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COMMENT

Percolation in a system of randomly distributed sticks

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Abstract. Arguments are given that a full description of any realistic model of randomly distributed conducting sticks in an insulating matrix must incorporate the history of system preparation. Different regimes of composite formation produce different dependences of the percolation threshold value on the width-to-length ratio ε: $\rho_c \sim \varepsilon^0$, $\rho_c \sim \varepsilon^1$ or even $\rho_c \sim \varepsilon^2$.

The electrical properties of composites with conducting (or superconducting) fibres embedded in an insulating matrix are usually interpreted in terms of percolation theory. This approach permits us to evaluate the threshold concentration and to predict the critical behaviour and crossover effects. The prediction of the percolation threshold concentration ρ_c is the most important problem from a practical point of view. On the other hand, this problem is the most difficult one. The above calculation of ρ_c demands the complete knowledge of microscopic structure and properties, which is often unattainable. Therefore analytical and Monte Carlo calculations [1-6] were aimed at determining the dependence of ρ_c on the width-to-length ratio ε. In the limit of small ε this dependence is found to be a linear one:

$$\rho_c \approx (1/v)\varepsilon. \tag{1}$$

Here v is the volume of a stick of length l and width d: $v \approx d^2 l$ and the width-to-length ratio $\varepsilon = d/l$ lies in the range 10^{-1}-10^{-3}. The estimation (1) is valid for a system of permeable sticks randomly distributed in an insulating media. Two sticks are assumed to be connected in this model if they intersect with each other.

If we consider the problem of absolutely rigid impenetrable sticks, their random distribution can be realised in different ways. For example, one could place sticks randomly without intersections with the previously placed sticks. In this case, at a concentration of the order of $\rho \approx (1/v)\varepsilon$ there will be no free space for any new stick. Nevertheless the sticks will be disconnected from each other. The point is that, in the case of hard inpenetrable sticks, the contact occurs only if the distance between the surfaces of the sticks is exactly zero, and this event has zero probability.

Another possibility is to thermalise the system after addition of each stick and to place new sticks in the casually formed cavities. In this case, at $\rho \approx (1/v)\varepsilon$ the nematic alignment of sticks becomes thermodynamically favoured and the process of addition of new sticks can be continued till $\rho \approx 1/v$. The percolation transition takes place in this model only in the dense packing limit:

$$\rho_c \approx 1/v. \tag{2}$$

One might think that the only way to make the model more realistic is to introduce a finite interaction length between the sticks. Indeed, this can be caused by finite conductance of the insulating matrix, by its electric breakdown, by finite rigidity of the sticks, inclusions, roughness of the surface of the sticks, etc. The 'hard core and permeable shell' model is free from the above peculiarities and is often used in explanations of experimental results.

The purpose of this comment is to point out that the 'thermodynamical' approach to the configurational statistics of sticks has a very restricted (if any at all) field of application.

The sticks are macroscopical objects and they cannot be thermalised via the processes of Brownian diffusion, if the processes of sedimentation or flocculation are present. For example, let us consider the case of a homogeneous suspension of depth 5 mm of fibres with typical size $l = 1$ mm, $d = 10\ \mu$m [7, 8]. Using the barometric formula it can easily be estimated that the density of the medium must be tuned to the density of the fibre to an accuracy of 10^{-9}. Thus any homogeneous system can be prepared only using external macroscopic intermixing. We shall show below that, during the intermixing, the sticks interact with each other and this determines the connectivity properties of the system. It is interesting to note that the finite interaction length, which is very important in the thermodynamical model, does not affect the percolation threshold criteria in the case of the intermixing system.

For educational purposes we shall start with some idealised procedure of addition of new sticks in a system. The new stick cannot simply appear in an unoccupied space; it must somehow be transported to that place. During this transportation it slides against other sticks, pushes them apart, sticks to them, carries them along, etc. If the transported stick comes into contact with another stick, this contact is retained during the displacement of the first stick along the length of order l. There are two possible regimes of transportation.

(a) The stick is transported linearly with random orientation (figure 1(a)). During this motion it intersects most of the sticks centred in a tube of cross section $\approx l^2$ along its line of motion. If the stick stops, the mean number of contacts with neighbouring sticks will be of the order of ρl^3. The percolation transition will take place at $\rho l^3 \approx 1$ with ρ_c independent of the thickness of the sticks:

$$\rho_c \approx 1/l^3 \approx (1/v)\varepsilon^2. \tag{3}$$

(b) The sticks move, aligned in the direction of motion (figure 1(b)). In this case the number of contacts reduces by a factor of d/l and we get for the threshold concentration the soft core estimation (1).

Both cases (a) and (b) look rather artificial but it is easy to connect them with the processes during mechanical intermixing of the system.

First, we consider a case of homogeneous large-scale intermixing of the viscous fluid with sticks immersed in it. The motion of the fluid on small scales can be represented as a shear flow (figure 2). If the inertial forces are negligible the stick is carried along with a flow and rotates in it. It is clear from figure 2 that the relative movement of the stick and medium is longitudinal (shown by the heavy arrow) and if there are other sticks in the flow their relative movement can be described by figure 1(b). The percolation threshold in such a system occurs at a concentration given by (1). If the intermixing is strongly inhomogeneous, with the scale of inhomogeneity of the order of l or smaller, then for each stick there is no possibility of joining up the flow as in the previous case. There will always exist some transverse relative motion

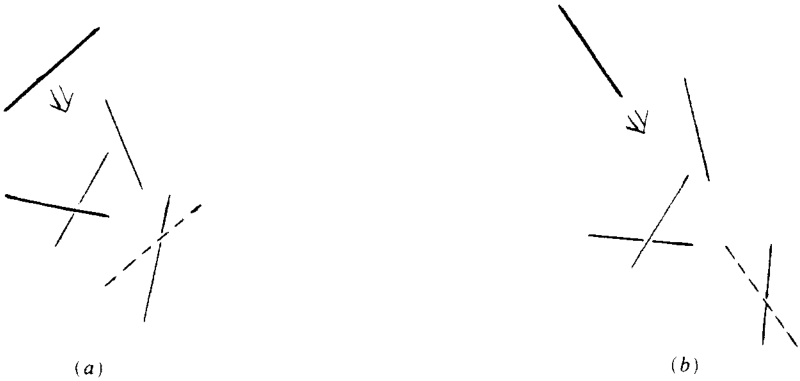

Figure 1. Two idealised ways of addition of a new stick (shown by a heavy line) in a system. The finite position of a stick is shown by a dotted line. (*a*) Stick is transported with random orientation. (*b*) Direction of a stick coincides with the direction of its motion.

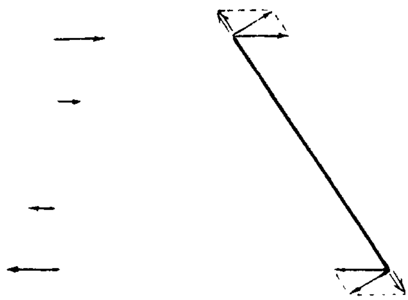

Figure 2. The motion of a stick trapped in a shear flow. The relative longitudinal movement of a stick and the medium is shown by a heavy arrow. If there are other sticks in the flow, the interaction with them will be as in figure 1(*b*).

of the stick and medium and the considered stick will hit other sticks trapped in the flow. The intersection with other sticks in the flow is then described by the situation in figure 1(*a*). The threshold concentration of sticks is given by (3). In the intermediate case of the scale of inhomogeneity of the flow L larger than l, the ratio between average transverse and longitudinal velocities of the stick and medium is of the order $(l/L)^2$ and the threshold concentration is given by

$$\rho_c \approx \frac{1}{v}\varepsilon^2\left(\frac{L}{l}\right)^2 \qquad \left(\frac{L}{l}\right)^2 < \varepsilon^{-1}. \tag{4}$$

After intermixing, the resulting configuration of sticks is usually frozen by cooling or by chemical processes. If this process is not rapid enough, the contacts which were formed during intermixing can be broken by Brownian diffusion of the sticks. At this point we must recall that there is a finite interaction length λ which determines the decay time for contacts formed during intermixing:

$$\tau_0 = D^{-1}\lambda^2 \tag{5}$$

where D is the transverse diffusion coefficient of a long stick $D \approx kT/\eta l$ and η is the viscosity. In the case of the time-dependent viscosity coefficient (5) will be somewhat more complicated. This process of decay of contacts after intermixing can, in principle, be observed through the electrical DC and AC measurements of the conductivity of the system.

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