Factors Influencing Percolation Threshold

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Abstract

Percolation theory is described as the study of connectivity through random networks. This mathematical theory is used to understand various systems, including disease spread, natural systems, and electrical connectivity. It studies how links between adjacent components are able to form a continuous path throughout the entire system once a critical concentration of components and driving force is reached — the percolation threshold [1]. If this threshold is not met, the connectivity throughout the system will not be continuous, remaining as small, isolated clusters. Different systems show different connectivity vs concentration curve shapes such as linear, non-linear, and exponential, as seen in many percolation graphs. This difference is seen across various materials and mediums in the electrical field, because particle shape, size distribution, and how components are arranged changes how easily a continuous conductive path can form. Additionally, when examining multiple percolation threshold results, the reported values vary widely across different studies. This paper aims to better understand why the percolation behavior of carbon nanotube composites vary by analyzing their conductivity patterns and percolation thresholds to understand how matrix properties, filler geometry and experimental set up determine conductivity behavior and why the percolation threshold differs from paper to paper.

Percolation Theory Basics

Fundamentally, electric percolation requires an understanding of how electrons flow within a conductor and what differentiates the material from an insulator. Electron movement in a conductor flows like water through a pipe. With metal conductors, the outer valence electrons do not have significant bonds to the atom itself, so when in a matrix, the electrons are able to become delocalized and move freely with the application of electrical potential. This applied potential drives the electrons to move in a certain direction, imitating a continuous stream of water. With insulators, the valence electrons have much stronger bonds and are not able to detach and become delocalized. Because of this, the electrons are unable to move freely and cannot conduct an electric current. The study of percolation is a bridge between the two identities, examining how adding conductive material to an insulator can promote the material to be able to conduct a current. The minimum amount of conductive composite to create a continuous pathway is called the percolation threshold. Assessment of percolation threshold fluctuation between molecular and geometric composites can be further explored by interpreting discrepancies among datasets of percolation experiments.

Percolation in Carbon Nanotubes

The percolation threshold in carbon nanotubes is broken down into two main types, electrical and geometrical percolation. Electrical percolation determines conductivity and is based on how freely electrons move throughout a material. Graphene is a two-dimensional sheet of carbon atoms arranged in a single layer lattice. In graphene conduction, the valence band (the highest energy range of filled electron orbitals) only touches at K points which are used to describe electron energy levels in a solid. K-space uses wavelength and direction to show how waves or repeating patterns (like electrons) behave in a material [2]. In the K-space, where the K points touch

determines how electrons move in a material and affect its electrical conduction and whether it behaves as a conductor, semi-conductor, or insulator [2]. Since graphene is a flat lattice, electrons can move freely across the crystal momentum space (k-space). When an electron's allowed energy wave fits with the specific momentum value (how the electron is moving) and lattice, it is referred to as hitting a K point. When an atom is in an allowed state and hits a K point, the valence bands touch allowing electrons to move freely and act like a metal conductor. If no allowed states hit a K point, then a small energy gap opens, and the material behaves like a semiconductor [3].

However, since carbon nanotubes (CNT) are cylindrically rolled graphene sheets, electrons can only move a certain way around and along the tube [8]. Having different circumference sizes force electrons to fit into certain spaces only allowing specific energy states. The diameter and helicity of a CNT determine the position and spacing of the K points [4]. When the nanotube has a small diameter, the curvature bends the carbon bonds causing sigma and pi orbitals to overlap allowing new electron states in and out of the band gap. Closing the gap can change the conductivity of the CNT changing it from a semiconductor to a conductor [5]. Figure 1 illustrates the conduction and valence band in both semiconducting and metallic nanotubes as then create a band gap or "hit" the K points respectively.

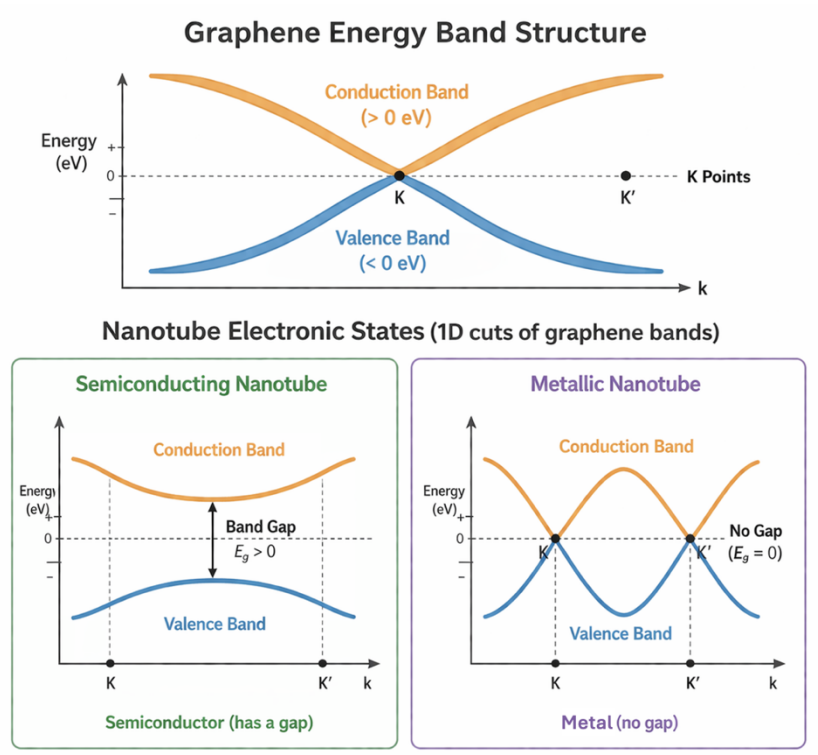

Figure 1: *Comparing graphene band structure with semiconducting and metallic nanotube 1D projections (generated using an AI text-to-image synthesis model)*

The two types of carbon nanotubes are single-walled (SWCNT) and multi-walled (MWCNT), where SWCNTs have one single cylinder of carbon atoms and MWCNTs have several concentric cylinders of carbon atoms. Having only one rolled up sheet of graphene and a diameter of ~1-2 nm the CNTs will have very high aspect ratios thus having high intrinsic conductivity [6]. On the other hand, MWCNTs have a diameter of 15-50 nm due to the concentric cylinders of graphene [7].

The main basis of geometric percolation is the aspect ratio of the individual tubes. The aspect ratio is the tube's proportional relationship between its length and diameter. Having a high aspect ratio (i.e. longer and thinner) usually results in a low percolation threshold since it can form interconnected networks much easier. While having a low aspect ratio (i.e. short and wide) gives a high percolation threshold [8]. Mechanically CNTs are very strong materials because of the

carbon-carbon sp^2 bonds that hold the structure together; which also behave elastically. This allows them to be excellent nanofillers for polymer composites through providing exceptional strength and electrical conductivity [9].

Graph and Growth

While the physical dimensions and aspect ratios of nanotubes establish the theoretical framework for connectivity, the most practical way to evaluate these differences in real-world application is through direct graphing. Understanding how to interpret a percolation graph is crucial for comparing datasets across various papers productively. In most cases, the x-axis represents the CNT filler concentration (ϕ), while the y-axis represents the conductive ability, or conductivity (σ), of the compound.

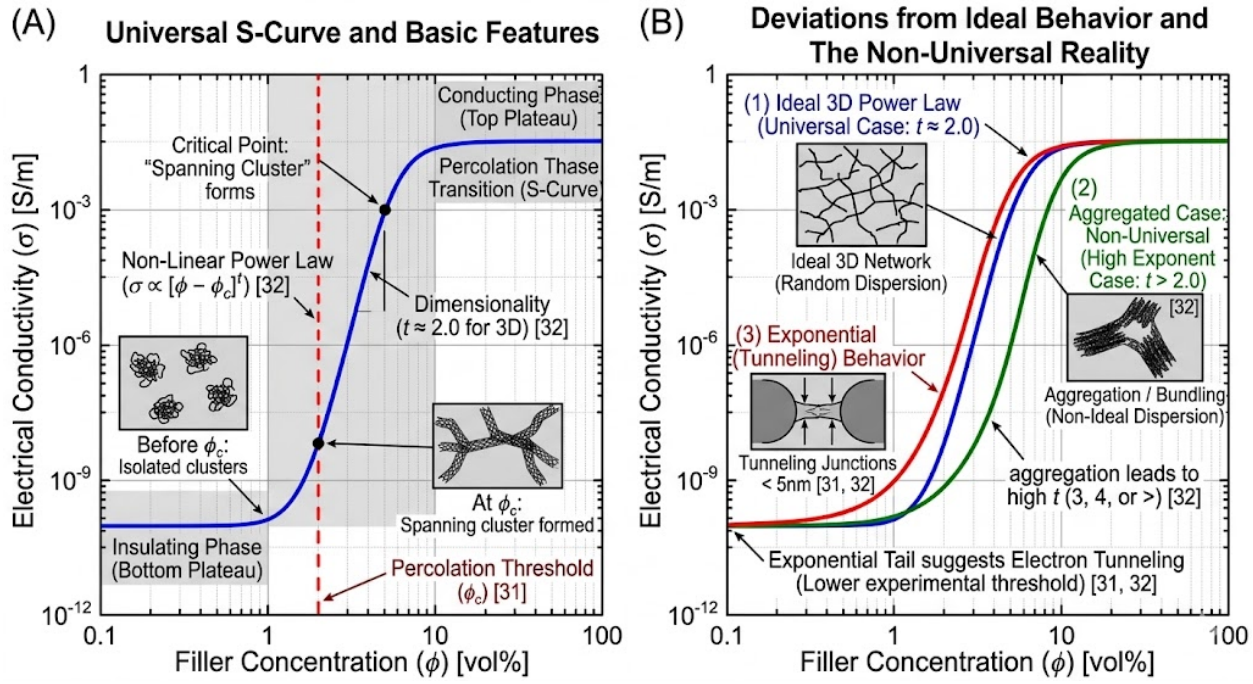

Figure 2 (A) Universal S-curve and basic features: following non-linear power law (B) Deviations from ideal behavior: exponential (tunneling) and aggregated/high exponent (generated using an AI text-to-image synthesis model)

When plotting these variables, the trend typically follows the power rule, which is visually characterized by a distinct "S" curve. This curve features two plateaus: a lower plateau representing

the insulating phase and a higher plateau representing the conducting phase. The sudden, sharp increase in slope between these two levels is the point at which the material sustains a continuous path of conductivity, known as the percolation threshold (ϕ_c) [31]. Although this "S-curve" is a universal visual, the mathematical behavior within that slope reveals the specific nature of the composite. Standard percolation theory dictates a Non-Linear Power Law

$$\sigma \alpha [\phi - \phi_c]^t \qquad \text{[Eq 1]}$$

representing the phase transition where the system moves from isolated clusters to a single "spanning cluster" [32]. However, in some studies, a linear behavior is observed after the threshold; once the network is fully formed, adding more filler simply adds redundant parallel paths, increasing conductivity in a predictable, additive way. Most notably for comparing theory to data, the most important mathematical relationship for polymer-nanotube research is exponential (tunneling) behavior. If a graph shows a gradual, exponential increase just before the main vertical jump, it suggests electron tunneling. In these systems, electrons move across thin polymer barriers—usually less than 5 nm—between nanotubes that are close but not physically touching. This mechanism often explains why experimental thresholds are lower than what purely geometric models predict [31,32].

The steepness of this upward slope is defined by the critical exponent (t), which identifies the dimensionality of the network and provides a window into the type of path the electrons are taking. In an ideal three-dimensional network, *t* is expected to be approximately 2.0, whereas a two-dimensional network yields a *t* of roughly 1.33 [32]. However, many research papers report "non-universal" *t* values significantly higher than 2.0, sometimes reaching 4.0 or higher. This discrepancy is a major source of conflict in literature. High *t* values typically point to a complex transport mechanism, such as a wide variety of tunneling distances or the presence of "sticky"

interactions that cause the nanotubes to aggregate into non-random clusters rather than an even distribution [32]. These graphical variations provide the empirical data necessary to apply more rigorous mathematical models, specifically the power and scaling laws used to define the threshold's behavior.

Power and Scaling Law

When measuring the percolation threshold, there are two different laws used: power and scaling. The power law explains how conductivity jumps when CNTs are added past the point of the percolation threshold, with the equation:

$$\sigma \propto [\, \varphi - \varphi_c \,]^t \qquad \text{[Eq 2]}$$

where σ = conductivity, φ = filler amount, φ_c = percolation threshold, and t = critical exponent [10]. This equation only takes into account how much filler is added to the polymer matrix. For the scaling law, it takes into account the geometry of the filler (CNT), using the equation

$$\sigma = [\varphi - \varphi_c\,(\alpha)]^{t(\alpha)} \qquad \text{[Eq 3]}$$

where *a* represents the aspect ratio of the nanotubes [11]. When calculating conductivity of carbon nanotubes, studies mostly apply the power law that does not include the aspect ratio. This is because researchers are using the standard percolation model which focuses only on conductivity changes with filler concentrations [12]. These different analyses give conflicting percolation threshold results in various papers.

The theoretical analysis that's based on the excluded volume theory, which predicts the critical threshold at which objects form a spanning network and where the excluded volume is the region around a particle where the center of another particle cannot enter without overlap, state that the percolation threshold of carbon nanotubes should decrease as the aspect ratio increases.

Thus, the longer the CNT, the easier the CNTs can form conductive networks at lower concentrations. Because of this universal assumption that this theory should work for different systems regardless of the particle geometry, the aspect ratio or alignment of the carbon nanotubes are not included in the equation. The aspect ratio is only assumed to affect the percolation threshold, with the standard of the higher the aspect ratio the lower the percolation threshold.

Also, a more idealized model was used in which the particles are stationary. As a result, many papers only use this equation:

$$\sigma \propto [\, \varphi - \varphi_c \,]^t \qquad \text{[Eq 2]}$$

to calculate the percolation threshold and critical exponent. This assumption that the particles are static is especially problematic because in making real CNT composites the nanotube can move, rotate and aggregate during the processing. After processing, CNTs are largely immobilized within the polymer matrix and do not move freely. Their motion is restricted because of the physically interconnected and often bonded or strong interactions with the surrounding polymer chains. The movement allows them to form clusters and networks leading to kinetic percolation, which is the conductive networks formed due to the particle's motion. Ignoring this behavior in calculations will only allow predictions of the static percolation threshold and not the threshold in real processing conditions. The authors in this study [13], found that the dispersion of the CNTs played a vital role in the conductivity and percolation threshold measurements. This means that two experiments can have the same CNT aspect ratio, but different dispersion quality, which leads to changes in the CNT contacts, tunneling and aggregation, creating different measured percolation thresholds.

Transport Mechanisms

In carbon nanotube composites, conduction can be formed through either direct contact or electron tunneling. Direct contact is when the nanotubes are physically touching each other, which is rare since the tubes are usually coated or separated by a thin layer of polymer. This restricts their ability to form conduction through only direct contact methods, and usually only occurs in high concentrations of CNTs where the network is dense [14]. On the other hand, tunneling happens when the CNTs are very close to each other without touching directly. The electrons move between the ~1-3 nm space by quantum tunneling, which allows them to move through the thin polymer layer. The electrons essentially "tunnel" through the barrier instead of physically travelling through a contact [15]. Tunneling in CNTs refers to electrons passing between nearby nanotubes through very thin insulating gaps, even if not physically touching, as is shown in Figure 3.

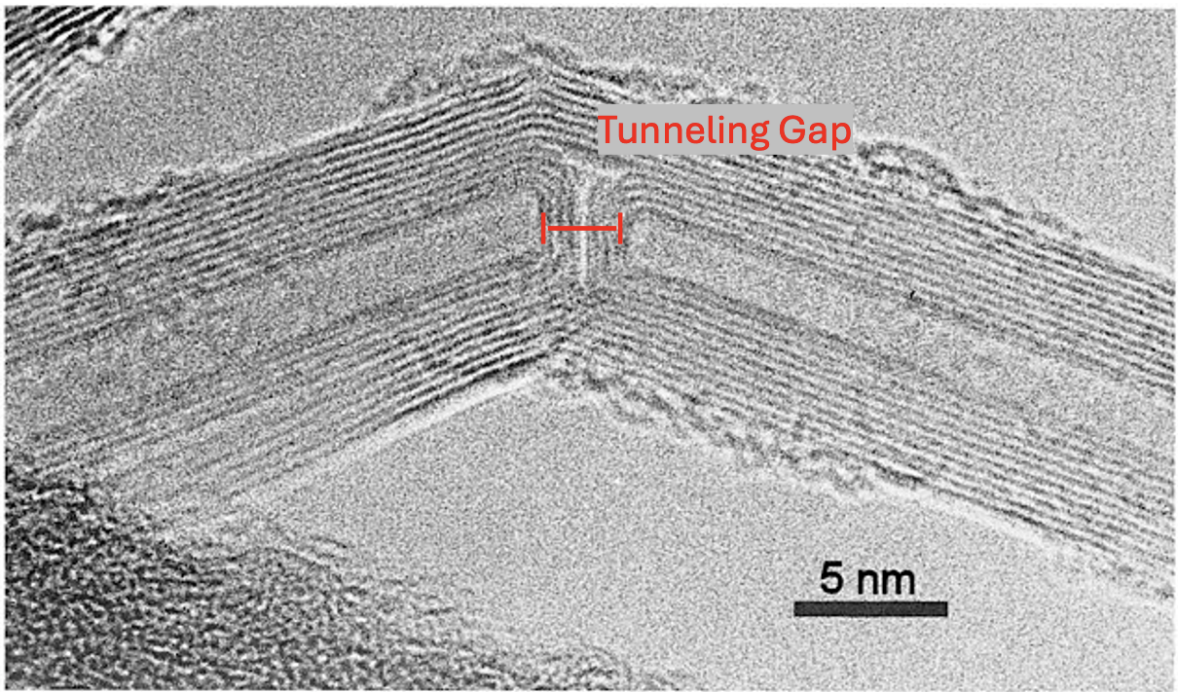

Figure 3: *Tunneling gap between multi-walled carbon nanotubes. (Image adapted from Wikimedia Commons, public domain)*

This process of electron tunneling lowers the percolation threshold, because the CNTs do not need to touch to conduct. In CNT polymer composites, the conduction of electricity is mainly through electron tunneling rather than direct contact, since perfect direct contact is more difficult to achieve [14]. Quantum tunneling is tightly related to the conductivity of the CNTs. The quantum process of electrons moving between CNTs has a probability which decreases exponentially with distance, which can be measured with the Simmons equation,

$$I = I_0 e^{-\beta d} \qquad \text{[Eq 4]}$$

where I is the tunneling current, I_0 is the current at zero separation, d is the distance between CNTs, β is the decay constant that depends on the polymers properties [33]. When looking at a percolation threshold graph the amount of tunneling in the composite is a cause of the lower threshold. As more electrons tunnel between nearby non-contacting particles, a continuous conductive network forms at lower CNT concentrations, reducing the percolation threshold [17]. There is also a contact resistance which is the sum of direct contact and tunneling resistance which can be measured using the Landauer-Büttiker formula:

$$R_{contact} = \frac{h}{2e^2} \cdot \frac{1}{MT} \qquad \text{[Eq 5]}$$

where h is Planck's constant, e is the charge of an electron, and M (the total number of conduction bands) and T (transmission probability of electron charges between contacting surfaces) are added to account for the tunneling effects. Both M and T are parameters for tunneling resistance. M will depend on the nanotube's structure, such as diameter and chirality. What was found, however, was that direct contact resistance was not the result of actual physical contact, but rather the minimum resistance that occurs when CNT's are separated by a small distance. Even at these close

separations, the electrons are still tunneling, implying that CNT conduction is the result of quantum tunneling [18].

SWCNT networks exhibit higher tunneling and contact resistance because electrons must cross high-energy barriers with only a single conduction pathway, making conductivity sensitive to barrier height and distance. In contrast, MWCNTs have multiple conductive shells and larger contact areas, which give alternative pathways and reduce the impact of tunneling barriers, resulting in a lower effective resistance [16,18]. When researchers use the words *direct contact,* it refers to how close CNT's can get before physically touching and theoretically no longer requiring quantum tunneling. This indicates the conductivity in CNT composites are controlled by electron tunneling, especially in systems with low CNT concentrations. As the concentration increases and more direct contact networks form, the system becomes more conductive as the electrons travel through the increasing number of pathways rather than through tunneling. On the other hand, tunneling resistance is usually higher than contact resistance, as a few gaps can limit the overall conductivity and the effectiveness of tunneling greatly depends on the type of material that surrounds the tube. Lower energy barriers in the material used while processing allows for easier electro transfer.

Polymers

Carbon nanotube composites are made by adding conductive CNT fillers dispersed within a matrix, typically a polymer, in order to enhance properties like electrical conductivity and mechanical strength [34]. In the processing of carbon nanotube composites, various materials are used to control dispersion, structure, electrical properties, and the balance between contact and electron tunneling. The most common materials used are polymer matrixes, more specifically thermoplastic polymers that include polyethylene, polystyrene, and polymethyl methacrylate

(PMMA) are used due to the ease of processing through melt blending, a process that applies high shear at temperatures above the polymer's melting point, and are light weight. However, this process does not show good CNT dispersion, and is an insulating polymer that creates high energy barriers that limit an electron's ability to tunnel which reduces conductivity.

Thermosetting polymers, such as epoxy, are shown to be most effective for high-performance applications as the stiffness, thermal stability, and conductivity of the CNTs is improved. Some conductive polymers— polyaniline and polypyrrole— are also used as the electrical performance of CNTs become enhanced, which is prominently used for electronic devices and sensors. Other less used polymers include functional polymers (polyvinyl alcohol) to improve dispersion and matrix interaction, as well as elastomeric polymers (rubber or polyurethane) to provide flexibility and durability [19,20,21].

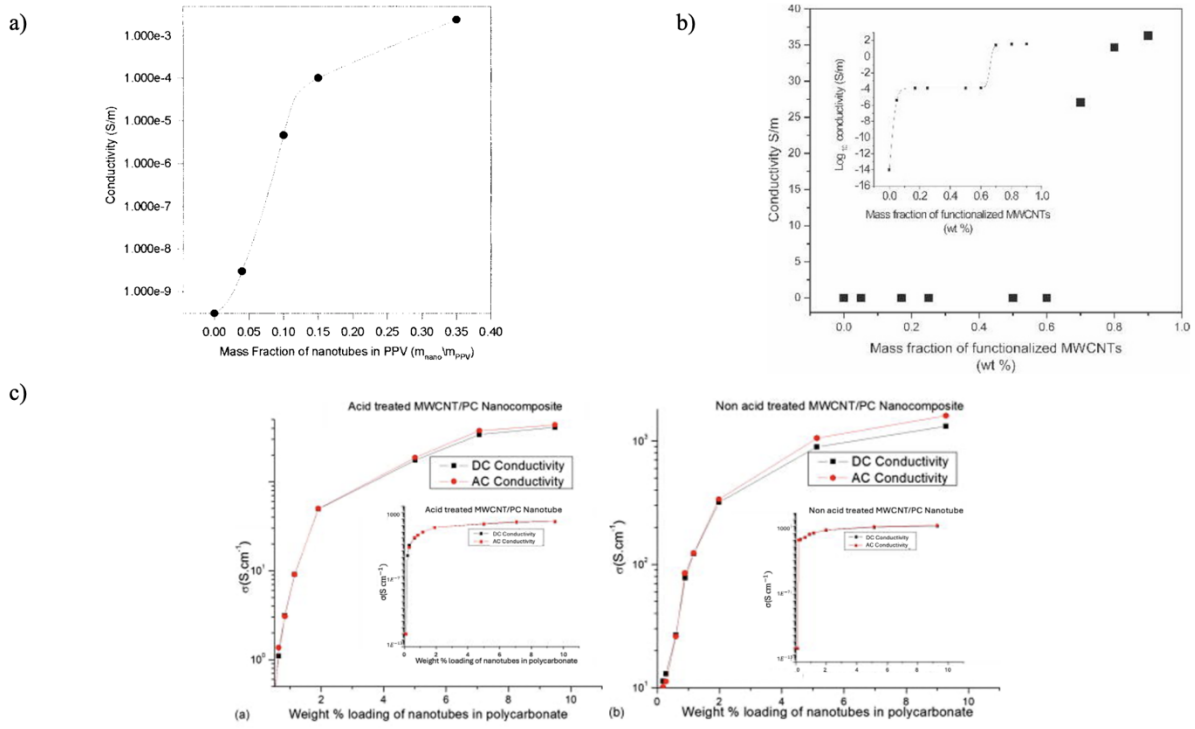

Figure 4. *Electrical conductivity as a function of nanotube loading in thermoplastic polymer composites: (a) PPV/multiwall carbon nanotube composites showing a sharp increase in conductivity due to the formation of percolating conductive networks [37]. (b) MWCNT/polystyrene composites demonstrating*

a clear percolation transition from insulating to conductive behavior with increasing nanotube content [35]. (c) MWCNT/polycarbonate composites, with acid-treated MWCNTs exhibiting higher conductivity than non acid-treated MWCNTs due to improved dispersion [36].

No polymer stands out as the best for CNT's, as each polymer has its own pros and cons: for easy processing, the use of thermoplastics are an attractive choice; for dispersion, however, functional polymers appear better suited. [21,22]. In terms of percolation threshold, the polymer used directly affects how high or low the threshold can be. In thermoplastic polymers, the percolation threshold is often relatively higher because CNTs are harder to disperse uniformly in viscous melts, leading to aggregation and fewer effective conductive pathways [21,22,23]. As demonstrated in Figures 4a, 4b and 4c, the specific chemical composition and filler treatment create vastly different graphical profiles even within the thermoplastic category.

Figure 4b illustrates the substantial surge in conductivity possible in a rigid thermoplastic matrix [35]. Because polystyrene is a glassy, rigid polymer, it locks the nanotubes into a static, "frozen" network after processing. This results in a remarkably sharp, well defined percolation transition (a high critical exponent, t). However, because polystyrene is highly insulating and the chains are bulky, it requires a higher concentration of nanotubes to overcome the physical separation between fillers compared to the PPV system. This polystyrene study was a pioneering work in the field, providing some of the first evidence that the sudden jump in conductivity was driven by quantum tunneling rather than just physical contact, establishing the "Tunneling Percolation Model" for rigid insulating matrices.

Figure 4a (PPV) exhibits a noticeably lower percolation threshold overall, specifically compared to Figure 4b (Polystyrene) despite both graphs being characterized by a very sharp, steep slope [37]. This suggests that while both matrices are rigid enough to "capture" a spanning cluster effectively, the PPV polymer facilitates better initial network formation. The reason for this is rooted in the conjugated, semiconductive backbone of PPV that consists of alternating single

and double bonds versus the unconjugated, bulky backbone of polystyrene that is purely single bonds. The presence of the double bonds not only lowers the electron tunneling energy barrier across nanotubes, but also allows π-π stacking and lowers contact resistance between the polymer and CNT due to the planar geometry of the bonds. This specific case was from the 1998 *Advanced Materials* paper, which was one of the first major demonstrations of π-π stacking that enabled the PPV polymer to "wrap" around CNTs. The discovery emphasized the importance of polymer and filler compatibility, or interfacial chemistry, as a factor affecting percolation threshold, rather than just filler volume as previously thought.

The MWCNTs in Figure 4c exemplify a more gradual slope in comparison to polystyrene and PPV, but its difference in surface treatment allows for the percolation threshold to still be altered. Acid-treating the MWCNTs introduces functional groups that act as chemical anchors, reducing the contact resistance between the nanotubes and the polycarbonate chains. This allows electrons to tunnel more efficiently across the polymer barriers, resulting in a lower threshold and higher overall conductivity levels compared to the untreated "slick" nanotubes that tend to aggregate [36]. This experiment was significant in proving that interfacial resistance could be lowered through chemical pre-treatment to optimize industrial grade plastics like polycarbonate and other difficult, viscous systems.

In contrast to the thermoplastic examples, thermosetting polymers like epoxy typically show a lower percolation threshold because their lower initial viscosity allows better CNT dispersion before curing, helping form conductive networks at lower CNT loadings. Elastomeric polymers can also achieve relatively low thresholds due to their flexibility, which allows CNTs to rearrange and maintain conductive contacts under deformation. Conductive polymers further reduce the percolation threshold because the polymer already carries charge, so fewer CNT

connections are needed to achieve conductivity. Overall, better dispersion, stronger CNT–polymer interactions, and the ability to form interconnected networks all contribute to lowering the percolation threshold, while poor dispersion and CNT agglomeration increase it [22,23].

However, it is important to note that percolation behavior is not a universal characteristic of all composite materials. A distinct percolation threshold only emerges when there is a significant conductivity contrast between the filler and the matrix. In systems where the matrix is already highly conductive (such as a metal-matrix composite), or where the filler geometry lacks a high aspect ratio (such as spherical particles), the conductivity may increase linearly rather than exhibiting the non-linear "S-curve" surge. Furthermore, the absence of a spanning cluster due to severe filler clumping or poor interfacial wetting can prevent percolation entirely, highlighting that the sharp transitions observed in pioneering research are the result of carefully optimized material parameters.

Solvents

In some cases, a solvent is added to help with dispersion. Dispersion is explained by the balance of interaction between CNTs, polymers, and solvents through Van der Waals forces, polarity, and π-interactions. Using molecular simulations, study results show that solvents such as dimethylformamide (DMF), dimethylacetamide (DMAc), and dimethyl sulfoxide (DMSO) improve dispersion in thermoplastic polymers as the nanotubes have a strong tendency to form clusters without solvent assistance, the addition of DMF or DMSO cause the CNTs to spread out [24]. Studies showed that DMF worked the best in dispersing the CNT over DMSO or DMAc because of its planar shape and stronger van der Waals forces [24,25]. Also showing that PMMA sticks to CNTs better than polyacrylonitrile (PAN), another thermoplastic polymer, because its shape allows more surface contact meaning CNTs are able to disperse more evenly. When the

concentration of polymer increased, the CNTs tended to stick to each other rather than the CNTs reducing the dispersion [24,25]. Another study assert that polarity is the main factor of whether the solvent is good at dispersing CNTs, since the interactions were strong with CNT surfaces and polymers, helping the liquid wet and penetrate the CNT network, which was evidence from testing DMF and DMSO over ethanol and acetone. The solvent that was found best was ethylene glycol due to its two polar hydroxyl (-OH) groups, making it especially good at penetrating the CNT network and interacting with the CNT surfaces [26]. Overall, polarity of the solvent is the main factor controlling how well CNTs disperse and using highly polar solvents improves CNT dispersion, which lowers the percolation threshold and strengthens the resulting fibers.

Processing

While the choice of polymer and solvent determine the chemical nature of the composite, the processing method plays a big role in how the percolation threshold fluctuates. Processing is divided into two stages: synthesis (creation of the raw carbon nanotubes) and fabrication (integration of final composite), with each stage influencing the percolation properties of the final composite independently. During synthesis, properties such as aspect ratio, purity, and defect density are determined, making the different processes critical points of establishing percolation properties [28,30]. Synthesis processes can be characterized by either high-temperature (arc discharge and laser ablation) or low-temperature (chemical vapor deposition/CVD). The high-temperature methods prioritize slower, high-energy growth of nanotubes, resulting in CNTs with higher crystallinity and fewer structural defects [28]. In regards to percolation, this causes a cleaner electronic path and a more polarizing spike in conductivity at the percolation threshold. With low-temperature CVD, the use of gaseous hydrocarbons over metal catalysts allows for the constant cycling of the process and therefore mass production [28,30]. While this benefit exists, CVD is

more prone to broader diameter distributions and structural defects. Compared to its high-temperature counterparts, CVD is less conductive in general, but these defect points can act as scattering sites for electrons and can lead to less ordered bundles and a more unpredictable threshold [27,28]. Following synthesis, fabrication determines the final structure of the system, as well as if the system utilizes statistical, with a random and uniform distribution, or kinetic percolation, where the CNTs move and self-assemble after dispersion [27]. Traditional approaches, like solution mixing, involve high-energy ultrasonication to break up CNT bundles through cavitation (microscopic bubbles forming and collapsing). The process is capable of producing high-quality dispersion; however, in its controlled evaporation phase, the system can shift to an aggregated state if the solvent is removed too slowly. This variable would result in a much higher percolation threshold than expected [27,29]. Another fabrication method is melt blending, using high-shear mechanical mixing to distribute the filler. Unfortunately, the intensity of the mixing commonly results in the breakage of tubes, lowering the aspect ratio, and therefore causing the percolation threshold to rise. The in-situ polymerization method disperses nanotubes with a liquid monomer prior to a chemical reaction. The process allows the nanotubes to be dispersed more effectively, typically allowing stable and highly connected systems held by polymer chains directly from the surfaces of the CNTs [29]. In comparison to solution mixing and melt blending, the in-situ polymerization process typically harbors the strongest filler-matrix interactions with less clumping and a lower percolation threshold respectively [27,29]. Some more advanced methods include layer by layer (LbL) deposition and bucky paper-based approaches, eliminating the randomness of the traditional processes [29]. LbL deposition uses electrostatic attraction to submerge substrates into alternately charged CNTs/polymers, creating a systematic, layered structure, and allowing a 2D-like connectivity with a high filler concentration [29,30].

Ultimately, every step by which a CNT is made is critical to the structural and kinetic properties of a network [27].

Conclusion

The percolation threshold in carbon nanotube systems is strongly influenced by a combination of geometrical, chemical, and processing factors. A high aspect ratio of CNTs will lower the threshold because fewer nanotubes are needed to form a connected network. While the geometry of the CNTs, diameter, single vs multi-wall, affects how well the CNTs conduct and contact each other, processing methods and solvent choice are also important as a solvent with high polarity will improve dispersion of the CNTs by disaggregating and distributing them throughout the matrix. Having good dispersion reduces aggregation and allows for an efficient forming of the network at lower concentrations. The polymer chosen can also increase the connectivity of the nanotubes by bridging gaps which can improve mechanical and electrical pathways. The lower the energy barrier the polymer has the easier the electron is able to move throughout the system, thus lowering the threshold. Finally, theoretical calculations and models help predict how these variables interact, guiding optimization of CNT systems. Altogether, controlling aspect ratio, processing conditions, solvent polarity, dispersion quality, CNT geometry, polymer interactions, and transport behavior is essential for minimizing the percolation threshold and maximizing material performance.

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