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# Controlling the Percolation Threshold of Conductor-Insulator Composites in a 2D Triangular Lattice by Introducing Binary Size Distributions of Conductor Particles

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When one attempts to modulate and control the characteristics of composite materials, the mathematical threshold of the percolation transition dictates the modulation limit. Using a series of computer simulations, we have been investigating the dependence of the percolation threshold on particle size distributions. However, one of the other factors that can deeply affect percolation behavior, the design of lattice, is always fixed to simple square or cubic lattices. This report presents the first calculation of a percolation threshold in a 2D triangular lattice with binary size distributions of conductor particles. Although a small quantitative difference was found, the results qualitatively matched to the results already reported for 2D square lattices, thus confirming our previous finding: the introduction of large conductor particles increases the percolation threshold in 2D. [doi:10.2320/matertrans.M2012029]

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## 1. Introduction

The percolation behavior[1] of composite materials has been explained accurately by a simple theoretical model, such as randomly occupied sites (or bonds) on a lattice in various lattice types. The sudden appearance of a large connected cluster at a certain threshold of the occupancy ratio, denoted as $p$, of lattice sites clearly coincides with the transition in macroscopic characteristics of the materials. This success, in turn, means that the limit of composite materials imposed by the percolation transition is a rather mathematical one, rooted in the randomness of material structures and difficult to overcome within the conventional assumptions used in material sciences. The percolation threshold, denoted as $p_c$, can be a strict limit on the mixture ratio of constituent materials, because sometimes, ensuring the connectedness of one of several constituent materials is fundamental to ensuring the desired performance of the material, such as electronic conductance.

It would be possible to systematically move the limit by stepping outside of the simple percolation model, where the configuration of the particles in a composite material is not completely randomized but to some degree correlated. To realize such a limited-ordered configuration of composite materials in reality, researchers have considered the use of particles modulated in their size distributions,[2–4] with shape differences,[5,6] and with controlled affinity between particles.[6] The core–shell particle designs[7–9] and direct control of the particle deposition process (if possible), are also promising approaches to realize correlated nanostructures and controlled percolation. As for cases of continuum percolation models, a simulation study showed[10] that even simpler size distributions (Gaussian, uniform, and log-normal distributions of particles, regardless of their conductivity) shows a clearly nontrivial effect. Even in simpler models,[11,12] for example, conductor sites arranged in length $X$ bars aligned vertically or horizontally in square lattice,[13] can exhibit quite complex effects and there actually is an optimum value of $X$, beyond which the effect on $p_c$ is inversed.

In particular, we have been investigating[14,15] the effects of the most basic type of correlations realizable simply by using particles with differentiated sizes on the percolation behavior of the system. Although this type of correlation seems to be quite useful for increasing connectedness among particles, it is important to investigate the effects of particle size differentiations in combination with other factors that can affect percolation characteristics such as the type of lattice used in the simulation.

To this end, here we extend the same basic binary size distribution model we investigated before on the square lattice model to a 2D triangular lattice model. This is interesting because, as we pointed out in one of our previous papers, the direct cause of the shift in the $p_c$ can be the change in the effective coordination number of the lattice (or the number of the neighboring sites).[8] Since the triangular lattice has a clearly different and increased coordination number (6) than the simple square lattice (4), the system may exhibit a different reaction to the same type of correlation. Conveniently, the $p_c$ of site-percolation on a 2D triangular lattice (0.5[16–18]) is rather close to that of the 2D square lattice (0.59[19]), while there is a drastic difference between these lattices in the $p_c$ of the bond percolation (0.5 and 0.35,[20] respectively). One of the minor and additional advantages of using a triangular lattice is that it can be a little closer to reality: relatively large particles on a triangular lattice are modeled by hexagons of various sizes; this is closer to a typical experimental situation involving spherical particles. Finally, in this report we introduce a further simplification: we confine ourselves to cases with large conductor particles.

## 2. Methods

In this calculation, we basically generate and evaluate a large number of ways to place numerous hexagons, some of which are bigger than normal and occupy $X > 1$ sites, onto a triangular lattice. Random particle configurations are generated as follows. First, non-overlapping *hexagons*

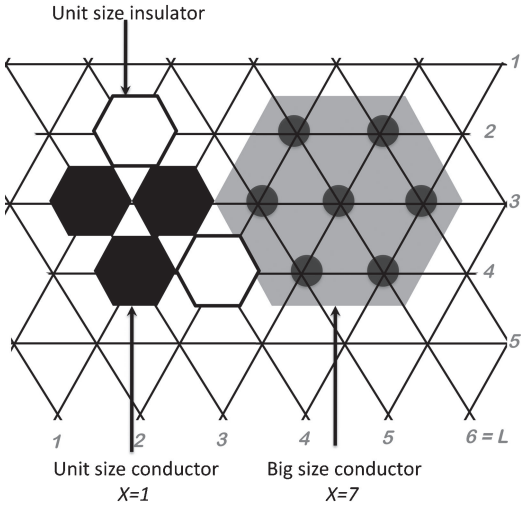


Fig. 1 In this work, a unit size insulator particle, a unit size conductor particle, and a conductor particles with an increased size shown in this figure are considered on a triangular lattice.

of the required number and size are randomly placed on the triangular lattice by means of a simple trial-and-error algorithm to decide the positions of large conductor particles. Then lattice sites not covered by these large particles are randomly classified into the conductor sites and insulator sites with a given probability. This simple scheme apparently samples all possible configurations with a given combination of the number and type of particles. About 20000 different random particle configurations are generated and analyzed for each condition by means of a standard cluster analysis method. For each condition introduced below, lattice sizes $L = 32$, 64, 128, 256 and 512, are simulated. Details on the lattice and particle design (as well as detailed definitions of $X$ and $L$) are given in Fig. 1. Although this percolation problem in a triangular lattice is algorithmically equivalent to that in a square lattice, we developed a new simulation code in C++, mainly because the excluded volume effect between hexagons is easier to program in C++.

In our previous works,[14,15] the number of enlarged particles is decided in such a way that 50% of the conductor sites are located in the enlarged particles. Here, this restriction is removed, and allowing us to calculate 9 different settings regarding the fractions of large conductor particles (see Fig. 1). In these conditions, 0 to 80% of conductor sites belong to large conductor particles. The volume fraction of the large particle over all conducting particles (not over all particles) is denoted as $q$. Therefore, $q = 0.0$–$0.8$, and when $q = 0.5$, the particle number ratios between enlarged conductor, unit size conductor, and unit size insulator are $p/14 : p/2 : 1 - p$. Whereas the sampling interval of $q$ is 0.1, the typical interval used for $p$ is 0.01. Note that all lattice sites are defined as either insulator or conductor sites ("void" is not considered, because in this simulation, a void is equivalent to insulator sites in all respects).

The output of these calculations is the mean finite cluster size, $S(L,p)$, defined as a summation taken for all clusters except the largest one,

$$S(L,p) = \sum_s s^2 n_s(p), \tag{1}$$

where $n_s(p)$ is the average number of clusters with size $s$. Roughly speaking, the peak of the finite cluster size coincides with the critical point.

However, due to the effect of the finite size of the simulation lattice, the observed critical densities depend on lattice size, therefore does not indicate the true critical density. The critical densities for infinite lattices are estimated by the standard method introduced in the following,[1] called finite size scaling (FSS). The correlation length of the system, which is basically the characteristic distance between two sites belonging to the same cluster of percolating sites, $\xi$, diverges near the critical point as

$$\xi \propto |p - p_c|^{-\nu}. \tag{2}$$

A scaling function for cluster size, $f(z)$, is introduced,

$$f(z) = L^{-\gamma/\nu} S(L,p). \tag{3}$$

In these equations, $\gamma$ and $\nu$ are values called *critical exponents*[21] that are, along with other critical exponents, describes how the corresponding physical values of the systems diverge at their critical point. The values of these exponents are known to be unchanged over very different systems if the dimensionalities of the systems are unchanged. Here, it is assumed that, at the critical point, this function is dependent only on the ratio of the current system size to the correlation length of the system. Therefore, it is natural to define $z = L/\xi = L/|p - p_c|^{-\nu}$. Naturally, the value of $z$ is constant at any critical point observed in systems having any $L$. Because of this constant nature of $z$, the critical points exhibited by $S(L,p)$ in various $L$ are denoted as $p_c(L)$, as in

$$|p_c(L) - p_c| \propto L^{-1/\nu}. \tag{4}$$

This formula tells that there is a value of $-1/\nu$ that can make the $L^{-1/\nu}$ versus $p_c(L)$ plot into a linear line, and the point where the line intersects the $y$-axis ($y = 0$, that is, $L \to \infty$) gives the true critical density in the ideally large system.

## 3. Results and Discussion

In Fig. 2, the location of (system size dependent) critical density is indicated by the peak of the plot, which shows the mean size of finite connected clusters in the lattice ($y$-axis) as a function of the density of conducting sites ($x$-axis). Each condition shows three peaks with different heights and grades, corresponding to different lattice sizes ($L = 512$ shows the highest and steepest plots). The basic correctness of the simulation is demonstrated by the location of the peak for $q = 0$, which coincides with the reported transition point in the conventional site percolation model (0.5)[20] in a 2D triangular lattice. The other peaks clearly show that, also in the case of a 2D triangular lattice, the enlarged conductor particle shows a weak but clear effect of increasing $p_c$. This is confirmed by the value of "true" $p_c$ estimated by a successful FSS analysis with $\nu = 4/3$ (the same as the value for normal 2D percolation[1]) also shown in Fig. 3. When

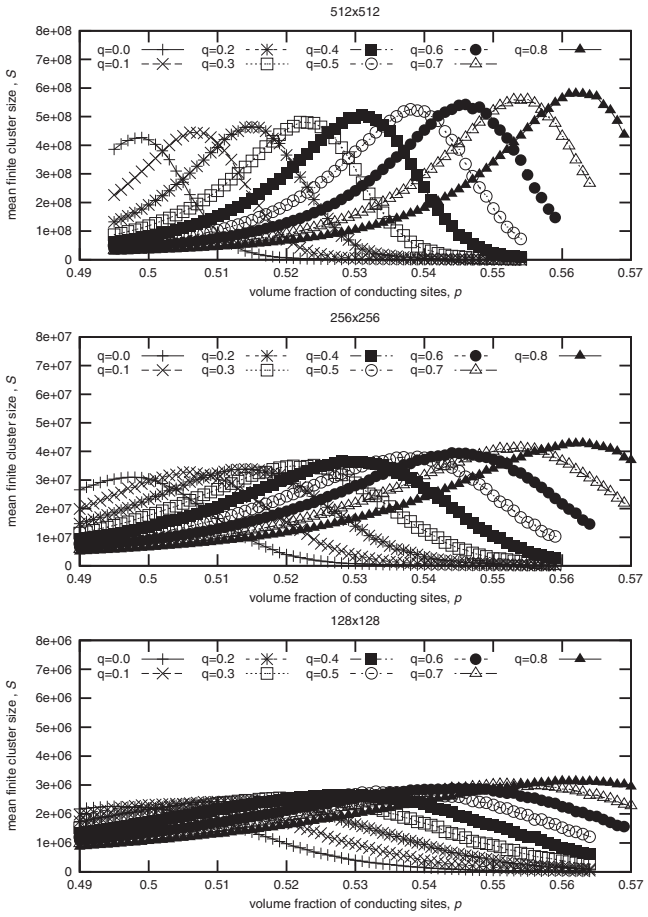


Fig. 2 The mean size of finite connected clusters as a function of the volume fraction of conducting sites on a 2D lattice, plotted for $L = 128$, 256 and 512. The leftward shift of the plots clearly indicates the $p_c$-increasing effect of enlarged insulator particles.

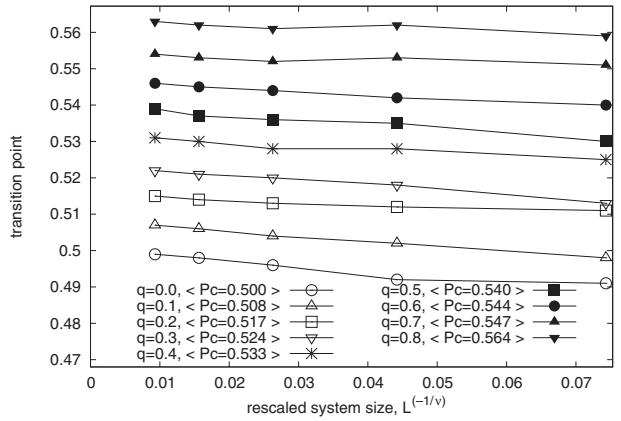


Fig. 3 Finite-size scaling analysis of $p_c$. The values of $p_c$ extrapolated to the infinite system for various values of $q$ are tabulated in the legend.

half of the conductor sites are in the enlarged particles, an approximately 8% ($0.5 \rightarrow 0.540$) increase in $p_c$ was observed. Interestingly, this is a much bigger change than that observed in the corresponding situation in a previous report on a square lattice.[5,22] The strongest effect on $p_c$ is observed for $q = 0.8$, where $p_c$ is increased by 12.8% ($0.5 \rightarrow 0.564$).

## 4. Discussion

Although the values of the critical exponents are strongly bound to the dimensionality of the space, the lattice strongly affects the detailed behavior of percolation phenomena. This point should not be denied when an application of computer simulation to real materials is considered. In that sense, the result introduced above is important because it proves that the increase in $p_c$ observed in the case of a 2D simple lattice is not an artifact but clearly a direct result of the poly-dispersity of the conductor particles in the system. It is also important to notice that the hexagonal particle in a triangular lattice is more realistic than a cubic particle in a simple cubic lattice. Real particles tend to be spherical, and spherical particles tend to take hexagonal configurations when they are closely packed. Note that this is more true in cases of enlarged particles; unlike hexagonal enlarged particles, square enlarged particles do not have to be packed in hexagonal configurations.

Our previous results have shown that the percolation behavior with particle size distributions may be very different in 3D cases. But the superiority of the hexagonal particle or lattice in modeling realistic particles is generally not changed in 3D cases, although 2D hexagonal packing will be replaced by 3D hexagonal close packing (hcp). Considering these points, we must extend the result of the current paper into 3D cases, presumably using hcp lattices. A continuous model with impermeable spherical particles (and, preferably, with continuous diameter distributions) will be a very interesting subject to investigate by means of large-scale computer simulations. The main difficulty in realizing this is the increased complexity of packing; realizing dense random packing in computer simulations is a much more difficult and complex problem than is generally understood.[23]

## 5. Conclusion

In this report, a series of the computer simulations on site percolation on 2D triangular lattice with particle size distribution is presented. The type of particle size distribution is limited to a binary distribution of conducting particles. The results of the simulations are in accordance with our previous simulation work about similar conditions on 2D square lattice, although in triangular lattice, the observed effect of the particle size distribution is stronger: a 12.8% increase of critical density is observed when 80% of conducting sites are belong to bigger particles. Considering that triangular lattices are more suitable than square lattices to model the conducting films in reality, the observed enhancement in the change of critical density is quite interesting.

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