



# A Review on Nanoparticle Filled Polymer Nanocomposites

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## Abstract

Nanoparticle filled polymer composites are a new class of engineering materials that are finding widespread applications in numerous fields owing to their tailorable mechanical, thermal and electrical properties that are superior to traditional monolithic materials. But, in order to realize the full potential of such nano composites, one has to ensure proper control over the uniform dispersion of nanoparticles within the polymeric matrices and should understand the interfacial reactions between the chemically distinct filler and the matrix. The present paper reviews the mechanical thermal and electrical properties of nanoparticle filled polymer nanocomposites. An effort is made to understand various fabrication processes employed to fabricate nanoparticle filled polymer nanocomposites.

**Keywords:** Glass Fibers; Nanocomposites; Nanoparticles; Nanotechnology; Polymers.

## 1. Introduction

Polymer Nanocomposites (NC) comprise of a group of three dimensional materials formed by combining different reinforcements with polymeric matrices in which one or more reinforcing materials are nanosized with lengths varying from 1-100 nm [1]. Nanoparticles (NP), due to their extremely small size tend to act as individual molecule that act as building blocks as such small molecules tend to play a significant role at the interfaces of a matrix and reinforcement. Thus, resulting in a material whose properties can be tailored to suit specific requirement [2]. It is, hence, imperative that the size of the NP play a significant role in determining the final properties of the composite material [3]. The nano-sized particles bring about chemical or physical interventions at the polymeric interfaces that yield a NC with improved properties in comparison with the un-reinforced polymer. Owing to their unique mechanical properties and tailorable thermal and electrical conductivities, the nanoparticle filled polymer NC find widespread applications in diverse areas which includes electronics, electrical, automotive, aerospace, biomedical and food processing industries.

### 1.1. Classification of nanomaterials

The primary factor which deciphers nanomaterials from others is their size [4]. Hence, the classification of nanomaterials is mainly done on the basis of their dimensional size [5]. They are broadly classified as follows:

Zero-dimensional nanomaterials: These particles possess length, breadth and width in nanoscale i.e., their dimension in all three axes is less than 100 nanometre. Examples include quantum dots and NP.

- One-dimensional nanomaterials: These materials have any one of their dimension, either length or breadth or width, in nano scale. This results in nanomaterials that are needle shaped.

- Two-dimensional nanomaterials: These materials have any two of their dimension in nano scale. Most of the nanofibers, including carbon fibres are two-dimensional nanomaterials.
- Three-dimensional nanomaterials: These materials have all three of their length, breadth and width in nanoscale. Nanoclays are good example of three-dimensional nanomaterial.

Researchers have tried to classify nanomaterials based on their potential end use and have grouped them in systematically listed risk categories [6]. According to risk-based classification, nanomaterials are classified as follows:

- High risk category: This class includes nanomaterials that are used in biomedical and toxicological studies.
- Low risk category: This class includes nanomaterials that are used in commercial applications such as automotive, aerospace, sport equipment, electrical and electronics.

### 1.2. Nanoparticles

Nanoparticles (NP) measuring less than 100 nano-meter (nm) have garnered a lot of attention from academic researchers as well as industrialists due to their adaptability in numerous application area spanning electrical, electronics, optics, automotive, aviation, biomedical, food technology, pharma and fast-moving-consumer-goods. The unique properties exhibited by nano-sized particles is due to their extremely high surface to volume ratio rendering them chemically very active. Thus, addition of even smaller quantities of NP to polymeric matrices can greatly alter the properties of the NC [7]. Metals and their oxides are employed for synthesizing the NP that find widespread application due to their excellent physical, thermal, electrical and mechanical properties. Thus, nanopowders of metals and metal oxides are generously incorporated as fillers in fabrication of polymer NC.

## 2. Nanoparticle filled polymer nanocomposites

Various types of NPs are used to fabricate polymer NC so as to improve mechanical and thermal properties. Inorganic NPs, due to their high surface to volume ratio, tend to setup strong reactions at the organic-inorganic interface when incorporated in polymeric matrices. This results in the fabrication of superior quality polymer NC with properties that can be tailored to suit specific needs. Effect of nanoparticle size and dispersion also plays an important role in determining the mechanical and thermal properties of polymer NC. NPs of a single uniform size are known to imitate isotropic materials allowing for easy incorporation of such NPs into molten polymeric resin that can then be extruded or injection moulded with a lot of convenience. Another important parameter that controls the uniform dispersion of NP in a polymeric matrix is its hydrophilic or hydrophobic nature. Hydrophilic NPs call for surface modification due to their extreme polarity which if overlooked can induce agglomeration of NPs in a polymeric matrix [8].

## 3. Fabrication methodologies of nanoparticle filled polymer nanocomposites

One of the primary factor that help in obtaining enhanced properties for polymer NC depends directly on the selected method of fabrication. Hence it is imperative to choose and employ the correct method of polymer NC processing. Broadly, the various fabrication techniques of polymer NC are classified into the following groups:

- In-situ polymerization
- Intercalation technique
- Sol-gel technique
- Mixing of NP with the resin

### 3.1. In-situ polymerization

In-situ polymerization involves the generation of NC through the onset of a chemical reaction. This technique involves the introduction of the NP within a monomer solution to form a homogeneous mixture. Upon addition of a suitable catalyst and under the application of heat, the polymerization of the homogeneous mixture of monomer and NP ensues forming a strong interfacial bond between the various phases of the NC [9]. Figure 1 illustrates the In-situ polymerization process.

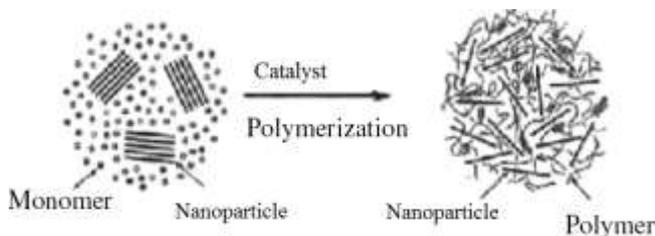


Fig. 1: In-situ polymerization.

### 3.2. Intercalation technique

Intercalation is a process of introducing a new molecule of one material into the crystal lattice of another material to generate a highly layered structure. Intercalation is most widely used fabrication technique for clay-based NC that resemble the properties of ceramic materials [10]. Intercalation is another method of fabrication employed for polymer NC wherein layered silicates act as reinforcement and a wide variety of monomers/polymers that fill in between the layered structure constitute the matrix. Direct intercalation method involves mixing of layered silicates into a polymer-solvent solution. This type of intercalation results in exfoliated silicate and up on solvent evaporation the polymer is thoroughly sandwiched between the silicate galleries yielding a highly

structured composite. In melt intercalation, the layered silicate compatible with the selected polymer is introduced into the molten polymer in the absence of solvent. The polymer intercalates between the layers of the silicate resulting in intercalated composite. Figure 2 illustrates the melt intercalation technique. Melt intercalation method is widely adapted owing to its compatibility with industrial mass production techniques such as extrusion and injection moulding. The absence of solvent renders the process safe and environment-friendly [11].

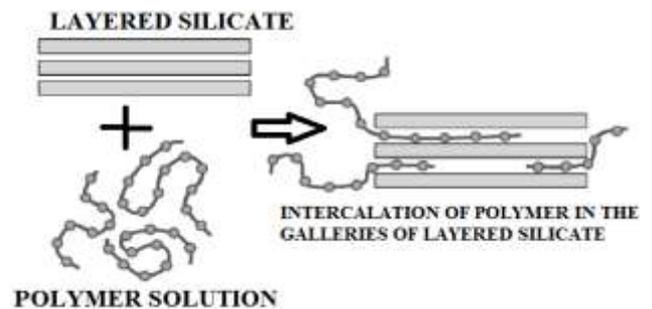


Fig. 2: Melt intercalation technique.

### 3.3. Sol-gel technique

Sol-gel technique involves the formation of a sol and a gel. Hydrolysis and condensation reactions of alkoxides result in the generation of a colloidal solution called Sol which contains angstrom to nanometer sized particles dispersed in the solution. The polymerization of the colloidal particles in the sol through the removal of stabilizing agents forms a continuous network of inorganic particles in a liquid phase which is termed as gel. The removal of liquid phase from the gel results in the formation of polymer NC filled with metal oxide NPs. The use of sol-gel processing techniques are relatively primitive that allows the formation of a varied geometrical structured materials, highly porous membranes, powders, thin-films and fibres. Sol-gel synthesis route enables the incorporation of both organic as well as inorganic materials to come up with a new class of engineering materials with tailorable properties [12].

### 3.4. Mixing of nanoparticle with the resin

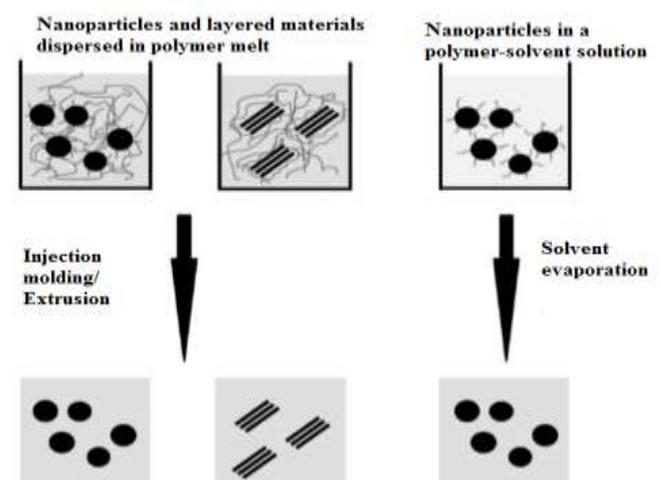


Fig. 3: Direct mixing technique.

A reasonably simple method of fabricating a polymer NC is through the direct mixing technique wherein the selected NPs are incorporated in the polymer at its glass transition temperature in the absence of solvent through mechanical means. This method is often termed as melt-compounding method. Another method is through the addition of NP in a solvent into which the polymer is then dissolved. The NC are obtained through the removal of solvent either through evaporation or via coagulation. This method of

fabricating the NC is called as solution-mixing method. Ultrasonication techniques help in uniform dispersion of the NPs within the polymer solution and is found effective in breaking down the agglomerated NPs thus resulting in a NC with superior properties [13]. Figure 3 depicts the melt-compounding and solution-mixing techniques for obtaining polymer NC.

#### 4. Mechanical properties of nanoparticle filled polymer nanocomposites

The mechanical properties of polymer NC is greatly influenced by the quantity of NP incorporated in the matrix, their size and the quality of wetting of NP with the polymer. The excellent stiffness possessed by the NP helps in enhancing the tensile modulus of the polymeric matrices that are known to be materials with poor tensile modulus. Another vital characteristic of NP is their ability to obtain a superior bonding with the polymer resulting in a strong interface. The strong interfacial bond allows smooth and easy transfer of stresses induced due to loading from the weak matrix to the strong reinforcements and thus resulting in a polymer NC with superior mechanical strength [14]. This fact also calls for thorough surface modification of NP as it is understood through various published sources that surface modification obtained from the employment of proper coupling agent reduces the hydrophilicity of the NP and in turn improves its wettability with the polymer matrix. Hussian et al. reported the improvement in tensile and flexural strength of the epoxy composite filled with surface modified titanium dioxide ( $\text{TiO}_2$ ) NP. In this study the treatment of the  $\text{TiO}_2$  particles was brought about by titanate and silane as two different coupling agents. The results favoured the use of titanate over saline coupling agent [15]. In another study ensued by Yoshida et al., saline proved to be a good coupling agent for silicon dioxide NPs that are filled in the epoxy matrix [16]. Rao et al. reported the effect of copper oxide ( $\text{CuO}$ ) NP on PVA NC. Authors observed that up to 2 weight percent (wt%), the tensile modulus and toughness showed a linear increment, while the trend reversed at higher weight percent addition of  $\text{CuO}$  NP. Higher concentration of  $\text{CuO}$  resulted in severe agglomeration of  $\text{CuO}$  NP within the polymer matrix resulting in the reduction of mechanical properties of the polymer NC [17]. Rithin kumar et al. found that incorporation of zinc oxide and tungsten trioxide NP have increased the tensile strength and Young's modulus of PVA composite films [18]. Another published report by Salehian and Jahromi also con-firms that higher concentration of NP beyond certain limit worsens the composite properties rather than improving it. The authors re-ported that  $\text{TiO}_2$  filled vinyl ester composites exhibited improved strength at lower concentration of  $\text{TiO}_2$  as opposed to its higher concentration within the matrix [19]. Carbon nanotubes (CNT) are now being widely incorporated as fillers in epoxy based composites owing to their excellent inherent properties [20]. But, the selection of the type of CNT bears a greater impact on the final mechanical properties of the composite. It is observed that the incorporation of multi-walled CNTs greatly enhance the mechanical properties of tensile and flexural strengths as compared to single-walled CNTs [21].

#### 5. Thermal properties of nanoparticle filled polymer nanocomposites

The percent composition of NPs within the matrix, their surface properties and the quality of dispersion of NPs in the given polymeric matrix bear a significant effect not only on the mechanical properties, but also on the thermal properties of the polymer NC. Kumar et al. investigated the thermal properties of titanium dioxide ( $\text{TiO}_2$ ) filled epoxy NC in which they reported through Thermo-Gravimetric Analysis (TGA) that epoxy matrix showcased good thermal stability with increasing  $\text{TiO}_2$  content ranging from 5% to 10% by weight, while the thermal stability of the epoxy resin decreased significantly 15 weight percent addition of  $\text{TiO}_2$ .

The Author's quoted the reason of extreme agglomeration of  $\text{TiO}_2$  NPs at higher particle loading in the resin [22]. Eksik et al. reported seven fold enhancement in the thermal conductivity of epoxy based composites through the incorporation of poly-methyl methacrylate nano-sized balls coated with graphene oxide as a core-shell filler. This significant improvement in the thermal conductivity of the NC over neat epoxy is attributed to the uniform dispersion of the core-shell filler within the matrix resin and also to the generation of extremely good pathways arising out of good interfacial bonding between the core-shell filler and the epoxy resin which facilitates conduction of photons at a faster rate [23]. Mandakini et al. observed that the addition of alumina NPs up to 5 wt% in an epoxy NC increases the glass transition temperature ( $T_g$ ) of the polymer NC [24]. NP are known to influence the thermal properties of many polymeric matrix composites through their close interaction with the base polymer matrix resulting in excellent interfacial bond. Haeri et al. studied the effect of addition of silicon dioxide ( $\text{SiO}_2$ ) NP along with graphene oxide (GO) to epoxy resin based hybrid NC. The  $\text{SiO}_2$  addition to GO sheets greatly improved the wettability of GO sheets resulting in a strong interfacial bonding with the epoxy matrix. This strong adhesion of  $\text{SiO}_2$ -GO NPs to the polymeric chain refrains the free movement of the NPs resulting in greater stability at higher operating temperatures [25]. Ramesan et al. observed the influence of silver-doped zinc oxide NP used as fillers in a matrix which is a blend of Poly (vinyl alcohol)/Poly (vinyl pyrrolidone). The composites exhibited excellent flame retarding capabilities as compared with the base polymeric blend. The glass transition temperature and thermal stability improved linearly with the weight percent loading of nano-filler into the polymer blend. The results indicated clustering of the NP at increased loading of NP resulting in the decrease of thermal stability of the composite [26]. Rajamanikam et al. prepared zinc oxide ( $\text{ZnO}$ ) reinforced polybenzoxazine NC and studied optical and thermomechanical behaviour. They observed that surface functionalization of  $\text{ZnO}$  NP greatly improved the interfacial adhesion with the polymer matrix. Authors concluded that  $T_g$  improved at higher loading of zinc oxide NP [27]. Martin-Gallego et al. investigated the thermal properties of carbon nanotube filled epoxy composites and concluded that uniform dispersion of NP within the polymer matrix has a direct bearing on the final properties of the composite and is always a big challenge as the NPs tend to agglomerate due to strong force of adhesion. In this study they found that the adoption of shear-mixing through the incorporation of a calender is a novel technique to achieve a uniform dispersion of the NP within the selected polymeric resin. Also uniform distribution of carbon nanotubes within the epoxy resin generates a network of conducting NP that greatly enhance the thermal conductivity of the composite [28]. Tessema et al. reported that the uniform distribution and optimum loading of silica ( $\text{SiO}_2$ ) and carbon nanotube (CNT) NP in epoxy matrix influences the thermal conductivity of the epoxy NC. The increment in  $\text{SiO}_2$  loading (up to 2 weight percent) into the epoxy resin results in approximately 35% increase in thermal conductivity of the composite. When the  $\text{SiO}_2$  loading was increased to 4 weight percent, there was a mere 14% increment in the thermal conductivity value for the composite in comparison with the base epoxy matrix which was presumed due to agglomeration of the NP at higher loading. In the next stage, when the  $\text{SiO}_2$  loading was taken to 6 weight percent, the thermal conductivity of the resulting composite increased by approximately 20% which is attributed to a close packing of particles ensuring a dense composite aiding in good conduction of heat. The investigators also reported the linear increment of the thermal conductivity of the CNT filled epoxy NC owing to the uniform dispersion of the CNTs within the matrix brought about by ultra-sonication method [29].

## 6. Electrical properties of nanoparticle filled polymer nanocomposites

NPs when used as fillers in polymeric matrices have resulted in the enhancement of thermal and mechanical properties without declining the dielectric strength and improves the order of magnitude of voltage endurance. Addition of even small quantity of carbon NP have shown to provide excellent electrical conductivity to most of the polymeric resins that are good insulators [30, 31]. The excellent electrical conductivity imparted to an otherwise insulating resin is achieved through a close control over particle loading. The exact particle loading that results in imparting good electrical conductivity is referred to as 'percolation threshold' which results in the formation of a continuous network of conductive NP within the resin matrix that transforms the insulator into an electrical conductor [32]. NP filled composites have the ability to respond positively to induced strain through elastic deformation before they fail. The induction of controlled strain in a NC can significantly alter its electrical and magnetic properties [33]. Studies involving inorganic NP fillers such as TiO<sub>2</sub> and ZnO, suggests an improvement in electrical permittivity of the epoxy based NC when the filler concentration is greater than 1 wt. % while the permittivity of the NC is at its lowest at the filler loading of 0.1 wt. % [34]. This proves the fact that the NC permittivity increases linearly with the filler concentration as ZnO and TiO<sub>2</sub> filler permittivity is higher than epoxy permittivity. Surface modification of NP fillers also has a direct bearing on the electrical properties of the polymer NC. In a study incorporating surface modified TiO<sub>2</sub> NP filled in a low density polyethylene matrix indicated the improvement in the dielectric breakdown strength of the NC resulting from the introduction of polar groups at the nanoparticle surface [35]. Surface modified ceramic NPs are used as a candidate fillers in epoxy based matrix in order to improve the thermal, physical and electrical properties of the NC. The surface modification of such ceramic NPs help in the uniform dispersion of the NPs within the matrix that in turn resulted in the pronounced effect of ceramic NPs which possess superior thermal conductivity coupled with an extremely low dielectric constant [36].

## 7. Conclusions

The paper outlines the fabrication, mechanical, thermal and electrical properties of nanoparticle filled polymer NC through an in-depth study of the available, published research works. It can be concluded that though nanoparticle filled polymer composites are an attractive and promising replacements to their conventional counterparts, it is needless to say that one needs to be cautious regarding the size of the selected nanoparticle, particle loading, surface modification techniques adapted and the fabrication method employed to ensure a polymer NC that exhibits superior and isotropic mechanical, thermal and electrical properties.

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