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Quasiuniversal Connectedness Percolation of Polydisperse Rod Systems

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The connectedness percolation threshold (η_c) and critical coordination number (Z_c) of systems of penetrable spherocylinders characterized by a length polydispersity are studied by way of Monte Carlo simulations for several aspect ratio distributions. We find that (i) η_c is a nearly universal function of the weight-averaged aspect ratio, with an approximate inverse dependence that extends to aspect ratios that are well below the slender rod limit and (ii) that percolation of impenetrable spherocylinders displays a similar quasiuniversal behavior. For systems with a sufficiently high degree of polydispersity, we find that Z_c can become smaller than unity, in analogy with observations reported for generalized and complex networks.

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Idealized elongated objects such as perfectly rigid cylinders, spherocylinders, and prolate spheroids are prototypical models for a wide array of technologically relevant systems that include liquid crystals, nanocomposites based on filamentous fillers, as well as fiber-reinforced materials. Percolation phenomena involving dramatic increases in, e.g., structural rigidity and electrical and thermal conductivities of composites with increasing filler loading are currently of particular interest [1]. These increases are caused by the formation of an infinite cluster of in some sense connected particles at the critical loading, i.e., the percolation threshold.

It has been established by analytical [1–3] and numerical [4–11] studies that for dispersions of sufficiently elongated objects of identical size and shape, i.e, "monodisperse" objects, the geometric percolation threshold expressed in terms of the critical volume fraction of particles is inversely proportional to the aspect ratio of the filler particles. This property is exploited in the fabrication of conducting polymeric composites with very low conducting filler contents. Depending on the production processes of the composites, however, the filler particles almost invariably exhibit a pronounced polydispersity in both size and shape [12,13]. Although it represents a possible factor behind the huge quantitative discrepancies between theory and experiments [14,15], such polydispersity has received relatively little attention in terms of theoretical modeling until fairly recently [1,16,17]. Achievement of a theoretical understanding of how the continuum percolation of fibrous fillers is affected by polydispersity is thus key to the controlled design of a large class of composite materials for practical particle size and shape distributions.

Recent analytical results obtained from integral equation methods [16] and heuristic mapping onto a generalized Bethe lattice [17] predict that in the slender rod limit, where the particles have asymptotically large values of the aspect ratio, the volume fraction at the percolation threshold is inversely proportional to the weighted average $L_w = \langle L^2\rangle/\langle L\rangle$ of the rod lengths, where the brackets imply number averages over the distribution of rod lengths L. This Letter presents Monte Carlo (MC) results for the percolation threshold of isotropically oriented spherocylindrical particles with length polydispersity and having aspect ratios ranging from ~ 1 to several hundreds. We show that the percolation threshold of polydisperse interpenetrable spherocylinders is a nearly universal function of L_w over the entire range of aspect ratios considered. In addition, the percolation threshold closely follows the predicted $1/L_w$ behavior even for particles with aspect ratios that are considerably smaller than the slender rod limit, thus generalizing the current theory.

For systems of *impenetrable* spherocylinders with fixed $\sqrt{\langle L^2\rangle}/D$, where D is the diameter of the hard core, we show that the percolation threshold is nearly independent of the length distribution. Finally, we find that the critical coordination number per particle at the percolation threshold (denoted Z_c) can be smaller than unity for polydisperse systems. Although similar observations have been reported for a number of complex networks and in systems of hyperspheres in high-dimensional spaces [18], this finding is novel in the context of the continuum percolation of three-dimensional objects.

We generate isotropically oriented distributions of penetrable rods by randomly placing N penetrable spherocylinders with a distribution of lengths L and identical diameter δ within a cubic box with periodic boundary conditions and side length $\mathcal{L}$. As a measure of the concentration of the spherocylinders, we shall use the dimensionless density $\eta = \rho\langle v\rangle$, where $\langle v\rangle = (\pi/6)\delta^3 + (\pi/4)\delta^2\langle L\rangle$ is the number-averaged volume of the spherocylinders, $\langle L\rangle = \int dL L f(L)$ is the mean (number-averaged) rod length for a given distribution $f(L)$ of lengths, and $\rho = N/\mathcal{L}^3$ is the number density of the particles [19].

We consider two spherocylinders connected if they overlap geometrically. The percolation threshold is identified by ascertaining the minimal diameter δ_c (for fixed value of ρ) for which a cluster of connected particles spans the entire cubic box. This definition is equivalent to the usual procedure of finding a critical density ρ_c of spherocylinders with fixed diameter and has the additional advantages of (i) being computationally more convenient and (ii) allowing a more direct relation to the conductivity σ of rods through the critical distance approximation $\sigma \propto \exp(-2\delta_c/\xi)$, where ξ is the tunneling decay length [11].

In the following, we shall use the critical distance δ_{c0}, defined as $\delta_{c0} = 2/\pi\rho\langle L\rangle^2$, as our unit of length for polydisperse systems of penetrable spherocylinders. This quantity corresponds to the critical distance obtained from the second virial approximation formula $\eta_c = (1/2)\delta_{c0}/L$ for the critical concentration of a system of monodisperse spherocylinders with identical lengths $L \gg \delta_{c0}$ chosen to coincide with $\langle L\rangle$.

To find δ_c, we employ the clustering method described in Ref. [20], which allows computation of the spanning probability as a function of the spherocylinder diameter δ for fixed density ρ (Supplemental Material [21]). Figure 1(a) shows the results obtained for polydisperse systems with a bimodal length distribution $f(L) = p\delta(L - L_1) + (1 - p)\delta(L - L_2)$ with $L_2 = 20$ and $L_1 > L_2$, where $0 \le p \le 1$ is the number fraction of long rods. In the figure, we display the ratio R (symbols) of the critical distances for the polydisperse rod system to those for monodisperse systems of spherocylinders with lengths equal to $\langle L\rangle = \int dL L f(L)$, which for the particular distribution considered corresponds to $\langle L\rangle = pL_1 + (1 - p)L_2$. The ratio R of the critical distances is systematically reduced by polydispersity and displays a minimum that becomes deeper and moves towards smaller values of p as L_1/L_2 is increased, implying that a small fraction of longer rods can substantially lower the percolation threshold.

This trend is in full agreement with the theory of Ref. [16] based on the second virial approximation to the connectedness Ornstein-Zernike equation, which predicts for $\langle L\rangle/\delta_c \gg 1$

$$\eta_c = \frac{1}{2}\frac{\delta_c}{L_w}, \tag{1}$$

where $L_w = \langle L^2\rangle/\langle L\rangle$ is the weight-averaged rod lengths. From this equation and by using $\eta_c \simeq \rho(\pi/4)\delta_c^2\langle L\rangle$ for $\langle L\rangle/\delta_c \gg 1$, the critical distance is predicted to follow $\delta_c = 2/\pi\rho\langle L^2\rangle$. The reduction factor $R_0 = \delta_c/\delta_{c0}$ predicted by the theory is thus

$$R_0 = \frac{\langle L\rangle^2}{\langle L^2\rangle} = \frac{(pL_1/L_2 + 1 - p)^2}{p(L_1/L_2)^2 + 1 - p}, \tag{2}$$

where the second equality applies for the bimodal length distribution. As shown in Fig. 1(a) the MC findings for R are in semiquantitative agreement with Eq. (2) (solid lines), although R is consistently slightly smaller than R_0. This discrepancy could arise from the circumstance that the values of L_1 and L_2 used in the simulations may be insufficiently large to achieve the slender rod limit, which is a prerequisite for the validity of Eq. (2). We examine this issue in Fig. 1(b), which shows R/R_0 as a function of L_2 (up to $L_2 = 150$ in units of δ_{c0}) for $L_1/L_2 = 3$ and selected values of p. For $L_2 \ge 20$, our MC results are less than 10% smaller than $R/R_0 = 1$. Furthermore, for $L_2 \ge 50$, R/R_0 appears to increase monotonically (albeit somewhat slowly), which may indicate that the slender rod length limit $R/R_0 = 1$ could ultimately be reached (for any p) only for very long rod lengths.

The MC results shown in Fig. 1 and the relatively small deviations from Eq. (2) suggest that, for rods with identical radii, L_w is the key quantity that controls the percolation threshold for mixtures of penetrable spherocylinders. This is demonstrated in Fig. 2 where η_c is shown as a function of L_w/δ_c for various bidisperse (open symbols) and monodisperse (+ signs) systems of spherocylinders (in the

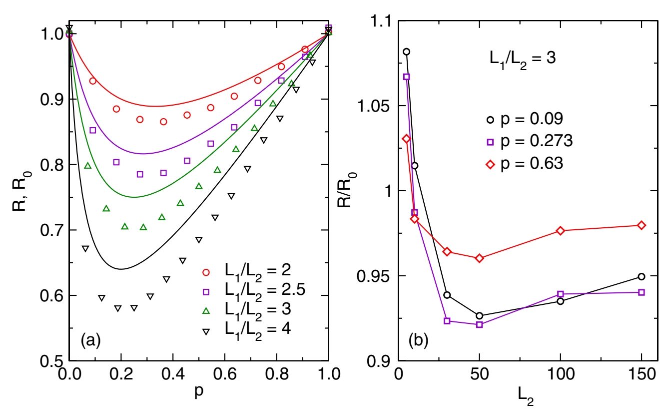

FIG. 1 (color online). (a) Critical distance ratio R between polydisperse and monodisperse spherocylinders as a function of the fractional occupancy p of the longer rods with shorter rod length fixed at $L_2 = 20$ in units of $\delta_{c0} = 2/\pi\rho\langle L^2\rangle$ (see text). The solid lines represent Eq. (2). (b) The critical distance ratio R in units of R_0 calculated from Eq. (2) for $L_1/L_2 = 3$ as a function of the length L_2 of the shorter rods and for selected values of p.

latter case L_w is identical to the unique particle length). Results for systems of spherocylinders for which the lengths follow Weibull and uniform distributions are shown in Fig. 2 by filled circles and squares, respectively (see the Supplemental Material [21] and Ref. [22]). Surprisingly, all of our data collapse onto a single curve over the entire range of $L_w/\delta_c > 1$, implying that η_c is a quasiuniversal function of L_w/δ_c independent of the particular distribution considered.

This finding is rather unexpected because the observed quasiuniversality extends well below the slender rod limit of Eq. (1) (solid line), which is approached by the MC data to within less than 10% only for $L_w/\delta_c \gtrsim 200$. Furthermore, even though Eq. (1) might be expected to apply only asymptotically for $L_w/\delta_c \gg 1$, we observe that the data for $L_w/\delta_c \gtrsim 10$ are well fitted by $a(L_w/\delta_c)^{-\beta}$ with $a = 0.165 \pm 0.009$ and $\beta \simeq 1.080 \pm 0.002$. The inverse scaling of η_c with L_w thus applies approximatively even for spherocylinders with very modest aspect ratios.

We have also examined the effects of length polydispersity on the percolation of impenetrable spherocylinders with identical hard-core diameters D. Two impenetrable spherocylinders ($D \neq 0$) are considered connected if their surfaces approach closer than δ. The percolation threshold for a given density ρ of the particles is identified, as before, by the critical distance δ_c. In the slender rod

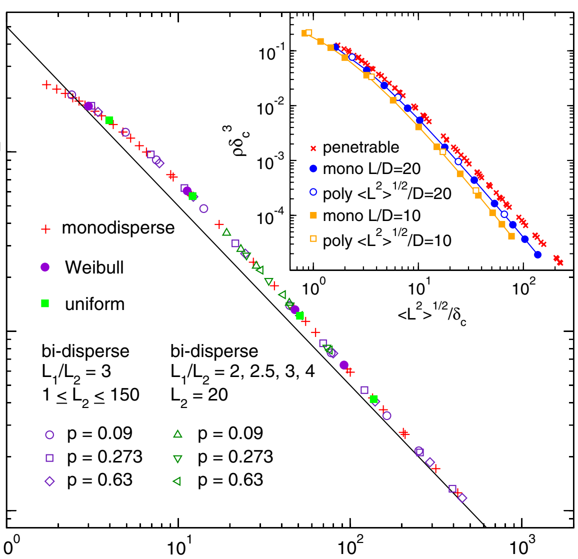

FIG. 2 (color online). Critical reduced density η_c as a function of L_w/δ_c for monodisperse (plus symbols), bidisperse (open symbols), Weibull (filled circles), and uniform (filled squares) distributions of spherocylinder lengths (see the Supplemental Material [21] and Ref. [22]). The solid line represents Eq. (1). Inset: $\rho\delta_c^3$ as a function of $\sqrt{\langle L^2\rangle}/\delta_c$ calculated for monodisperse and bidisperse impenetrable spherocylinders with hard-core diameter D. The x symbols are the results for both the monodisperse and polydisperse penetrable rods of the main panel.

limit, the percolation threshold of impenetrable rods is predicted to follow $\phi_c = D^2/(2\delta_c L_w)$ [16,17], where $\phi_c \simeq \rho(\pi/4)D^2\langle L\rangle$ is the critical volume fraction for the hard-core particles. By noting that Eq. (1) is the percolation threshold for penetrable rods and since $\eta_c \simeq \rho(\pi/4)\delta_c^2\langle L\rangle$, we see that for sufficiently elongated rods the percolation relation

$$\rho\delta_c^3 = (2/\pi)\delta_c^2/\langle L^2\rangle \tag{3}$$

is predicted to be satisfied by both hard and penetrable rods, independent of their length distribution.

We have generated by MC simulations equilibrium dispersions of impenetrable spherocylinders with different length (L) distributions. The inset of Fig. 2 shows $\rho\delta_c^3$ as a function of $\sqrt{\langle L^2\rangle}/\delta_c$ for monodisperse systems with $L/D = 10$ and 20 (filled symbols) and for two bidisperse cases with L_1, L_2, and p chosen as to give $\sqrt{\langle L^2\rangle}/D = 10$ and 20 (open symbols) (see Supplemental Material [21]). Although for computational reasons the rod lengths considered by us are not large enough for our results to fulfill Eq. (3), we see nevertheless that for a given $\sqrt{\langle L^2\rangle}/D$, $\rho\delta_c^3$ is essentially independent of the particular rod length distribution. Furthermore, the calculated $\rho\delta_c^3$ values for increasing $\sqrt{\langle L^2\rangle}/D$ tend to follow the same functional behavior of the interpenetrable spherocylinders (x signs in the inset of Fig. 2) [23]. This latter feature suggests that for sufficiently large $\sqrt{\langle L^2\rangle}/D$ there exists a universal relation of the form $\rho\delta_c^3 = F(\sqrt{\langle L^2\rangle}/\delta_c)$, which is expected to reduce to Eq. (3) for $\sqrt{\langle L^2\rangle}/\delta_c \gg 1$ and that applies to both penetrable and interpenetrable spherocylinders over a wide range of $\sqrt{\langle L^2\rangle}/\delta_c$ values.

Although currently there is no theoretical explanation for the quasiuniversal dependence reported in Fig. 2, a partial understanding may be achieved by following the method developed in Ref. [16]. In this formalism, applied here for simplicity to penetrable rods, the overall cluster size S satisfies $S = \langle T(L)\rangle_L$ where $T(L) - \rho\langle\hat{C}^+(L, L', \delta_c)T(L')\rangle_{L'} = 1$ and $\hat{C}^+(L, L', \delta_c)$ is the orientation-averaged connectedness direct correlation function at zero wave vector. Within the plausible ansatz $\hat{C}^+(L, L', \delta_c) = LL'c_{11} + (L + L')\delta_c^2 c_{10} + \delta_c^3 c_{00}$ [24], where the coefficients $\{c_{ij}\}$ are assumed to depend only upon the packing fraction, it is found that for systems with different length distributions but equal values of L_w/δ_c, S diverges at percolation thresholds that differ by $\sim\sigma_s^2\delta_c/L_w$ for $L_w/\delta_c \gg 1$ and $\sim(L_w/\delta_c)\sigma_s^2/(1 + \sigma_s^2)$ for $L_w/\delta_c \ll 1$, where $\sigma_s^2 = \langle L^2\rangle/\langle L\rangle^2 - 1$ is the scaled variance. Since the scaled variances for all length distributions considered in this work were always smaller than $\sim$50% (see Supplemental Material [21]), for $L_w/\delta_c \gtrsim 10$ the expected deviation from universal behavior is thus only $\sim\sigma_s^2\delta_c/L_w \lesssim 5\%$, which is consistent with the results of Fig. 2. Although we expect that systems with values of σ_s^2

much larger than those considered by us would imply a stronger deviation from universality, $\sigma_s^2 \lesssim 0.5$ is nevertheless representative of the scaled variances observed in several real polydisperse systems of rodlike particles [12,13].

The quasiuniversal dependence of the percolation threshold upon L_w implies a general nonuniversality of the critical coordination number Z_c, where Z_c denotes the average number of contacts per rod at the percolation threshold. This is best viewed for the case of randomly placed and oriented overlapping objects for which $Z_c = \eta_c\langle v_{\rm ex}\rangle/\langle v\rangle$, where $\langle v_{\rm ex}\rangle$ is the excluded volume averaged over the orientations and the rod lengths. Given that η_c depends on L_w/δ_c, while

$$\frac{\langle v_{\rm ex}\rangle}{\langle v\rangle} = 8 + 3\frac{(\langle L\rangle/\delta_c)^2}{1+(3/2)\langle L\rangle/\delta_c} \tag{4}$$

depends on the rod lengths through $\langle L\rangle/\delta_c$, we see that mixtures of rods with equal η_c values (i.e., equal L_w) may have rather different Z_c values if the distribution $f(L)$ of the rod lengths is such that $\langle L\rangle \neq L_w$.

Figure 3 shows the critical average number of connections per rod calculated from $Z_c = \eta_c\langle v_{\rm ex}\rangle/\langle v\rangle$ for the same mixtures of penetrable spherocylinders considered in Fig. 2. We have verified that the configurational average of the connection per rods at δ_c coincides with the excluded volume formula, as expected. We see that in general, Z_c is sensitive to the extent of polydispersity, although in the limit $L_w/\delta_c \to 0$ it can be expected that Z_c should coincide with the result for identical overlapping spheres, namely, $Z_c \simeq 2.74$ [25]. In particular, Z_c is always larger than unity and approaches $Z_c \to 1$ asymptotically in the slender rod limit for monodisperse systems. In contrast, for distributions with sufficiently large values of the variance and of L_w/δ_c, polydisperse systems of rods may display fewer than one connection per particle at the threshold, i.e., $Z_c < 1$.

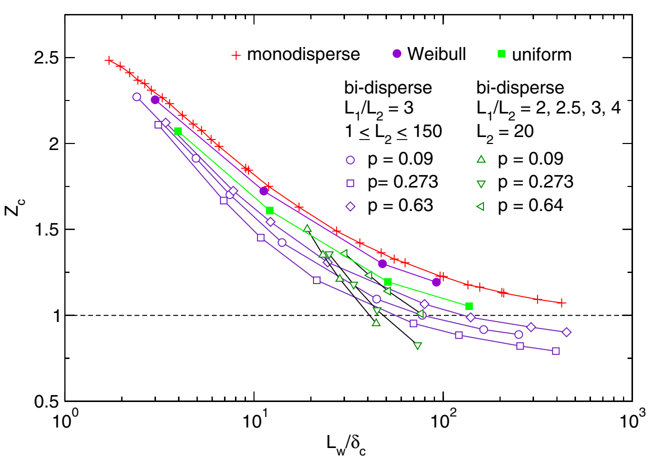

FIG. 3 (color online). Critical coordination number Z_c as a function of L_w/δ_c for polydisperse and monodisperse spherocylinders. The symbols have the same meaning as in the main panel of Fig. 2.

This latter feature is somewhat novel since the continuum percolation of objects randomly dispersed in a three-dimensional space is usually characterized by the condition $Z_c \geq 1$ [18]. Indeed, to the best of our knowledge, percolation occurring with $Z_c < 1$ has been reported only for penetrable identical hyperspheres in spaces of dimensionality exceeding 12 [18] and in random or complex networks that are *not* embedded in a physical space. For example, given an uncorrelated network with nodes having a distribution of coordination numbers z, upon random removal of nodes the network becomes disconnected at a critical node occupation probability $p_c = \langle z\rangle/(\langle z^2\rangle - \langle z\rangle)$ [26,27], which results from the irrelevance of closed loops [28]. The critical coordination number $Z_c = p_c\langle z\rangle$ is thus

$$Z_c = \frac{\langle z\rangle^2}{\langle z^2\rangle - \langle z\rangle}, \tag{5}$$

which can be smaller than unity when the node degree distribution is such that $\langle z^2\rangle/\langle z\rangle - \langle z\rangle > 1$.

The results of Fig. 3 that show that polydisperse rod mixtures may display $Z_c < 1$ suggest that these systems may relate to such classes of generalized graphs that can exhibit the same feature [29]. Indeed, as shown in Ref. [17], Eq. (5) is also the critical coordination number of a generalized Bethe lattice that by construction lacks closed loops. Hence, by applying the mapping $\langle z\rangle \to 2\langle L\rangle/\delta_c$, $\langle z^2\rangle \to 4\langle L^2\rangle/\delta_c^2$ formulated for polydisperse slender rods in Ref. [17], we find

$$Z_c \to \frac{\langle L\rangle^2}{\langle L^2\rangle} = \frac{\langle L\rangle}{L_w} \leq 1, \tag{6}$$

which is qualitatively consistent with the behavior of Z_c seen in Fig. 3. A physical explanation for the observation that $Z_c < 1$ for sufficiently polydisperse rod systems is provided by the fact that the percolating cluster is predominantly comprised of the longer rods in the system, and the shorter rods have a greater likelihood of being isolated [17]. An interesting corollary arising from this interpretation is that, as in generalized random networks where targeted removal of highly connected nodes enhances the percolation threshold [26,27], preferential removal of the longer rods from the system may lead to similar enhancement of the critical concentration.

In conclusion, we have studied by MC simulations the effects of length polydispersity on the percolation threshold of penetrable spherocylinders. We find a quasiuniversal dependence of the percolation threshold on L_w that extends well below the slender rod limit considered in Refs. [16,17]. For systems of impenetrable spherocylinders, we find that universality is fulfilled for a given $\sqrt{\langle L^2\rangle}/D$, where D is the hard-core diameter. The predicted quasiuniversality could be tested by experiments in systems of conducting fibrous fillers by altering the distribution of the rod lengths, e.g., by sonication, and measuring the resulting change in the percolation threshold. Furthermore, we have demonstrated that the average number of connections per rod at the percolation threshold can be smaller than unity for random distributions of rods that are sufficiently slender and polydisperse. This finding reveals an intriguing analogy with the case of random percolation in complex networks.

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[22] The systems with Weibull distribution have been generated by considering rods with lengths $L_i = i$ ($i = 1, 2, 3, \ldots$) distributed according to the discretized Weibull probability function $f(L_i) = \exp[-(L_i/\lambda)^k] - \exp[-(L_{i+1}/\lambda)/^k]$ with $k = 6$ and $\lambda = 3$, 15, 60, and 110. Uniform distributions of rods have been constructed from $f(L) = 1/(L_1 - L_2)$ for $L_2 \le L \le L_1$ and $f(L) = 0$ otherwise, with $L_1/L_2 = 4$ and $L_2 = 1$, 5, 20, and 50.

[23] When $\rho\delta_c^3$ is plotted as a function of $\sqrt{\langle L^2\rangle}/\delta_c$, the system of penetrable rods displays a (quasi-) independence on the length distribution similar to that observed from the η_c versus L_w/δ_c plot of the main panel of Fig. 2.

[24] The form of this ansatz is suggested by dimensional considerations and by the fact that the direct correlation function is a local quantity. The coefficients c_{11}, c_{10}, and c_{00} represent respectively the contributions arising from cylinder-cylinder, cylinder-cap, and cap-cap correlations. For $\eta \to 0$, they reduce to the second virial approximation $c_{11} = \pi/2$, $c_{10} = \pi$, and $c_{00} = 4\pi/3$.

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