Percolation on patchwise heterogeneous surfaces under equilibrium conditions

M.C. Giménez, A.J. Ramirez-Pastor, F. Nieto *

Departamento de Física, Instituto de Física Aplicada, Universidad Nacional de San Luis - CONICET, Chacabuco 917, D5700BWS, San Luis, Argentina

A B S T R A C T

The site percolation problem on square lattices whose sites are grouped in two types of energetically different patches is studied. Several lattices formed by collections of either randomly or orderly localized and no overlapped patches of different sizes are generated. The system is characterized by two parameters, namely, the size of each patch, l, and the energy difference between the two kind of sites, ΔE. Particles are adsorbed at equilibrium on the lattice. The critical coverage is determined by means of Monte Carlo simulations and finite-size scaling analysis. The percolative behavior of the system as a function of the parameters characterizing the heterogeneity of the energetic surface topography is presented and discussed.

© 2008 Elsevier B.V. All rights reserved.

1. Introduction

Even though the problem of percolation has been known for many years and its various aspects have been dealt with by relevant contributions, there are still interesting open questions to be discussed [1–7]. Percolation deals with the effect of random dilution of elementary geometrical object located in lattices or in the continuum. Upon dilution of the elements, a sharp transition is found to occur in the connectivity of the system. The percolation threshold is defined as the minimum concentration or density at which an infinite cluster of occupied elements spans the system. Studies of percolation processes are of interest not only for their potential utility in understanding a range of physical phenomena where the connectivity is a dominant feature, but also because the geometric critical phenomena exemplified by percolation possess many striking parallels with the thermally driven critical phenomena, as it is provided by the Fortuin–Kasteleyn mapping [8].

Most of the studies in the field are devoted to the percolation of particles that are irreversibly deposited. In part this is due to the fact that the deposition (or irreversible adsorption) of particles on solid surfaces is a subject of considerable practical importance. In many experimental systems, the relaxation timescales are much longer than the times of the formation of the deposit. In such processes, the temperature of the system does not play a relevant role and it is not considered. However, in numerous systems of both theoretical and practical importance, where the adsorbed particles are in thermodynamic equilibrium, the spatial distribution of the particles might be characterized by using the percolation model [9,10]. In these cases, the temperature governs the phase in the system and can be an important controlling factor in the percolation process. By means of a percolation study, in Refs. [11,12] it was shown that the spatial configuration of the adlayer (particles adsorbed on a solid surface) **at equilibrium** recognizes peculiarities related to the phase transitions occurring in the adsorbate. Such

---

* Corresponding author. Tel.: +54 2652 431080; fax: +54 2652 430224.
  E-mail addresses: cecigime@unsl.edu.ar (M.C. Giménez), antorami@unsl.edu.ar (A.J. Ramirez-Pastor), fnieto@unsl.edu.ar (F. Nieto).

a study implied the determination of the critical parameters as a function of the concentration (surface coverage) and temperature. The main aim of the present paper is to go one step further and to include in the analysis the influence of the energetic surface heterogeneities on the percolation of particles adsorbed at equilibrium onto such surfaces.

In recent years, the progress in the experimental techniques for surface analysis at atomic scales have improved our knowledge about the energetic surface topography. These facts have encouraged the development of more refined atomistic models for heterogeneous surfaces which in principle are capable to include the energetic surface topography in the statistical description [13–19]. One of the aspects requiring a better understanding is the way in which the energetic topography affects different adsorption processes. In fact, even though it has been shown that many molecular surface processes are indeed dependent on the way adsorptive energy is spatially distributed on the surface (topography) [20–22], a systematic study showing how the main characteristics determining the topography act on the percolation of adsorbed particles at equilibrium is far to be exhausted. The present work intends to be a new step in this direction. Unlike adsorption, where lateral interactions – necessary in order to get adsorption isotherms sensitive to the topography – compete with topography in such a way to obscure its effects [23,24], percolation of monomers at equilibrium is found to have such a sensitivity even for non-interacting particles. This is a great advantage and for this reason we restrict our study to the case of null adsorbate–adsorbate interactions, in order to put forward with greater clarity the pure topographic effects.

The aim of this paper is to investigate the site percolation problem in a particular lattice with restricted geometry, i.e. the patchwise lattice. These lattices, largely used to model adsorptive heterogeneous surfaces, are composed of two different types of sites which in turn are grouped into homogeneous patches or finite domains. Our main motivation to study the percolation properties of non-interacting particles at thermodynamical equilibrium on these structures is based on the fact that previous results of adsorption, diffusion and reaction on both random [25–27] and ordered patchwise lattices [28–30], have demonstrated to be very interesting from a theoretical and practical point of view.

This paper is organized as follows. In Section 2 we describe the lattice-gas model and the Monte Carlo simulation technique used to obtain the desired quantities for describing the percolation phase transition. Results are presented and discussed in Section 3. Finally, some conclusions are drawn in Section 4. In the Appendix we establish a mapping between this formulation of the problem (reversible adsorption of non-interacting particles on heterogeneous surfaces) and that informed in Ref. [31] (irreversible deposition of particles on simple patchwise surfaces).

2. Model and calculation method

Let us consider that the substrate is represented by a two-dimensional square lattice of $M = L \times L$ adsorption sites, with periodic boundary conditions. In order to describe the system of N monomers adsorbed on M sites at a given temperature T, let us introduce the occupation variable c_i which can take the following values:

$$c_i = \begin{cases} 1, & \text{if site } i \text{ is occupied} \\ 0, & \text{if site } i \text{ is vacant.} \end{cases} \tag{1}$$

Particles can be adsorbed on the substrate with the restriction of at most one adsorbed particle per site and we consider a null nearest neighbor (NN) interaction energy among them. If ε_i is the adsorption energy of one given surface site, the Hamiltonian of the system is given by

$$H = \sum_i^M \varepsilon_i c_i. \tag{2}$$

Now, we describe the model used for introducing the surface heterogeneities. Let us consider a square lattice of $L \times L$ sites, as a board divided in shallow (S) and deep (D) square patches with $l \times l$ sites each one and energies E_S and E_D, respectively. We assume that the number of S squares is the same as the number of D squares. S and D patches can be put in order onto a square lattice, in such a way that any S (D) patch has four nearest neighbor D (S) patches, so the resulting structure will be a perfect chessboard lattice or regular patchwise lattice. But if the squares in a chessboard are randomly occupied by S or D patches, we have a random patchwise lattice. These lattices can be easily simulated on the computer. We suppose that the patches are touched together, sharing common borders, and any overlap between them is entirely forbidden.

For fixed values of surface coverage, $\theta = N/M$, and temperature T, the thermodynamic equilibrium is reached in the canonical ensemble by using a standard Kawasaki algorithm [32]. The procedure is as follows. An initial arbitrary configuration of N adsorbed monomers with the desired surface coverage is generated. Two sites are randomly selected, and their position are established. Then an attempt is made to interchange its occupancy state with probability given by the Metropolis rule [33]:

$$P = \min\{1, \exp(-\Delta H/k_B T)\} \tag{3}$$

where $\Delta H = H_f - H_i$ is the difference between the Hamiltonians at the final and initial states and k_B is the Boltzmann constant.

A Monte Carlo Step (MCS) is achieved when M pair of sites have been tested to exchange its occupancy state. The equilibrium state can be well reproduced after discarding the first n' MCS. Then, a set of n samples in thermal equilibrium is generated by taking configurations separated from each other by a few MCS in order to avoid memory effects.

The central idea of the percolation theory is based on finding the minimum coverage degree θ for which at least a cluster [a group of occupied sites in such a way that each site has at least one occupied nearest neighbor site] extends from one side to the opposite one of the system. This particular value of the coverage degree is named *critical concentration* or *percolation threshold*, and it determines a phase transition in the system.

As the scaling theory predicts [6], the larger the system size to study, the more accurate the values of the threshold obtained therefrom. Thus, the finite-size scaling theory give us the basis to achieve the percolation threshold and the critical exponents of a system with a reasonable accuracy. For this purpose, the probability $R = R_L^X(\theta)$ that a lattice composed of $L \times L$ sites percolates at concentration θ can be defined [2,34,35]. Here, the following definitions can be given according to the meaning of X: (a) $R_L^{R(D)}(\theta) = $ the probability of finding a rightward (downward) percolating cluster; (b) $R_L^I(\theta) = $ the probability that we find a cluster which percolates both in a rightward **and** in a downward direction; (c) $R_L^U(\theta) = $ the probability of finding either a rightward **or** a downward percolating cluster and (d) $R_L^A(\theta) \equiv \frac{1}{2}\left[R_L^R(\theta) + R_L^D(\theta)\right] \equiv \frac{1}{2}\left[R_L^I(\theta) + R_L^U(\theta)\right]$. The first step for determining the percolation threshold consists in evaluating the effective threshold $\theta_c^X(L)$ for a lattice of finite size L. Once the temperature, the energy difference between the two kind of sites (ΔE), the size of the patches (l) and the patch distribution (for the ordered case) are fixed the following procedure is used: (a) the construction of n samples for a given coverage (according to the scheme presented before) and (b) the cluster analysis by using the Hoshen and Kopelman algorithm [36]. n runs of such two steps are carried out for obtaining the number m^X of them for which a percolating cluster of the desired criterion X is found. Then, $R_L^X(\theta) = m^X/n$ is defined and the procedure is repeated for different values of θ. For the case of random distribution of patches, the procedure is very similar, but the n samples involve different random distributions of patches, in order to take an average over different surface structures.

The second step is the extrapolation of $\theta_c^X(L)$ towards the limit $L \to \infty$ by using the scaling hypothesis. Thus, the correlation length, ξ, can be expressed as

$$\xi \propto |\theta - \theta_c|^{-\nu} \tag{4}$$

where the critical exponent ν is analytically shown to be equal to $\nu = 4/3$ in the case of random percolation [2,4–6]. As $\theta = \theta_c^X(L)$ the correlation length reaches the linear dimension L of the lattice. Thus, we have

$$\theta_c^X(L) = \theta_c(\infty) + A^X L^{-\frac{1}{\nu}} \tag{5}$$

where A^X is a non-universal constant. The maximum of the differences between $|\theta_c^I(\infty) - \theta_c^A(\infty)|$ and $|\theta_c^U(\infty) - \theta_c^A(\infty)|$ gives the error bar for each determination of θ_c. The interested reader is invited to see Ref. [11] for a more detailed analysis.

3. Results and discussion

The present percolation study involves adsorbed particles on heterogeneous surfaces under equilibrium conditions (which makes the difference with the traditional percolation problem being concerned with irreversible adsorption). In our calculations we have used values of L ranging between $L_{\min} \approx 40$ and $L_{\max} \approx 400$. With these lattice sizes we verified that finite-size effects are negligible. Note, however, that the linear dimension L has to be properly chosen such that it is a multiple of $2l$.

Without any loss of generality, we consider that all energies are measured in units of $k_B T$ (or $k_B T = 1$), and that $E_S = 0$ and $\Delta E = E_S - E_D$, in such a way that the adsorptive energy is characterized by the single adimensional parameter ΔE (energy difference between the two kinds of adsorption sites).

Fig. 1 shows the adsorption isotherms (surface coverage, θ, as a function of chemical potential, μ) for three values of ΔE. The isotherms were calculated theoretically using the following relationships:

$$\theta_S = \frac{e^{-\beta(E_S - \mu)}}{1 + e^{-\beta(E_S - \mu)}} \tag{6}$$

$$\theta_D = \frac{e^{-\beta(E_D - \mu)}}{1 + e^{-\beta(E_D - \mu)}} \tag{7}$$

$$\theta = f_S \theta_S + f_D \theta_D \tag{8}$$

where θ_S (θ_D) is the coverage of the shallow (deep) sites and f_i are the fraction of sites of type i present on the surface (in this case, $f_S = f_D = 1/2$).

Due to the absence of lateral interactions, the shape of the isotherms is independent of the distribution of the adsorption sites (i.e. independent of l and if the patches are ordered or at random). As it can be clearly seen, a plateau appears which gets wider as the temperature decreases or as the energy difference ΔE grows. This plateau corresponds to the situation where half of the sites are occupied $\theta = 0.5$, precisely the deeper ones. In the isotherms, for each chemical potential there is a coverage degree associated and we study the percolation state of the system for each point.

Fig. 2 shows the frames corresponding to four of the studied situations with $l = 1$: (a) ordered patches and $\Delta E = 1.0$; (b) ordered patches and $\Delta E = 10$; (c) random distribution and $\Delta E = 1.0$; and (d) random distribution and $\Delta E = 10$; In all cases we choose $\theta = 0.6$ because this value is close to the critical coverage degree for the random percolation case
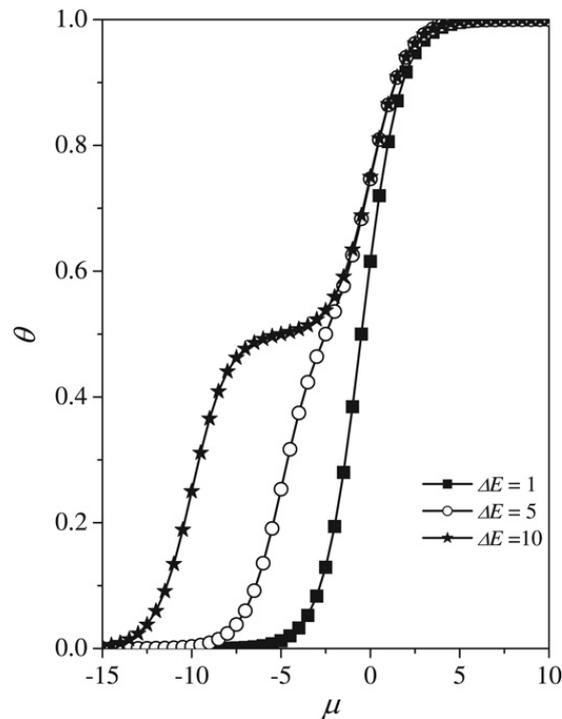
Fig. 1. Adsorption isotherms for three values of ΔE (energy difference between the two kinds of adsorption sites). The shape of the isotherms is independent of both the size and the distribution of the patches due to the absence of lateral interactions between the adsorbed particles. Lines and symbols represent theoretical calculations by using Eqs. (6)–(8).
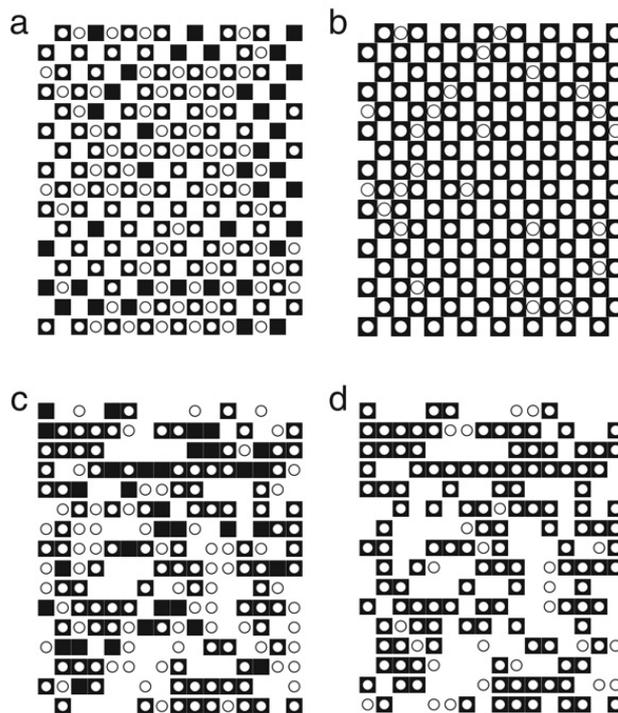
Fig. 2. Snapshots of typical equilibrium states for $l = 1$ and coverage $\theta = 0.6$ in the following cases: (a) ordered patches, $\Delta E = 1$, (b) ordered patches, $\Delta E = 10$, (c) random patches, $\Delta E = 1$, (d) random patches, $\Delta E = 10$. Black squares: deep sites; white squares: shallow sites; circles: adsorbed particles. The snapshots are a single Monte Carlo realization of the system.

($\theta_c \approx 0.5927\ldots$), which would be equivalent to the homogeneous surface ($\Delta E = 0.0$) in the studied model. The snapshots shown in Fig. 2 are a single Monte Carlo realization of the system in each case. We can see that for $\Delta E = 10.0$, all deep adsorption sites are occupied and the remaining particles are distributed at random between the shallow sites.

Fig. 3 shows the percolation threshold, θ_c, as a function of the energy difference between the two kind of sites, ΔE, for several values of patch size, l, in the case of ordered distribution. In all cases, θ_c grows upon increasing ΔE until it reaches a
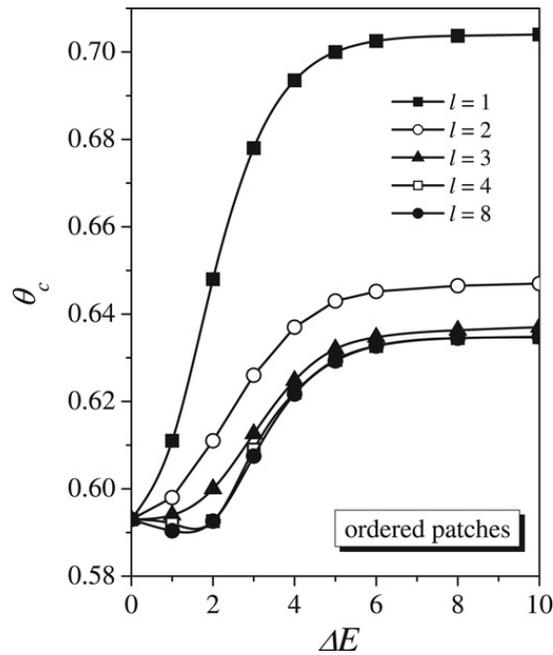
Fig. 3. Percolation threshold as a function of ΔE for different patch sizes in the case of ordered distribution of patches. Symbols represent Monte Carlo simulation data and lines are a guide to the eyes.

saturation value, approximately close to $\Delta E \approx 6$. This saturation value can be explained for the case of $l = 1$ as follows. When ΔE is large enough compared to the thermal energy [see Fig. 2(b)], all the deep sites are occupied. Then, we can imagine an effective lattice formed by the shallow sites where each element has eight effective neighbors (for more details see Ref. [31]). For this kind of lattice, it is found that the percolation threshold for the irreversible adsorption is $\theta_c(z_{eff} = 8) \approx 0.41$ [4]. As shallow sites constitute half of the sites, this corresponds to a fraction of occupied sites of $\theta \approx 0.2$, which added to the fraction of deep sites, $\theta = 0.5$, that are completely occupied, gives a total coverage of 0.7, which is the observed value of θ_c for large enough values of ΔE, in the case of $l = 1$.

For larger values of l, the saturation value can also be explained in terms of z_{eff}. From another perspective, once all the deep sites are occupied, the percolation problem becomes in finding the percolation threshold on the shallow patches, which are embedded in a set of occupied sites belonging to the deep patches. The combination of this critical value, which is expected to be smaller than the corresponding to a square lattice with periodic boundary conditions, and the fraction of deep occupied sites lead to the observed saturation value $\theta_c \approx 0.64$ for $l = 2$ and $\theta_c \approx 0.63$ for $l > 2$.

Finally, for large values of l and small values of ΔE we can see a small decrease of the curves, with respect to the homogeneous case ($\Delta E = 0$). This effect will be explained in detail in connection with Fig. 5.

Now, the results in Fig. 3 are presented in an alternative way. Thus, Fig. 4 shows the percolation threshold, θ_c, as a function of the patch size, l, for several values of ΔE, in the case of ordered distribution. In all the cases considered, θ_c decreases as l increases. The solid lines in the figure are fitting curves by using an exponential decay. The curves corresponding to higher values of ΔE produce larger values of the percolation threshold, θ_c.

Fig. 5 shows the percolation threshold, θ_c, as a function of the energy difference between the two kind of sites, ΔE, for several values of patch size, l, in the case of random distribution. For $l = 1$ the value of θ_c is almost constant regardless the value of ΔE. This can be explained as follows. Let us consider the case of large values of ΔE. If the deep sites are distributed at random, the first group of incoming particles, up to $\theta = 0.5$, occupies these sites. This is equivalent to the random distribution of such particles. Then, the rest of the particles distribute at random in the remaining sites until percolation occurs. All this is equivalent to the well known random deposition, with $\theta_c \approx 0.5927$. For small or intermediate values of ΔE, the situation is not very different, since even thought not necessarily deep sites occupy first, the distribution of the particles is completely at random. On the other hand, for values of l larger than 1, it can be observed that the curves initially descend to values lower than θ_c ($l = 1$) and then rise until they reach a saturation value, which is higher upon increasing l. The existence of a minimum in the curves for $l > 1$ can be easily understood from the analysis of Fig. 6. In the figure, the curves of θ_c vs. ΔE for $l = 2$ and $l = 4$ are plotted along with the corresponding coverage of deep and shallow sites, θ_D^c and θ_S^c, respectively. The starting point of the curves corresponds to the case of random percolation ($\Delta E = 0$, $\theta_c \approx 0.5927$), and there are not differences between deep and shallow sites, being $\theta_D^c = \theta_S^c \approx 0.5927$. Then, the coverage of the deep sites increases monotonically until the filling of the strong sites is completed. On the other hand, the coverage of the shallow sites decreases as ΔE is increased up to reaching a saturation value. This value corresponds to the minimum fraction of shallow sites which is necessary to reach the connection between the extremes of the lattice. Several conclusions can be drawn from the figure: (i) the curves of θ_D^c vs. ΔE do not depend on the patch size l, and consequently, the percolation process is governed by the shallow sites; (ii) the coverage of the shallow sites for large enough values of ΔE increases with the patch size l. This
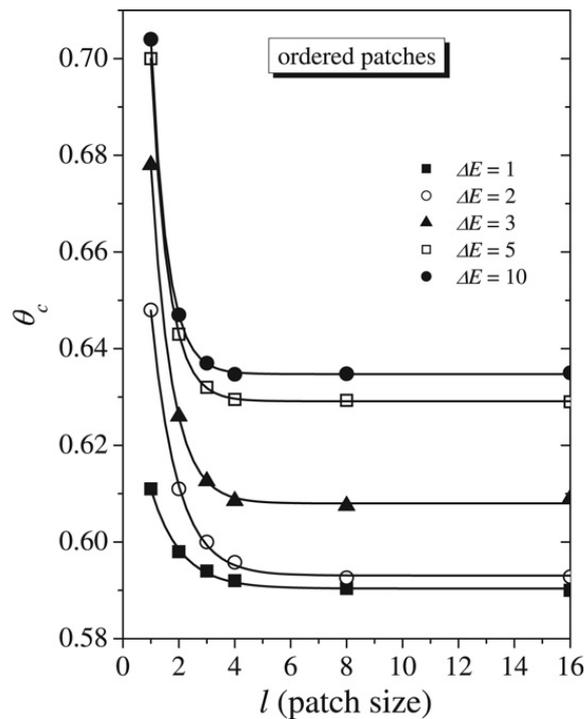
Fig. 4. Percolation threshold as a function of the patch size, *l*, for several energy differences between the two kind of sites, ΔE, in the case of ordered distribution of patches. Symbols represent Monte Carlo simulation data and lines are a guide to the eyes.
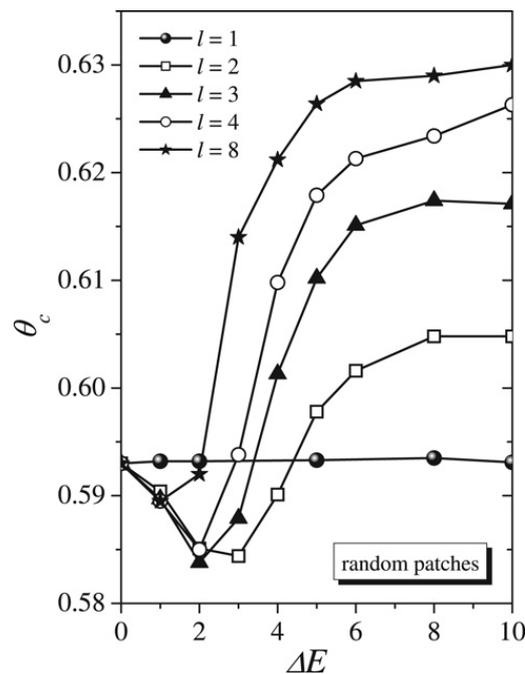
Fig. 5. Idem Fig. 3 for the case of random distribution of patches.

effect is responsible for the different saturation values observed in Fig. 5; and (iii) for a given *l* and $\Delta E \leq 4$, the slope of the curve of θ_D^c vs. ΔE is smaller than the absolute value of the slope of θ_S^c vs. ΔE, so that the total $\theta_c = \frac{1}{2}\left(\theta_D^c + \theta_S^c\right)$ coverage decreases with respect to the homogeneous case and shows a minimum.

Fig. 7 shows the percolation threshold, θ_c, as a function of the patch size, *l*, for several values of ΔE, in the case of random distribution. For $\Delta E = 1$, the percolation threshold decreases as *l* increases in agreement with the minimum shown in Fig. 5. The opposite occurs for $\Delta E = 5$ and $\Delta E = 10$.

In order to compare the two cases studied in the paper (ordered and random distribution of patches), Fig. 8 shows the percolation threshold, θ_c, as a function of the patch size, *l*, for a fixed value of $\Delta E = 10$. It can be noticed that in the ordered (random) case, θ_c goes slowly downwards (upwards) and reaches a minimum (maximum) value at $\theta_c \approx 0.63$. Both curves
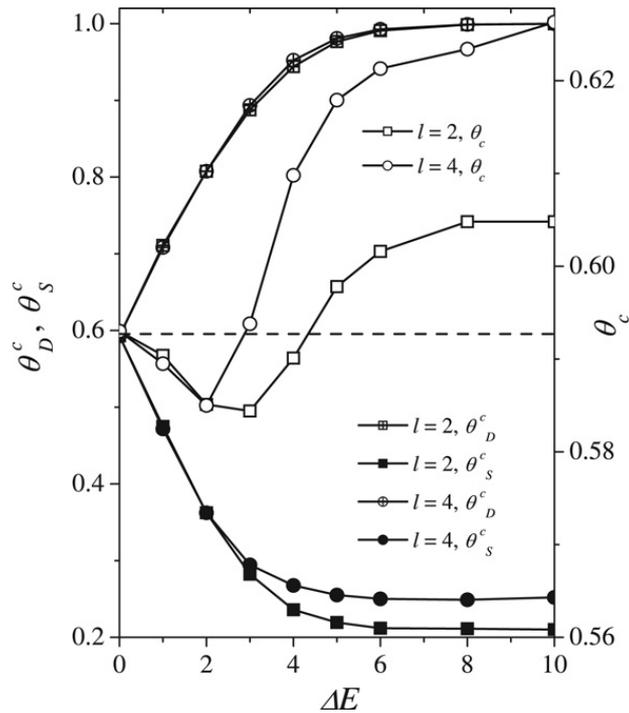
Fig. 6. Total and partial percolation thresholds as a function of ΔE for the case of random distribution of patches and two different values of patch sizes as indicated. Symbols represent Monte Carlo simulation data and lines are a guide to the eyes.
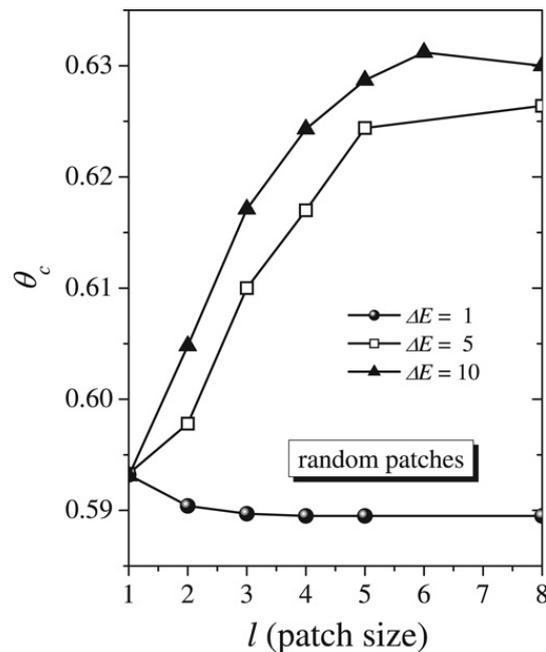
Fig. 7. Idem Fig. 4 for the case of random distribution of patches.

seem to converge to the same value for large l. Taking into account that larger connectivity of the lattice implies smaller percolation thresholds [12], these observations can be explained in terms of the effective neighbors [31]. In the case of ordered patches, the effective coordination number increases as l becomes larger, whereas the opposite occurs for random distribution [31].

4. Conclusions

In the present paper we have studied the percolation behavior of the adsorbed monolayer composed by non-interacting particles on a simple heterogeneous surface. In the framework of this model, the surface is assumed to be formed by a set of homogeneous patches. Every adsorption site within a given patch has the same adsorption energy. However, different patches have different adsorption energies. We have considered only two kinds of square patches with different energies,
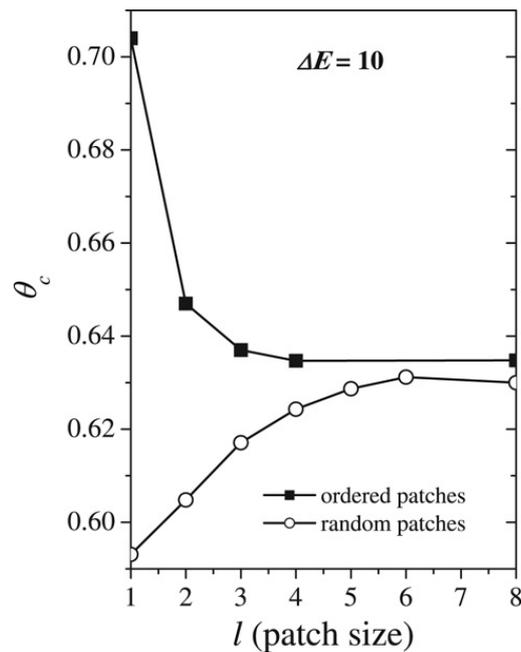
Fig. 8. Percolation threshold as a function of the patch size, l, for a fixed value of $\Delta E = 10$, in the case of ordered (squares) and random distribution (circles) of patches. Symbols represent Monte Carlo simulation data and lines are a guide to the eyes.

i.e. deep and shallow patches, which are (i) arranged in a chessboard-like ordered structure or (ii) randomly located. In detail, on the basis of this so-called bivariate trap model, simple energetic topographies are created by placing square patches of $l \times l$ deep or shallow adsorption sites onto the lattice, which is represented by a two-dimensional array of $L \times L$ sites with periodic boundary conditions. For simplicity we assume that the number of deep adsorption sites is equal to the number of shallow adsorption sites, and we do not allow overlap of different patches.

We have used the Monte Carlo technique in order to determine the equilibrium conditions of the lattice once l, L, T an θ are fixed. The thermodynamic equilibrium is reached in the canonical ensemble by using a standard Kawasaki algorithm [32]. Once the equilibrium is established, then we consider each independent state in order to analyze the percolation transition. The numerical scheme used for that is based on the Hoshen and Kopelman algorithm [36]. Finally, the determination of $\theta_c(l, \Delta E)$ is done by using extensively the finite-size scaling analysis.

We conclude this work remarking the distinctive qualities of the site percolation problem on simple patchwise lattices. $\theta_c(\Delta E)$ for fixed values of l slowly grows up to a saturation value as the energy difference between shallow and deep patches increases for ordered distribution of patches. In the case of random distribution, the same limiting behavior is observed for large values of ΔE but the curves pass through a minimum for $l > 1$. A singular behavior is observed for $l = 1$ where $\theta_c = 0.5927$. $\theta_c(l)$ for fixed values of ΔE presents a quite different behavior depending the spatial distribution of patches. In fact, for ordered structures $\theta_c(l)$ exponentially decreases upon increasing l. For larger values of ΔE the decay is more pronounced. In contrast, when the patches are randomly distributed a slow decay is shown for weak values of ΔE. However, for larger values of ΔE the curves raise up to a saturation value. It is important to remark that for fixed values of ΔE and in the limit $l \to \infty$ the percolation threshold becomes independent of the spatial distribution of energetic surface topography.

Finally, we would like to emphasize that the studied problem (percolation of the adsorbed monolayer at equilibrium) has a clear link with the random deposition of monomers on the same surface; we consider two different deposition probabilities depending on whether the particle is dropped on deep or shallow sites. This point is clarified in the Appendix. The present work encourages the analysis of the competitive effect of the ad–ad interactions with the surface heterogeneities on the percolative behavior of the system. Such a study is in progress.

Acknowledgements

Appendix

In this article we have studied the percolation behavior of adsorbed particles at equilibrium. On the other hand, in Ref. [31] the authors have performed a similar study but by considering the irreversible deposition of particles on the same

heterogeneous lattice. In that version of the problem, two occupation probabilities, p and q, for a site belonging to the shallow and deep sites, respectively, have been defined. The state of the system is specified by these two independent variables p and q. If the network is very large and p and q are sufficiently small, the size of any cluster is likely to be small. But if p and q are close to unity the network should be entirely connected and an infinite cluster of occupied sites spans the network. In this Appendix we shall show that there exists a mapping between both versions of the problem.

In order to explain such mapping, it is interesting to know how the particles are distributed over the deep and shallow patches. Thus, we define the site specific surface coverage, θ_D and θ_S, as the fraction of occupied deep and shallow trap sites, respectively. Such partial specific concentrations, θ_D and θ_S, can be also interpreted as the probabilities p and q for each kind of sites. Thus, it can be established that:

$$\theta_S = \frac{e^{\beta\mu}}{1 + e^{\beta\mu}} \tag{9}$$

$$\theta_D = \frac{e^{\beta(\mu+\Delta E)}}{1 + e^{\beta(\mu+\Delta E)}} \tag{10}$$

$$\theta = \frac{\theta_S + \theta_D}{2}. \tag{11}$$

From Eqs. (9) and (10), and by using $\beta = 1$, we can easily obtain ΔE as:

$$\Delta E = \ln\left[\frac{\theta_D(1 - \theta_S)}{\theta_S(1 - \theta_D)}\right]. \tag{12}$$

Thus, we identify θ_S and θ_D with the values of p and q of Ref. [31]. From the above equations, both partial surface concentrations can be obtained from knowing θ and ΔE. Therefore, p and q (or θ_S and θ_D) are the corresponding probabilities for each kind of site, associated to each pair $(\Delta E, \theta)$ of the present paper.

References

[1] J.M. Hammersley, Proc. Cambridge Phil. Soc. 53 (1957) 642.
[2] D. Stauffer, A. Aharony, Introduction to Percolation Theory, 2nd ed., Taylor & Francis, London, 1992.
[3] M. Sahimi, Application of percolation theory, Taylor & Francis, London, 1992.
[4] R. Zallen, The Physics of Amorphous Solids, John Willey & Sons, NY, 1983.
[5] J.W. Essam, Rep. Progr. Phys. 43 (1980) 833.
[6] K. Binder, Rep. Progr. Phys. 60 (1997) 488.
[7] C. Lorenz, R. May, R. Ziff, J. Stat. Phys. 98 (2000) 961.
[8] C.M. Fortuin, P.W. Kasteleyn, Physica 57 (1972) 536.
[9] Z. Gao, Z.R. Yang, Physica A 255 (1998) 242.
[10] K.Y. Gao, B.X. Liu, J. Phys.: Condens. Matter 11 (1999) 39.
[11] M.C. Giménez, F. Nieto, A.J. Ramirez-Pastor, J. Phys. A 38 (2005) 3253.
[12] M.C. Giménez, F. Nieto, A.J. Ramirez-Pastor, J. Chem. Phys. 125 (2006) 184707.
[13] R. Gomer, Rep. Progr. Phys. 53 (1990) 917.
[14] G. Zgrablich, in: W.W. Rudzinski, G. Zgrablich (Eds.), Equilibria and dynamics of gas adsorption on heterogeneous solid surfaces, Elsevier, Amsterdam, 1996.
[15] M. Tringides, R. Gomer, Surface Sci. 145 (1984) 121.
[16] M. Tringides, R. Gomer, Surface Sci. 155 (1985) 254.
[17] A. Sadiq, K. Binder, Surface Sci. 128 (1983) 350.
[18] V.P. Zhdanov, Surf. Sci. Lett. 149 (1985) L13.
[19] C. Uebing, R. Gomer, Surf. Sci. 331-333 (1995) 930.
[20] F. Nieto, C. Uebing, Ber. Bunsenges. Phys. Chem. 102 (1998) 156.
[21] F. Nieto, C. Uebing, V.D Pereyra, R. Faccio, Surf. Sci. 423 (1999) 256.
[22] F. Bulnes, F. Nieto, V. Pereyra, G. Zgrablich, C. Uebing, Langmuir 15 (1999) 5990.
[23] M. Ciacera, C. Zuppa, G. Zgrablich, J.L. Riccardo, W.A. Steele, Surf. Sci. 356 (1996) 257.
[24] F. Nieto, C. Uebing, Vacuum 54 (1999) 119.
[25] I. Morgenstern, K. Binder, R.M. Hornreich, Phys. Rev. B 23 (1981) 287.
[26] K. Binder, Phys. Rev. B 29 (1984) 5184.
[27] M. Aizenman, J. Wehr, Phys. Rev. Lett. 62 (1989) 2503.
[28] S. Prager, H.L. Frisch, J. Chem. Phys. 72 (1980) 2941.
[29] A. Patrykiejew, Thin Solid Films 223 (1993) 39.
[30] C. Uebing, V. Pereyra, G. Zgrablich, Surf. Sci. 366 (1996) 185.
[31] F. Nieto, A.J. Ramirez Cuesta, R.J. Faccio, Phys. Rev. E 59 (1999) 3706.
[32] K. Kawasaki, in: C. Domb, M. Green (Eds.), Phase Transitions and Critical Phenomena, vol. 2, Academic, London, 1972.
[33] N. Metropolis, A.W Rosenbluth, M.N. Rosenbluth, A.H. Teller, E. Teller, J. Chem. Phys. 21 (1953) 1087.
[34] F. Yonezawa, S. Sakamoto, M. Hori, Phys. Rev. B 40 (1989) 636.
[35] F. Yonezawa, S. Sakamoto, M. Hori, Phys. Rev. B 40 (1989) 650.
[36] J. Hoshen, R. Kopelman, Phys. Rev. B 14 (1976) 3428.