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The percolation of fibres with random orientations: a Monte Carlo study

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Abstract. Monte Carlo numerical simulations of the percolation of sticks with random orientations on a cubic lattice are reported. Finite size scaling and the position space renormalisation group are used. As the length of the sticks is increased it is found that the critical probability decreases whereas the correlation length exponent remains, within experimental errors, the same as in classical 3D percolation. Comparison with previous experimental and numerical results on related systems leads to the emphasis of the importance of the excluded-volume condition.

1. Introduction

The percolation problem has been much studied on theoretical, numerical and experimental grounds (for extensive reviews see Stauffer 1979, Essam 1980, Adler *et al* 1982). Various kinds of problems can be defined according to the connection laws and occupation rules for the basic elements (sites or bonds); in the usual random percolation these elements are each occupied with a probability p independent of the other elements; in 'correlated percolation' this probability depends on the occupancy of the surrounding elements and in 'anisotropic percolation' on the spatial orientation of the bonds. Moreover, a distinction must be made between 'discrete percolation' in which the basic elements are placed on a lattice and 'continuous percolation' in which they are allowed to take any position compatible with the considered problem. Among the most investigated topics are the determination of the percolation threshold p_c and the critical exponents, especially the 'correlation length exponent' ν , and, when a conductance is associated with the bonds, the 'conductivity exponent' t .

These theories have also been shown to be relevant for describing the conductivity of binary mixtures of conducting and insulating materials (Kirkpatrick 1979, Deutscher 1981). The usual random percolation applies to heterogeneous materials constituted with spherical, monodisperse, isotropic conducting particles, either randomly distributed in a continuous insulating matrix (Fug *et al* 1978) or mixed with identical insulating particles (Ottavi *et al* 1978).

Some interest has been focused recently on the conductivity of random media in which the conducting particles are non-spherical and/or have anisotropic conductivity. We can quote a theoretical study (Shklovskii 1978) and model experiments (Smith and Lobb 1979, Blanc *et al* 1980). All these studies are limited to 2D systems and consider only the case of oriented symmetry or principal axis of the particles. At least over a domain of concentration in conducting material, such a medium must exhibit

an anisotropic macroscopic conductivity. Anisotropic percolation (Redner and Stanley 1979) has been shown (Blanc *et al* 1980, Lobb *et al* 1981) to apply to such situations.

The large development of fibrous composite materials arouses interest about the percolative properties of random assemblies of elongated elements characterised by large aspect ratios (the ratio of the length to the transversal size). Carmona *et al* (1980, 1981) have studied experimentally the electrical properties of short conducting carbon fibres embedded in an insulating resin and have proposed some theoretical interpretations of the fibre length dependence of the conductivity threshold. The purpose of the present work is to perform a Monte Carlo simulation of a 3D random dispersion of elongated elements with various aspect ratios and to compare the results on the one hand with the former experimental and theoretical results and on the other hand with the different 2D simulation approach of Pike and Seager (1974).

Two different problems are to be distinguished.

(1) The sticks or rods are dispersed with random position and orientation. The medium is always macroscopically isotropic and the problem is only a matter of a certain type of 'correlated percolation', due to both the shape of the sticks and the excluded-volume effects.

(2) The sticks or rods are partially or completely oriented parallel to a fixed direction or a fixed plane. The medium becomes anisotropic on a macroscopic scale and we are then facing an 'anisotropic percolation' in addition to other possible correlations.

This paper is only concerned with the first of these problems. In § 2 we define the model and describe briefly the methods that are used. In § 3, we give the results of the simulation and discuss their validity. In § 4, these results are compared with experiments and previous theory, and we then propose some explanation of the observed discrepancies and outline future possible developments.

2. Simulation model and methods

(a) Representation

We use a 3D simple cubic lattice of sites with lattice spacing unity. Each conducting fibre is represented by a group of n adjacent occupied sites, lined up along one of the three principal axes of the lattice. Two different fibres are not allowed to have a common site, so that steric exclusion effects are introduced. The conduction rule is the same as for usual random percolation: two sites are said to be connected if they are nearest neighbours (each site has six nearest neighbours in the cubic lattice). If one imagines that each elementary cube of side unity centred on an occupied site is filled with conducting material, the fibre is clearly a parallelepipedic conducting element of length n , with transversal dimensions unity, so that the aspect ratio is n . Conduction occurs between two different sticks when they touch along a non-zero surface. Figure 1 gives a 2D example of the representation in the case $n = 4$: the current goes between 1, 2 and 3 but not between 1 and 4. The density of the medium is equal to the proportion of occupied sites p .

(b) Sample generation

For each sample, cubic shaped with linear size b , the sticks are introduced at random, starting with an empty lattice and giving an equal probability to all positions still

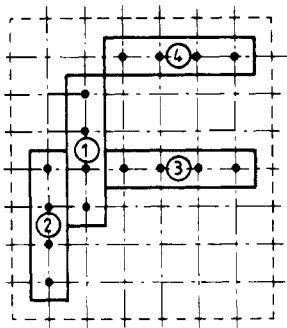


Figure 1. Two-dimensional scheme of the generation of sticks in the case $n = 4$: — — — —, the square lattice; ●, the occupied sites; —, the limits of the simulated sticks. According to the connection criterion (see text) stick 1 is connected to sticks 2 and 3 and not connected to stick 4.

available, that is compatible with the steric exclusion rule. Thus a new stick is rejected if at least one of the sites belonging to it would overlap a site already occupied. In order to avoid wall effects in this correlated problem, periodic boundary conditions are used. Several rules equivalent in the limit of infinite samples can be chosen to define the infinite cluster (Reynolds *et al* 1980). We choose the following one: the sample is said to percolate if it is spanned by a conducting path in a given direction (say 'vertical') and, because of the periodic boundary conditions, if the final occupied site is identical to the initial one. Note that, due to the spatial correlations and exclusion-volume effects, the complete sample must be kept in memory. A process like the one used by Hoshen and Kopelman (1976) cannot be used to reduce the computational requirements.

For each sample, the building process is stopped when percolation first occurs and the corresponding density is registered. In practice this point is not tested for each added stick but with a predetermined precision by means of a bipartition process. Since the sample is finite, this value is not exactly equal to p_c , but distributed according to a probability law $L(b, p) dp$ to find the percolation point between p and $p + dp$. $L(b, p)$ exhibits a maximum for $p = p_{max}$ which sharpens and approaches p_c for very large samples (Reynolds *et al* 1980).

By generating many different samples, in practice 2000 to 15 000, of a given linear size b , one gets an experimental distribution $L(b, p)$. The critical behaviour of the infinite lattice is determined by convenient extrapolation of the results obtained on samples of increasing size according to the following methods.

(c) *Extrapolation procedure*

The two methods employed for determining p_c and the correlation length exponents ν are finite size scaling (FSS) and position space renormalisation group (PSRG).

FSS: When b is not too small, $L(b, p)$ can be approximated, at least in the vicinity of the maximum, by a Gaussian and the mean value $\langle p \rangle$ by p_{max} . Then, following the scaling theory of Levinshtein *et al* (1975), $\langle p \rangle$ is expected to approach p_c according to the relation

$$|\langle p \rangle - p_c| \sim b^{-1/\nu}. \tag{1}$$

The same proportionality law holds for the width w of the distribution:

$$w \sim b^{-1/\nu}.$$

For a Gaussian it becomes

$$L_{\max}^G \sim b^{1/\nu} \quad (2)$$

where L_{\max}^G is the maximum of the distribution.

PSRG: On the other hand, the probability for a cell of size b and density p to involve an infinite cluster is given by

$$R(b, p) = \int_0^p L(b, \tilde{p}) d\tilde{p}.$$

Reynolds *et al* (1978) and Reynolds *et al* (1980) have shown that the recursion relation $p' = R(b, p)$ defines a renormalisation group transformation which has a fixed point p^* , defined by $p^* = R(b, p^*)$. p^* depends on b and approaches p_c when $b \rightarrow \infty$ according to the relation

$$|p^*(b) - p_c| \sim b^{-1/\nu}. \quad (3)$$

$p^*(b)$ may be calculated exactly in the case of very small values of b and numerically otherwise. Moreover, the eigenvalue of the transformation is given by the slope λ_b of $R(b, p)$ at $p = p^*$

$$\lambda_b = (dR(b, p)/dp)_{p=p^*} = L(b, p^*).$$

On the basis of standard renormalisation arguments it can be inferred that

$$y_b = \log \lambda_b / \log b \rightarrow y = 1/\nu \quad \text{when } b \rightarrow \infty \quad (\text{or } 1/\log b \rightarrow 0). \quad (4)$$

In recent articles, Tsallis (1982) and Family and Reynolds (1982) have shown, in particular cases, that a linear dependence of the form $y_b \sim 1/\log b$ is asymptotically exact in the large b limit.

Both relations (1) (FSS) and (3) (PSRG) can be used to extrapolate the results to infinite samples. If b is large enough to approach the asymptotic regime a plot of $b^{-1/\nu}$ against $\langle p \rangle$ (resp p^*) is a straight line, the intercept of which with the $\langle p \rangle$ axis (resp p^* axis) being p_c . Due to uncertainties ν cannot be determined in this way with good accuracy; but an accurate determination of p_c can be achieved by using an estimated value of ν . In two dimensions, Reynolds *et al* (1980) have shown that the resulting p_c value is not very sensitive to small errors in ν .

ν can be better determined by a similar extrapolation of y_b according to (4). In two dimensions an accurate determination of ν has been done in such a way by Eschbach *et al* (1981) who showed that the result depends slightly on the extrapolation procedure and that the residual curvature cannot be ignored. In three dimensions, the more accurate value of ν has been obtained by Heermann and Stauffer (1981) using FSS.

In order to obtain good determinations of the p_c and evaluations of ν we use simultaneously FSS and PSRG. We describe here the procedure in the case of the usual random site percolation on a cubic lattice ($n = 1$). The same one is used for $n > 1$, but will not be repeated in the sequel. The experimental distribution $L(b, p)$, for b sizes ranging between 4 and 75, was determined on a Digital VAX 11/780 computer as described above, with the number of trials going from 10 000 for the smallest

samples to 2000 for the largest ones. In order to save time and increase precision close to the maximum, the search was limited to the central peak of $L(b, p)$, ignoring the wings, and the former was fitted to a Gaussian (figure 2(a)). Grouping several points smoothes the fluctuations in the histogram but does not give significant differences for the p_{\max} values. The best compromise has been retained finally for p_{\max} . The experimental histogram $L(b, p)$ is integrated in order to get $R(b, p)$ and the point $p^* = R(b, p^*)$ calculated for each value of b (figure 2(b)). The results for p_{\max} and p^* (without uncertainties) are displayed in the first column of table 1. The actual number of trials in each case is given in parentheses.

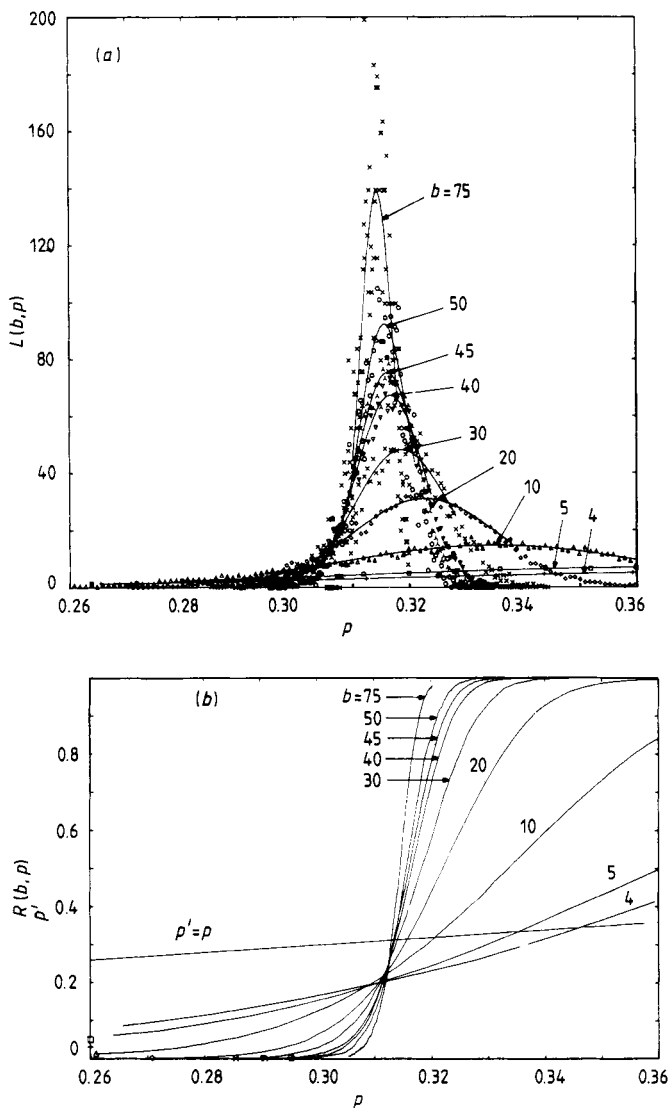


Figure 2. Experimental results in the case $n = 1$ (isotropic case). (a) Experimental histogram of $L(b, p)$ for b ranging between 4 and 75. Full curves are fits to a Gaussian in a restricted range of p values. (b) Smoothed $R(b, p)$ curves for the different b values as calculated from the data of (a). The intersections with $p' = p$ give the fixed points $p^*(b)$.

Table 1. Monte Carlo results n ranging between 1 and 15, for various sample sizes b ranging between 4 and 75. In each cell the upper figure is for $\langle p \rangle$, the lower for p^* ; the figure in parentheses is the number of samples. The row $b = \infty$ gives the extrapolated percolation threshold p_c . The row ν gives the values of the correlation length exponent as obtained using FSS and relation (2) of text. The last row gives the estimated value ν_e of ν for $n = 1, 5$ and 10 using the 'intersection' criterion (FSS and PSRG).

$b \backslash n$	1	2	3	5	8	10	15
4	0.3877 0.3515 (10 000)						
5	0.3651 0.3320 (10 000)						
10	0.3348 0.3205 (15 000)	0.2841 0.2610 (10 000)	0.2419 0.2115 (10 000)				
20	0.3228 0.3159 (15 000)	0.2695 0.2585 (5000)	0.2259 0.2115 (10 000)	0.1698 0.1503 (5000)			
30	0.3189 0.3137 (15 000)	0.2649 0.2552 (5000)	0.2212 0.2124 (5000)	0.1646 0.1524 (5000)	0.1180 0.1016 (5000)	0.0994 0.0816 (5000)	
40	0.3168 0.3134 (7500)		0.2188 0.2124 (5000)	0.1620 0.1529 (5000)			
45	0.3163 0.3132 (7500)						
50	0.3157 0.3131 (10 000)	0.2613 0.2573 (5000)	0.2174 0.2123 (10 000)	0.1606 0.1538 (10 000)	0.1142 0.1056 (5000)	0.0954 0.0856 (5000)	0.0665 0.0564 (5000)
60							0.8436 0.0574 (5000)
75	0.3142 0.3126 (2000)	0.2598 0.2575 (2000)	0.2158 0.2125 (2000)	0.1586 0.1541 (2000)	0.1120 0.1065 (2000)	0.0936 0.0873 (2000)	0.0656 0.0588 (5000)
∞ (extrapolated)	0.3119 ± 0.0005	0.2572 ± 0.0005	0.2128 ± 0.0005	0.1554 ± 0.0003	0.1084 ± 0.0006	0.0904 ± 0.0003	0.0635 ± 0.0010
ν	0.875 ± 0.030	0.88 ± 0.04	0.885 ± 0.020	0.925 ± 0.050	(0.88)	(0.86)	(0.87)
ν_e (estimated)	0.85 ± 0.05			0.87 ± 0.05		0.85 ± 0.05	

Now, we assume that $\langle p \rangle = p_{\max}$ or, strictly speaking, that $\langle p \rangle$ follows the same scaling laws as p_{\max} and use simultaneously (1) and (3) with a trial value ν_t . If ν_t is the exact value of ν , the two extrapolated straight lines for b^{-1}/ν_t as a function of $\langle p \rangle$ or $\langle p^* \rangle$ must cross exactly on the p axis at the point $p = p_c$.

The use of this criterion with several trial values of ν_t leads to a more accurate determination of p_c and to an estimated value ν_e of ν . A better determination of ν can be performed by means of FSS and relation (2), as a log-log plot of L_{\max}^G against b must approach a straight line for large values of b , with slope $1/\nu$.

Figures 3(a) and 3(b) show the extrapolation using both methods when $n = 1$ (pure random site percolation). The first procedure (figure 3(a)) gives the value $p_c = 0.3119 \pm 0.0005$ and the estimated correlation length exponent $\nu_e = 0.85 \pm 0.05$. This value of p_c is in good agreement with the value 0.3117 ± 0.0003 obtained by Heermann and Stauffer (1981) with fss procedure and sample sizes up to $b = 100$. The second extrapolation procedure (figure 3(b)) provides the more accurate value $\nu = 0.875 \pm 0.030$ in better agreement with the value $\nu = 0.89 \pm 0.01$ of Heermann and Stauffer (1981) than with the previous one $\nu = 0.845 \pm 0.015$ obtained by Kirkpatrick (1979).

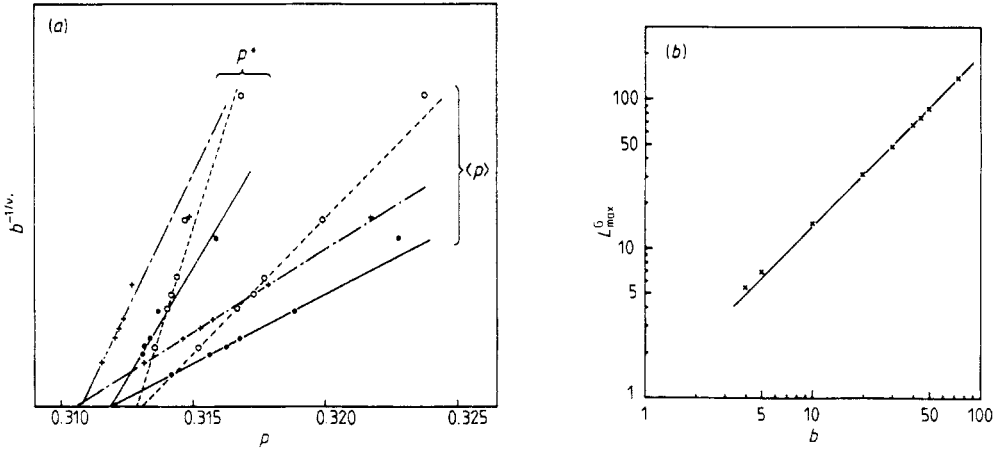


Figure 3. Extrapolation procedures for obtaining p_c and in the case $n = 1$: (a) Fits of b^{-1/ν_i} against $\langle p \rangle$ and p^* to straight lines for three trial values of ν_i . The vertical scale is arbitrary and the sets of data for $\nu_i = 0.90$ and $\nu_i = 0.80$ have been translated along the p axis by 0.002 for clarity. Notice that only the lines for $\nu_i = 0.85$ intersect on the p axis, +, $\nu_i = 0.90$; ●, $\nu_i = 0.85$; ○, $\nu_i = 0.80$. (b) Log-log plot of L_{\max}^G against b . A linear variation is made evident for large values of b . From the slope of the straight line the value $\nu = 0.875 \pm 0.03$ is obtained.

For increasing values of n , p^* is shifted into the wings of the distribution $L(b, p)$ where the Gaussian approximation is expected to break down. Then the second extrapolation procedure using fss and relation (2) becomes less accurate. Fortunately, at the same time, the criterion of the first procedure becomes more sensitive to slight changes in ν_i , since for $n > 2$, the two straight lines of b^{-1/ν_i} against p^* and p_{\max} have slopes of opposite signs.

3. Results

The former analysis has been performed for increasing values of n up to 15. It must be noticed that for so large an aspect ratio the linear size 75 of the largest sample is only five times the length of the element. So the uncertainties and fluctuations in the experimental values of $L(b, p)$ increase with n . Nevertheless, the determination of p_c remains rather accurate up to these values since the asymptotic behaviour seems to be approached in a faster way.

As an example, the extrapolation curves using the 'intersection' criterion are given in figure 4 for $n = 5$. The complete numerical results $\langle p \rangle$, $\langle p^* \rangle$ and the extrapolated values of p_c are summarised in table 1 for all n and b values. We have represented p_c as a function of n with logarithmic scales in figure 5. $p_c(n)$ does not approach a proportionality law in n^{-2} as could have been expected from the work of Carmona *et al* (1981) but rather approximately as asymptotic law in n^{-1} . Nevertheless, although according to the curvature n^{-2} seems to be definitively ruled out, larger aspect ratios would be necessary to ascertain the n^{-1} behaviour.

The exponent ν has been determined in order to evidence eventual deviations to the correlation length exponent of the pure 3D random-site problem. The reliability of such determination becomes poor at large values of n because of the small number of experimental points and limited range of b values. In addition, a careful evaluation

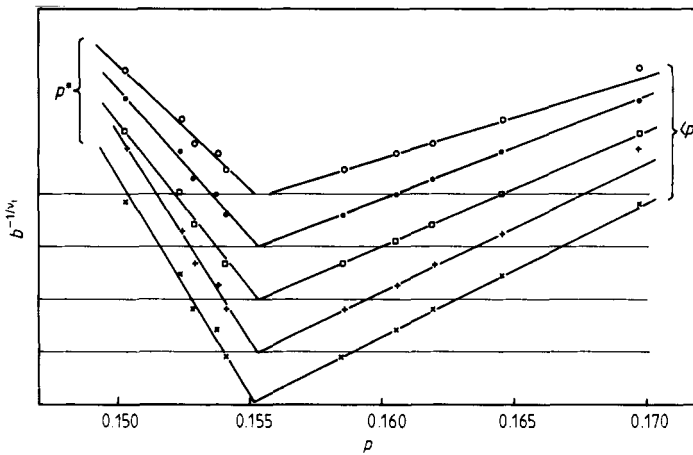


Figure 4. The same as in figure 3(a) in the case $n = 5$; five trial values for ν_i have been used. Ordinate origins have been translated for clarity. Notice that the intersection of lines occurs on the p axis for three values of ν_i : the values of ν_i are denoted by \circ (0.80), \bullet (0.85), \square (0.87), $+$ (0.90), \times (0.92).

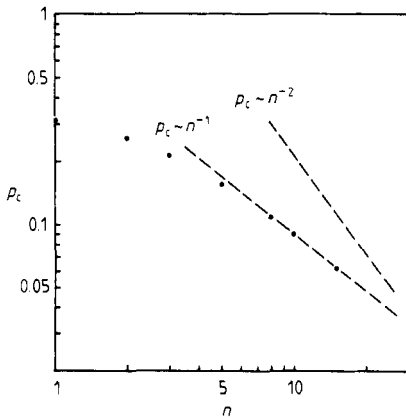


Figure 5. Log-log plot of the variation of percolation threshold p_c with sticks length n . Broken lines give, for comparison, n^{-1} and n^{-2} dependences.

of ν_c using the 'intersection' criterion (FSS and PSRG) has been done for the two values $n = 5$ and $n = 10$. These values have been chosen since, on the one hand, we expect that eventual deviations will be greater the larger n is, but on the other hand, when n is too large there is too small a set of reliable experimental points (those for which the cell size to stick length ratio is large enough). All these results are reported in table 1.

Values of ν exactly equal to 1 or 0.5 (the 'classical' exponent) can be ruled out. Moreover, in the limits of the accuracy, all the systems studied seem to belong to the same universality class as the pure site percolation problem for which we found $\nu = 0.875 \pm 0.030$.

4. Discussion

Our two main results are thus:

- (i) $p_c(n)$ follows an asymptotic law close to $1/n$ at large n .
- (ii) The universality class, defined through the ν exponent, seems to be the same as for usual 3D random percolation.

These results contradict the prediction by Carmona *et al* (1980) $p_c \sim l^{-2}$ on the basis of dimensional arguments for very large aspect ratios l of fibres stating that the probability of contact between two sticks depends, for a given distance between their centres of symmetry, only on their lengths. In a 2D system, the same arguments give $p_c \sim l^{-1}$, a result also obtained by Pike and Seager (1974) in a 2D Monte Carlo simulation of sticks of length l , without any thickness, centred on the nodes of a square lattice, and taking random orientations. Moreover Carmona *et al* (1981) have found experimentally a conductivity threshold for carbon fibres with aspect ratios larger than 100 embedded in an epoxy resin following the proportionality law n^{-2} and a critical conductivity exponent $t = 3.1$, close to the mean-field theory exponent $t = 3$ in place of $t = 1.6$ (Kirkpatrick 1976) for the random usual percolation, which could indicate a different universality class.

We notice that our model is a discrete one, while the other works are concerned with continuous percolation; such an argument does not seem very relevant to the asymptotic behaviour and universality-class problems, as has been shown for the usual random percolation by means of experiments (Fug *et al* 1978) and Monte Carlo simulations (Haan and Zwanzig 1977, Gawlinski and Stanley 1981, Kertesz and Vicsek 1982). Let us now consider these discrepancies, first with the previous 2D simulation and the dimensionality arguments, then with the experiments.

If we randomly disperse needles on a table, we get a system of sticks analogous to the Pike and Seager system; that is, the sticks can cross at any point. Geometrically, the system appears to be quasi two dimensional because the needles are very thin compared with the size of the area over which they are dispersed. However, from a topological point of view, the system is a 3D assembly of elements piled up under the action of gravity and the statistics of contacts are not those of a real 2D assembly of solid elements for which the steric-exclusion conditions holds in the plane. Taking into account this physical restriction leads to a different percolation problem. Similar arguments can also be applied to the already recalled dimensional analysis by Carmona *et al* (1980). Strictly speaking, assuming zero thickness (or infinite aspect ratio) in a random system of sticks gives zero probability of the contacts! So in the development of the argument, even if scale transformation leaves the geometry unchanged, the

transformation of the contact conditions remains unclear. This argument then seems to be more relevant to sticks displayed on a lattice but without the excluded-volume condition. This leads us to emphasise the importance of this condition in the present calculations.

In preliminary simulations performed on a 2D square lattice, it was found that, due to this effect, large clusters of parallel sticks tend to form. This effect increases rapidly with the length of the 2D sticks and results in a non-monotonous variation of the percolation threshold with n . p_c starts decreasing, goes to a minimum and then increases slowly for very large values of n . In a sense, one can say that a 'nematic-like transition' occurs before the 'true percolative transition' which moves toward larger p values. In 3D systems, the densities at the threshold, at the same n , are lower and the number of accessible configurations is higher than in 2D systems, so this effect should be less important. Nevertheless, for large values of n , we can expect a slower decrease of $p_c(n)$ than predicted as the present results seem to indicate.

Coming back to the results obtained on composites, we must consider that according to the aspect ratio (up to 300) and the continuous nature of the problem (all positions and orientations are accessible), the excluded-volume effect might be smaller than for a lattice problem. But according to the cylindrical shape of the rods, which allows only for points contacts, the probability of such contacts would again be close to zero and no conduction would occur. In our opinion, this system is not a fully random one but the 3D extension of the quasi-two-dimensional needles system described above. The 'piling process' is not performed here by gravity, but rather by the shear forces exerted inside the matrix, still fluid, during the preparation of the sample. These shear forces bring some elements into contact and keep them until the mixing process is stopped and the resin allowed to harden. If the mixing is homogeneous enough, we may expect percolative properties close to the 'random piled' system defined above in 2D case, that is a random assembly of elongated elements without excluded volume. It remains to be understood if such a system could behave relative to the critical properties like a Cayley tree and exhibit a conductivity exponent $t = 3$; this is still questionable and might be investigated carefully.

Conclusion

We have performed Monte Carlo simulation for the correlated 3D site problem equivalent to an assembly of identical parallelepipedic conducting elements of length n lattice spacing displayed randomly on a lattice according to the steric exclusion condition. Accurate determinations of p_c as a function of n are given, up to an aspect ratio equal to 15. From evaluation of the critical exponent ν , we infer that all these systems remain in the same universality class as the ordinary isotropic percolation.

The discrepancies with former papers on similar systems seem to be related to the consideration of steric exclusion. Systematic analysis of the critical properties, both with and without this condition, would be suitable, as well as the extension to the case of oriented elements. Such work is currently in progress.

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