



**REVIEW**

# A Review of Graphene Oxide Crosslinking as Enhanced Corrosion Shield Application

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## ABSTRACT

Nowadays, corrosion is not only undesirable, but it also has a significant influence on the industrial sectors and technical innovations that have demand for metals. The global economic damage is expected to reach \$2.5 trillion, equivalent to more than 3.4% of the world's GDP in 2013. It is linked with significant financial harm, manufacturing pollution, and safety issues. An electrochemical process primarily induces metal corrosion at the metal-electrolyte interface region, caused by steel oxidation and the reduction of oxygen, protons, and water. Therefore, organic and epoxy coatings can be applied as protective coatings. Additionally, it can prevent metal corrosion in various fields due to its unique properties, like the ability to manage its curing process over a long period. This review paper focuses on improving the physicochemical properties of modified graphene oxide (GO) coating by attaching a polymer, epoxy resin (EP). Moreover, we reviewed recent achievements for different methods in minimizing a corrosion phenomenon. Further, we considered the modification of epoxy resin using the curing agent as a bridge to the polymer to strengthen the functionalization of nanocomposites during the reaction. Afterward, we discussed the relationship between the modified GO/EP anticorrosive coating and physicochemical properties since it is currently being questioned and remains unresolved. As a result, extensive studies on the mechanism of synthesis of the modified GO/EP anticorrosive coating were highlighted.

## KEYWORDS

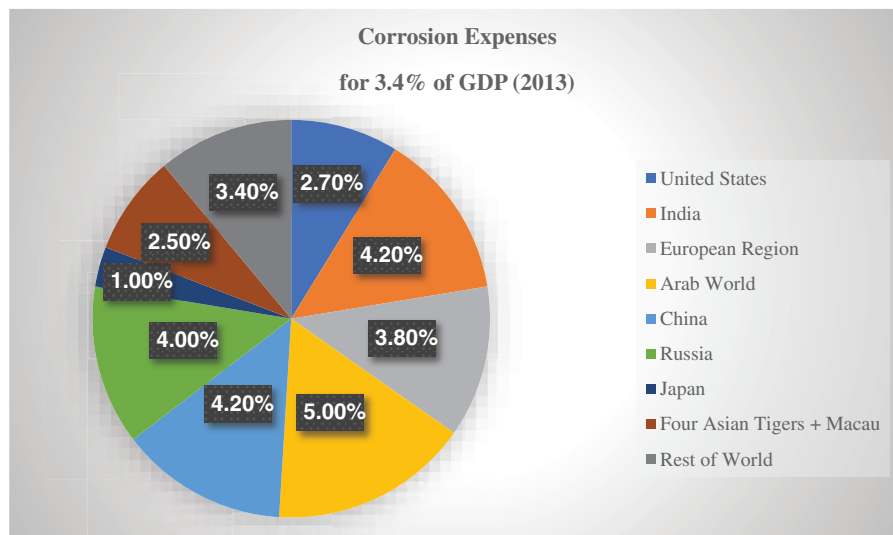
Corrosion; anticorrosive; graphene; polymer; epoxy resins

## 1 Introduction

Corrosion is not only unfavorable, but it also has a huge impact on the economy of industrial zones, resulting in substantial losses of profits [1–4]. According to reports, due to corrosion, one-third of total metal production declined for industrial purposes [5]. Corrosion can be classified into numerous types, including general corrosion, localized corrosion, pit corrosion, galvanic corrosion, and corrosion chemicals. According to a prior publication citing a study from the National Association of Corrosion Engineers (NACE) International, corrosion damage led to an estimated 2.5 trillion dollars in financial damage globally, accounting for almost 3.4% of the world's GDP during 2013 [6]. Both safety and effects on the environment are often excluded from such expenses. Generally, the high cost of corrosion is seen in several fields, such as agriculture, industry, and services. Some sectors have discovered that a shortage of corrosion control may be costly due to close encounters, accidents, compulsory closures, and



disasters. thus, efficient corrosion control can result in considerable financial savings throughout the lifespan of the property [7]. As shown in Fig. 1, corrosion is a significant cost strain and among the country's highest costs. Meanwhile, Table 1 lists the approach's benefits and drawbacks in minimizing the corrosive phenomena. Such phenomena should be dealt with by researchers and technologists worldwide as they are connected to significant damage to the economy, production pollution, and hazard risks. It would be worth mentioning that coatings are commonly employed to shield steel from corrosion, as are other accepted procedures for creating quality protective coatings. It can be noted that metal corrosion is a widespread concern, as it is one of the most severe difficulties that people have encountered, especially ever since metals have been used in everyday life [8].

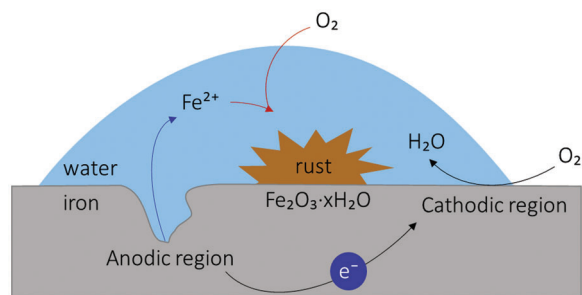


**Figure 1:** Corrosion expenses for 3.4% of GDP during 2013

Metal corrosion is typically induced through an electrochemical or chemical process at a metal-electrolyte interface region and leads to steel oxidation and the reduction of oxygen, protons, and water [9–11]. The alignment of cathodic and anodic reactions and the substance interaction occur at specific points. At the cathode, oxygen is reduced chiefly along with an active catalytic layer of oxidized metal. At the same time, many chemical reactions happen at the anode, culminating in the formation of ferrous ions when iron (Fe) atoms disperse through the wet layer and leave a negative charge in the metal, as in Fig. 2 [12]. Hence, as a depolarizer takes electrons from the metal, corrosion occurs. For example, aluminum and its alloys corrode when they begin to interact with corrosion attacks involving chlorine, water, oxygen, and many more. As a result, the essential way to avoid metal corrosion is to minimize direct interaction between metal surfaces and corrosive environments [13].

Corrosion has become a leading detrimental problem and a long-term process throughout many industry sectors and technology advances involving the need for metals [14,15]. Presently, deteriorated coatings are generally replaced or cured, which is pricey and time-consuming, which has evolved into products with self-healing characteristics [16–18]. Various solutions have been presented in recent days to address this issue and increase the anticorrosion properties of coatings [19]. Several difficulties have pushed experts toward perfecting numerous tactics for controlling corrosion mechanisms and inventing an appropriate way to overcome these issues. Metal barrier technologies have been classified as (i) corrosion-resistant materials, (ii) corrosion inhibitors, (iii) cathodic protection, (iv) anodic protection, (v) conversion coatings and organic coating protection, (vi) protective polymeric films, and (vii) paint coatings as shown in Table 1

[20]. Therefore, to sum up, an organic coating is employed as one of the most anticorrosive barriers, which can be a protective layer that delays the corrosion of metals throughout a range of areas owing to its fascinating features, including the capability to regulate the corrosion process over a lengthy amount of time [21,22].



**Figure 2:** Graphic figure of the corrosion mechanism

Recently, there has been a surge of interest in using graphene as an active layer, possibly as in addition to becoming a good corrosion protection coating additive. Compared to untreated graphene anticorrosive coatings, anticorrosive graphene-composite coatings are not just graphene's outstanding chemical durability and efficient electrical properties but also its good results owing to its characteristics. Graphene's overall corrosion protection effectiveness by self-healing, superior mechanical, impermeability, and self-cleaning capabilities [23–26]. Therefore, protective coatings based on graphene-composite can be a new entity as fillers in polymer composites in corrosion protection coating materials for various applications [27,28]. Hence, graphene substitutes like graphene oxide (GO), reduced graphene oxide (rGO), and other materials that are employed and other corrosion protection additives in self-healing coating materials to enhance corrosion resistance qualities, including adherence, durability, and long drying rate [29–31]. The synthesis of GO-polymer-functionalized nanocomposites is a promising technique for improving the coatings' corrosion protection and physicochemical performances [32,33]. As a result, GO might well be employed for various purposes and shows potential as an indispensable product in developing new materials [34]. Furthermore, GO can also act as a barrier, which is critical for long-term protection [18]. The low conductivity of GO, in comparison to graphene, reduces charge transport within the coating, enhancing protection against corrosion [35].

**Table 1:** The benefits and weaknesses of various approaches employed in minimizing the corrosion phenomenon

Methods	Benefits	Weaknesses	Ref.
Corrosion resistance materials	Exhibits superior corrosion resistance during prolonged immersion.	The corrosive medium gradually penetrates the coating	[36]
Corrosion inhibitors	Reduced the corrosion current density and acted as a mixed type of inhibitor.	The hydrogen evolution mechanism remains the same	[37]
Cathodic protection	The corrosion-promoting effect is inhibited.	The risk of hydrogen embrittlement failure	[38]
Anodic protection	Better corrosion resistance is obtained.	Formation of a porous corrosion layer	[39]

(Continued)

Table 1 (continued)			
Methods	Benefits	Weaknesses	Ref.
Conversion coatings	Advancement with a more efficient and sustainable substitute.	The sensitivity to localized damage and the efficiency of film deposition was reduced	[40,41]
Organic coating protection	Promising corrosion protection of condensing economizer elements.	A slight decolorization	[42]
Protective polymeric films	Increased effectiveness of the coating due to the synergistic effect and good adhesion to the metal.	Poor wettability	[43]
Paint coatings	It can simulate and accelerate corrosion damage of the substrate.	The coating could be damaged if the electric charge reached 0.1 C	[44]

Recent studies have indicated that organic coating protection has emerged as the leading candidate method. It has been attracting increasing interest because of the efficacy of its composites and the ease of its synthesis process for keeping metals from corrosion [45]. Coating an organic coating onto a metal surface is a cost-effective technique of shielding the steel surface while retaining the ideal mechanical characteristics of the base material.

The corrosion resistance materials method was reported to have a better lifespan and low environmental toxicity but involves a high-cost operation. A solution was discovered when nickel aluminide,  $Ni_3Al$ , was micro-alloyed with boron [46]. The use of  $Ni_3Al$ -based alloys for high-temperature applications is predicated on their generally excellent mechanical, thermal, and physical features.

Next, the use of corrosion inhibitors is a low-cost technique and an expert one. Unfortunately, it may lead to the pollution of the environment. Graphene can create a protective barrier between the metal and a corrosive environment. According to cyclic voltammetry tests, it significantly inhibits metal oxidation and oxygen reductions [47]. Tafel's research showed that metal with a multi-layered graphene sheet corroded 20 times slower than a raw nickel.

The cathodic protection method is easy to implement and has a low cathodic interruption, but it has inadequate performance. Dong et al. used localized electrochemical impedance spectroscopy (LEIS) to investigate localized steel corrosion at a defect [48]. Metal corrosion was shown to be dependent on cathodic protection potential and flaw shape. The rate-limiting stage in steel corrosion is the bulk of oxygen going through the flaw with a narrow, deep shape.

Through anodic protection, the corrosion behavior can be improved, but the surroundings will be easier to become corrosive due to the dissolution of oxide. Anodized coatings can efficiently reduce the corrosion of magnesium (Mg) materials by covering the substrate contact area and slowing the infiltration of the corrosive solution [49]. Coating thickness affects the number of through-pores, and as a result, anodized Mg alloys have high corrosion resistance.

Conversion coatings can improve efficiency and be a sustainable substitute. Nevertheless, the sensitivity of localization can easily be affected, and the deposition efficiency of the film will be reduced. Following a suitable process, the extracts were analyzed through the phosphate permanganate procedure, and the cathodic epoxy was electro coated [50]. The foremost serious aspect of generating high-quality conversion coating materials was pH stabilization.

Protective polymeric films were reported to be effective due to their practical synergistic effect and good adhesion to metal. However, they were also shown to have poor wettability. According to electrochemical impedance spectroscopy (EIS), for the anticorrosion coating of E32 marine steel developed under curing conditions, the polyurethane acrylic paint had positive results when using the epoxy base coat coating [51]. The paint coating method has excellent coating protection and anticorrosive properties. The resulting adherence is weak due to the increment of color concentration. Truong et al. [52] reported that acid acrylic paint incorporating electrically conductive polypyrrole (PPy) showed significant cathodic open circuit potential scans and EIS for the PPy-coated sample. The large cathodic current densities were attributed to the cathodic reactions with low voltage spikes at the PPy particles.

## 2 Graphene's Properties and Characterization

Graphene is a recently discovered source of carbon allotrope that has stimulated research interest and broadened the research field on composite material uses due to its exceptionally developed surface area and mechanical, electro-optical, chemical inertness, and thermal properties [53–57]. Owing to its unique features, including solid thermal properties (3000–5000 W/mK), large surface area (2630 m<sup>2</sup>/g), high charge carrier mobility (200000 cm<sup>2</sup>/V), and inherent toughness (130 GPa) as a flat layer [58–60], graphene has piqued the curiosity of many researchers. These magnetic properties inspire substantial efforts to utilize graphene in various applications involving hydrogen generation, detectors, batteries, ultracapacitors, photovoltaic cells, and bioproducts. As a result, several studies and approaches have been explored to achieve a reliable graphene process.

Geim and Novoselov extracted graphene, a minor substance in history with scientific value, a single sheet of carbon atoms, exfoliated from graphite in 2004 by applying the “Scotch tape” technique [61]. Graphene can be defined as a one-atom-thick, uniform single-layer structure made of carbon atoms with a zero-gap two-dimensional (2D) atomic lattice with strong sp<sup>2</sup> carbon-carbon interaction in a hexagonal structure shape and large aspect ratio [28,62,63].

Graphene is distinguished by various aspects that make it a suitable protective layer. Surfaces covered with graphene, for instance, might keep their visual aspect owing to the excellent transmitted light of graphene (97%) in a broad range of wavelengths [64]. In addition, because graphene is adaptable, graphene coatings may adapt to such stiffness and distortion of the surface layer. The aromatic C=C bond structure in graphene spreads throughout the whole basal plane due to the delocalization of the outer electrons, giving graphene thermal stability. This chemical inertness is required for its application as a protective layer. Once exposed to intense pressure and overheated water, graphene is extremely durable than diamond [65].

To understand more about graphite's composition, British scientist B. C. Brodie researched the reactivity of flaky graphite in 1859. He concluded that the molecular weight of graphite should be 33 [66]. Graphite was recognized as a natural resource after about 500 years. The use of graphite in the physical science industries began about 150 years after that. Graphene is an incredibly thin layer of carbon atoms with a surface thickness of 0.5–1 nanometer (nm), owing to the chemical imbalance between graphene and its substrate. Raman spectroscopy is among numerous methods that can characterize graphene and is now commonly used. In contact or tapping mode, surface topology, flaws, and bending characteristics can all be investigated using atomic force microscopy (AFM).

Graphene is hypothesized to be composed of pure aromatic ‘islands’ of different diameters that are not oxidized. They may be separated by aliphatic areas, including hydroxyl and epoxide groups, and double bonds [67–70]. Besides, nuclear magnetic resonance (NMR) spectroscopy has been employed for centuries to link spatial features of diverse non-crystalline materials [71]. X-ray diffraction (XRD) is also a method for characterizing several types of graphene. Raman spectroscopy is used to measure the conversion of sp<sup>3</sup> polarized carbons, which returns to sp<sup>2</sup> during GO reduction.

Transmission electron microscopy (TEM) can precisely determine the number of layers of graphene using. Graphene layers are not perfectly flat and experience static disturbances from the plane with a size of 1 nm. The hanging graphene layers represent this in the TEM, which may provide a picture of the graphite structure and the well-defined electron diffraction patterns. In addition, contrary to a few of the preconceived notions about graphene layers' performance, there is no inclination for them to be spiral or bent., Naebe et al. [72] said that the tensile properties of composite coatings with even a small amount (0.1–1.0 wt.%) of graphene loadings in the epoxy coating could improve the tensile strength, failure strain, and Young's modulus.

Naebe et al. [72] discovered that the electrically charged graphene/polymer nanocomposite's resistance constantly decreased as the graphene nanosheet concentration increased. Once the graphene nanosheets reached a specific number, the resistivity dropped precipitously. The resistance was steadily lowered as the loading of graphene nanosheets increased. It can be highlighted here that the number of graphene nanosheets may be less than 0.1 vol percent, implying that graphene/polymer nanocomposite with no corrosion-promotion activity must have a small number of graphene nanosheets. As a result, identifying the peak loading of graphene nanosheets is critical in developing graphene nanosheet-based corrosion protection coatings.

Various techniques can be employed for synthesizing graphenes, such as micromechanical exfoliation, chemical vapor deposition (CVD), graphitization, solvothermal, liquid-phase exfoliation, thermal and electrochemical exfoliation, and chemical reduction processes. Most studies have relied on the fabrication of graphene layers using various techniques. Graphene was initially created using micromechanical exfoliation, which included peeling away a coating of graphite powder with sticky tape while tailoring it to a millimeter size [73]. Nonetheless, the approach possesses yield limitations, rendering it unsuitable for sizable deployment.

Chemical vapor deposition (CVD) is a well-known way of depositing or growing nanostructured graphene, either crystalline or amorphous, from solid, liquid, or gaseous substrates. One method produces large single-crystal graphene and seems to have poor output; this output can be used in electrical devices [74]. Another technique uses porous flakes or layers to create amorphous graphene; despite its small volume, it has high productivity. Graphene has a large specific surface area (SSA) and excellent stability, so it is suitable as electrode material in supercapacitors.

Polymer graphitization is an affordable method for producing nanostructured graphene in various materials with configurable shapes, diameters, and properties. The precursor fabrication yields crystallite sizes of several ranges in nanometers, which are substantially smaller than conventionally generated polycrystalline graphene [75]. This method allows for the flexibility of graphene growing on multiple surfaces with fixed thicknesses and sequentially obtaining various forms. Solvothermal production is a typical humid process that uses liquid or an organic phase as the precursor solution in a closed chamber at temperatures higher than the solvents' boiling points [76]. The approach is utilized to form extremely thin 2D nanomaterials, particularly for inorganic materials.

Liquid-phase exfoliation is the most flexible procedure for obtaining colloidally stable dispersions from diverse, layered materials. The energy content of the ultrasonic treatment or shearing is utilized to resist the van der Waals attraction within the massive, layered material. There are still challenges in handling graphene, particularly the graphene's low water solubility and dispersion stability in certain conventional solvents [77]. A thermal exfoliation is an efficient tool for exfoliating graphite flakes. In a broad sense, it aids the incorporation through covalent or noncovalent functionalization, expansion, and swelling, organic molecule adsorption in gaseous form, solid nanoparticle implantation, or direct molecular peeling [78]. Thermal investigations have shown that some functional groups may be eliminated at temperatures lower than 300°C.



The electrochemical approach can scale graphene synthesis, but numerous downsides must be addressed. The process employs a liquid mixture (electrolyte) and an electric current to form a graphite electrode. The graphite-based electrode undergoes anodic oxidation or cathodic reaction during this procedure. Cathodic reactivity techniques are more suited for producing a few high-quality layers of conductive graphene [79].

A reducing agent and, in some instances, an additive is used in the chemical reduction of GO [80]. A substantial reduction in the capacity is required for the elimination of oxygen-containing compounds from GO. However, the procedure must leave zero residues that can harm the output. The creation of graphene-like substances from reducing GO can be expanded. The imperfection of GO and the inevitable formation of defects throughout the oxidation process may be obstacles.

### **2.1 Graphene Chemical Crosslinking**

The most significant graphene derivative is graphene oxide (GO). Because of the presence of multiple polar groups, GO exhibits a high-water dissolution rate. According to Lerf and Klinowisk, the basal planes are populated by hydroxyl oxide and epoxy groups, while carboxyl groups are found on the edges of GO sheets [81]. These characteristics improve the compatibility of GO, where it can be easily dispersed in solvents or polymers.

Chemical functionalization, or crosslinking, is among the most common approaches for manipulating the physicochemical characteristics of different properties and studying the processes involved in engaging other properties with the surroundings [82]. There are two major forms of functionalization: covalent, through covalent bond creation, and noncovalent, through van der Waals forces alone. Such interactions are intended to reduce the polymer matrix's dispersion features and improve the polymer's solubility and GO nanostructures [45,83,84].

#### **2.1.1 Covalent Bond**

The covalent functionalization of graphene is chosen for its lasting efficiency and constancy. As a result, this may be the most advantageous method for producing graphene-based composite materials. The polymer matrix's characteristics are often enhanced to satisfy the demands. Furthermore, graphene-polymer composite structures are adaptable and beneficial for additional functionalization, including integration with nanomaterials [85]. It offers many uses for composite materials, such as corrosion protection coatings.

Hence, covalent functionalization significantly alters the geometrical and electrical features of graphene. Meanwhile, several density functional codes applied today need to consider the inclusion of the consequences of the van der Waals force, which contribute significantly to noncovalent functionalization [86]. The density-functional theory (DFT) is today's modern gold standard, and the typical local (LDA) and semi-local density approximations (GGA) have an extensive variety of applications [87]. Chemical functionalization in graphene can be associated with ionic covalent bonding.

Seifert et al. [88] discovered that graphene's level of electrolysis of water had a direct influence on anchoring density. Styrene compounds can be bonded to copper-supported graphene mechanically. As a result, Poly(styrene-co-acrylate) copolymer brushes are created by copolymerizing styrene and acrylates and consecutive crosslinking between component groups. It is possible to achieve a perfect balance between the density of surface groups and the density of defective areas on graphene. It can also be used to construct graphene-based block copolymers for corrosion protection coatings quickly.

Meanwhile, Yu et al. used a monolayer of (3-aminopropyl) triethoxysilane (APTES) to modify GO and then coated it with TiO<sub>2</sub> to create TiO<sub>2</sub>-GO anticorrosion epoxy coatings [89]. Covalently reacting the GO with the silanol moieties of the 3-glycidoxypropyl-trimethoxysilane (GPTMS) monomer, Xue et al. formed silane functionalized GO connected by C-O-Si structures [90]. Xu et al. [91] reported that they crosslinked functionalized GO to pentaerythritol (PER) in water and used it in a flame-retardant polypropylene (PP)

solution. The GO and CNTs were bonded with 3-amino phenoxy phthalonitrile, and the resultant composite substance was employed to enhance the efficacy of the anticorrosive coating [92]. Water-stable dispersions of imidazole ionic liquids were effectively covalently bonded to the surface of GO nanosheets for use in polymer composite anticorrosive coatings [93].

### 2.1.2 Non-Covalent Bond

The noncovalent functionalization of graphene is accomplished primarily through  $\pi$ - $\pi$  stacking, hydrophobic links, van der Waals forces, and electrostatic connections [94,95]. This approach is intriguing as it does not disrupt the attached  $sp^2$  bonds while retaining its excellent thermal and electrical conductivity. The  $\pi$ - $\pi$  reactions have two parameters, one being the presence of a structure and the next being the presence of a correlation between the two interconnected elements, which is generally encouraged by the polarizability of the two components.

Xu et al. used a water-soluble pyrene compound, 1-pyrenebutyrate (PB), as a preservative to create stable aqueous suspensions of nanocomposite [96]. The pyrene moiety is shown to have a significant affinity for the graphite lattice by stacking [97]. GO was successfully fabricated using pyrene butyric acid without a base, and the resultant product was decreased with hydrazine. Wang et al. noncovalently altered a graphene sheet using pyrene butanoic acid succinimidyl ester (PBSA) to enhance the power efficiency of graphene [98]. The graphene reactions and PBSA do not influence the visible area of the optical absorption of the graphene layer. Compared to pristine graphene (0.21%), this enhanced power conversion efficiency to 1.71% for functionalized graphene.

Graphene oxide, moderately reduced graphene oxide (rGO), and functionalized graphene is used as reinforcing filler to the waterborne polyurethane (PU) substrate to make corrosion protection coatings with stable loadings as significant as 20% [99]. Tamilarasan et al. [100] created electroplating Ni-P-rGO coatings on low-carbon metal by adding various amounts of rGO into the electroplating Ni-P bath. Raghupathy et al. electrochemically formed Cu-GO composite coatings on mild steel substrates by scattering GO in the solution to achieve a homogenous and tight structure [101].

## 3 Graphene-Based Polymers as an Anticorrosive Coating

Based polymer coatings are now among the highly efficient and emerging trends used to inhibit the development of corrosion products owing to their excellent corrosion protection, significant cost efficiency, and limited usage restrictions [102–105]. Strengthening polymers has increasingly become the focus of study and scalable methods against corrosive media in polymer materials research [106]. Introducing flexible or elastic polymers such as elastomers and rubber has commonly been regarded as an effective method of toughening polymeric materials. Graphene and its derivatives efficiency in reinforcing polymers vary depending on the polymer chain for the polymer composites' improvement [107–111]. The gains in hardness of various polymeric matrices assessed using that method vary. For instance, the strengthening efficacy of GO on epoxy, nylon, polyurethane, and polyamide, for example, differs greatly even when the GO concentration is the same [112]. As a result, incorporating functional GO into the barrier property of polymer coatings is a potential approach improving their corrosion resistance [31,113–119].

### 3.1 Epoxy Resin as a Polymer

Epoxy resin (EP) has become a very valuable polymer matrix that is utilized in a wide range of applications in industries for building components, cell replacements, anticorrosion coatings, adhesion, semiconductors implementation, and insulation in production [58,89,120,121]. Owing to its good chemical resistance, outstanding barrier qualities, minimal cure shrinking, and superior insulation, epoxy resin has been developed as a long-lasting anticorrosive organic coating material for several uses [20,122,123]. However, numerous solutions, such as nanofillers, have enhanced surface coatings' barrier



and corrosive resistance of surface coatings [124]. Naebe et al. [72] revealed that introducing only 0.1 wt.% of functionalized graphene platelets into an epoxy resin, increased the accelerated condition and storage elasticity to 22% and 18%, respectively. Even though epoxy resin offers high chemical resistance, excellent adherence to metallic substrates, great electricity generation, and other properties, it can be fragile as it has brittle nature and poor heat resistance [121,125–127].

Due to the rich hydrophilic chemical groups, the metal beneath it suffers a reduction in protection when epoxy attracts water from the environment to which it is exposed [128]. Several attempts have been made to enhance the epoxies' protectiveness using different nanofillers to address this issue [125]. As a result, the epoxy resin becomes a common solvent-borne dispersion medium that includes a few volatile organic compounds (VOCs) because it contains a specific amount of volatile organic chemicals. An epoxy matrix is considered undesirable for the environment and health [129].

Unbleached epoxies are distinguished primarily by their epoxy concentration, color density, hydrolyzable chlorine, and volatility substance. Epoxide equivalent weight or weight per epoxy (WPE) is commonly used to express the epoxy content of aqueous polymers. WPE is characterized by the resin accumulating 1 g of epoxide equivalent weight. The standard molecular weight of commercial-grade liquid epoxy resins is around 370. At 25°C, the viscosity is 11–15 Pa·s, and the weight per epoxide (wpe) is approximately 188. FT-IR and NMR spectroscopy approaches can identify hydroxyl reactivity. Thermogravimetric analysis (TGA) and differential thermal analysis (DTA) are employed for measuring heat transfer characteristics. DSC and DMTA analyses can investigate curing behavior and glass transition.

### 3.1.1 Curing Agent for Epoxy Resin

Curing agents and hardeners are relatively similar in that they are required to promote or regulate the hardening of the polymers. A hardener added to the epoxy polymer allows it to be bonded. Major transitions occur inside the epoxy resin throughout the hardening process. The composition of curing chemicals determines the speed and temperature of the epoxies' cure process. Due to their chemical composition, strengthening agents are classified as amino groups, alkaline, anhydrides, or catalyst-curing agents.

Some curing agents enhance curing while others directly participate in the reaction while remaining bonded covalently to the resin, such as in Table 2. The coated material can be a homo epoxy polymer, a heteropolymer composed of epoxy resin and curing agent, or a combination of the two. Promising epoxy curing agents with appropriate adaptive group functions and extra reinforcing functions in epoxy composites have piqued the curiosity of many researchers. Thus, a curing procedure with relevant functionalities can result in higher bond strength. The adhesive strength of epoxy resins is determined based on the curing environment and temperature, where low temperatures lead to weak strength, while high temperatures increase the strength.

Along with its extended lattice structure, epoxy is sensitive to various curing agents, including amines, anhydrides, carboxylic acids, alcohols, and thiols [130]. Since sensitivity seems unlikely to be an issue, the choice of hardener has mainly been influenced by the predicted form and characteristics. Co-reactive curing agents generally produce polymer chains, whereas catalyzed curing agents produce chain-growth polymers.

**Table 2:** Curing agents and their enhancements

Types of EP	Curing agents	Types of curing agents	Remarks	Ref.
Epoxy resin	Dodecyl succinic anhydride (DDSA)	Co-reactive	The presence of a lengthy hydrocarbon chain increased tensile properties.	[131]

(Continued)

Table 2 (continued)				
Types of EP	Curing agents	Types of curing agents	Remarks	Ref.
Glycidyl amine epoxy (GDAE)	Diaminodiphenyl sulfone	Amine	Resin dispersed in uniform dispersion.	[132]
Cobalt acrylate (CoA <sub>2</sub> ) modified epoxy resins	Ylide-Pyridiniumdieyanomethylide (PDMY), P-acetylbenzilidenetriphenylarsoniumylide (p-ABTAY)	Catalytic	Greater molecular weight, hydrolyzable chlorine content, and improved electrical conductivity.	[133]
Methyl hydrophthalic anhydrite (MTHPA)	Cycloaliphatic epoxy resins	Catalytic	Hydroxyl groups catalyzed the epoxy-anhydrite reaction.	[134]
Bisphenol-A based benzoxazine (Ba) and phenol-novolac-based benzoxazine (Na)	Latent curing agent	-	Strong heat resistance as well as mechanical qualities.	[135]

### 3.1.2 Epoxy Resin-Based Composite Materials

Strongly crosslinked epoxy resins are often rigid and robust while having low resistance to crack formation, which limits their utilization across numerous applications. Numerous methods have been utilized for enhancing physical qualities, including toughening or reinforcing chemicals, as shown in Table 3.

As for the elastomer-modified epoxy, it typically results in many forfeitures in modulus and strength to attain high toughness. Elastomers in epoxy have long been considered a viable alternative to rubber toughening to resolve epoxy's brittleness. Elastomer components, including poly (ether sulfone) (PES), poly (phenylene oxide) (PPO), poly (ether imide) (PEI), poly(acrylonitrile-co-butadiene-co-styrene) (ABS), polysulfones (PSF), poly (ether ketone) (PEK), and polyimides (PI), are added into the epoxy to form elastomer modified epoxy [136,137].

Hence, the functionalization of epoxy/inorganic nanocomposites can increase the impact strength, where certain inorganic particles are also used for epoxy resins. Using of inorganic materials offers resistance to deformation and fracture initiation via hardening processes. Inorganic nanoparticles added to epoxy resins can enhance elasticity, stiffness, and impact resistance. Carbon fibers with excellent mechanical properties can be absorbed into a range of polymer matrices. If a mass is added to a carbon fiber composite, the stress is mainly transmitted via the polymer composite from one carbon fiber toward another carbon fiber. If somehow, the binding between the epoxy and the fibers is poor, this mass transmission will diminish or break down the interface. Sized, a polymer mixture that enhances carbon composite adherence to the polymer resin, is typically used for fibers [138–140].

The synthesis of polymer/clay composite materials may lead to clay aggregation, leading to high structural and hardness degradation. Different composites are formed when surface-treated ceramics are incorporated into substrate polymer matrices, intercalated or disintegrated composite structures [141,142]. Compared to pure epoxy matrices and conventional, tiny hybrids, epoxy/clay nanoparticles with minimal clay content demonstrate significantly better tensile properties, thermal properties, susceptibility, and heat

resistance. The improved method for epoxy/clay composites results from the nanocomposites' robust interface/volume relation, which is related to the nanoscale-level clay particle dimensions [143,144].

Numerous researchers focus on carbon nanotubes (CNTs) due to their sizeable interfacial region and intrinsic properties for formulating a more efficient polymeric matrix with excellent mechanical properties, better stiffening efficiency, and greater thermal conductivity [145]. There are mainly three types of CNTs, which are single-walled (SW), double-walled (DW), and multiple-walled (MW) CNTs.

**Table 3:** Enhancement of its properties with various epoxy-based composites

Epoxy-based composites	Systems	% increment/decrement in properties				Remarks	Ref.
		Glass transition temperature, $T_g$ (°C)	Modulus, E(GPa)	Tensile strength, $\sigma$ (MPa)	Fracture toughness, $K_{Ic}$ (MPa·m <sup>1/2</sup> )		
Elastomer modified epoxy	Epoxy/13 wt.% PES	156	1.6	-	2.1	Had better properties, which implied a shorter processing cycle.	[146]
Epoxy/inorganic composites	0.133 wt.% Silica/polyether-amine cured DGEBA/F	68	1.67	82	1.76 ± 0.16	Ready formation and stabilization.	[147]
Epoxy/carbon fiber composites	1 wt.% carbon nanofibers (CNFs)/Epoxy	-	1.22	74.4	-	High tensile strength.	[148]
Epoxy/clay nanocomposites	Epoxy/1.0 wt.% clay (Cloisite 30B)	-	3.4	73.3	0.82	Polishing.	[149]
Epoxy/carbon nanotube composites	Epoxy/0.1 wt.% DWCNT (Nanocyl) + NH <sub>2</sub>	80	3.28	63.8	0.65	Cured and post-cured.	[150]

### 3.1.3 Application Fields of Epoxy

#### Paints and Coatings

Due to their various advantages, including ease of handling, good safety, excellent solvent, and chemical stability, high hardness, good dimensional stability on healing, good structural and strength properties, and excellent adhesion to several substrates, epoxy materials are commonly used as corrosion protection coatings

for heavily loaded items. Steel cartons and bottles are routinely coated using epoxies to prevent corrosion, especially while preparing low-pH foods such as tomatoes. Epoxy resins are used in textured and somewhat raised flooring applications.

#### *Adhesive*

The adhesive is a vital characteristic of the gluing form known as “adhesive tapes”. These improved adhesives are exploited in manufacturing airplanes, vehicles, bicycles, boats, golf clubs, skis, skateboards, and other interfacial uses. For it to be utilized as a sealant in cryogenic engineering fields, the epoxy tensile toughness must be optimized in both cryogenic and ambient environments. The toughness of manufactured epoxy additives is engineered by inserting sequence thermoplastics, rubber particles, or stiff inorganic materials through the substrate. Silicone additives have often been treated at high temperatures to improve their strength and initiate complexation at the surface contact.

#### *Industrial Tooling*

Throughout advanced manufacturing equipment operations, epoxy systems create molds, master approaches, fiberboard, castings, fittings, and several essential manufacturing tools. Such “plastic tooling” can become substitutes for metal, wood, and some conventional items and enhance operational efficiencies by decreasing expenses or cutting timeframes for numerous production lines. Fiber-reinforced epoxy polymers are helpful in the restoration of metal materials and cylindrical pipelines.

#### *Aerospace Industry*

Owing to their outstanding adhesion characteristics and minimal cost, epoxy resins are already widely employed for systemic adhesive purposes in the aircraft sector. The aircraft industry is most interested in epoxy resins that are strengthened with extra-strong silica, carbon, Kevlar, or tungsten fibers.

#### *Electronic Material*

The semiconductor industry uses epoxy resin compositions in engines, turbines, transformers, switchgear, bearings, and capacitors. Epoxy resins are good electrical conductors that shield circuit boards from short circuits, dust, and moisture. Electromagnetic interference protection is a common need for steel composites. Epoxy molding compounds (EMCs) are widely utilized as encapsulating materials for electronic components to protect them from humidity, portable pollutants, and adverse climatic factors such as temperature, ultraviolet light, humidity, and physicochemical harm. In electronic packaging applications, epoxy composites incorporating particle additives such as quartz glass, marble powder, and mineral silica are also employed as matrix materials.

#### *Biomedical Systems*

In the clinical field, epoxy resins are commonly employed. For human therapeutic uses, collagen-based substances are utilized as bandages for wounds, bone grafts, and aortic heart valves. Elastic polymer foams, such as embolic sponges, have great potential as biomaterials. Due to their extreme toughness, remarkable chemical inertness, poor electrical and solid thermal resistance, broad spectral opacity, and several specific characteristics, nanodiamond epoxy substitutes have been significantly used in biomedical fields.

## **4 Graphene Oxide/Epoxy Nanocomposites**

### **4.1 Processing Methods**

One of the thorniest issues in the fabrication of GO/epoxy composite materials is achieving a proper dispersion of the nanostructure. A well-dispersed phase implies that the largest surface area of the filler is accessible, which also influences the surrounding crosslinks and, as a result, the characteristics of the nanocomposite. The fabrication methods used for epoxy or any matrix considerably impact dispersion. Extensive studies have been undertaken regarding fabrication approaches for generating a homogenous

and evenly distributed system. Solvent blending and epoxy bonding are the most commonly utilized GO/epoxy composite material approaches.

One of the primary and typically preferred approaches for synthesizing GO/epoxy nanocomposites involves capitalizing on the availability of hydroxyl groups linked to the graphene sheets, allowing for the spontaneous dispersion of graphene in various chemical solvents. This approach also correlates the nanostructured graphene's extensive organic or inorganic reactivity with different polymers. Usually, synthesized graphene is dissolved using a specific solvent, such as via ultrasonic treatment, after which it is coated into the epoxy matrix, where the solution is evaporated under specified parameters. A guiding idea will be to use an organic solvent for the surface hydroxyl groups of graphene and a chemical bonding that is ideal for the polymer matrix. Numerous solvents are being explored to improve the dispersion of nanocomposites, as in [Table 4](#).

**Table 4:** Differences in graphene oxide dispersion and characteristics enhancement

Solvent	Filler	Dispersion method	Char residue %	Increase in $T_g$ (°C)	Remarks	Ref.
Acetone	0.5 wt.% as-prepared (aGO)	Ice-bath sonication + mechanical mix	-	128.7	wide band assigned to O-H, C = O, and -COH bending.	[151]
Dimethyl formamide (DMF)	3 wt.% silica-dendrimer attach GO (GSD)	Sonication + mechanical mix	60.9	322.9	The appearance of the bands assigned to Si-O and C = O groups.	[152]
Dimethyl formamide (DMF)	Low density 1-((4-aminobutyl) amino)-2-methyl-1-oxopropan-2-yl-dodecyl-carbonotrithioate (OR)-functionalized GO	Sonication + three-necked jacketed reactor + mechanical mix	44.7	234.8	The band's occurrence in the C-H, C = S, and N-H groups.	[153]
Ethanol	Polyoxyethylene octyl phenyl ether (POPE)-functionalized GO	Ice-bath sonication + ball milling	57	-	Increase of C-OH, C-O-C, and C = O groups.	[154]
Water	2.0 wt.% rGO	Ice-bath sonication + mechanical mix	-	81.6	Increase in zeta potential and an upshift of the $\pi$ - $\pi^*$ peak.	[155]

#### 4.2 Water-Based Epoxy (WEP) Resin Coating

Water-based epoxy (WEP) coating has recently piqued scientists' interest as it satisfies pollution control requirements [156]. Furthermore, it offers several benefits over solvent-borne epoxy coating, including good adaptability and firm adherence to a wide range of steel substrates [156]. Researchers have discovered that nanofillers can be employed to increase corrosion resistance and fix flaws. As a result, fillers have demonstrated the highest permeation since they have a capable shield for nanopores [89].

To remedy these shortcomings, GO has attracted attention as it can be exploited as an ideal additive for WEP coatings, mainly because of its polar functional groups, such as hydroxyl, carbonyl, carboxyl, and

epoxy [157]. However, the performance of GO as a filler in corrosion inhibitors is still being debated [158]. Furthermore, owing to the intrinsic properties of water-based coatings, deformation, fractures, and micro-cracks are probable to occur inside WEP following layer formation [159,160]. In theory, the surface modification or functionalization of GO not only enhances the adhesion of the WEP coating method but also considerably improves the epoxy coating's mechanical, electrical, thermal, and barrier qualities.

## 5 Graphene Oxide/Nanofiller Composites Materials

Recently, adding nanofiller has been an excellent technique to promote corrosion resistance [161]. Ongoing efforts are made to enhance the physicomechanical and anticorrosive characteristics of protective coatings by using an optimal number of crosslinking agents and additives, as shown in Table 5. Ramezanzadeh et al. [162] stated that moist transition was used to introduce polyisocyanate nanostructured GO nanosheets through the polyurethane substrate. Hence, the use of 0.1 wt.% GO, and PI-GO significantly improves the coating's anticorrosive capabilities and electrical resistance. The study on this issue is in its initial phases and is rapidly developing.

Zheng et al. [163] demonstrated that GO sheets significantly increased the protection against corrosion of epoxy coatings over carbon steel substrates. The excellent dispersal of modified GO films in the polymer matrix was maintained, resulting in the improved reactivity of a prepolymer of urea–formaldehyde (UF) resin with the polymer matrix. It can be concluded that TiO<sub>2</sub>-GO hybrids can be proactively applied as nanofillers for anticorrosive epoxy coats, according to Yu et al. [89].

Both graphene oxide layers and polymer materials have lower utilization fillers for WEP coatings as they have low water dissolving rates and linking, which may result in free spaces and reduced coating barrier efficiency [158]. Nevertheless, graphene with strong electron conductivity has a better capacity than others and offers long-term barrier properties in WEP coatings [129].

Because of its weak water dissolution rate and crosslinking, both graphene oxide layers and organic polymers possess ineffective compliance fillers for WEP coating, which may generate free spaces and subsequently degrade the barrier efficacy of the coatings [158]. However, graphene with strong electron conductivity possesses a better ability than others and can have long-term corrosive resistance in WEP coatings [129].

**Table 5:** Related research on composite coating against the corrosion phenomenon

Coating agents of composite coating	Electrochemical Impedance Spectroscopy	Solution of action	Ref.
PI-GO/PU sample	The PI-GO coating showed the lowest breakpoint frequency changes after 14 days of immersion.	Improved barrier performance and electrical impedance.	[162]
UF-GO/EP resin	The coating resistance ( $R_C$ ) of GUF/EP coatings remained high and steadily varied, which is $3.79 \times 10^9 \Omega\text{cm}^2$ .	Enhancement of the corrosion resistance of composite materials on carbon steel surfaces.	[163]
TiO <sub>2</sub> -GO/EP resin	TiO <sub>2</sub> -GO sheet hybrids have better enhancement than GO and nano- TiO <sub>2</sub> .	Improved nanofillers' efficacy in corrosion protection epoxy coatings.	[89]
GO-polydopamine (PDA)/EP resin	The GO-PDA/EP coating exhibits the shielding performance of the blank EP and GO/EP coating, where the impedance modulus is about $2.46 \times 10^7 \Omega\text{cm}^2$ .	Enhancement of the structural applications of a waterborne EP coating.	[129]

(Continued)



Table 5 (continued)			
Coating agents of composite coating	Electrochemical Impedance Spectroscopy	Solution of action	Ref.
GO–alumina (Al <sub>2</sub> O <sub>3</sub> )/EP resin	The GO-Al <sub>2</sub> O <sub>3</sub> nanocomposites exhibited a higher anticorrosion performance of epoxy coatings than Al <sub>2</sub> O <sub>3</sub> and GO.	At a modest level (2 wt.%), it improved the corrosion protection of protective coatings and was recognized as a viable choice for epoxy corrosion protection coatings.	[157]
Silica (SiO <sub>2</sub> )-GO/EP composite	The EP samples loaded with SiO <sub>2</sub> -GO nanosheets showed higher $R_C$ .	Development of its dispersion and anticorrosion properties.	[164]
GO- polyaniline (PANI)/WEP coating	1.5 wt.% GO/PANI has a better anticorrosive property since it has a higher value of corrosion potential ( $E_{corr}$ ), lower value of corrosion current density ( $I_{corr}$ ), and polarization resistance ( $R_p$ ).	GO/conducting composite materials in aqueous corrosion protection coatings were being enhanced.	[165]
GO-polypyrrole (PPy)/WEP coating	GP <sub>0.05%</sub> coating showed the highest impedance modulus $ Z _{0.01 Hz}$ value where $E_{corr}$ is -625.2 mV and $I_{corr}$ is $2.677 \times 10^{-8}$ A/cm <sup>2</sup> .	With GP <sub>0.05%</sub> coatings, the corrosive prevention and impermeable properties were improved.	[84]
GO/Hexagonal boron nitride (h-BN) coating	$R_C$ of GO/h - BN <sub>(1:1 w/w)</sub> /WBE is maximum ( $4.05 \times 10^6 \pm 6.61 \times 10^5$ Ωcm <sup>2</sup> ).	The composite coating exhibited excellent barrier and corrosion protection properties owing to the synergistic Impermeable performance of h-bn and go.	[161]
Functionalized GO/ Polycaprolactone (FGO-PCL) coating	Large $R_C$ value of FGO-PC which is $2.186 \times 10^6$ Ωcm <sup>2</sup> and larger $ Z $ .	The incorporation of FGO nanosheets makes PCL more hydrophobic and provides additional adhesion strength to the polymer.	[166]

## 6 Green Chemistry Method

Several countries are currently employing green chemistry frequently to drastically reduce the overall usage of harmful substances and thereby repair the ecosystem to encourage ecologically friendly corrosive corrective actions [167]. Functional biofilms have recently grown in popularity as an environmentally safe corrosion protection technique [168]. It is critical to highlight those specific stabilizers, toxic-free reagents, and sustainable reducing agents have become the leading and crucial components in the production and stabilization of nanoparticles. Chen et al. [169] have reportedly devised an eco-friendly manufacturing method epoxidized biomass *Eucommia* gum (EEUG), as a nanocomposite in epoxy polymer composites by utilizing water as a surfactant. The authors discovered

that high functionalization densities with 1 wt.% EEUG led to a significant improvement in corrosion prevention efficacy.

## 7 Future Prospects

We can infer from our review research on the evolution of graphene that its transport characteristics have been effectively established. However, there is still space for improvement regarding its mechanical qualities. Relevant suggestions that bridged the gap between theoretical behavior and the actual achievement of mechanical behavior has already received much interest. These will be summed up as follows.

Due to the scale involved, the most widely investigated methods to produce graphene are through GO. Although all processes begin with the same source, the structural and surface properties of the GO can vary dramatically depending on how it is exfoliated and decreased. Graphite oxidation and heat or chemical reduction cause severe distortion of graphitic carbons, altering their micromechanical and electrical transport characteristics. Nonetheless, the processes of defect development and their impact on nanocomposite characteristics are still unexplored. To optimize the capability and gain of polymeric materials, synthetic methods to produce graphene sheets that prevent structural distortion or excessive functionalization, as well as post-processes that can recover graphitic planar domains, must be pursued.

The adhesion of graphene to polymers is another significant issue to address when creating composite materials. Carbon nanofillers' improved dynamics can be wholly exploited when tightly bound to matrix polymers. Enhanced adherence may reduce phonon contact dispersion and lead to better composite thermal conductivity and a reduction in surface-free volume, which impacts gas penetration. Thermal conductivity over the graphene/hydrocarbon junction can be improved by chemically adding alkyl groups to the graphene margins.

Graphene/polymer nanocomposites can satisfy thermal or energy storage requirements, static charge disposal, and electromagnetic pulse reflecting medium with electrical solid or thermal conductivity. Composite resistance can differ wildly with temperature, chemical assault, and external pressure, specifically around the threshold. This externally driven on-off phenomenon in electrical characteristics is used to identify stress and electric shifting. If microscopic sheets of highly charged graphene are put on glass or polymer materials, elastic screens, thin-film transistors, and photovoltaic, and crystalline systems will become viable.

## 8 Conclusion

Even though graphene has anticorrosion characteristics, imperfections in graphene can accelerate and intensify corrosion. Adding additional components to graphene or its compounds, including graphene oxide, improves its corrosion resistance over time. As a result of its flexibility, such approaches can be commonly applied by researchers in academic and industrial fields to create graphene-based polymer composite coatings. However, combining corrosion inhibitors covalently with polymer-functionalized graphene flakes or metal oxide nanoparticles can aid in increasing corrosion attacks. These composite materials are predicted to exhibit synergistic effects, improving corrosion protection characteristics. Once reliable sources of chemical bonding are employed for chemical functionalization, additional properties such as self-healing qualities, can be exploited to address any downsides. Using polydopamine, polyaniline, polypyrrole, and metallic nanoparticles such as  $\text{TiO}_2$ ,  $\text{Al}_2\text{O}_3$ , or  $\text{SiO}_2$  are examples of such applications. Hence, these composite materials' synergistic chemical resistance can lead to effective barriers that inhibit the penetration of corrosion attacks from going further into the coating. As a result, exceptional corrosion protection qualities, long-term rigidity, and toughness are guaranteed.

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