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1992 J. Phys.: Condens. Matter 4 5757

(http://iopscience.iop.org/0953-8984/4/26/009)

J. Phys.: Condens. Matter 4 (1992) 5757–5762. Printed in the UK

Bond percolation and conduction in random and regular networks: topological effects at low coordination numbers

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Received 11 February 1992, in final form 13 May 1992

Abstract. Earlier workers have shown that percolation and conductivity properties of 14-coordinated random and regular networks in 3D are essentially indistinguishable. This work examines the generality of this result for networks of low coordination (≥ 3). The critical percolation exponents, determined by finite-size scaling, are found to be independent of topology and coordination in agreement with existing literature values. However, differences in the percolation threshold, conductivity, and cluster distributions increase with decreasing coordination suggesting that the transferability of these properties between networks of different topology but the same coordination is not valid in general.

Regular grid pore networks are being used increasingly to model transport and reaction processes occurring in porous media (reviewed by Sahimi *et al* 1990). The structures of the media are, in general, disordered and the work of Jerauld *et al* (1984b) is the most commonly cited justification for using a regular square or cubic grid to represent them (Sahimi and Tsotsis 1985, Sahimi *et al* 1990, Sahimi and Stauffer 1991). Jerauld *et al* determined the percolation thresholds and critical exponents as well as other percolation properties for a BCC2 network (coordination number, $z = 14$) and compared these results with a modified Voronoi tessellation of the same mean coordination number. For all the properties evaluated the differences between the random and regular grids were small and often within the uncertainty of the estimated value. It was concluded that the mean coordination number is much more important in determining the properties of the network than are topological considerations.

Recently, Hollewand and Gladden (1992) have compared the behaviour of 3D random and regular networks in the modelling of diffusion and diffusion with simultaneous reaction occurring in porous media. The two different topologies exhibited significantly different behaviour. Moreover, it was shown that lattice coordination numbers in the range 3 to 6 are required to obtain effective diffusivities in agreement with experimental measurements reported by other workers. The aim of the work presented here is to examine whether the effect of network topology remains small at these lower coordination numbers, i.e. whether the conclusions of Jerauld *et al* (1984a, b) are valid for all coordination numbers. Throughout this work, the definitions and nomenclature used by Jerauld *et al* (1984a, b) and Kirkpatrick (1979) will be employed.

0953-8984/92/265757+06$04.50

Jerauld *et al* (1984b) obtained a random network by randomly deleting bonds from a Voronoi tessellation until the average coordination number was 14. In order to obtain random networks with coordinations in the range 3 to 14, we have used a different method of constructing the network. The network is generated by distributing N points at random in a 3D space. Nearest neighbours are connected until the specified coordination number is satisfied at every point. Two parallel planes are then imposed on the network and the intersections of bonds with these planes define the network surfaces. The grid is periodic in the other two orthogonal directions.

The regular grids for $z = 3, 4, 5, 6$ were based on the simple cubic structure while the BCC2 network was used for $z = 14$. To obtain coordination numbers of 3, 4 and 5, bonds were deleted from the simple cubic structure in a regular fashion until each node had the required coordination number. This gives rise to unit cells of length 6, 3 and 6 for the coordination numbers, 3, 4 and 5, respectively. Larger, periodic networks were constructed out of these unit cells, each having the property that they consist of a single cluster when the conducting fraction $p = 1$. Random networks containing up to 18000 points and regular grids up to $42 \times 42 \times 42$ points were generated. All simulations were performed using SUN SPARC work-stations. The number of realizations of each network ranged from 4000 for the smallest network ($6 \times 6 \times 6$) to 800 for the largest ($42 \times 42 \times 42$). For the random networks, several bond assignments were made for every realization of the network and all calculated properties were based on the accessible fraction at $p = 1$ (since a few isolated clusters are formed in generating the random model).

Since the bond percolation problem is more appropriate in the study of transport in porous media we restrict our discussion to this case. For each coordination number considered, bonds were assigned as present or absent with probabilities p and $1-p$, respectively, in the usual manner. For a network of length L, where $L = N^{1/3}$, the proportion of conducting networks, $R(L, p)$ and the percolation probability, $P(L, p)$ were determined as defined in Kirkpatrick (1979). Previously, Reynolds *et al* (1980) have found that $R(L, p)$ is described accurately by a cumulative beta distribution. For both random and regular networks at low coordinations, we have obtained a better fit for $z \leqslant 6$ using a three-parameter exponential or Weibull distribution of the form

$$F(x) = 1 - \exp\left(-\left(\frac{x-c}{b}\right)^{a}\right) \qquad x > c \tag{1}$$

although it is observed that this function has an unbounded domain for $x > p_c$. In all cases the difference in fit between (1) and the beta distribution in the range $0.1 < R(L, p) < 0.9$ is small and does not affect the calculated properties within the estimated uncertainties. Figure 1 gives results of $R(L, p)$ for the 3-coordinated random network.

Following Kirkpatrick (1979) the percolation thresholds, p_c, were estimated by using the fact that for a suitable $x < p_c$, $p(R(L) = x)$ increases with L and for $x > p_c$, $p(R(L) = x)$ decreases with L. Both tend to p_c for large L and so p_c is estimated by extrapolating the two curves. Values of x of 0.2 and 0.8 were found to be suitable for all simulations. The calculated thresholds are given in table 1. Considering firstly the networks with $z = 14$ it is clear that the results for the random and regular grids agree within the quoted uncertainty. As the coordination number is decreased, the difference between the percolation thresholds in the random and regular grids increases from 0.035 at $z = 6$ to 0.103 at $z = 3$. In all cases, the

threshold in the random network is greater than in the regular grid. The threshold for the regular grid at $z = 4$ agrees well with the reported value for the diamond structure suggesting that the bond arrangement in the regular grid is not important.

Table 1. Percolation thresholds for random and regular networks.

z	Random grid p_c	Regular grid p_c	Literature regular grid p_c
3	0.646 ± 0.005	0.543 ± 0.004	—
4	0.459 ± 0.003	0.382 ± 0.003	0.388[a]
5	0.350 ± 0.003	0.302 ± 0.003	—
6	0.283 ± 0.002	0.248 ± 0.002	0.2495 ± 0.0005[b]
14	0.103 ± 0.002	0.099 ± 0.002	0.0991 ± 0.0005[c]

[a] Data for a diamond 3D grid, from Stauffer (1985).
[b] From Kirkpatrick (1979).
[c] From Jerauld *et al* (1984b).

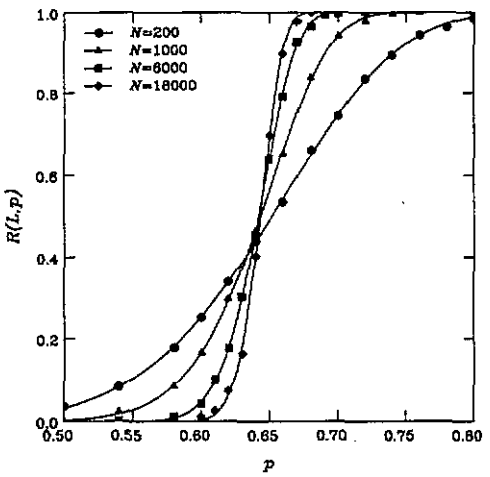

Figure 1. *$R(L, p)$ data and a fit to (1) for the* random network ($z = 3$).

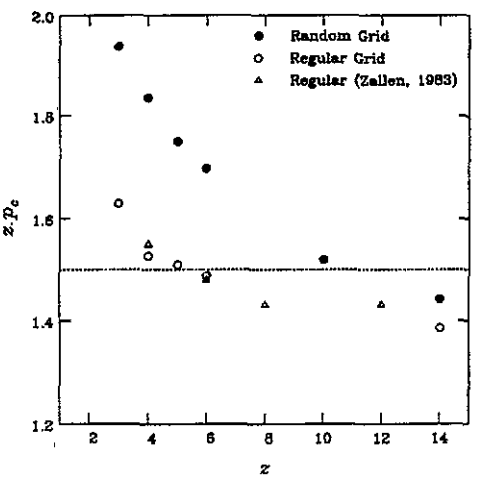

Figure 2. Comparison of random and regular lattice results with the hypothesis $zp_c = 1.5 \pm 0.1$.

The dependence of the percolation threshold on coordination number is well known. Scher and Zallen (1970) first suggested that the critical probability, defined as the fraction of occupied space in the continuum percolation problem, is independent of lattice structure. For site and bond percolation, the product zp_c has been proposed as the appropriate quasi-invariant group. For bond percolation in 3D, $zp_c = 1.5 \pm 0.1$ has been proposed (Zallen 1983, Guyon 1987). Figure 2 shows the values of this group for the random and regular lattices obtained in this study; the results quoted by Zallen (1983) are also given. Not only does this emphasize the difference between the random and regular lattices, but clearly in both cases zp_c is a decreasing function of z. This result combined with the absence of a formal justification for the invariance of zp_c and the critical density illustrates the limitations of these heuristics.

Table 2. Critical exponents for random and regular networks. Networks of coordination 3, 4, 5, 6 and 14 were studied.

	Random model	Regular model	Literature
β	0.42 ± 0.02	0.42 ± 0.02	0.42 ± 0.006[a]
ν	0.91 ±0.03	0.91 ± 0.02	0.906 ± 0.03[b]
t	1.9 ± 0.1	1.9 ± 0.1	1.94 ± 0.1[c]

[a] From Nakanishi and Stanley (1981).
[b] From Jerauld *et al* (1984b).
[c] From Derrida *et al* (1983).

Finite-size scaling was used to determine the critical constants which are given in table 2. Following Reynolds *et al* (1978) the correlation length exponent, ν, was determined from

$$[\mathrm{d}R(L,\ p)/\mathrm{d}p]_{p=p^*} = L^{1/\nu} \tag{2}$$

where p^* is near the percolation threshold. Since for a finite network the threshold is not uniquely defined we chose p^* such that $R(L,\ p^*) = 0.5$. The percolation probability exponent β was determined from

$$P(L,\ p) \propto L^{-\beta/\nu} f\left(L^{1/\nu}(p - p_c)\right) \tag{3}$$

where the values of p_c and ν were taken from the previous determinations and assumed to be constant (Kirkpatrick 1979). The exponent β was taken as that value for which all the data lie on a single universal curve given by equation (3). It was possible to estimate β to within ±0.02 using this method. Within the quoted uncertainties, the critical exponents are independent of the network structure and coordination number and agree with commonly accepted values. This supports the current weight of evidence suggesting that these parameters are functions of the network dimensionality only.

The conductivity $G(L,\ p)$ was determined at the thresholds given in table 1 for both random and regular grids of coordination $z = 3$ and $z = 6$. Regular grids with $L = 6$, 12 18, 24 for $z = 3$, $L = 6$, 9, 12, 15, 18 for $z = 6$ were used. The random grid sizes used were $N = 200$, 500, 1000, 3000, 6000. The scaling law (Jerauld *et al* 1984b and references therein)

$$G(L,\ p_c) \propto L^{-t/\nu} \tag{4}$$

was used to calculate the conductivity exponent and a result of $t = 1.9 \pm 0.1$ was obtained for both coordination numbers and network types.

The conduction problem is of interest in the study of transport in porous materials and a universal conductivity exponent is not sufficient to describe transport in the material. Figure 3 shows the calculated conductivities for $z = 3$ and $z = 6$ for the random and regular networks ($L = 12$). This demonstrates the effect of the different percolation thresholds on the conductivity at $p > p_c$. At the lower coordination number the conductivity of the random network is significantly lower than in the regular grid. The conductivities approach each other as the coordination number increases.

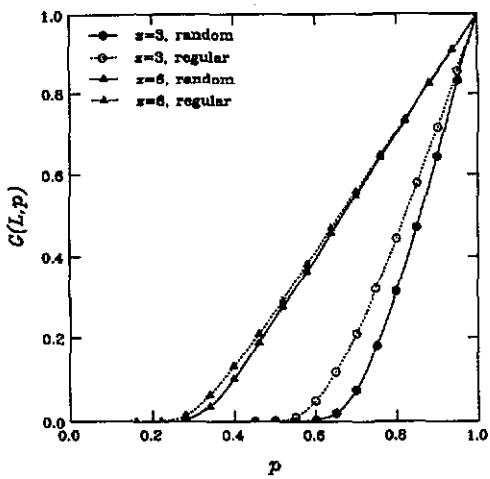

Figure 3. Random and regular network conductivities for $z = 3$ and $z = 6$ ($L = 12$).

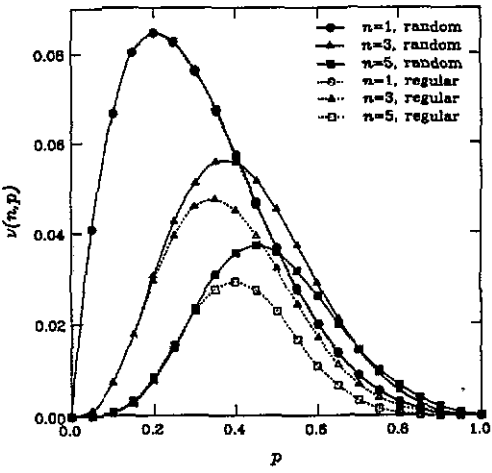

Figure 4. Bond cluster distributions for random and regular grids $z = 3$, $n = 1, 3, 5$.

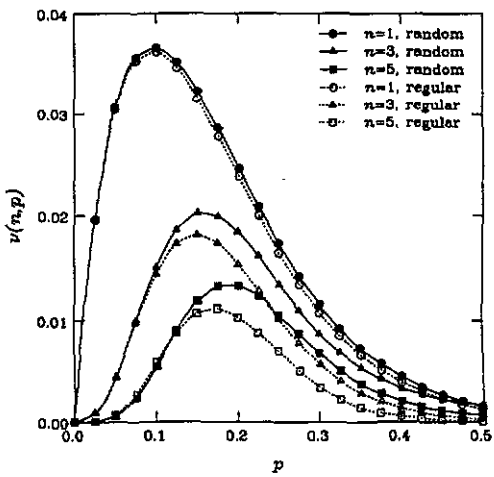

Figure 5. Bond cluster distributions for random and regular grids $z = 6$, $n = 1, 3, 5$.

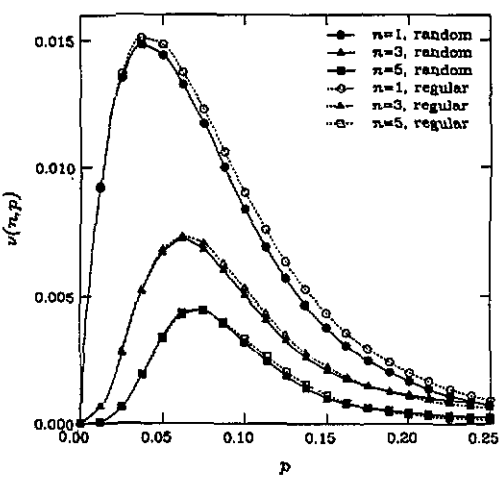

Figure 6. Bond cluster distributions for random and regular grids $z = 14$, $n = 1, 3, 5$.

A useful means of visualizing the difference in the structures of the networks is the cluster distribution $\nu(n, p)$ defined as the probability that a bond belongs to a cluster of n bonds (Winterfield *et al* 1981). Figures 4, 5 and 6 compare the cluster distributions for random and regular networks with $z = 3$, 6 and 14, respectively. For $z = 3$ and 6 the cluster distributions obtained for the random grids are broader than those found in the regular grids, consistent with the results obtained by Jerauld *et al* (1984a) for 2D grids. The relative difference between the random and regular distributions is largest for $z = 3$. In the cases of $z = 3$ and 6, the random networks have a greater tendency to form small clusters which is consistent with the higher percolation thresholds of these networks compared to the regular grids. At $z = 14$

the differences in the distributions are small.

Although it has been shown previously that at high coordination numbers the effect of topology on the percolation properties of a network is very small, we have shown that this general conclusion does not extend to small coordination numbers. The effect of topology becomes more important as the network coordination number decreases. This difference is not observed in the critical percolation exponents and the values of these obtained are in agreement with currently accepted universal values.

Acknowledgment

We thank a referee for bringing some interesting points to our attention.

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