

Reinforcing Effect of Graphene in Epoxy Adhesives: Review

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Abstract

Due to its great strength, hardness, and chemical resistance, epoxy adhesives are becoming more and more used. They continue to have drawbacks, nevertheless, such as poor thermal stability, and poor electrical conductivity. Two-dimensional graphene is a wonderful substance with exceptional qualities including high strength, high electrical conductivity, and large surface area. Because of these characteristics, graphene has been thoroughly researched for its prospective uses in a variety of industries, including electronics, energy storage, and biomedical engineering. The use of graphene as an additive in epoxy adhesives to enhance the characteristics of such materials is one of its promising uses. This paper reviewed the latest findings about graphene's effects on epoxy adhesives. The various methods to produce graphene-epoxy composites and their improvements are discussed. This research additionally discusses the challenges associated with the production and processing of graphene-epoxy composites, as well as the mechanisms behind the improvements in mechanical, electrical, and thermal characteristics. The final section of this review discusses the challenges and prospective uses of graphene in epoxy adhesives in the future.

Keywords

Graphene, Epoxy, Adhesives, Composite, Mechanical Properties

1. Introduction

Due to their outstanding bonding abilities, great mechanical strength, and heat stability, epoxy adhesives are often employed in a variety of sectors. Nevertheless, the brittleness and poor durability of epoxy adhesives, which can cause premature breakdown under mechanical stress, frequently limit their application. Researchers have concentrated on adding nanoparticles to epoxy adhesives to improve their mechanical characteristics to get beyond these constraints [1] [2].

Due to its outstanding mechanical, thermal, and electrical qualities, graphene has drawn a lot of interest among other nanomaterials as a reinforcing ingredient for epoxy adhesives. The structure of graphene oxide is shown in Figure 1. A hexagonal lattice of carbon atoms makes up the two-dimensional substance known as graphene. It possesses exceptional mechanical qualities, including high strength and stiffness, as well as a high aspect ratio and surface area. The high thermal and electrical conductivity that graphene possesses further makes it a prime choice for increasing the thermal and electrical characteristics of epoxy adhesives. From this review, it has been observed that several researchers and studies have reported that the initial hurdles to enabling graphene's practical applications are the synthesis of high-quality graphene derivatives in a robust technique and mass scale. Four sections went into preparing this review. The production of the graphene epoxy composite was covered in the first section. The impact of graphene on epoxy adhesives was the focus of the second section. The dispersion of graphene in epoxy adhesives was the subject of the third section, and the use of GNPs composites was covered in the final one. Consequently, a wide range of detailed information about graphene nanoplatelet composites was provided by this review study.



Figure 1. Structure of graphene oxide [2].

2. Preparation of Graphene-Epoxy Composites

The preparation process utilised to include graphene into epoxy adhesives plays a crucial part in establishing the physical and chemical characteristics of the composites that are produced. In-situ polymerization, electrochemical breakdown, mechanical mixing, and sonication are a few of the techniques that may be used to introduce graphene into epoxy adhesives. Using a high-shear mixer, mechanical mixing involves combining graphene with epoxy resin and hardener. In sonication, graphene is combined with epoxy resin and a hardener to create a homogenous dispersion by first being dispersed in a solvent and then sonicated to create that dispersion [3] [4]. In in-situ polymerization, graphene is first mixed with epoxy resin before the hardener is added, and then the resin is polymerized. Graphene and epoxy resin are both polymerized simultaneously during in situ polymerization. Electrochemical deposition involves the electrochemical reduction of graphene oxide to produce graphene in the epoxy resin. The kind of graphene employed the technique of synthesis, the concentration of graphene, and the curing conditions are only a few of the variables that affect the physical and chemical characteristics of the resultant composites. The characteristics of the resultant composites are significantly influenced by the amount of graphene present in the epoxy resin. Better mechanical, thermal, and electrical characteristics of the resultant composites are frequently brought about by higher graphene concentrations. The mechanical characteristics of graphene-epoxy composites are greatly influenced by the process used to prepare them. For instance, it was discovered in a study by Gao et al. (2017) that the tensile characteristics of graphene-epoxy composites made via in-situ polymerization were superior to those prepared by mechanical mixing. This was attributable to the in-situ polymerization process' improved graphene dispersion in the epoxy resin [5].

3. Effect of Graphene on Epoxy Adhesives

3.1. Mechanical Properties

It has been demonstrated that adding graphene to epoxy adhesives enhances their mechanical qualities, such as stiffness, strength, and toughness. In a research, it was discovered that adding 1 weight percent (wt%) graphene nano particle (GNP) to an epoxy adhesives increased its stiffness and toughness by 20% and 40%, respectively [6]. Similar results were obtained in a research which showed that adding 0.5 weight percent of Graphene oxide (GO) to an epoxy adhesives increased its toughness and tensile strength by 30% and 40%, respectively [2]. As comparison to the pure epoxy adhesives, the 0.5% graphene additive raised the tensile strength by 36.84% and the Young's modulus by 82.07% [7]. Covalent functionalized epoxy-graphene nanocomposites were created by Naebe *et al.*, who observed increases in flexural strength and modulus of 18% and 23%, respectively. When 1 weight percent of non-functionalized GNP was added, the greatest increases in tensile strength and elastic modulus were seen to be 13% and 39%, respectively [8]. According to a study by A. Heydari *et al.*, adding 0.5

wt% GONPs increased stiffness, strength, and toughness by 33.4%, 45%, and 25.6%, respectively [9] [10] [11].

The exceptional qualities of graphene, including its high strength and large surface area, are responsible for the advancements in mechanical characteristics. Graphene can reinforce the epoxy matrix by forming a strong interface between the graphene and epoxy molecules, graphene can strengthen the epoxy matrix and improve load transmission between the two materials. Since graphene has a high surface area, it has a lot of surface area in contact with the epoxy matrix, which enhances the interfacial adhesion between the two materials. Tensile strength increasing with increasing in the concentration of GO is shown in **Figure 2**.

Moreover, the use of graphene in the epoxy matrix can result in crack deflection and bridging, increasing the composite's ability to withstand fracture. In adhesive applications, where the composite's capacity to resist fracture propagation is vital, this impact is particularly significant [12].

3.2. Thermal Stability and Conductivity

Graphene has excellent thermal stability. The thermal resilience of epoxy adhesives can be increased by addition of graphene. Epoxy adhesives are suited for high-temperature applications because to their ability to drastically raise their glass transition temperature when graphene is added. In a research by Shen *et al.* (2021), it was discovered that 0.1 wt% of GO increased the Thermal conductivity of an epoxy adhesive by 50% [13]. The impact of non-functionalized GNP addition in various percentages from 1 to 10 wt% on heat conductivity of bonded joints was examined in the study by R. Moriche *et al.* At greater weight percentage composition, the author found a significant increase (306%) in thermal conductivity [14]. This is linked to graphene's high thermal conductivity, which can enhance the composite's ability to dissipate heat. Using Thermogravimetric analysis (TGA), it was discovered that adding 0.5% GNPs increased thermal



Figure 2. Tensile strength of graphene oxide/epoxy composite at different contents of graphene oxide [15].

stability in the 360°C - 580°C range [16]. By adding 3 wt% of GNP, epoxy resins Limiting oxygen index value went from 15.7 to 21.0 and its overall heat release went from 33.37 to 28.20 KJ/m² [17]. An analysis of the impact of graphene oxide (GO) on the thermal stability of epoxy adhesives was reported in the journal Polymers. The study showed that 3% GO addition to an epoxy glue enhanced its thermal stability and raised the decomposition temperature which is shown in **Figure 3** [8]-[14] [16] [17] [18].

3.3. Electrical Conductivity

Because of its superior electrical conductivity, graphene may considerably improve the electrical conductivity of epoxy adhesives. Because of this, electrical applications, such as those in the electronics sector, may be appropriate for the epoxy glue. The electrical conductivity of epoxy adhesives was increased by using GO. The outcomes demonstrated that the inclusion of GO greatly enhanced the electrical conductivity of the epoxy adhesive. The epoxy adhesive containing 0.5 wt% GO had an electrical conductivity of 7.84 S/m, which was 200 times more than that of pure epoxy adhesives [19]. In a study, electrical conductivity of epoxy adhesives was increased by using functionalized graphene. The results demonstrated that the addition of functionalized graphene enhanced the electrical conductivity of the epoxy adhesive, and the electrical conductivity was determined to be 5.5 S/m at a loading of 1.5 wt% [8]-[14] [16] [17] [18] [19] [20]. In order to increase the electrical conductivity of epoxy adhesives, GNPs were utilized. High electrical conductivity (525 S/cm) in graphene platelets has a notable impact on adding conductivity to nanocomposite adhesives. Also, the low percolation threshold is greatly influenced by the high aspect ratio and decent dispersion quality of graphene platelets in epoxy matrix. It was discovered that the epoxy adhesives containing 2 wt% graphene nanoplatelets had an electrical conductivity of 7×10^{-6} S/cm, which was almost 3.5 times greater than that of the pure epoxy adhesive [21].



Figure 3. Thermal conductivity of neat epoxy and its GNP based nano composites adhesives [8].

3.4. Adhesion Properties

Epoxy adhesives' adhesion capabilities may be enhanced by the inclusion of graphene. Since graphene has a huge surface area, it can enhance the contact area between the substrate and the adhesive, improving adhesion. When compared to plain epoxy adhesive, the mode I fracture toughness of nanocomposites shows a 5 times improvement with a graphene level of only 0.25 wt% [8]. The addition of graphene oxide increased the shear strength at the interface between the adhesive and substrate. This was due to the strong interactions between the oxygen-containing functional groups on the graphene oxide and the epoxy resin. The graphene oxides large surface area and high aspect ratio gave the epoxy adhesive additional surface area to bond to. The shear strength of the adhesive bond was dramatically increased using Reduced Graphene Oxide. The increased surface area and excellent dispersion of Reduced Graphene Oxide in the epoxy matrix allowed for more efficient stress transmission from the adhesive to the substrate, which enhanced adhesion. Due to the enhanced mechanical characteristics of the epoxy matrix brought on by the presence of GNPs, the inclusion of GNPs increased the lap shear strength of the adhesive junction [15] [22] The mechanical, electrical, and thermal properties of graphene-epoxy composites can be further enhanced by the addition of other nanoparticles, such as silica nanoparticles, carbon nanotubes, and metal nanoparticles. The combination of graphene with other nanoparticles can lead to synergistic effects that further enhance the properties of the resulting composites.

4. Dispersion in Epoxy Adhesive

Effective filler dispersion plays a major role in the efficient application of polymer composite. Graphene is frequently utilised as a polymer matrix filler. Its mechanical, electrical, morphological, and other properties make good dispersion desirable, contingent upon the requirements of the final use. In graphite, the weak Van der Waals force bonds the graphene layers together [23]. Since dispersion and distribution during the processing/manufacturing of the composite are necessary for the delamination of graphene layers from stacked aggregates and its orientation in the composite, the processing method is also important in this regard. Due to the exceptionally high aspect ratios and enormous surface areas of graphene, which produce strong Van der Waals forces on the surface, as well as the comparatively high viscosity of epoxy resin, it is difficult to disperse graphene uniformly and evenly throughout the matrix [24]. Many techniques have been developed for improving the dispersion of GO into epoxy resin, even though the homogeneity of graphene dispersion was still far from adequate. Three categories comprise most used techniques: chemical, physical surface treatments, and mechanical mixing [24]. Breaking apart graphene clusters using mechanical mixing, such as ultrasonic mixing and three-roll milling, was the most common technique to enhance the dispersion [25]. Graphene is hydrophobic, which makes it difficult for epoxy adhesives to disperse. However, functionalized graphene can enhance graphene's dispersion in epoxy adhesives. A study using scanning electron microscopy showed that 0.5% wt GO added to an epoxy glue resulted in a more homogeneous dispersion [26].

5. Potential Applications of Graphene-Epoxy Composites

5.1. Aerospace Applications

Aerospace applications would benefit from the superior mechanical and thermal characteristics of graphene-epoxy composites. They can be utilised to create lightweight parts for satellites, spacecraft, and aircraft. Along with helping to dissipate heat produced during space missions, graphene's excellent thermal conductivity helps lower the likelihood of equipment failure.

5.2. Electronics

Due to their excellent electrical conductivity, graphene-epoxy composites are appropriate for use in electronic applications. They can be used to create electromagnetic shields, sensors, and antennae, among other conductive components. Graphene is a desirable material for energy storage systems like batteries and supercapacitors due to its large surface area.

5.3. Biomedical Applications

Graphene-epoxy composites have potential applications in biomedical engineering due to their biocompatibility, high surface area, and mechanical strength. They can be used to make implantable devices, drug delivery systems, and tissue engineering scaffolds.

5.4. Automotive Applications

Vehicle body panels, engine components, and suspension systems can all be made from lightweight, durable graphene-epoxy composites. Moreover, the high heat conductivity of graphene can aid increase engine performance and cut emissions.

5.5. Colour Paints

Epoxy adhesives based on graphene have been demonstrated to greatly enhance the adhesion of coatings to surfaces, which is especially important in the context of paints. In order to give adequate coverage and endurance, paint must be able to stick strongly to the surface when applied. Epoxy adhesives made with graphene can strengthen the bond between the substrate and the paint to increase adhesion.

Epoxy adhesives based on graphene can improve adhesion as well as coating longevity by making paint more resistant to abrasion, UV rays, and other environmental variables that can cause paint to deteriorate over time. Excellent mechanical strength and durability are two qualities of graphene that can be used in the creation of new materials. Since graphene is known for having exceptional mechanical strength and endurance, these qualities can be used to create coatings that are more durable and long-lasting. The exceptional colour stability of graphene-based adhesives in paints is another possible advantage. The extraordinary colour stability of graphene-based materials has been demonstrated, which may be useful in paints. This might aid in preventing fading or colour changes over time, producing a finish that is more constant and long-lasting. It has been demonstrated that materials based on graphene have improved scratch resistance, which may be useful for paints used on surfaces that are prone to scratches or abrasion. Epoxy adhesives made with graphene can help the coating last longer and keep its appearance over time by supplying greater scratch resistance. The improvement of epoxy composites' mechanical, thermal, and electrical properties has shown encouraging results when using graphene-epoxy composites. They are excellent for a variety of applications in many fields thanks to their special qualities. To fully realise these composites' potential and enhance their performance for applications, more study is necessary [23]-[32].

6. Challenges and Future Perspectives in the Synthesis and Processing of Graphene-Epoxy Composites

There are still difficulties in the manufacturing and processing of graphene-epoxy composites, despite the encouraging outcomes from the addition of graphene to epoxy adhesives. The homogeneous dispersion of graphene in the epoxy matrix is one of the difficulties. Because of its high surface energy, graphene tends to aggregate, which can cause poor dispersion and diminished mechanical characteristics in the composite. To increase the dispersion of graphene, several techniques have been suggested, including functionalization with surfactants and ultrasonic processing. The scalability of the synthesis of composites made of graphene and epoxy presents another difficulty. Most current preparation techniques for graphene-epoxy composites are restricted to small-scale laboratory synthesis. For large-scale graphene-epoxy composites to be used in practical applications, scalable and affordable production techniques must be developed. Careful thought must be given to graphene's compatibility with epoxy adhesives also. The addition of graphene may have an impact on how well the epoxy resin cures, which could lead to diminished mechanical properties and incomplete curing. Additionally, the compatibility of graphene with the epoxy adhesive's hardener must be considered [27].

Epoxy adhesives have improved in terms of their mechanical properties thanks to the use of graphene. To fully utilise the capabilities of graphene-epoxy composites, however, several obstacles still need to be overcome. One of the difficulties is optimising the type and quantity of graphene used in the composite. Depending on the application and the type of epoxy adhesive being used, a different graphene concentration may be best. To compare the effects of various types of graphene on epoxy adhesives, more research is required. The type of graphene used can also influence the resulting mechanical properties [31]. The creation of multifunctional graphene-epoxy composites, which can enhance not only the mechanical properties but also other properties like electrical conductivity, thermal conductivity, and corrosion resistance, represents another challenge. Graphene-epoxy composites can be used in a variety of applications due to the combination of multiple functionalities in a single material [32].

7. Conclusion

We have reviewed the most recent research on the impact of graphene on epoxy adhesives in this paper. It has been demonstrated that adding graphene to epoxy adhesives enhances their mechanical properties, Dispersion and Adhesion properties, Electrical conductivity and thermal stability. The distinctive characteristics of graphene, including its high strength, large surface area, and crack deflection and bridging, are thought to be responsible for the improvements in mechanical properties. The uniform dispersion of graphene, scalability of production, and compatibility with the epoxy resin and hardener are still issues in the synthesis and processing of graphene-epoxy composites. Future research is required to develop multifunctional graphene-epoxy composites for various applications and to optimise the type and amount of graphene used in the composite.

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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