Finite-Size Scaling Features of Electric Conductivity Percolation in Nanocomposites

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Abstract Using conductive nanocomposites for bipolar plates in fuel cells can improve their performance. Percolation is the mechanism for nanocomposite conductivity. When the volume fraction of fillers in a composite material reaches a critical value, percolation starts to happen. If the composite material has an infinitesize, the probability of conductivity jumps from zero to 100% at the critical volume fraction. However, for finite-size composites, the probability would rise in a Gaussian-error-like smooth curve function. This research uses Monte Carlo simulations to study the percolation probability of finite-size nanocomposites cubes. The analyses show that there are two critical volume fractions. As the cube size approaches infinity, the two critical volume rates converge and should be equal to the theoretical value. Using the normal-cumulated-distribution function model, a power-law equation is obtained to estimate the critical volume fraction from the corresponding nanocomposite cube size. However, the Monte-Carlo simulations in this research have been based on a uniform distribution of nanotubes in the composite cube. In practical implementation, the CVF obtained from this research can be treated as the lower bound of possible real values.

Keywords Nanocomposites, percolation, finite-size scaling, Monte-Carlo Simulation

1. Introduction

Energy sustainability is one of the major areas that researchers have focused on in the past decades. In a United States Department of Energy [1] report on nanoscience for energy needs, research targets having the greatest impact are identified; one of these targets is to build low-cost fuel cells using nanostructured materials.

Bipolar plates play a crucial role in a fuel cell's long-term performance. The development of bipolar plates is one of the major factors that limit the commercialization of fuel cells

[2]. Researchers have suggested the use of nanocomposite materials, which have better mechanical strength, smaller volume and weight, and lower cost compared with other materials [2-4], for bipolar plates.

The bipolar plates in a fuel cell need good conductivity, but polymer matrices in nanocomposites have very low conductivities. This electrical conductivity can be increased by increasing the amount of conductive nano-sized fillers in the materials from which plates are made[3]. However, there is a limit on adding fillers into matrices. To achieve the best results, it is important to understand percolation theory, a theory that explains why a non-conductive polymer matrix can be conductive.

Percolation is a physical phenomenon of the filtration of fluid through porous materials. It has been expanded into many other areas throughout the decades. Broadbent first suggested a model of the phenomena with Monte Carlo simulations [5]; it has since sparked persistent studies among researchers and resulted in numerous applications not only in material science [6, 7], but also in geophysics [8], environmental science [9], medical research [10], and even social science as well [11].

The problem of a percolated material's electric conductivity rose naturally in earlier years [12] and remains a focus of intense experimental, theoretical, and simulation studies. After the 1990's, due to the robust properties discovered about carbon nanotubes (CNTs) dispersed in polymers [13], the validity of various percolation models to account for the behavior of CNT via Monte Carlo simulations has also drawn significant attention [14-18]. Thus far, the simplistic soft-core, monodisperse spherocylinder model has been argued and shown to be adequate for comparison with experimental results and will be the approach this work is based on [19-21].

2. Percolation Theory

The theory of percolation was originally developed for studying the gelation of colloidal suspensions [22]. Simon

R. Broadbent [5], after a short note he made during the 1954 symposium on Monte Carlo methods, popularized this problem in a work with Hammersley [23]. The studies that followed in the 1960's established some formal definitions and suggested exact solutions [24-26]. But even to this day, mathematically rigorous treatments have gained only limited success [27, 28]. The models involve the bonds (or sites) of a regular lattice (2D square, 2D hexagonal, 3D face centered cubic (FCC), etc) being 'opened' independently with the identical probability p , and percolation is defined to be the appearance of an infinitely large open-bond (site) cluster. As p increases from zero toward a critical value φ_c , the percolation probability (the probability of any infinite cluster to exist), denoted as $\pi(p)$, makes a jump from zero to one as if it is a step-function. The exact value of φ_c and the behavior of $\pi(p)$ as well as other geometric quantities such as mean cluster size, correlation length, and conductivity are of interest.

As regular lattices may not serve to model many physical systems that are continuous in nature, the critical volume fraction (CVF), denoted as φ_c , was introduced in 1970 as an invariant for the lattice problem [29]. It can be viewed as a natural link between lattice percolation and continuum percolation, which was developed in parallel and not directly related.

In the continuum case, fillers of a certain shape (for examples, discs, spheres, rods, etc.) are randomly inserted into another wise homogeneous continuum (medium). The insertions may connect or overlap with each other and form pathways in a manner similar to that of open-bond (sites) clusters in the lattice case. With more and more fillers, the onset of any huge pathways that span to infinity is recognized as the critical point of percolation. A measure of the amount of fillers therein, such as the volume fraction, is of interest, along with the percolation probability $\pi(p)$ that depends on it. While exact solutions have remained elusive for both the lattice and the continuum cases, approximations and effective theory in the 1970's provided as much insights as quantitatively accurate results for both.

One of them is the critical-phenomena theory and related techniques developed for physical phase transition in the 60's. It was naturally applied to percolation problems as critical phenomena often share similar traits [30, 31]. The resultant finite size scaling theory has laid the analytical foundation for a number of power laws and conjectures for the relevant exponents. While the mathematical community may only accept them as heuristic [27], no significant exceptions have been reported so far to the best of the authors' knowledge. The lattice version of the scaling laws has also been widely accepted [32, 33].

3. Finite-size Scaling Theory

When the system is finite, percolation is defined as the appearance of any large cluster that spans the system. One of the analytical consequences, and actually one basis to

estimate critical exponents, is that the percolation probability $\pi(p)$ is no longer a step function, but rather a Gaussian-error-like smooth curve function [34-36]. This behavior has been reported explicitly or can be implied from many simulation works regarding different configurations of fillers (lattice, continuum, shapes, aspect ratios etc) in two or three dimensions [37-41]. However, simulation studies regarding monodisperse isotropic 3D cylinders (capped or uncapped) have not reported this behavior; they have mainly concentrated on the critical exponents, the conjectured excluded volume law and their dependence on the aspect ratio of the fillers [42, 43]. This work reports explicitly for the first time a systematic simulation result for that matter.

To conclude this section, a minor point that is often neglected about the definition of volume fraction φ is worthy of note. The practically convenient and intuitively straight forward way to define volume fraction is to take the ratio of the total volume of the fillers to the total volume of the system:

$$\varphi \equiv \frac{N \cdot V_{cyl}}{V} \quad (1)$$

where N is the number of fillers present, V_{cyl} is the volume of a single filler (spherocylinder), and V is the total volume of the system. Equation (1) serves perfectly as the definition for hard-core (completely impermeable) fillers as the volumes do not overlap; on the other hand, for the soft-core objects, the standard probability theory results in:

$$\varphi_{soft} \equiv 1 - e^{-\varphi} \quad (2)$$

where φ should be interpreted equivalently as the uniform volume density of the fillers [44-46]. Note that Equation (2) represents the basic idea of Monte Carlo methods: estimate the volume fraction (or any variable of interest with corresponding modification) as the probability of any given point being inside the filler.

Nonetheless, in this work, the definition of Equation (1) will be used throughout for the following three reasons: (i) There is a one-to-one correspondence between φ and φ_{soft} ; comparing φ is equivalent to comparing φ_{soft} . (ii) The nanocomposites of concern usually exhibits CVF lower than a few percentage points, thus in this limit: $\varphi \ll 1, \varphi_{soft} \approx \varphi$. (iii) Many works have ignored this fact, and the above reasons have validated the use of Equation (1) to compare our results with others directly.

4. Problem Statement

Conductive nanotubes are dispersed in the non-conductive polymer to form a nanocomposite three-dimensional cube. The nanotubes that are in contact form a conductive cluster. If any cluster spans from one wall to the opposite wall, the cube becomes conductive.

The goal is to fabricate the nanocomposites and achieve percolation conductivity with high certainty.

The minimum volume fraction of nanotubes in the composite that is sufficient to reach conductivity is called the critical volume fraction (CVF). In theory, with volume fraction below CVF, the nanocomposite exhibits no conductivity; on the other hand, a volume fraction above CVF implies conductivity. In reality, however, due to the randomness nature of the nanotube dispersions, the percolation conductivity depends on the relative positions of the nanotubes. One can therefore argue that CVF by itself is a random variable. One of the objectives of this research is to use Monte Carlo simulations to study the statistical properties of CVF.

Also, randomness can cause uncertainties in production. Assume that the theoretical CVF of a certain nanocomposite can be decided. However, using a nanotube fraction ratio above CVF to fabricate the nanocomposite may not guarantee percolation. Therefore, it is important to obtain the theoretical probability of the percolation conductivities in various nanotube fraction ratios. In production, those values will be the upper limit of the confidence level of achieving percolation conductivity.

5. Geometric Modeling

The nanotube is modeled as a spherocylinder, or a cylinder capped with two hemispheres on both ends. As shown in Figure 1, L is the length of the cylinder alone and R is the radius of each hemisphere cap as well as the radius of the cylinder. There are three modes of nanotube contacts: side-to-side, cap-to-cap, and cap-to-side, as shown in Figure 2.

Figure 1. Nanotube model

Figure 2. Nanotube contact models

Referring to Figure 3, the two nanotubes are represented

by $\overline{AB}$ and $\overline{PQ}$, where A , B , P , and Q are at the centers of the respective hemispherical caps. Using the soft-core model, if nanotubes $\overline{AB}$ and $\overline{PQ}$ are in contact, then the shortest distance between them is less than or equal to $2R$. Hence, the algorithms that determine whether two nanotubes are in contact will need the calculations of the shortest distance between the nanotubes in the three-dimensional space. The detailed procedures are discussed below.

From an arbitrary point O on $\overline{AB}$ draw an arrow $\overrightarrow{OW}$ that is parallel to $\overline{PQ}$ (see Figure 3). The plane defined by $\overline{AB}$ and $\overrightarrow{OW}$ is denoted as plane H in Figure 3, which contains $\overline{AB}$ and is parallel with $\overline{PQ}$. Line $\overline{CD}$ is the projection of $\overline{PQ}$ on plane H . Let X and Z be two arbitrary points on line $\overline{AB}$ and line $\overline{PQ}$, respectively, and Y be the projection of point Z on plane H . The distance between X and Z is equal to $\sqrt{(ZY)^2 + (YX)^2}$, where ZY and YX are the lengths of $\overline{ZY}$ and $\overline{YX}$, respectively. Since $\overline{CD}$ is in parallel with $\overline{PQ}$, the length of $\overline{ZY}$ is the same no matter where point Z is on $\overline{PQ}$. Now the problem has been simplified to “finding points X and Y so that the distance YX between two coplanar lines $\overline{AB}$ and $\overline{CD}$ is the shortest.” This is a relatively simple two-dimensional problem and will be omitted here for brevity.

After the shortest distance has been obtained, whether the two nanotubes are in contact can be determined by comparing it to $2R$, as discussed above. Next, list the nanotube pairs that are in contact and identify the conductive clusters from it. If at least one cluster spans in at least one dimension of the cube from wall to wall, the cube has percolation conductivity.

Figure 3. Two nanotubes in a 3-D space

6. Critical Volume Fraction Study

The followings are the procedures for finding the critical volume fraction (CVF) for a cube of known size:

- Add one nanotube to the polymer cube at a random position;
- Check if there is percolation conductivity;

- a. If no, go back to step (i);
- b. If yes, calculate CVF and exit the procedure.

The random positions of the nanotubes are generated from a random number generator. If the seed of the random number generator changes, the positions generated will be different; hence, the CVF obtained from the above procedure will also be different. The dependence of CVF on a random seed implies that CVF behaves like a random variable.

A Matlab program is coded using the procedures in this section. Using 1,000 different random seeds to obtain 1,000 CVF values, the results are shown in Figure 4. Figure 5 shows the histogram of the data in 100 bins.

The data in Figure 4 shows that the critical volume fractions, ϕ_c , are random and Figure 5 shows a distribution close to the normal distribution.

Figure 4. Critical volume fractions for 1,000 random seeds

Figure 5. Histogram of critical volume fractions for 1,000 random seeds

7. Percolation Conductivity Probability Study

In this section, Monte Carlo simulations are used to estimate the probabilities of reaching percolation conductivity. A typical result is shown in Figure 6, which are the results of 1,000 runs for each volume fraction. The horizontal axis is the volume fraction (in percent) and the vertical axis represents the corresponding probability of percolation (in %). When the volume fraction is below 0.45%, there is very little possibility of getting percolation. As the volume fraction increases, the probability starts to increase, first slowly and later, above 0.55%, faster. At 0.65% volume fraction, over 99% of the runs result in percolation.

The curve in Figure 6 is similar to the “Gauss-error-function-like smooth curve”, which has been also reported in other papers for different configurations. The classical concept of critical volume fraction (CVF) is based on the assumption that when percolation starts to happen, the probability jumps from 0 to 100%, acting like a step function; therefore, CVF is a single value. From the above example, one can see two possible values can be associated with CVF, at 0.5%, when percolation “could” happen (with a very low probability), and at 0.65%, when percolation “most likely” will happen (with a very high probability). From a manufacturing point of view, the second value should be more significant.

Figure 6. Probability of conductive percolation in different volume fractions

Figure 7 shows the percolation probability curves for various aspect ratios L/D , where L is length and $D = 2R$ is the diameter of the tubes. The results show that using higher aspect ratio nanotubes can reach conductivity percolation at lower fraction ratios. Most nanotubes have aspect ratios higher than 100 and hence even lower CVF's can be expected. However, it is very hard to achieve a uniform distribution outside of simulations. Also, nanotubes usually have non-uniform lengths and diameters, and hence non-uniform aspect ratios. The CVF's obtained

in simulation can thus be used as the upper-limit in the design of the conductive nanocomposites.

Figure 7. Probability of conductive percolation for different aspect ratios

8. Percolation Conductivity Probability Study

The Monte Carlo simulation results of the percolation probabilities for different cube sizes are shown in Figure 8. From the results of the three sizes tested, the trend shows that as the sizes increase, it will approach a hypothetical step function for an infinitely large cube. The simulations of nanotubes with other aspect ratios also show a similar trend; this is shown in Figure 8-11. In all cases, the length of each nanotube $L = 0.01$ and each figure has a different diameter D .

From those figures, it is reasonable to assume that as the size of the cube approaches infinity, the curves will approach a step function with the CVF somewhere between the volume fractions when the probability becomes non-zero and reaches 100%. Figure 12 also shows three other papers' estimations of CVF. They will be described in the next paragraph.

Figure 8. Probability for cube sizes 0.025, 0.05, and 0.1, $L/D = 100$

Figure 9. Probability for cube sizes 0.025, 0.05, and 0.1, $L/D = 50$

Figure 10. Probability for cube sizes 0.025, 0.05, and 0.1, $L/D = 20$

Figure 11. Probability for cube sizes 0.025, 0.05, and 0.1, $L/D = 10$

Figure 12. CVF estimated using the model in other papers, $L/D = 100$

Berhan and Sastry [47] have used numerical experiments to determine the parameters in the formulas modified from the excluded volume approach. For aspect ratio 100, the soft-core model formula gives CVF = 0.608%, which agrees with the results in this paper the best. Hu et al [49] also used numerical simulation to come up with a simple heuristic formula that gives $CVF = (L/D)^{-1.1} = 100^{-1.1} = 0.631\%$. Foygel et al [19] calculate CVF based on nanotube loading that gives $CVF = (V/V_{ex}) * 1.2 = 0.581\%$, where V is the nanotube volume, V_{ex} is the excluded volume. From Figure 12, the simulation data match the theoretical models in those papers reasonably well.

9. Normal Cumulated Distribution Function Model

In the previous sections, it has been demonstrated that the percolation probability versus volume fraction curves have the features of a Gaussian error function (Figure 6). Comparing curves for cubes in different sizes, as in Figure 8-11, it is obvious that as the cube size increases, the curve becomes steeper after percolation starts to happen. Therefore, it is reasonable to hypothesize that as the cube size approaches infinity, the curve turns into a step function. This is consistent with the infinitively-large-cluster percolation definition of CVF: the volume fraction at which the percolation probability jumps from 0% to 100%. For the finite size cubes, the CVF definition becomes “when percolation starts to happen”, as discussed in Section 7.

To model those percolation probability curves, the cumulated distributed function (CDF) of the normal (Gaussian) distribution is used. A normal probability distribution $\phi = N(\mu, \sigma)$ with mean μ and standard deviation σ has a cumulated distribution function Φ given by

$$\Phi(x) = \frac{1}{2} \left[1 + \operatorname{erf} \left(\frac{x - \mu}{\sigma\sqrt{2}} \right) \right] \quad (3)$$

Here erf is a special function called error function. As σ approaches zero, the accumulated function becomes a step function.

Monte Carlo simulations as described in Section 7 are run with aspect ratio $L/D = 50$ and cube sizes ranging from 0.02 to 0.11. The relationship of percolation probability versus cube size data is shown in Figure 13, where only three curves are shown for clarity. Using Equation (3) to curve-fit the percolation probability data and obtain the mean μ and standard deviation σ for each cube size. The results are shown in Figure 14 and 15.

Figure 14 and 15 show that as the cube size increases, the means start to converge to a certain value and the standard deviations approach zero. For finite-size cubes, the means represent 50% percolation probability according to the normal distribution model.

Figure 13. Probability for different cube sizes, $L/D = 50$

Figure 14. Mean μ versus cube size

Figure 15. Standard deviation σ versus cube size

Next, the critical volume fraction for the infinitely large cube can be obtained by using the power-law model reported by Stauffer [36], which is given by

$$P_c - P_c(X) \propto X^{-\frac{1}{\nu}} \quad (4)$$

Here P_c is the theoretical CVF with size X approaching infinity, $P_c(X)$ is the CVF with size X , and the constant $\nu = 0.876$ has been suggested by a number of authors.

The “Means” shown in Figure 14 are used as the $P_c(X)$ for various X cube sizes. Figure 15 shows the inverse power of cube sizes versus $P_c(X)$. Based on the linear relationship in the model represented by Equation (4), a straight-line is used to curve fit the data, as shown in Figure 16. The equation obtained is

$$X^{-1/0.786} = -64036 P_c(X) + 825.27 \quad (5)$$

In Equation (4), $P_c = 740.18/57229 = 0.01293$ or 1.293%; this represents the CVF when cube size approaches infinity. In with Figure 14, when the size approaches infinity, μ approaches 0.0128.

Figure 16. Inverse power of cube sizes $X^{-1/0.876}$ versus $P_c(X)$

10. Conclusion

The above analyses have shown that finite-size nanocomposite cubes have a major difference in how percolations start to happen. When the volume fraction exceeds the first threshold, CVF1, the percolation could happen; whereas, when the volume fraction exceeds the second threshold, CVF2, the percolation most likely will happen. As the cube size increases, CVF1 increases too, as shown in Figures 8-11. This result is expected. On the other hand, Figures 8-11 also show that CVF2 decreases with the increasing cube size. This is an interesting result that is not expected. Nevertheless, as the cube size approaches infinity, the two critical volume rates CVF1 and CVF2 converge and should be equal to the theoretical value.

Using the normal-cumulated-distribution function model, a power-law equation, Equation (5), is obtained from the Monte-Carlo simulation data. This formula can be used to estimate the critical volume fraction (CVF) from the nanocomposite cube size. However, the Monte-Carlo simulations in this research have been based on the uniform distributions of the nanotubes in the composite cube. In practical implementation, the current technology still cannot fabricate truly uniform nanocomposites. Therefore the CVF obtained from this research can be treated as the lower bound of possible real values.

Acknowledgements

This work was partially supported by the National Research Foundation of Korea Grant funded by the Korean Government (MEST) (KRF-2009-220-D00034).

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