with different structures on the microscopic level) that the inhomogeneous properties of matrices substantially affect the percolation process. The introduction of the obstacles causes strong influence on all percolation parameters in both two- and three-dimensional spaces. Increase of the relative area of the obstacles leads to increase of the critical concentration value, increase of the critical exponent γ value, increase of the growth rate of percolation probability with increase of the concentration and decrease of the fractal dimension of percolation cluster. In turn, increase of the linear size of the obstacles (with the same relative area of them) results in a fall in the critical concentration value, increase in the critical exponent γ value, a fall in the growth rate of percolation probability with increase of the concentration and decrease of the fractal dimension of percolation cluster. As a result, our investigation clearly shows that the critical exponents and the fractal dimension depend on the inhomogeneous macroscopic structure (linear size and concentration of the obstacles) of the system. Only the spectral dimension of the percolation cluster does not depend on the presence of the obstacles in lattice, which accords with the concept of super universality for this parameter.

In general, our research demonstrates that all percolation parameters essentially depend on the linear size and relative area of the obstacles. Nevertheless, the question about the influence of the finite size of the systems on the precision of the results, especially on the critical exponents, is still open, because only investigations for the systems with various linear

sizes (it is better to have bigger ones) can give a more exact answer, and we hope that our work only opens discussion about this problem.

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