# Polymer Nanocomposites

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Guest Editors

## Abstract

Polymer nanocomposites (PNCs)—that is, nanoparticles (spheres, rods, plates) dispersed in a polymer matrix—have garnered substantial academic and industrial interest since their inception, circa 1990. This is due in large part to the incredible promise demonstrated by these early efforts: PNCs will not only expand the performance space of traditional filled polymers, but introduce completely new combinations of properties and thus enable new applications for plastics. Low volume additions (1–5%) of nanoparticles, such as layered silicates or carbon nanotubes, provide property enhancements with respect to the neat resin that are comparable to those achieved by conventional loadings (15–40%) of traditional fillers. The lower loadings facilitate processing and reduce component weight. Most important, though, is the unique value-added properties not normally possible with traditional fillers, such as reduced permeability, optical clarity, self-passivation, and increased resistance to oxidation and ablation. These characteristics have been transformed into numerous commercial successes, including automotive parts, coatings, and flame retardants. This issue of the *MRS Bulletin* provides a snapshot of these exemplary successes, future opportunities, and the critical scientific challenges still to be addressed for these nanoscale multiphase systems. In addition, these articles provide a perspective on the current status and future directions of polymer nanocomposite science and technology and their potential to move beyond additive concepts to designed materials and devices with prescribed nanoscale composition and morphology.

## Introduction

Polymers have been a part of life since the beginning of humankind. From tar and shellac, tortoise shell and horns, to today's synthetic offerings such as polyolefins, epoxies, and engineering resins, polymers provide crucial materials for construction, commerce, transportation, and entertainment across the globe. Estimates of global polymer production range from 250 billion pounds to more than 400 billion pounds (approximately 114–181 billion kg) annually.

In the majority of their diverse applications, polymeric materials are not chemically or molecularly homogenous but are multicomponent systems. By adding fillers, such as minerals, ceramics, metals, or even air, materials scientists can generate an infinite variety of materials with unique physical properties and competitive production costs. For example, adding filler to a commodity thermoplastic such as polypropylene can achieve performance levels that would otherwise require a much more expensive engineering plastic. Similarly, combining different polymers to form a polymer blend or resin can increase the value of existing polymers.

Polymer nanocomposites incorporate a new spectrum of fillers that extend the function and utility of polymers while maintaining the manufacturing and processing flexibility inherent to plastics, thermosets, and resins. In particular, polymer nanocomposites have been successful with regard to overcoming traditionally antagonistic combinations of properties.

Since the first reports in the late 1980s,[1–6] the term "polymer nanocomposite" has evolved to refer to a multicomponent system in which the major constituent is a polymer or blend thereof and the minor constituent has at least one dimension below 100 nm. Polymer nanocomposite is an appropriate synonym for inorganic–organic hybrids and molecular composites and also encompasses mature commercial products such as polymers containing carbon black or fumed silica. This issue of *MRS Bulletin* focuses primarily on polymer nanocomposites containing nanoscale clays and various carbon nanotubes to illustrate the status of this rapidly evolving research and development enterprise.

The numerous reports of large property changes with very small additions of nanoparticles (<1–5 wt%) have fueled the view that nanoparticles are a magic pixie dust that delivers huge dividends. In fact, recent market surveys have estimated global consumption of polymer nanocomposites at tens of millions of pounds (~\$250 million), with a potential annual average growth rate of 24%, to almost 100 million pounds in 2011 at a value exceeding \$500–800 million.[7–9] Major revenues are forecast from large commercial opportunities such as automobile parts, coatings, flame retardants, and packaging, where lower-cost, higher-performance materials would improve durability and design flexibility while lowering unit price and life cycle cost.

Whatever the case for the long-term economic growth of polymer nanocomposites, the opportunities to deliver targeted material performance reside with the implications associated with nanoscale multiphase systems. There are important differences when the fillers shrink from microscale to nanoscale. This issue of the *MRS Bulletin* provides a snapshot of exemplary successes, future opportunities, and the critical scientific challenges still to be addressed for these nanoscale multiphase systems. In addition, these articles provide a perspective on the current status of polymer nanocomposite science and technology as well as future directions to move it beyond additive concepts to designed materials and devices with prescribed nanoscale composition and morphology.

## The Nano Advantage

When fillers are nanoscopic, there are advantages afforded to filled polymers and composites that lead to performance enhancements. These advantages result primarily from filler size reduction and the concomitant increase in surface area. The size of the additive might drop by up to three orders of magnitude relative to conventional alternatives. In contrast, many nanotechnologies associated with electrical or optical properties benefit from new physical phenomena arising from quantum confinement effects induced by the nanoscale dimensions of the material. The literature about polymer nanocomposites contains many discussions about the implications and physical manifestations of the reduction in filler length scale.[10–14]

For example, compare a microcomposite and a nanocomposite with the same volume fraction of a secondary constituent (filler), where the spherical particles have volumes of $1\,\mu m^3$ or $1\,nm^3$ per particle, respectively. The mean particle–particle separation is smaller by three orders of magnitude, the total internal interfacial area increases by six orders of magnitude, and the number density of constituents increases by nine orders of magnitude in the nanocomposite. Although these numbers alone are impressive, the filler size must be viewed relative to the size of polymer molecules to capture the full potential impact of nanoscale fillers on composite properties.

Many properties are related to the size of the polymer chain, which can be expressed as the radius of gyration $R_g$ (the second moment of the three-dimensional distribution of the monomers of the polymer chain—approximately the expanse of the molecule). $R_g$ is on the order of 3–30 nm. Depending on the strength of interaction between the filler surface and the matrix, the polymer chains in close proximity to the filler will be perturbed with respect to those in the bulk (i.e., away from the interface). The thickness $t$ of this interfacial region that surrounds the particle is, to first order, independent of the size of the particle. Thus, as the particle size decreases, the relative volume of this interfacial material, $V_{interface}$, with respect to the volume of the particle, $V_{particle}$, will increase.

Figure 1 shows this ratio, $V_{interface}/V_{particle}$, as a function of particle aspect ratio from plates (aspect ratio <1) to spheres to rods (aspect ratio >1). The filler size is expressed as $\delta$, the ratio of the thickness of the interface to the smallest dimension of the particle. Micrometer-sized fillers have $\delta \sim 0.01$, so that at any aspect ratio, the volume of the particles exceeds that of the interfacial region. However, when the fillers are reduced to the nanoscale and $\delta \sim 1$–10, the volume of the interfacial region exceeds the volume of the particle. In addition, at a fixed value of $\delta$, the aspect ratio has an effect on $V_{interface}/V_{particle}$, showing an expected increase from plates to rods to spheres as the fillers change from two-dimensional (plate) to one-dimensional (rod) to zero-dimensional (sphere) objects. The magnitude of this change increases dramatically as the filler size drops; for example, at $\delta$ 10, $V_{interface}/V_{particle}$ increases by two orders of magnitude between plates and spheres.

Furthermore, these calculations demonstrate the impact that even a small volume fraction of filler has on the surrounding polymers. For example, by dispersing a mere 1 vol% of a nanosphere (radius ~2 nm) in a polymer (interfacial thickness ~6 nm), the volume fraction occupied by
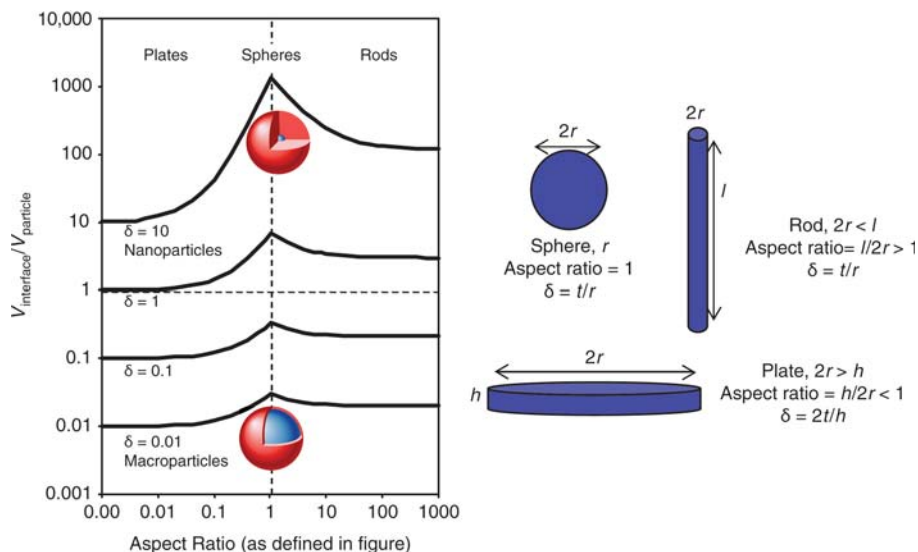


Figure 1. The ratio of interfacial volume to the particle volume ($V_{interface}/V_{particle}$) as a function of the particle aspect ratio and the ratio of the interfacial thickness to the particle size ($\delta$). The aspect ratio and $\delta$ are defined in the schematic at right ($r$ is radius, $l$ is length, $h$ is height). The interfacial thickness (red shell, $t$) is assumed to be independent of the particle size. As particles decrease in size to less than 100 nm, the interfacial volume around a particle can dominate the physical properties, and this is particularly evident for spheres and rods.

the interfacial region is ~63 vol%, suggesting that more than half of the composite is affected by the presence of the second-phase particles. If the particle is increased to 20 nm in radius without changing the interfacial thickness or particle loading, the volume occupied by the interfacial region would be only ~1.2 vol%.

The importance of polymer–particle interactions is amplified in polymer nanocomposites such that the interface and the cooperativity between particles dominate the macroscopic properties. For example, weak forces between particles, such as van der Waals, are more pronounced for nano-sized particles because of lower surface roughness, smaller average particle separations, and thus higher dispersive forces. Also, because of the nanoscopic dimensions of the particles, the accessible aspect ratio of discrete secondary constituents can approach 100 or more. These high-aspect-ratio, nanoscale fillers can reach percolation thresholds at <1–5 vol% and thereby exhibit large increases in bulk mechanical and transport properties at these low loadings. The percolation threshold is the filler concentration at which the electrical conductivity increases sharply by orders of magnitude, indicating that conductive pathways span the macroscopic sample. Thus, the casual observation that nanofillers act as pixie dust is firmly rooted in the implications of reducing the size of the fillers by up to three orders of magnitude.

## Scope and Impact

As with traditional filled plastics, an infinite variety of possible polymer–nanoparticle combinations conceptually affords tunability. Thus, given the diversity of possible properties and tolerable costs, there is no universal "best" nanoparticle filler for polymer nanocomposites. The best nanoparticle filler (or traditional filler, for that matter) is determined by meeting both a specific set of physical properties and a price point associated with a particular part or product. Table I compares the size, shape, properties, and uses of traditional fillers and newer nanoscale fillers. As noted earlier, a few traditional fillers have sizes below 100 nm, and nanoscale fillers can access high aspect ratios. Although there is considerable overlap in the elastic moduli and thermal conductivities between the traditional and nanoscale fillers, the electrical properties of the carbon-based nanofillers are in a class by themselves, with conductivities more than 100 times higher. This summary of fillers encourages one to imagine many possibilities for remarkable properties within the broad materials class of polymer nanocomposites.

The first key demonstration of polymer nanocomposites was provided by the pioneering work of Okada and co-workers at Toyota Central Research in the late 1980s.[1–4] By combining inclusion and colloidal chemistry of mica-type layered silicates (nanoclay) with surface-initiated

**Table I: Characteristics of Traditional and Nanoscale Fillers: Shape, Size, Properties, Dimensions, and Uses.**

| | Approximate Shape[a] | Smallest Dimension (nm)[a] | Aspect Ratio[b] | Elastic Modulus (GPa) | Electrical Conductivity (S/cm) | Thermal Conductivity (W/m K) | Commercial Uses |
|---|---|---|---|---|---|---|---|
| **Traditional Fillers** | | | | | | | |
| Carbon black[32] | agglomerate of spheres | 10–100 | 1–5 | ... | 10–100 | 0.1–0.4 | tires, hoses, shoes, elastomers |
| Carbon fiber[33] | rods | 5,000–20,000 | 10–50 | 300–800 | 0.1–10 | 100–1000 | aerospace, automotive, marine, sporting, medical |
| Carbon graphite[34] | plate | 250–500 | 15–50 | 500–600 | 1–10 | 100–500 | gaskets, seals |
| E-glass[35] | rod | 10,000–20,000 | 20–30 | 75 | ... | ... | marine, automotive, construction, filtration |
| Mineral: CaCO$_3$[36] | sphere platelet | 45–70 600–4,000 | ~1 1–30 | 35 | ... | 3–5 | paper, paint, rubber, plastics |
| Mineral: silica[37,38] | agglomerate of spheres | 8,000–30,000 | 5–10 | 30–200 | ... | 1–10 | reinforced plastics, thermal insulator, paint, rubber reinforcing agent |
| Mineral: talc, china clay[37,39] | platelet | 5,000–20,000 | 5–10 | 1–70 | ... | 1–10 | paper, consumer goods, construction |
| **Nanoscale Fillers** | | | | | | | |
| Carbon nanofiber[40] | rod | 50–100 | 50–200 | 500 | 700–1000 | 10–20 | hoses, aerospace, ESD/EMI shielding, adhesives |
| Carbon MWNT[41] | rod | 5–50 | 100–10,000 | 1,000 | 500–10,000 | 100–1000 | automotive, sporting, ESD/EMI shielding |
| Carbon SWNT[42] | rod | 0.6–1.8 | 100–10,000 | 1,500 | 1000–10,000 | 1000 | filters, ESD/EMI shielding |
| Aluminosilicate nanoclay[43] | plate | 1–10 | 50–1000 | 200–250 | ... | 1–10 | automotive, packaging, sporting, tires, aerospace |
| Nano-TiO$_2$[37,44] | sphere | 10–40 | ~1 | 230,000 | $10^{-11}$–$10^{-12}$ | 12 | photocatalysis, gas sensors, paint |
| Nano-Al$_2$O$_3$[37,45] | sphere | 300 | ~1 | 50 | $10^{-14}$ | 20–30 | seal rings, furnace liner tubes, gas laser tubs, wear pads |

[a] Dispersible unit.
[b] Aspect ratio is defined as the long axis to short axis irrespective of shape. Note that this differs from Figure 1.
ESD is electrostatic discharge; EMI is electromagnetic interference; MWNT is multiwall carbon nanotube; SWNT is single-wall carbon nanotube.

polymerization, they demonstrated that only ~2–4 vol% of layered silicate sufficiently improved the mechanical properties of nylon-6 polymer at elevated temperatures to enable its use inside an automotive engine compartment.

Since then, the patent and literature activity has been astonishing (Figure 2).[15] From 1992 to 2004, the number of citations for polymer nanocomposites has doubled every two years, indicating that these materials are still on the steep part of the technology S-curve (Figure 2a). Since 2001, polymer nanocomposites represent ~43% of the broader nanocomposite field, which includes metals, ceramics, and thin films (Figure 2b). Of the publicly available literature on polymer nanocomposites, the majority (80%) is in peer-reviewed journals, whereas patents have maintained a constant fraction (8–10%). Together, layered silicates (nanoclays) and carbon nanotubes represent almost 50% of the ongoing investigations. Polymer–clay nanocomposites, however, might be reaching saturation, as evidenced by a diminishing growth rate in publications and patents per year. In contrast, polymer–carbon nanotube composites have rapidly accelerated since the availability of carbon nanotubes became widespread in the late 1990s and are still exhibiting a steady growth rate (Figure 2b).

After almost 20 years, the diversity in scientific investigations, technology advancements, processing innovations, and product development is staggering. A significant number of excellent review papers (e.g., clays[16–23] and carbon nanotubes[22–26]) and books[27–30] are available that chronicle and summarize the status of various nanoparticle–polymer combinations and the broad scientific and technological challenges that still need to be overcome.

This issue of *MRS Bulletin* provides six articles to illustrate the breadth of activity in polymer nanocomposites. Hunter et al. highlight the issues in polymer–nanoclay composites, where the most commercial activity currently exists. Baur and Silverman consider the opportunities in adding nanofillers to traditional engineering polymer composites that use continuous fiber reinforcements. Schadler and co-workers focus on the implications and engineering possibilities of larger interfacial areas per unit volume. Krishnamoorti addresses the issues of weak forces becoming significant for nano-sized components and strategies for overcoming their tendency to agglomerate. Winey et al. explore opportunities for nanofillers to modify electrical and thermal properties of polymers. Finally, Hule and Pochan consider the opportunities of polymer nanocomposites in the medical arena.
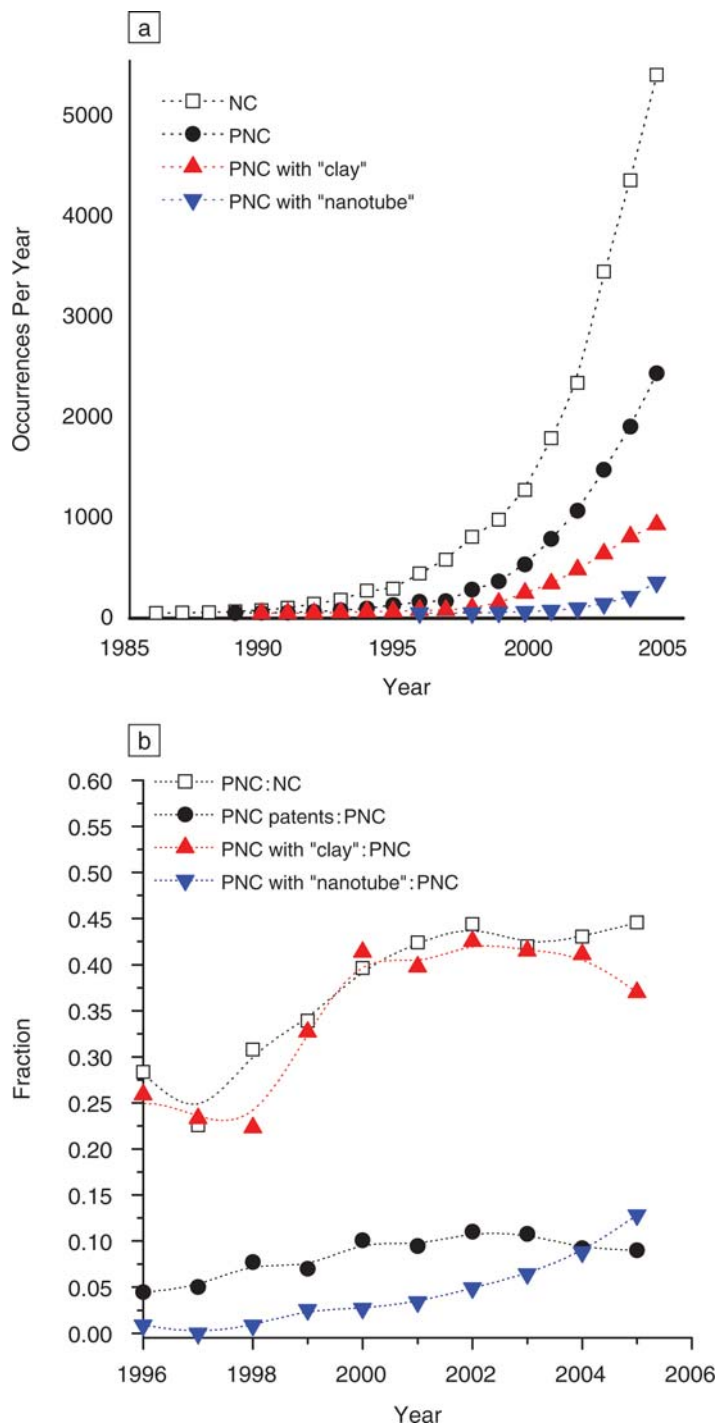


Figure 2. Growth trends of the polymer nanocomposite enterprise based on yearly publications catalogued in the CAPLUS and MEDLINE databases of the American Chemical Society.[15] (a) Number of occurrences per year of the term "nanocomposite" (NC, open squares) and "nanocomposite" appearing with "polymer" (PNC, solid circles). "Polymer nanocomposites" (PNC) is further refined to those discussing "clay" PNC (red symbols) and "nanotube" PNC (blue symbols). (b) Analysis of the number of citations per year, showing the total fraction of "nanocomposite" occurrences that discuss polymer nanocomposites (PNC:NC, open squares), as well as the total fraction of "polymer nanocomposite" occurrences that are patents (PNC patents, solid circles) that discuss clay-based PNCs (PNC with "clay," red symbols) and that discuss nanotube-containing PNCs (PNC with "nanotube," blue symbols).

## Future Outlook

What's next? Where are the groundbreaking opportunities? What are the challenges that pervade polymer nanocomposites? Of extreme importance in all the potential markets is the establishment of a better, quantitative understanding of the occupational health risks.[31] For polymer nanocomposites, this is particularly important during the production of nanosized fillers and composite fabrication, as well as during recycling, incineration, or combustion.

Whereas the recent increased availability of the new nanoscale fillers has been a major contributor to the rapid development of polymer nanocomposites, robust structure–property–processing relationships are critical to further market infiltration. Relationships that provide *a priori* predictions of macroscopic properties for a given polymer, a specific nanoscale filler (or fillers), and a particular spatial arrangement of the filler are still in their infancy. For example, to what extent can existing continuum composite theories be modified to account for the implications that arise when the filler is comparable to the polymer in size? Are the properties currently being achieved in polymer nanocomposites as high as we can expect to obtain?

However, approaches to these challenges are not without precedent. The underlying science and constitutive relationships for these nanoscopic materials should share commonality with collections of nanoscopic polymer chains, whose framework has been developed through nearly a century of chemistry and physics and is the foundation of the global polymer industry. Future developments toward the full potential of nanoscale multicomponent polymer blends rely on these previous insights to tackle the ambiguities associated with smaller filler sizes, where the distinction between filler and polymer fade into filler-molecules and polymer-molecules.

Economically, given the current diversity in nanoparticle cost (carbon black and montmorillonite versus single-wall carbon nanotubes), two approaches are developing based on potential markets. The lower-cost nanoparticles provide competition to traditional filler technologies and have important advantages in commodity applications, whereas the higher-cost nanoparticles target higher-value industrial sectors such as medical and electronics. Rather than replacing existing materials and traditional filled plastics, a common business strategy is to develop new applications based on the uniqueness of polymer nanocomposites, such as shape-memory materials for morphing aircraft, self-passivating films for satellites, and piezoresistive materials for MEMs-based sensors. In addition, new processing tools and on-line controls are being developed to either (1) uniformly distribute nanofiller to produce homogeneous bulk properties or (2) spatially vary the nanofiller concentration to meet specific design criteria. One might refer to these two classes of polymer nanocomposites as nano-"filled" systems and nano-"composite" systems, respectively. By drawing inspiration from biology and engineered fiber-reinforced composites, polymer nanocomposites with spatially controlled morphology are beginning to provide viable options to critical components of active devices, such as fuel cell membranes, batteries, photovoltaics, sensors, and actuators.

Polymer nanocomposites have recently become part of established modern technologies, but the most significant accomplishments of these materials are still ahead of us. As an increasing number of scientists and engineers contribute to the understanding of polymer nanocomposites, what remains to be seen is which products will be critically enhanced and enabled by this broad and evolving class of materials.

## Acknowledgments

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# Polymer Nanocomposites

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