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Percolation of binary mixtures adsorbed on square lattices

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HIGHLIGHTS

- Percolation properties of equilibrium adsorbed phases.
- Interacting binary mixtures adsorbed on square lattices.
- A rich variety of structural orderings were observed in the adlayer.
- Percolation thresholds and percolation phase diagrams were obtained.

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ABSTRACT

In this paper, the adsorption of interacting binary mixtures on square lattices has been studied. By using Monte Carlo simulation and finite-size scaling analysis, the connection between the surface ordered phases and the percolating properties of the adsorbed phase has been investigated. A rich phase diagram separating a percolating from a non-percolating region has been determined. The main features of the phase diagram have been discussed in terms of simple considerations related to the interactions present in the problem.

1. Introduction

Percolation theory [1] is a powerful tool for modeling diversity of phenomena, such as: fire propagation, fire spreading in multi-compartmented structures, spreading of computer viruses, network failures, gel formation, infectious disease epidemics, granular mixture and medical research. Due to its wide applicability [2], this theory has attracted the attention of different researchers, mathematicians, physicists, programmers, engineers, physicians, etc.

The percolation theory was derived from studying the random action of fluid-like materials as they migrate through a latticework of channels. The distribution of these channels determines the probability that they will become linked together in a *great network* that allows the flow from one end of the lattice to the other. Broadbent and Hammersley [3] gave the first mathematical formulation which was able to relate the emergence of the *great network* with the minimum concentration of channels (called by the authors *percolation threshold*) by a simplified *lattice percolation model*.

Most of the studies of percolation have taken into account that the state of sites on lattice changes irreversibly from empty to filled (occupied). This scheme of filling is known as Random Sequential Adsorption (*RSA*) model [4,5]. In the framework of

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Nomenclature

L	Lattice side
M	Total number of lattice sites
T	Temperature
w_{xy} $(x, y = A, B)$	Nearest-neighbor interaction energy
H	Hamiltonian of the system
c_i	Occupation number of the site i ($c_i = 0$ if empty; $c_i = 1$ if occupied by A and $c_i = 2$ if occupied by B)
P	Metropolis transition probability
$\Delta H = H_f - H_i$	Difference between the Hamiltonians of the final state and initial states
k_B	Boltzmann constant
MCS	Monte Carlo step
m	Number of MCS to calculate the averages of the adsorption quantities
r	Number of samples to calculate the averages of the percolation quantities
$N_{A(B)}$	Number of adsorbed A(B) particles
N	Total number of adsorbed particles
$R_L^{R(D)}$	Probability of finding a rightward (downward) percolating cluster, formed by A and B particles
$Q_L^{R(D)}$	Probability of finding a rightward (downward) percolating cluster, formed by A particles
R_L^I	Probability of finding a cluster, formed by A and B particles, which percolates both in a rightward **and** in a downward direction
Q_L^I	Probability of finding a cluster, formed by A particles, which percolates both in a rightward **and** in a downward direction
R_L^U	Probability of finding either a rightward **or** a downward percolating cluster, formed by A and B particles
Q_L^U	Probability of finding either a rightward **or** a downward percolating cluster, formed by A particles
R_L^A	$\equiv \frac{1}{2}\left[R_L^I + R_L^U\right]$
Q_L^A	$\equiv \frac{1}{2}\left[Q_L^I + Q_L^U\right]$

Greek symbols

$\mu_{A(B)}$	Chemical potential of A(B) particles
δ's	Kronecker delta functions
ϵ_0	Interaction energy between a monomer (type A or B) and a lattice site
$\theta_{A(B)}$	Partial surface coverage corresponding to the A(B) species
θ	Total surface coverage
θ_c	Percolation threshold

adsorption, the *RSA* has been used to modeling the electro-oxidation of ethanol, characterizing the coverage and percolation properties [6], or to study deposition of proteins and colloids from solution onto solid surfaces [7].

Outside the framework of lattice and *RSA* model, the percolation concepts have been applied to study systems composed by a binary mixture of patchy colloidal particles where each species has three coupling of two types, one of which promotes bonding of particles of the same species while the other promotes bonding of different species. Then, depending on the values of the parameters of the system four distinct percolating phases can be found: two gels where only one of the species is percolated, a mixed gel where the two species are percolated but neither species percolates by itself, and a bicontinuous gel where the two species percolate independently, forming two interpenetrating spanning networks [8]. Another interesting instance, in which the percolation concepts are applied is for analyzing a mixture of liquids, Dougan et al. [9] has shown the formation of clustering structure of both species for a mixture of methanol and water as a function of concentration.

Returning to *RSA* model, the temperature does not play any relevant role and it is not considered. This model is appropriate for many physical, chemical, and biological processes where the microscopic steps are irreversible, and where equilibration is not possible on the time scale of the experiment [4]. However, in numerous systems of both theoretical and practice importance, where the adsorbed particles are in thermodynamic equilibrium, the spatial distribution of the adsorbate might be characterized by using the percolation model [10,11]. In these cases, the temperature governs the phase in the system and can be an important controlling factor in the percolation process. In this context, Giménez et al. [12,13] introduced a model in which they studied the percolation properties of the adsorbed phase of interacting monomers on a square lattice.

In this paper, the percolation behavior of an adsorbed binary mixture has been investigated by using Monte Carlo (MC) simulation and finite-size scaling analysis. For this purpose, a square substrate is exposed to an ideal A–B mixed-gas phase, at temperature T and chemical potentials μ_A and μ_B. Then, the main percolation properties of the adsorbed monolayer are obtained regarding different repulsive interactions between the adparticles.

The adsorption thermodynamics of the present model was studied in a recent paper by our group [14]. The calculations were carried out by combining theoretical modeling and MC simulations in grand canonical ensemble. Two theoretical