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"Universal" percolation-threshold limits in the continuum

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The existing criteria for the onset of percolation in the continuum are limited to systems of parallel-aligned, equal-size, penetrable, convex objects. We present here criteria for the much more general case of macroscopically isotropic or anisotropic systems in which the objects may also be of variable sizes and of random orientations. It is found that the critical fractional occupied area (or the equivalent average critical total excluded area $\langle A_{ex} \rangle$) as well as the critical fractional occupied volume (or the equivalent $\langle V_{ex} \rangle$) are confined within two limits. The upper limit is that of the "parallel objects" systems and the lower limit is that of the (newly introduced) "horizontal-vertical" system. These limits are $3.2 \leq \langle A_{ex} \rangle \leq 4.5$ and $0.7 \leq \langle V_{ex} \rangle \leq 2.8$.

In 1970 Scher and Zallen[1] presented criteria for the onset of percolation in lattices in which the sites are occupied by hard-core circles or hard-core spheres. It was later shown[2,3] that to a reasonable accuracy their criteria of critical fractional occupied area or volume are valid for continuum systems. Similar criteria have also been found[2,4] for soft-core (interpenetrable) circles or spheres. The critical fractional occupied area for the soft-core circles was found to be $s_c = 0.68$ and the critical fractional occupied volume for spheres was found to be $\tau_c = 0.29$. In the many studies that followed[5-9] it was shown that these values applied to systems of parallel aligned objects, such as parallel squares or cubes. Recently, it was further demonstrated[10] that within the accuracy of available data[2] these values are also obtained for systems of variable-radius circles or variable-radius spheres.

The above results have not been presented only in terms of s_c or τ_c. In some studies the critical radius r_c was found for a given ensemble of circles or spheres, while in others the critical total area $N_c a$ or the critical total volume $N_c v$ have been determined. In the latter cases the area of the object, a, or the volume of the object, v, were given while the critical concentration for the onset of percolation, N_c, was obtained from the computations. In the most recent works,[10,11] the average critical total excluded area $\langle A_{ex} \rangle$, or the average critical total excluded volume $\langle V_{ex} \rangle$, has been found. All the results were shown to be in agreement with the above s_c and τ_c values. Since the one object geometrical picture is easier to comprehend and since the most recent data[10,11] are given in terms of $\langle A_{ex} \rangle$ and $\langle V_{ex} \rangle$ we shall use these quantities, rather than s_c and τ_c, in our discussion. The relations between these two sets of quantities were established[2,4,10] for parallel objects and thus one can simply use the equations

$$s_c = 1 - \exp(-\langle A_{ex} \rangle/4) \ ,$$
$$\tau_c = 1 - \exp(-\langle V_{ex} \rangle/8) \ , \tag{1}$$

to relate the above quantities. The $s_c = 0.68$ and $\tau_c = 0.29$ values correspond then to $\langle A_{ex} \rangle = 4.5$ and $\langle V_{ex} \rangle = 2.8$, as was confirmed by the Monte Carlo studies.[9-12] A further discussion of the applicability of Eqs. (1) will be given elsewhere.[12]

For many years the above s_c and τ_c values have been considered to be "dimensional invariants"[4,10] in the sense that, independent of how and with what objects one fills the continuum, those values are the ones which determine the onset of percolation. Recent findings show, however, that this invariance is not conserved if randomness in the orientation of the objects is put into the system.[10] For example, in the macroscopically isotropic system of randomly oriented capped rectangles (with length ten times the radius) it was found[12] that $\langle A_{ex} \rangle = 4.1$ and for a macroscopically isotropic system of randomly aligned long capped cylinders it was found[11] that $\langle V_{ex} \rangle = 1.4$. These lower values of $\langle A_{ex} \rangle$ and $\langle V_{ex} \rangle$ have not been explained and no empirical rules that can predict their dependence on system parameters have been presented. Since we have shown[10-12] that once values for the isotropic system are known, the effects of macroscopic anisotropy (in an anisotropic system made of the same objects) can be accounted for by the excluded volume theory, we will concern ourselves in this Rapid Communication with isotropic systems. (See Table I.)

Now that it is apparent that the values of $\langle A_{ex} \rangle = 4.5$ (or $s_c = 0.68$) and $\langle V_{ex} \rangle = 2.8$ (or $\tau_c = 0.29$) are restricted to the class of systems in which all the objects are parallel aligned and of equal size, the question arises whether or not one can find a more general criterion for the onset of percolation in the continuum. The criterion has to apply not only to the above class but also to the many other systems for which these two restrictions are removed. This extension is

TABLE I. Monte Carlo values of $\langle A_{ex} \rangle$ and $\langle V_{ex} \rangle$ for isotropic systems of equal size objects. From the computations involved one may conclude that the values given in this work have an accuracy of ± 0.1. Hence, values the difference between which is larger than 0.2, should be considered different. The significance of the systems listed here is explained in the text.

System	$\langle A_{ex} \rangle$ or $\langle V_{ex} \rangle$
Parallel objects (2D)	4.5
Horizontal-vertical (2D)	3.2
Capped-rectangles (randomly aligned)	4.1
Parallel objects (3D)	2.8
Horizontal-vertical (3D)	0.7
Capped-cylinders (randomly aligned)	1.4

expected to be useful for a variety of systems in the continuum, such as those encountered in porous media[13] (e.g., cracks in rocks[14,15]), polymers,[16] composites,[17,18] heavily doped[9] or disordered[19] semiconductors and even nuclear matter.[20] As an illustration we shall discuss the "strange" low percolation threshold observed[17] in one of these systems.

In our search for a general criterion we started from the well known[2,6] macroscopically (or globally) isotropic system of parallel squares and followed the variation of $\langle A_{\rm ex}\rangle$ as this system was changed, in a controllable way, into a system of variable size perpendicular rectangles. This was done so that the system retained the macroscopic characteristics of the parallel-squares system while its microscopic characteristics changed considerably. Specifically, consider a system of rectangles in which rectangle i has a side of length L_{xi} (in the sample's side length unit[11,21]) parallel to the x axis, and a side of length L_{yi} parallel to the y axis. In the original system of squares, $L_{xi}=L_{yi}=L_{xj}=L_{yj}$ for all i and j. Now, introducing a given length distribution for both L_{xi} and L_{yi} one gets that generally $L_{xi}\neq L_{yi}$ (i.e., a system of rectangles). In order to maintain the macroscopic characteristics of the squares system, such as the isotropy and the $\langle L_{xi}\rangle=\langle L_{yi}\rangle$ relation, one has to pick the values of L_{xi} and L_{yi} independently but to use the same distribution function for both L_{xi} and L_{yi}. Microscopically this means that in a large enough sample, for each rectangle i there is a rectangle j such that $L_{xi}\approx L_{yj}$ and $L_{yi}\approx L_{xj}$.

The first distribution checked in our Monte Carlo computation[12] was the uniform distribution[10,14] in the range

$$L_M-f\leq L_{xi},L_{yi}\leq L_M+f \quad , \tag{2}$$

where L_M is the center of the distribution and $f(\leq L_M)$ is the half-width of the distribution. We have followed the critical concentration of rectangles N_c with increasing f and have thus determined

$$\langle A_{\rm ex}\rangle=4\langle L_{xi}\rangle\langle L_{yi}\rangle N_c \quad , \tag{3}$$

as a function of f. We found that $\langle A_{\rm ex}\rangle$ has decreased from the squares value of 4.5 (for $f=0$) to a value of 3.6 (for $f=L_M$). The samples used for this study[12] consisted of a few thousand rectangles each. For example, in the $f=L_M$ case, the value of L_M (which is equal to $\langle L_{xi}\rangle=\langle L_{yi}\rangle$) was taken to be 0.015 and the onset of percolation was found at $N_c=3930$.

The questions to be answered in view of the above results are the following: What changes have taken place in the system upon the broadening of the distribution and how do these changes bring about the decrease in the value of $\langle A_{\rm ex}\rangle$? Since the macroscopic characteristics were retained we should examine the microscopic changes and their effect on $\langle A_{\rm ex}\rangle$. The most noticeable transition is from a system dominated by squares to a system dominated by long but narrow rectangles. If this is an important reason then the limiting isotropic system made of horizontal and vertical widthless equal-length line segments ("sticks"[10]) should have a value of $\langle A_{\rm ex}\rangle$ which is lower than the above 3.6. The isotropy of the latter system is achieved by implanting randomly one horizontal stick for one vertical stick. Since the excluded area is zero for parallel sticks and is L^2 for perpendicular sticks of length L, the average excluded area per stick in this system is $L^2/2$. Carrying out a Monte Carlo computation for such a system[12] ($L=0.046$, $N_c=3000$) we found that $\langle A_{\rm ex}\rangle=(L^2N_c)/2=3.2$. Since this value is lower than the above 3.6 value, the expected importance of the large aspect ratio is confirmed.

This value of $\langle A_{\rm ex}\rangle=3.2$ has further significance if one recalls that in the isotropic system of randomly aligned widthless sticks the value of $\langle A_{\rm ex}\rangle=(2/\pi)L^2N_c$ was found[10,21] to be 3.57 (we use this accuracy[10] here in order to distinguish this value from the variable rectangles 3.6 value). This latter value, being smaller than the 4.1 value found for the randomly aligned capped rectangles (see above), reconfirms the importance of the large aspect ratio. On the other hand, the fact that this 3.57 value is larger than the 3.2 value of the horizontal-vertical system is probably associated with, what one may call, a higher degree of "microscopic" or "local" orientational anisotropy in the latter system. The rationale behind this is, that in all other isotropic sticks systems each stick has components in both the x and the y directions, while in the horizontal-vertical system each stick is aligned only in one direction. Since it appears from the above data that the large aspect ratio and the "local" anisotropy determine the value $\langle A_{\rm ex}\rangle$, we must conclude that the corresponding $\langle A_{\rm ex}\rangle$ value is the lowest for macroscopically isotropic systems. We expect also that every distortion of given convex objects which will leave them convex will render the microscopic characteristics of a system intermediate between those of the "parallel objects" system and those of the "horizontal-vertical" system. Hence, we can have a general percolation-threshold criterion for the continuum two-dimensional isotropic systems, made of convex, soft-core, objects:

$$3.2\leq\langle A_{\rm ex}\rangle\leq 4.5 \quad . \tag{4}$$

While the criterion (4) is consistent with the above value of 3.6, obtained for variable-size rectangles, and while it is clear that the larger aspect ratio and the larger "local anisotropy" are responsible for lowering this value with respect to 4.5, there may be still one other factor which should be considered as a potential contributor to the $\langle A_{\rm ex}\rangle$ value lowering. The factor is the length and width distributions of the rectangles which are not well accounted for by the above widthless sticks systems. This is due to the fact that in the widthless sticks system there is only one length parameter (the length of the ith stick, L_i) per object, rather than the two parameters (L_{xi} and L_{yi}) which exist in the system of variable rectangles. We have shown[10] that when there is only one such parameter (a length of a stick or a radius of a circle) the value of $\langle L_i^2\rangle N_c$ is an invariant under variations in the distribution function, while here we have to consider the rectangles quantity $\langle L_{xi}\rangle^2N_c$ [see Eq. (3)]. From the point of view of this difference the horizontal-vertical widthless sticks system (and thus the value of 3.2) may not be the limit of the system of variable rectangles. It is important to check then, for a given distribution, whether or not the limits given by Eq. (4) are still maintained. In the above example of the widest uniform distribution function ($f=L_M$) we found that $\langle A_{\rm ex}\rangle=3.6$ (which is larger than 3.2) and thus we conclude that the limits given by Eq. (4) apply for all uniform distributions. Trying other distribution functions (e.g., normal distribution) for which the mean of the distribution $\langle L_{xi}\rangle$ equals the "center," or the median, of the distribution, L_M, we reached the same conclusion. Hence, the effect of the width of such distributions seems to be of little importance. This makes Eq. (4) a very general criterion suitable for all practical purposes.

However, another group of extremely wide distribution functions has the property that $\langle L_{xi} \rangle \neq L_M$ and it may be that for such functions the difference due to the different $\langle L_i^2 \rangle$ and $\langle L_{xi} \rangle^2$ averages will show up by an $\langle A_{\rm ex} \rangle$ value which is lower than 3.2. To check this possibility we have applied a very wide log-normal distribution function[10,21] to the sides of the rectangles. The center of the distribution was $L_M = 0.005$ and its width was $\sigma = \frac{1}{2}\ln(10)$. The percolation threshold found was $N_c = 7762$. For this distribution it can be easily shown[10] that $\langle L_{xi} \rangle = L_M[\exp(\sigma^2/2)]$ and thus we find that $\langle A_{\rm ex} \rangle = 4\langle L_{xi} \rangle^2 N_c = 2.9$ which is somewhat lower than our lower limit of 3.2. Hence, even though the effect of the difference between the $\langle L_{xi} \rangle^2$ average and the $\langle L_{xi}^2 \rangle$ average $\{= L_M^2[\exp(2\sigma^2)]$ for the above distribution$\}$ is relatively small for such a wide distribution a correction of the 3.2 value may be required. The simplest correction to consider is using the value of 3.2 times $(\langle L_{xi} \rangle^2 / \langle L_{xi}^2 \rangle)$ as the lower limit in Eq. (4). All we can say at present is that the true lower limit is much larger than this estimate. A finer determination of this lower limit for such extremely wide distributions (in which the mean is not equal to the median) will require further studies.

The latter finding sheds light on the "surprisingly" very low critical fractional area ($s_c = 0.4$) found in the isotropic system of metal islands produced by laser speckles.[17] This value, which "corresponds to" $\langle A_{\rm ex} \rangle = 2.1$, is lower than the above 3.2 limit. This apparent disagreement with Eq. (4) would suggest that either Eq. (4) is not good enough or that this system of metal islands is different from the systems for which Eq. (4) was derived. Indeed, it has been realized already[19] that the metal islands tend to be concave rather than convex, as assumed here. We note[12] that if one insists[22] on describing the metal island system as made of convex metal particles which are deposited *randomly,* one has to assume that the metal islands are made of deposited subparticles which are not only elongated but have also an extremely wide size distribution.

Applying the above considerations to continuum systems in three dimensions we have derived similar conclusions. The "parallel-objects" limit, $\langle V_{\rm ex} \rangle = 2.8$, was found previously for many such systems[9,11] while a horizontal-vertical system was developed for the present work.[12] The "sticks" used here were capped cylinders[10] (length L radius r) which were taken to be elongated ($L/r = 30$) in order to approximate a "widthless" sticks system. For every stick put parallel to the x axis and the y axis, n sticks were put parallel to the z axis. As an example of the results for such a system we give here the result of present interest, i.e., the one obtained for the isotropic ($n = 1$) case. For $L = 0.15$ and $r = 0.005$ we found[12] that $N_c = 4330$. This yields[12] an average total critical excluded volume $\langle V_{\rm ex} \rangle$ of 0.7. The latter volume is to be compared with $\langle V_{\rm ex} \rangle = 1.4$, found for the capped cylinders which had their orientations distributed randomly.[10,11] As was the case for the two-dimensional sticks systems, we see that the higher "local orientational anisotropy" lowers the value of $\langle V_{\rm ex} \rangle$. Following this we conclude that the percolation threshold limits in three-dimensional systems of convex objects will be

$$0.7 \leq \langle V_{\rm ex} \rangle \leq 2.8 \quad . \tag{5}$$

For very wide object-size distributions we may further expect that the lower limit will be larger than $0.7(\langle L_{xi} \rangle^3 / \langle L_{xi}^3 \rangle)$.

In conclusion, new percolation criteria are presented for two- and three-dimensional systems in the continuum where the percolation is obtained by a path of overlapping convex objects. The lower limits of the criteria are determined by the new, horizontal-vertical, sticks systems.

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