

Chapter

Synthesis and Properties of Epoxy-Based Composites

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Abstract

Epoxy-based composites are of great interest among academic and industrial researchers owing to their low cost, superior mechanical properties, large specific strength, super adhesiveness with good thermal and solvent resistance in recent times. However, the effect of carbon-based nanofiller reinforced epoxy composites is of prime focus due to their significant mechanical, dielectric and electrical performances for technological applications in broad fields of nanoscience and technology. There is a greater influence on the properties of the nanofiller reinforced epoxy matrix composites depending on the concentration of various types of nanofillers. The processing techniques play a crucial role in the prediction of attractive and suitable properties of the various nanofiller reinforced epoxy composites. There are several processing methods that have been employed to accomplish a superior degree of dispersion of nanofillers in the epoxy matrix. This current chapter portrays the simultaneous focus on their preparation techniques and effect of the dielectric, electrical and mechanical properties of various carbon nanofillers (such as fullerene, carbon nanotubes (CNTs), carbon nanofibers (CNFs) & graphene) filled epoxy resin composites for a broad spectrum of technological applications. We hope this chapter will facilitate the concrete in-depth ideas to the readers on the progress of various synthesis techniques and properties of different nanofiller reinforced epoxy composite systems.

Keywords: epoxy, nanofiller, carbon nanotubes, graphene, composites, mechanical, dielectric, electrical, properties

1. Introduction

The emergence of new technological fields is associated with the development of new hybrid polymeric composite materials with high-performance practical applications. These composite materials have several interesting multifunctional properties including superior strength, high stiffness or modulus of elasticity, durability, corrosion resistance, better thermal stability, enhanced electrical and electronic properties, lightweight with highest specific stiffness and strength along the direction of the reinforcing fiber, dimensional stability, good temperature, chemical resistance, flex performance and ease of processing with cost-effectiveness in contrast to other types of material [1–3]. However, there are different composite systems, which are used in the field of manufacturing technology. Among, epoxides, phenolics, polyurethane

and polyimides are commonly used as the matrix of the materials for the progress of advanced hybrid composite materials. The epoxy resin is one of the significant polymeric materials among academic and industrial researchers due to its remarkable versatile properties like thermal, electrical, mechanical performances, superior thermal and chemical resistance [4–7]. The thermoset resin such as epoxy is currently in wide use for the composite industry. This is because of their significant chemical, corrosion resistance, excellent adhesion performance, low shrinkage and lesser price with their challenging applications. Generally, the cured epoxy gives rise to large modulus, strength, good resistance to creep and high performance at elevated temperature due to its extreme cross-linked microstructure. The improved toughness of cured epoxy resin is commonly advantageous owing to its poor ductility. The betterment of toughness is a proficient approach for incorporating rigid or reactive rubbery particles into the epoxy network. It is the consequence at the expense of glass transition temperature and strength of this polymeric matrix [8–10]. Epoxy is intensely associated with daily life in the form of packaging, coating, adhesive and electrical insulating materials as well as applications in electrical appliances, semi-conductors, etc. [10, 11].

In the descended decade's, aluminum is one of the common metals around the globe, which is about 8% of abundance on earth's crust and seen in the form of oxides including karolinite, bauxite, nepheline and alunite. Sir Humprey Davy in the year 1808 revealed the existence of aluminum and further Oersted in 1825 formed its tiny pellets. However, Wohler a German scientist (1845) verified the specific gravity and aluminum lightness. They also discovered certain performances such as ease of deformation, air stability and its melting with a blow torch. The metals are characterized by high corrosion resistance, superb machining performances, superior thermal and electrical properties, large ductility, low strength, hardness and wear resistance [12, 13]. Due to high corrosion resistance and its lightness, aluminum and its alloy based materials played a vital role for the production of equipment (panels, roofs and frames) of packaging materials in the area of food and transportation (vehicle and aircraft parts) [13, 14]. Due to the broad spectrum of attractive properties and potential applicability of aluminum academic and industrial researchers have converged their focus on increasing the strength of aluminum and its hardness through solid solution and hardening. Also, they have progressed the aluminum-based metal matrix composite materials by the reinforcement of various fillers in the matrix of aluminum [12–14]. Recently, various researchers are putting their efforts to attempt for developing appropriate materials using aluminum-based alloys for end used applicability in the field of aerospace application, cast aluminum engine is used on flier, the manufacture of wind ribs (aluminum 2050). The high static strength of aluminum 7079 and 7075 are used to give sufficient toughness and corrosion resistances [3, 15, 16]. The composite of aluminum-graphite shows superior thermal conductivity as a result of the appreciable contribution coming out of the metal matrix. The aluminum-based metal matrix composite systems preserve the advantageous properties of both the reinforcement and matrix by associating the vital strength of the reinforcement with the ductility of the aluminum matrix [17, 18]. Many researchers have reported about the synthesis of carbon black powders by using agricultural byproducts including coconut shell, apricot stones, sugarcane, bagasse, nutshells, tobacco stems and forest residues. It is observed that the coconut shell shows costly disposal and cause environmental problem. Thus, they adopted an appropriate technique for the synthesis of carbon black using coconut shell owing to their superior natural structure and small ash contents *via* pyrolysis route (carbonization of coconut shell) with the application of temperature ranging from 550°C to 900°C. [3, 19–22]. However, activated carbon

developed through the conversion of coconut shell can be used as filler in processing the composites, which have potential utility in significant adsorbent for purification of water or industrial treatment and municipal effluents. The addition of these filler may also diminish the cost for waste disposal with a cheap alternative as compared to the commercial carbons [3, 21–24]. The byproducts (barley husk and coconut shell) reinforced thermoplastic is a better alternative for wood fibre-based hybrid composite materials. The experimental results revealed that barley husk and coconut shell are thermally stable at high temperature than that of soft wood fiber with different percentages of cellulose content (50% barley husk and 34% coconut shell) [25, 26]. Both coconut shell and barley husk are of large carbon-rich layers on their surface as compared to soft wood fiber. The superior tensile strength is observed in the barley husk fiber reinforced composite than that of the soft wood fibre-based composite. Moreover, the coconut shell and barley husk reinforced composite exhibited 80% and 40% improved elongation at break, 20 and 35% superior impact strength as compared to soft wood fiber composite systems [22, 27, 28].

The synthesis of composites is the combination of two materials such as matrix and reinforcement to form a hybrid composite material with excellent electrical and mechanical performances. The matrix is in the bulk form, which employs reinforcement with a strong bond [29–31]. However, the reinforcement is normally embedding additives for enhancing the properties of the material. The thermoplastic and thermosetting polymers are usually used as a matrix in the polymer composites. On the other hand, thermosetting polymer matrix exhibits better stiffness and superior strength than that of the thermoplastic polymeric materials [32]. Epoxy resin is one of the most commonly used thermosetting polymer matrix with better mechanical performances and good adhesive property with the incorporation of reinforcement particles [33, 34]. Thus, epoxy resin reinforced filler-based composites have immense interest among scientist, researchers for the development of hybrid composites for their use in the field of composite manufacturing industries, automobiles, paints and coating industries [35, 36]. The epoxy-based materials have common shortcomings including low impact strength and weak wear resistance, which can be overcome by selecting proper reinforcement of filler particles in the matrix; this will be useful in the field of tribological application. In epoxy-based composites, the epoxy reinforced nanoparticles show the large surface area with substantial interaction between matrix and filler particles [35–39]. However, nanocomposites are generally lightweight than that of the micro composites due to the relatively high density of the micro-additive fillers [40]. The enhancement of wear resistance and mechanical performances is due to the incorporation of hard oxide and carbide-based nanoparticles (including silicon oxide, alumina and tungsten) into the matrix [41–43]. A significant technique for the preparation of nanocomposites is needed for the homogeneous dispersion of the nanoparticles reinforced epoxy matrix *via* sonication technique [44]. The most crucial factors which influence the properties of the composites are curing conditions, molecular bonding between reinforcement and epoxy matrix with ratio of curing agent [45, 46]. There are various researchers have made to synthesize epoxy resin reinforced nano-filler-based hybrid composites for improving the mechanical and electrical performance of the composite systems. The different nanofillers (such as alumina, fullerene, graphene, carbon nanotubes (CNTs), carbon fibers (CFs), etc.) reinforced epoxy matrix composites result superior thermal conductivity, large thermal stability and better wearable resistance [44–47].

This chapter is organized on the basis of various filler-based nanomaterials used in epoxy resin composites, which includes carbon-based nanomaterials, fullerene,

graphene, CNTs, CFs, nanoclay reinforced epoxy resin composites have been presented in minute details in Section I. This section comprises of a brief introduction of the specific filler materials and its effect on epoxy resin followed by detailed discussion on the filler reinforced epoxy composite systems. Section II portrays the vivid insight into the synthesis techniques of carbon-based nano-filler reinforced epoxy composites. In Section III, we have especially emphasized on dielectric and mechanical properties of different filler reinforced epoxy resin composite systems.

2. Different carbon based nanofillers reinforced epoxy composites

Carbon-based epoxy composites are considered to be one of the most promising groups among the advanced materials of current times due to their distinctive physical and chemical properties. These materials exhibit significant properties as a result of the introduction of nanofillers into the matrix materials gives rise to unexpected properties, which make them distinguished due to unparallel design possibilities. In this context, Kroto et al. [48] in 1985 have discovered fullerene, which is the allotrope of carbon, where in fullerene the molecules of carbon atoms are well connected by single and double bonds. The family of fullerene is then extended with other forms of synthesis of carbon-based nanomaterials like single-walled carbon nanotubes (SWCNTs) (1991), multiwalled carbon nanotubes (MWCNTs) (1993) and graphene

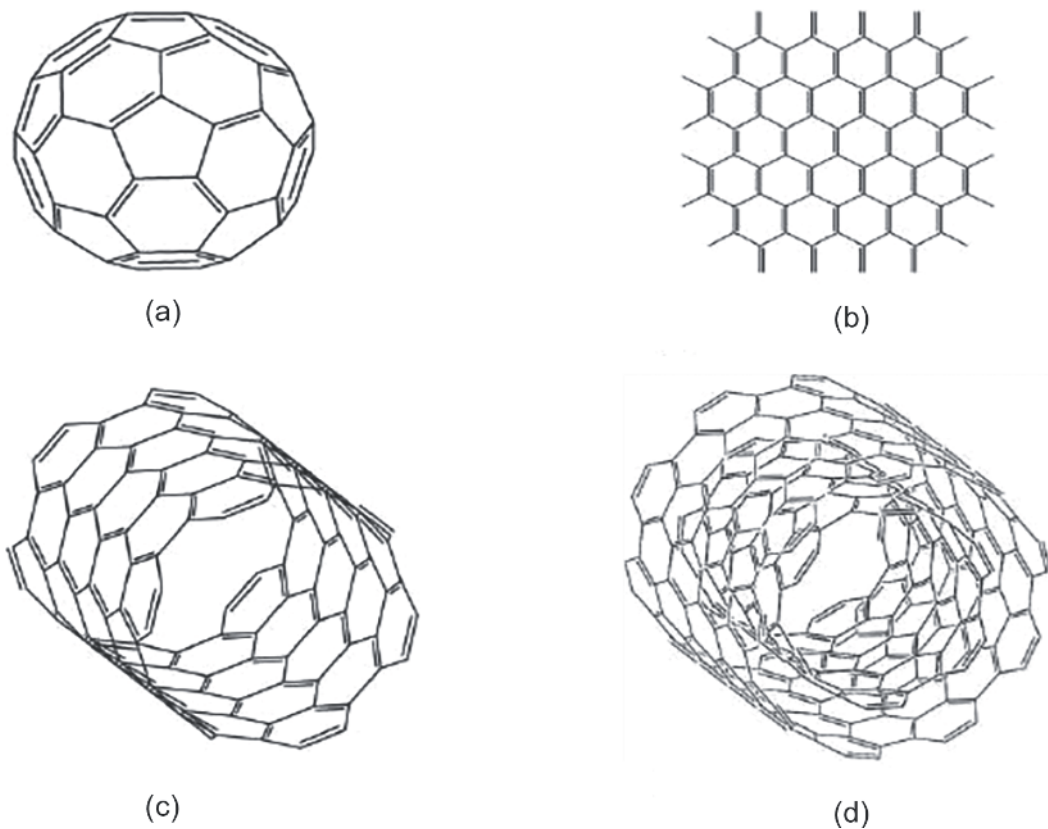


Figure 1. Carbon-based nanoparticles (a) graphene, (b) fullerene, (c) SWCNTs and (d) MWCNTs. Reprinted with permission from Ref. [49].

(2004) (as shown in **Figure 1**) have gained immense attention to the scientists for further research [49–55].

The carbon-based graphene nanofillers [49, 53–55] play an important role for the synthesis of composites in the field of nanoscience and technology. A single layer carbon sheet of graphene with sp^2 hybridization (two dimensional) is arranged in a hexagonally packed lattice structure analogous to a honey comb. They have unique performances including high charge mobility at room temperature, high surface area, good optical transparency, large young's modulus and superior conductivity. However, it is observed that fullerene may be considered as fascinating reinforcements in comparison to CNT or graphene due to their zero-dimensional structure of carbon molecules, which exist in the form of spherical, tube shape and ellipsoid. The fullerene related to C_{60} is called Bucky ball or Buckminster fullerene spherical in shape, which corresponds to a soccer ball [49, 54–56]. The carbon nanotubes (CNTs) [57–61] a new allotrope of carbon is normally thin hollow cylindrical fullerene structure in the diameter of nano-scale range and little micron length with significant properties. Recently, CNTs are considered to be the most talented candidates as reinforcement for polymer composites due to their high aspect ratio, high young's modulus, tensile strength, large thermal conductivity and approximately 1000 times larger than that of the electrical capacity than copper and thermally stable in vacuum at 2800°C [60, 61].

According to the number of concentric layers of carbon atoms, CNT is available at single-walled carbon nanotubes (SWCNTs) and multiwalled carbon nanotubes (MWCNTs), which shows multiple SWCNTs arranged in a concentric and coaxial manner as well as more flexible in nature than that of the MWCNTs. Moreover, CNTs appeared as outstanding materials and can be used as conducting nanofillers in the polymer matrix to give high-performance composites. Similarly, one dimensional carbon nanofibers (CNFs) [8] show a hollow cylindrical structure with lower cost and ease of processing than that of the pure CNTs. However, the carbon-based materials with a diameter less than 500 nm, do not exhibit structure of CNT falls into the class of CNFs and CNT, which exhibits clear structure as compared to CNFs. In this section, there are various studies have been made to achieve significant properties in epoxy-based carbon nanofillers composites [8, 57–61]. For instance, Kim et al. [62] have reported surface modified epoxy-based CNTs composites with superior homogeneity of CNTs in the epoxy matrix. They also studied their effect on the rheological and mechanical performances of the resultant composites. The multiwalled carbon nanotubes (MWCNTs) reinforced epoxy resin composites are of uniform dispersion in the matrix through the ultrasonication technique. The synthesized MWCNTs reinforced epoxy composites have superior Young's modulus and strength with optimized parameters were reported by Montazeri and his co-workers [63]. Allaoui et al. [64] have fabricated MWCNTs based rubber epoxy matrix composites with different concentrations of MWCNTs contents. It is observed that the resultant composite system has enhanced electrical performance with the increase of filler concentration in the epoxy matrix. However, the lower weight percentage (0.5 wt%) of MWCNTs reinforced epoxy composites exhibits a considerable increase of tensile strength and Young's modulus were reported by Montazeri and his group [65]. The incorporation of MWCNTs into the epoxy composites showed significant improvement of the thermal and mechanical performances *via* oxidation, acylation and amidation. It is also noticed that the enhancement of the electrical conductivity of the silane modified MWCNT-epoxy composites as compared to unmodified MWCNT-epoxy composites has been reported by Shen et al. [66]. Similarly, Choi et al. [67] have studied

the effect of silver-plated MWCNTs into the epoxy matrix. It is also noticed that the synthesized composites are of improved thermal conductivity with the increase of filler contents and time duration.

2.1 Fullerene reinforced epoxy composites

The contemporary demand for the development of lightweight hybrid composite materials with high specific strength, stiffness and improved tribological performance for end used applicability in the areas of aerospace and automotive industries [49, 56, 68]. During the past few times, fullerene and fullerene-based composites have been synthesized and used in various applications particularly in the thin films, organic polymers and hybrid organic-inorganic composites in the field of microelectronics [68]. There are various established techniques for the preparation of fullerene-based hybrid composites including solid-state reaction technique, liquid state techniques, deposition and spraying technique [56]. Rafiee et al. [69] have fabricated fullerene reinforced epoxy composites with various concentrations of filler content. It is revealed that the resultant composites with Young's modulus, fracture toughness, ultimate tensile strength were significantly improved in the epoxy matrix. It is reported that the incorporation of fullerene into the epoxy matrix shows superior properties as compared to neat nanosilica, nano-alumina and nano-titania filled epoxy composites; this may be due to the hollow structure, which results in increasing surface area and reduced weight simultaneously. However, the dispersion of fullerene into polymer matrix is easier than that of the other carbon based nanofillers which includes CNTs and CNFs, etc. The 1D and 2D filler-based materials are more prone to entanglement to each other than spherical fullerene particles. There are various research works related to fullerene modified polymer composites were reported by Ayesha et al. [70, 71]. However, the reports have been made based on fullerene modified epoxy-fiber composites. For example, fullerene modified epoxy carbon fiber reinforced polymer composites were fabricated and studied their effect of various mechanical performance of fullerene concentrations in the matrix. It is observed that 0.5% of epoxy matrix composites have improved interlaminar fracture toughness (60%) and also enhanced tension and compression up to 12% were reported by Ogasawara et al. [72]. Similarly, Jiang et al. [73] have reported about fullerene modified epoxy (1–3% concentration) reinforced fiber composites show improvement in the bonding strength between unidirectional carbon fiber and matrix. This improvement may be due to the suppression of fracture at interfacial layers of fibers, which is attributed to the presence of fullerene nanoparticles in the polymer matrix.

2.2 Carbon nanotube and carbon nanofiber reinforced epoxy composites

Several techniques including chemical vapor deposition (CVD), arc discharge and laser ablation techniques have been used to synthesize carbon nanotubes and CNT reinforced polymer composites in the past few decades [74–84]. The improvement of physical properties of the epoxy composites by the incorporation of different carbon-based nanofillers was reported by Liu et al. [85]. One of the key problems for the preparation of carbon nanotubes-epoxy composite system is the agglomeration or aggregation in the matrix. Thus, various techniques and studies related to CNT-based epoxy composites have been adopted to enhance the better dispersion and reduction of agglomeration or aggregation in the matrix [86, 87]. Moreover, the direct use of CNT in the epoxy matrix without surface treatment may give a

marginal improvement in the composites. Besides, the unmodified CNT embedded into the epoxy resin with one dimensional structure forms poor bonding, which is the high tendency to give entanglement with each other results in some problems. The structural performance is enhanced with the application of strong acids to convert the C–C bonds into various functional groups of amine, amide, etc. on the side surface of the CNT *via* plasma treatment or UV/ozone treatments [88]. In addition, the attachment of CNTs into the metal particles with enhanced homogeneity and interfacial adhesion results for improving the structural properties as compared to neat CNT-epoxy composites [89]. Similarly, carbon nanofibers are also creating some difficulties in uniform dispersion of these particles in the epoxy matrix owing to their structure. There are various physical (sonication, mechanical stirring, plasma treatment, high-temperature heat treatment) and chemical (surface functionalization through modifying agents, surfactants, etc.) methods are employed to synthesize homogeneous dispersion of CNF in the epoxy matrix. It is also observed that, using these techniques for preparation may affect the properties of the bulk composites. For example, the high-temperature heating technique for preparation of composites may reduce the interfacial strength, this lead to decrease the structural properties [90–93]. The high concentration of filler in the polymeric matrix may affect the final properties of the composites. To overcome this shortcoming, the incorporation of lower filler loading into the matrix gives better homogeneity as well as enhance structural properties of the composite systems. Moreover, for the utilities of CNTs various optimized parameters are essential to achieve better homogeneity, desired orientation and functionalization to extract the utmost benefits of this marvel material.

2.3 Graphene reinforced epoxy composites

The two-dimensional macromolecule graphene and its derivative have extensively been explored due to their application as nanofillers in graphene reinforced composites. The production of nanofillers must be attainable at a large scale with low cost for the progress of graphitic fillers from natural graphite, which make them suitable nano-filler-based materials for reinforcement in the field of composites *via* simple processing technique. These graphene-based composites are of immense interest among academic and industrial researchers owing to the excellent electrical, thermal and mechanical performance of graphene [49, 54, 55, 94–97]. However, there are various nanofillers including graphene nano platelets or sheets (GNPs or GNs) and graphene oxide (GO) shows additional flexibility at the nano-scale due to the stability of macromolecule and multitude of alternatives for more functionalization for the composite systems. An enormous modified forms of carbon fiber and polymer phase of carbon fiber reinforced composites (CFRC) are found in various literatures. The ultrafine GNP has been utilized to exfoliate graphite flake producing graphite nanoplatelets (GNP), on the other hand graphene oxide (GO) *via* modified hummer's technique exhibits oxidation followed by exfoliation of bulk graphite [96, 97]. Graphite is of strong planar structure with each carbon atom forming three covalent bonds with adjacent atoms. Whereas graphene oxide (GO) contains different functional groups (hydroxyl, epoxy and carboxylic acid groups) joined through sp^3 hybridized carbon atom but it may partially retain a number of sp^2 hybridized carbon depending on their reaction conditions. Moreover, it is also observed that the presence of polar functional group present in the surface of the graphene oxide, which results in bonding with polymer matrix to form strong interfaces. It is reported that the significant properties of graphene play an important role in the scientific research communities to develop

graphene-based epoxy matrix for the application in structural composites. Besides, these fillers are applied for additional needs to develop composites with attractive properties like thermal, electrical conductivity and mechanical strength [98–101]. Alexopoulo et al. [102] reported on the fabrication of epoxy-based GNP composite systems and studied their various properties on the effect of size on the GNP particle embedded into the epoxy matrix. It has also been noticed that the synthesized composites show the formation of agglomeration at higher concentrations (>5 wt%) within the matrix. Further, at lower concentration (0.25 wt%) of GNs, it is revealed that there is considerable improvement in properties (toughness, flexural strength and flexural modulus) of the composites.

The epoxy-based composites using reduced graphene oxide act as fillers, which give rise to significant improvement in properties (tensile, impact and flexural) of the resultant composite systems. However, it is interesting to observe that the functionalised graphene nano-sheet (GNS) reinforced epoxy composite at lower filler concentration shows superior fracture toughness, fracture energy, stiffness, strength, and fatigue resistance than that of the neat CNT based epoxy composites [103–106]. The advantages of GNS as compared to CNTs raise the structural properties of epoxy, which may be attributed to the larger surface area with improved adhesion of filler-matrix owing to the wrinkled surface and the two-dimensional geometry of GNS [105, 106]. Graphite continues to attract considerable attention among researchers due to its excellent mechanical, electrical properties, low density with ease of processing and low cost. However, graphite commonly exists as a layered material and these layers are closely packed through Van der Waals force. For the efficient use of graphite as filler in polymer composites, its layer must be separated partly to achieve expanded graphite (EG). It is difficult to intercalate monomers into the interlayer of graphite to produce composites if the raw graphite is used as reinforcement; also, it is not possible to disperse graphite layers in the epoxy matrix. To overcome this adversity, the preparation of expanded graphite (EG) from raw graphite is exposed to a strong oxidizer (e.g., HNO_3 , H_2SO_4 , KMNO_4). The EG and GO both are the derivatives of graphite and are considered to be ultimate nano-filler materials for epoxy matrix. It is due to the presence of covalent bonds having hydroxyl, phenolics and epoxide functional groups on their basal planes and also it is located on the carboxyl and carbonyl group at the sheet edges. The presence of these functional groups enables them to strongly hydrophilic in nature. The EG is readily dispersed in water and reinforced into matrix with the help of these functional groups for the synthesis of composites [107–109]. However, two-dimensional graphene-based nanomaterials show little agglomeration because of their high aspect ratio, which affects the mechanical properties of the resultant composites. Therefore, suitable dispersion and exfoliation techniques are essential to facilitate better structural properties.

2.4 Nanoclay reinforced composites

The nano-clay reinforced polymer-based composites have earned much attention among both academic as well as industrial sectors due to incorporation in a small amount of nanoclay considerably improves the mechanical performance of the neat polymers. The nanoclay based two dimensional nano-material are naturally occurring in the form of platelets, which include a few to 1000 sheets. These are mainly silicate and comparatively inexpensive than that of the other nanomaterials. The other types of nanomaterial possess a larger surface area with a high aspect ratio (>50) and are thermally stable. Usually, the use of nanoclay in the matrix improves their properties

with good optical transparency for a suitable selection of nano-filler reinforced in the polymer composites [49, 110].

Montmorillonite (MMT) is an aluminosilicate [111], which is extensively used in the clay based nanofillers (**Figure 2**). The thickness and lateral dimensions are observed in the montmorillonite around 5 nm and 500 nm, respectively. The single sheet of montmorillonite has a stiffness of about 250 GPa. Moreover, the large stiffness was observed in the clay minerals, which makes them appropriate for improving the structural performance of the polymer-based composites [111–113]. In recent few times, it has been reported about the epoxy-based glass and carbon composites, which may be embedded in the glass reinforced polymer composites. In addition, a montmorillonite (Nanomer I.30E) based clay mineral was used in this research [49]. They have reported each platelet are approximately 1 nm thickness and internal dimension around 300–600 nm with a high aspect ratio. Moreover, the compression strength is improved about 15–20% for particularly 5 wt% of nanoclay reinforced epoxy matrix composite systems. It is also observed that the incorporation of a small amount of nanoclay into the epoxy composites enhanced the mechanical performance using reinforcement of clay particles in the polymer matrix, which results the increase in impact and interfacial properties of the composite systems. Especially, the current research is based on nanoclay composites, which consist of the polymer as matrix material and nanoclay as the reinforcement particle (act as nanofiller) in the composite systems. Several properties (including mechanical, thermal and electrical) and structural aspects of the polymer matrix are enhanced due to the incorporation of nanoclay [114, 115]. These nanoclay based materials have been used as cost effective substituents with significant strength characteristics and considerable enhancement of properties on the nanoclay reinforced polymer-based hybrid composite systems. Several studies have been carried out on nanoclay reinforced polymer composites [113–119]. For instance; Hussain et al. [120] have reported that the natural fiber composite is robustly dependent on the optimum fiber length and weight percentage

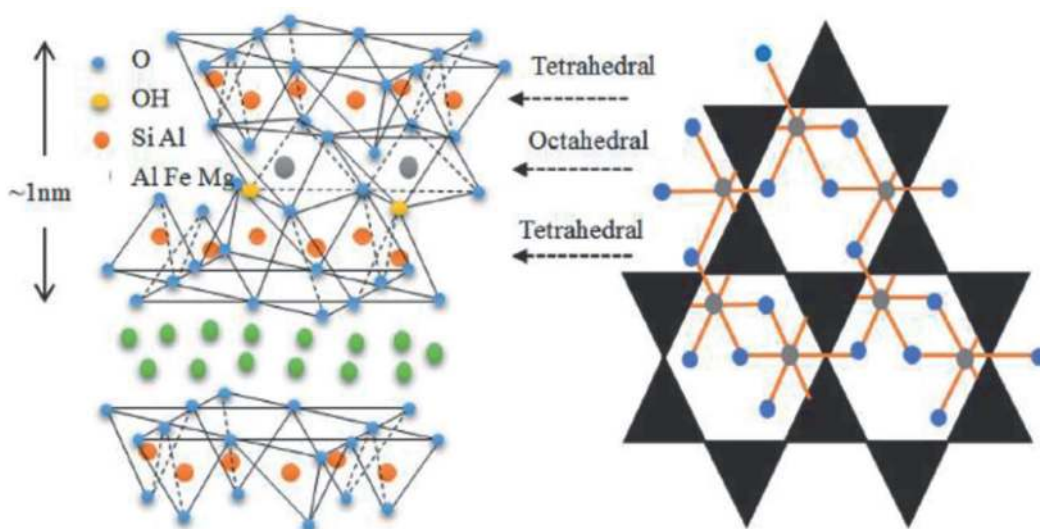


Figure 2. Schematic illustration of the montmorillonite clay (MMT) structure. Side view of tetrahedron units of MMT assembled through weak van der Waals and electrostatic forces to form the primary particles and top view of MMT shows hexagonal structure of oxygen and hydroxyl ligands of the octahedral layer. Reprinted with permission from Ref. [111].

of fiber. It is also reported that the mechanical properties of the polymer composite system are improved by the incorporation of a little amount of nanoclay in to the matrix [121].

3. Synthesis of carbon-based nano-filler reinforced epoxy composites

Several methods have been employed for the synthesis of various nano-filler reinforced epoxy resin composites in recent few times. In this section, we have emphasized mainly on three methods (including in-situ intercalative polymerization, solution mixing and melt blending) for preparation of composites, which are extensively discussed in details.

3.1 *In-situ* intercalative polymerization method

In situ polymerization is a very effective method that allows carbon-based filler particles to be uniformly dispersed in the matrix and therefore gives strong interaction between polymeric matrix and reinforcing filler in the composite systems. This technique generally involves the polymerization of monomers in the presence of dispersed filler particles. The polymerization is initiated through the incorporation of filler and suitable initiator with the application of heat or radiation after diffusion during the synthesis of the polymeric materials [122, 123]. Several researchers have been reported about the preparation of composites by using *in situ* polymerization methods and achieving superior mechanical properties with the low value of percolation threshold as compared to the other techniques (like solution compounding or melt blending method) [124–127]. However, in-situ polymerization is also been used to give non-covalent composites based on different polymeric matrices including polyethylene (PE) [128] and PMMA [129], etc. Ray and Okamoto [130] reported that the *in-situ* polymerization method referred as intercalation polymerization method is applied for the preparation of GDs based nanocomposites. Zhang et al. [124] have fabricated graphene sheet reinforced epoxy resin composites by in-situ polymerization method. It is observed that the prepared composites have significant improvement in the Young's modulus and thermal stability at 0.7 wt% of GNs content in the matrix. It is also noticed that there is an extensive enhancement in the thermal conductivity of the resultant graphene sheet-epoxy composite systems using in-situ polymerization method [131]. Ying and his co-workers [132, 133] have fabricated surfactant incorporated CNT into the epoxy composites *via* in-situ polymerization technique. It is observed that the tensile strength and ultimate strain are enhanced with an increase in the modified filler contents in the CNT reinforced epoxy composite systems.

Kotsilkova et al. [134] have reported on amine and epoxy grafted MWCNTs based composites. They have successfully synthesized and measured various performances (including rheological, dc conductivity, radio frequency and microwave properties) of the composites. The enhancement of the thermal and mechanical properties of the MWCNTs reinforced epoxy composites were utilized by in-situ polymerization method, which is reported by Theodore et al. [135]. The preparation of the epoxy-based nanocomposites using functionalized vapor grown carbon nanofiber (VGCNF) by in-situ polymerization technique. It is revealed that the functionalized VGCNF-epoxy composite systems have superior tensile modulus and strength than that of the neat VGCNF and epoxy matrix with enhanced thermal stability [136]. The carbon

nanofiber (CNF) based epoxy composites with various weight percentages of silane-modified CNF and unmodified CNF contents *via* in-situ polymerization technique were prepared. It is revealed that the synthesized composites with different properties (thermal, mechanical and electrical) were analyzed as a function of different weight percentages of modified and unmodified CNF contents [66, 137]. The key features of this technique offer covalent bonding between surface-functionalized CNTs and polymeric matrix, which result enhancement of mechanical performance of the composites with strong interfacial bond [86, 138]. On the other hand, the shortcomings of this method are that the dispersion of filler particles in the polymeric matrix occurred during the preparation of nanocomposites, so the plenty of energy is necessary, which might affect the large production of polymer-based nanocomposites. Similarly, Ramezanzadeh et al. [139] have prepared conducting polypyrrole (PPy) and zinc doped polypyrrole functionalized graphene oxide (GO) nano-sheets for the development of high-performance epoxy-based composite systems with enhanced thermal and mechanical performance. The results showed that the nanocomposites are enhanced about 54% for elongation at break and 115% for the energy at break, and there is an improvement of tensile strength and energy (21% and 32.44%) at break for the zinc doped GO-PPy in the epoxy composites.

3.2 Solution mixing/solution blending/solvent casting

Solution-induced intercalation technique is a simple, ease of processing and efficient approach to produce polymer-based nanocomposites. This technique is more suitable for both small- and large-scale preparation of the nanocomposites. In this technique, initially an appropriate proportion of polymeric mixtures were dissolved in a proper solvent. Then the different types of filler particles (such as graphene, carbon nanotubes (CNTs), carbon nanofibers (CNFs), etc.) at various weight percentages are reinforced into the polymeric solution with constant stirring either by magnetic stirrer or mechanically or by using ultrasonication technique for dissolution of the particles in the matrix. Once the complete mixing of solution is over, then the mixtures were dried to eliminate the solvent and ultimately resultant composite materials are molded with suitable mold to give necessary shape and making suitable for characterization and further properties measurement. Similarly, the solvent casting method has been used to synthesize composites in the form of thin films. This method deals with the uniform dissolution of ingredients in a suitable solvent and then evaporated through a drying device. However, the solvent casting technique shows higher mixing quality, thinner film, high purity and better clarity than that of the melt mixing technique [140]. Besides, the solution or polymer film is exposed to moderately low thermal or mechanical stress during the preparation technique, which results insignificant degradation or side reactions.

There are various literatures available with epoxy-based composites by using this technique and then studied several properties of those composites as a function of different weight percentage of filler contents. Similar to epoxy resin, other various polymers (such as PS, PMMA, PVDF and PI) and its co-polymers [P(VDF-HFP), P(VDF-TrFE), etc.] are used to fabricate nanocomposites with the addition of several fillers into matrix using this technique [141–146]. For instance; Lv et al. [147], Prolongo et al. [148] and Allaoui et al. [64] have fabricated carbon nano-fibers reinforced epoxy-based composites with different weight percentages of CNF contents and studied their properties. Similarly, Choi et al. [149] have synthesized polycarbonate-based carbon nanofiber composites. They have also examined mechanical

properties and electrical resistivity with the incorporation of various percentages of CNTs contents. Moreover, the limitation of this technique is the requirement of the high amount of solvent in the preparation of nanocomposites for industrial point of view, which may not be environmentally friendly and cost-effective.

3.3 Melt blending method

Melt blending is the facile, cost-effective, eco-friendly and conventional technique for the synthesis of various thermoplastic polymer-based composites. In this process, there is no such solvent used for preparation. Also, it is one of the most efficient approaches for industries in large scale production of the composite materials. During the last few times, most of the industrial researchers have preferred this synthesis technique for preparing nanocomposites. In this technique, initially, polymeric materials and filler particles are normally mixed systematically and then the homogeneous mixture is subjected to annealing greater than the melting point of the polymeric material. Besides, the polymeric material is melted and combined with apposite amount of the filler particles using an extruder. The process of melt blending is carried out in the presence of an inert gas including argon, nitrogen or neon. However, this melt blending technique has enormous benefits over in-situ intercalative polymerization or polymer solution intercalation method. This technique is compatible with present industrial processes including extrusion and injection molding techniques and thus it is a most popular method. Various academic and industrial researchers have fabricated different polymer-based nanocomposites (PP-CNT, nylon 6-MWCNT, PC-MWCNT and PS-MWCNTs, etc.) using this technique [150–153]. For instance; Jin and his co-workers [154] have fabricated PMMA-MWCNTs based composites by using this technique. In the synthesized composite system CNTs shows no obvious damage or breakage as well as uniformly dispersed within the polymeric matrix. The preparation of polylactide (PLA) exfoliated graphite (EG) based nanocomposites was synthesized using this technique. It is seen that the composite exhibits a considerable increase in the thermal degradation temperature with the increase in the EG contents. The mechanical and electrical performances are also improved with the continuous increase of graphite contents. A similar work on carbon-based material (such as graphite nano-sheet and MWCNTs, etc.) reinforced epoxy composites is fabricated by Kim et al. and his group [155, 156] using this technique and also studied their mechanical and rheological properties of the resultant composites.

4. Different properties of filler reinforced epoxy composites

Recently, epoxy-based composites are of great importance for their significant dielectric, electrical, thermal and mechanical performances. In this section, we have especially emphasized on properties (dielectric, electrical and mechanical) of the various filler reinforced epoxy-based composites.

4.1 Dielectric and electrical properties of epoxy-based polymer composites

Varma et al. [157] have fabricated calcium copper titanate ($\text{CaCu}_3\text{Ti}_4\text{O}_{12}$; CCTO) and metallic aluminum (Al) powder-filled epoxy-based tri-phase composites with various weight percentages of filler contents. It is observed that the dielectric constant

of the composites is greatly improved near the percolation threshold. The maximum dielectric constant was achieved (≈ 700) of the three-phase epoxy composites, which is much larger than that of the two-phase epoxy-CCTO (≈ 70) and neat epoxy matrix. These flexible three-phase composites are potential candidates for practical application in the field of energy storage devices [158]. The perovskite-type ceramic (BaTiO_3) based epoxy composites with a different weight percentage of filler contents have been reported by Kuo et al. [159]. They studied the dielectric properties of these composites and obtained a high dielectric constant (≈ 44) and negligible dielectric loss (< 1) at 40 wt% of filler content in the epoxy matrix. The synthesized three-phase composites have been uniformly dispersed in the polymeric matrix. However, the dielectric constant of these composites is proportional to the volume ratio of the ceramic contents and remained constant with the application of temperature and frequencies. The fabricated composites have larger than that of the commercial ceramic-filled polymer composites. Bhattacharya and Tummala [160] have fabricated PMN-PT filled epoxy composites with various weight percentages of filler contents. It is revealed that the high dielectric constant (≈ 29) was achieved in the 40 wt% of filler-filled epoxy composites with superior homogeneity within the epoxy matrix. The dielectric performance of Ni particle doped epoxy composites at 40 and 55 wt% of filler contents have been analyzed in the various frequency regions from 1 to 10^7 Hz and temperature range of -20 to 200°C . The dielectric constant of these Ni particle filled epoxy composites is improved with the increase of Ni particle content and simultaneously reduces the frequency at room temperature. The value of the dielectric constant is increased with the increase of filler contents due to the improved dipole and interfacial polarization effect. On the other hand, the dielectric loss value was reduced with the higher concentration of filler contents has reported by Chen and his co-workers [161]. The multifunctional polymer-based composites comprising of Fe_3O_4 fillers and epoxy resin as the matrix was analyzed in different concentration of magnetite Fe_3O_4 contents. It is observed that the composites with larger dielectric constant and suppressed dielectric loss at low frequencies. However, the synthesized composites with a larger volume fraction of filler contents appeared at the percolation threshold with an increase in the value of conductivity [162]. Xie et al. [163] have fabricated the modified hexagonal boron nitride reinforced epoxy composites with enhanced dielectric properties *via* simple free radical polymerization technique. It is observed that PGMA grafted h-BN reinforced epoxy matrix composite has improved thermal conductivity with increase of filler contents. The modified composite shows larger dielectric constant than that of the unmodified one and pristine epoxy matrix. Similarly, the dielectric loss of the resultant composites was achieved negligible at suitable frequencies. Zhang et al. [164] have fabricated core-shell satellite structured BaTiO_3 nanoparticles with polydopamine (PDA) layers and silver (Ag) nanoparticles incorporated into the epoxy matrix. It is observed that the epoxy nanocomposites with BT-PDA and BT-PDA-Ag fillers showed enhanced dielectric constant (≈ 9) and negligible dielectric loss (0.024) at microwave frequencies (10 GHz) for 20 vol% of filler contents. However, it is also noticed that the composite shows homogeneous dispersion with uniform particle size, due to strong interfacial interaction between modified particles and polymer matrix. Meng [165] and his co-workers have developed thermally stable honokiol derived epoxy resin nanocomposites with excellent thermal and dielectric properties. The results showed that the fabricated composites have excellent dielectric constant (9.74) and minimized dielectric loss values (0.026) at 1 KHz. Moreover, it is also confirmed the better thermal stability, thermal conductivity and high specific heat in the composites.

4.2 Mechanical properties of epoxy-based composites

Zhao et al. [166] have synthesized hyper branched graphene oxide structured based epoxy nanocomposites. The results showed excellent engineering application performances of the composite systems. The synthesized nanocomposites have uniform dispersion in the epoxy resin matrix and are combined with the matrix through chemical bonds, which shows strong interfacial active force and enhancing the load transfer efficiency of the matrix to hyper-branched polymer-graphene composites. However, these composites exhibited excellent mechanical properties [impact strength (58.53%), tensile strength (83.29%), and compression strength (57%)] with considerable increase of 0.2 wt% for HPB-GO contents than that of the neat epoxy matrix. Also, it is noticed that there is 80% increase in thermal conductivity ($0.32 \text{ W m}^{-1} \text{ K}^{-1}$) of the synthesized nanocomposites. The incorporation of rGO in the epoxy matrix in the composites improved the strength and Young's modulus is about 500 and 70%. It is observed that rGO plays a significant role in strengthening the epoxy than that of the GO. However, rGO efficiency in the improvement of modulus and strength is about 10–35% than those of GO. The significant results for r-GO composite are due to the efficiency of interfacial adhesion between r-GO sheet and epoxy molecules [167]. The enhancement of strength can be ascribed to the outstanding load bearing capacity of reduced GO sheets as well as excellent load transfer from matrix to reinforcements. They also suggest an increase in the value of modulus and strength in the composites for 1 wt% rGO reinforced polymeric matrix. The fabrication of graphene- nano-alumina based epoxy composites reported by Osman and his co-workers [168]. It is observed that both tensile strength and storage modulus are improved by 22.56% and 4.6%, which is much larger in contrast to the neat epoxy matrix. The thermal conductivity of the resultant composites is also improved by 23.4% with increase in the filler contents. The incorporation of alumina particles on the surface of the graphene not only reduces the electron transfer but also eliminates the agglomeration of graphene. Khan [169] and his co-workers have reported improvement of thermo-mechanical performance of carbon fiber and glass fiber reinforced epoxy composites. These composites were characterized by using universal testing machine (UTM) with tensile strength and Young's modulus. The tensile strength of carbon fiber reinforced epoxy composites is enhanced to 844.44%, 951.11% and 1122.22% with selected 40, 50 and 60 wt% of carbon fiber contents. On the other hand, the tensile strength of glass fiber reinforced epoxy composites is also enhanced about 156.66%, 171.10% and 197.77% for 40, 50 and 60 wt% of glass fiber contents. Karle et al. [170] have systematically studied CaSiO_3 particulate fillers reinforced epoxy-based composites and analyzed their hardness, flexural strength and impact resistance through mechanical performance. It is found that the addition of CaSiO_3 particles (1–2 wt%) in the matrix results effective enhancement of mechanical properties than that of the neat epoxy. Park [171] and his co-workers have investigated the reinforcement effect of molybdenum sulfide (MoS_2) nano-sheets on the mechanical performance of the epoxy-based composites. The fabricated high performance epoxy composites are extremely enhanced the fracture toughness (55–81%), flexural strength (25–66%), modulus (0.7–6%), impact strength (31–118%) and strong interfacial interaction (1–21% surface free energy) than that of the pristine epoxy matrix. The thermal and mechanical performance of the epoxy reinforced modified iron oxide nanoparticles reported by Baghdadi et al. [172]. It is revealed that the PDA modified Fe_3O_4 based epoxy composites normally improved as compared to neat epoxy matrix and unmodified one. The maximum enhancement in

tensile strength (34%) and fracture toughness (13%) is observed in the epoxy-based composites. The graphene oxide-epoxy composites with improved failure strength (48.3%) and toughness (1185.2%) for 0.0375 wt% of GO within the epoxy matrix. The fabricated composites with enhanced properties may be due to the uniform dispersion of the GO in the epoxy matrix through two-phase extraction technique using an aqueous dispersion of the GO were reported by Yang and his co-workers [173]. Bortz et al. [174] have reported helical carbon nanofibers to achieve graphene nanoribbon and then oxidized to get GO. This composite exhibit improved tensile strength (7.57 MPa) and modulus (3.32 GPa) at 0.5 and 0.1 wt% of GO loading into the polymer matrix. Moreover, the flexural strength was also improved by 12% and 23% for the addition of fillers into the matrix.

Munoz et al. [175] have synthesized GO based epoxy composites and studied their mechanical properties for various wt% of GO contents. It is revealed that the composites with enhanced elastic modulus and flexural modulus at 0.3 wt% of GO contents in the epoxy matrix. Fang et al. [176] have reported methylene dianiline (MDA) modified rGO-epoxy composites with different weight percentages of filler contents. In these composite systems, the fracture toughness and flexural strength is enhanced by 94% and 92% for 0.6 wt% filler loading. Seong et al. [177] have fabricated MDA modified GNP-epoxy composites and investigated their mechanical properties. The 1.5 phr MDA modified GNP content shows 120% and 63% enhancement in impact toughness and storage modulus in the epoxy-based composites. Naebe [178] and his co-workers have reported the effect of thermally reduced graphene oxide (TRG) with bingel reaction (FG) based epoxy composites. It is observed that 0.1 wt% of filler content in the FG-epoxy composite shows larger flexural strength (15% and 22%) and higher storage modulus (6% and 16%) than that of the pristine epoxy matrix. Similarly, Guo et al. [179] have synthesized GO modified triazine derivatives (GO-TCT-DETA) show homogeneous dispersion in the epoxy matrix. It is also exhibited higher flexural strength (49%) and modulus (15%) for 0.1 wt% of GO-TCT-DETA-based epoxy composites than that of the pristine epoxy matrix.

5. Summary

The epoxy-based composites have become potential candidates for the application in different technological fields owing to their excellent physical, chemical and electrical performances. The present chapter reviews on the research work related to carbon based nanofillers (e.g., fullerene, CNTs, CNFs, graphene & nanoclay) reinforced epoxy-based composites. However, several researchers have reported the variety of reinforcement in order to explore mechanical, dielectric & electrical performances to predict their behavior suitable for technological viability of the epoxy-based composites. According to the literatures, it has been found that the use of nanofillers into the epoxy matrix, which help to enhance the mechanical and electrical performance of the composite systems than that of the micron sized filler particles. Moreover, with the increase of concentration of nano-filler content results in the enhancement of certain properties of the epoxy-based composites up to the threshold value, in most of the cases after attaining the threshold value the properties again initiate to fall, this may be ascribed to the presence of large clusters into the epoxy resin. The incorporation of minimum amount of nano-filler contents is indispensable into the matrix to avoid agglomeration in the composite systems. Therefore, we have especially emphasized on most suitable synthesis techniques for the processing of nano-filler

reinforced epoxy-based composites. Further, our efforts have been made for providing higher insight on the properties (like dielectric, electrical and mechanical) of various nanofillers reinforced epoxy composites. The unique properties of nanofiller reinforced epoxy composites also reflected as potential applicability in the field of electronics, energy storage, gas sensors and aerospace.

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Conflicts of interest

The authors declare no conflict of interest.

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