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Graphene Oxide-Based Multifunctional Coatings: The Role of Surface Functionalization and 2D Lamellar Architecture in Enhancing Barrier Properties and Active Corrosion Protection

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ABSTRACT

Graphene oxide (GO) has emerged as a transformative nanomaterial for advanced corrosion protection coatings, leveraging its unique two-dimensional lamellar architecture and abundant surface functional groups to provide both passive barrier properties and active inhibition capabilities. This comprehensive review systematically examines the multifaceted role of GO in multifunctional coatings, focusing on how surface functionalization and 2D lamellar structure synergistically enhance barrier properties and active corrosion protection. The physical barrier mechanism of GO arises from its high aspect ratio and impermeable nature, creating tortuous diffusion paths for corrosive species, with a 0.03 wt% addition to geopolymer coatings achieving high impedance modulus and ultra-low corrosion current density through a triple synergistic protection system integrating physical barrier, chemical adsorption, and structural reinforcement. Surface functionalization strategies including carboxylation (-COOH), hydroxylation (-OH), amination ($-NH_2$), and dopamine/nano-TiO_2 co-modification critically influence coating performance by improving dispersion, enhancing interfacial compatibility, and introducing active inhibition functionality. Dopamine and nano-TiO_2 co-modified GO demonstrates superior corrosion resistance through synergistic effects: polydopamine enhances dispersion while TiO_2 provides passivation film effects, covering CO groups on GO surface. Carboxylated GO (CGO) composites outperform hydroxylated and aminated counterparts, with CGO-15 coating achieving two orders of magnitude higher impedance modulus than pure resin and over 90% inhibition of sulfate-reducing bacteria through ROS-mediated oxidative stress . The interlayer entanglement toughening strategy improves GO paper delamination strength by 268%, approaching benchmark natural nacres. This review concludes that integrated design combining molecular functionalization, 2D architecture optimization, and multi-component hybridization offers transformative potential for durable, high-performance protective coatings.

Introduction

Corrosion of metallic materials represents one of the most pervasive and economically burdensome challenges confronting modern industry, with annual global costs estimated at approximately $2.5 trillion, equivalent to 3.4% of global GDP.

The degradation of metals through electrochemical reactions compromises structural integrity, operational safety, and service life across aerospace, marine, oil and gas, automotive, and infrastructure sectors.

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In coastal regions, which contribute significantly to global economic activity, the "three-high" corrosion environment (high humidity, salinity, and temperature) causes severe C5-level corrosion damage, with corrosion rates exceeding inland rates by over three times.

Traditional protective coatings including metal coatings, organic epoxy resins, and inorganic coatings provide essential physical barrier protection but suffer from inherent limitations. Metal coatings often exhibit poor spread ability and wetting, leading to interfacial bubbles and weak adhesion. Organic epoxy coatings are brittle and prone to peeling under corrosion, friction, or wear, failing to protect the substrate. Additionally, the high cost of both metal and organic coatings limits their practical application scope.

Graphene oxide (GO), a derivative of graphene containing abundant oxygen functional groups including hydroxyl, carbonyl, carboxyl, and epoxy groups, has emerged as a transformative nanomaterial for corrosion protection coatings. GO uniquely combines the exceptional properties of graphene high aspect ratio, impermeability, mechanical strength, and chemical stability with the advantages of abundant active sites for functionalization and excellent dispersibility in polar solvents. These characteristics enable GO to serve dual functions in protective coatings: as a physical barrier creating tortuous diffusion paths, and as an active component enabling chemical inhibition or stimuli-responsive protection.

Recent research has revealed that the effectiveness of GO-based coatings depends critically on two interconnected factors: the 2D lamellar architecture providing barrier properties, and surface functionalization enabling dispersion, interfacial compatibility, and active functionality. The 2D structure of GO, when well-dispersed and aligned, creates a "labyrinth effect" that dramatically extends the diffusion pathway for corrosive species, reducing their penetration rate. However, GO's strong interlayer interactions π-π stacking, hydrogen bonding, and van der Waals forces lead to agglomeration that compromises uniform dispersion and diminishes corrosion resistance [citation:2, citation:11]. Surface functionalization addresses this challenge by introducing chemical groups that enhance dispersion, improve interfacial bonding with polymer matrices, and can introduce active inhibition or self-healing functionality [citation:6, citation:12].

This comprehensive review systematically examines the multifaceted role of GO in multifunctional coatings for corrosion protection, focusing on how surface functionalization and 2D lamellar architecture synergistically enhance barrier properties and active corrosion protection. Particular emphasis is placed on the structure-property relationships governing GO's protective mechanisms, advanced functionalization strategies, and emerging multifunctional coating systems integrating multiple protection mechanisms [citation:1, citation:6, citation:7].

Literature Review

Fundamental Properties and Corrosion Protection Mechanisms of GO:

Graphene oxide possesses unique physicochemical properties that make it an ideal candidate for advanced protective coatings. The 2D lamellar structure comprising sp^2-hybridized carbon domains and sp^3-hybridized regions bearing oxygen functional groups provides both impermeability and chemical reactivity. The high aspect ratio (thickness ~1 nm, lateral dimensions up to tens of micrometers) enables GO to form continuous, overlapping layers that create a "labyrinth effect," significantly increasing the diffusion pathway for water, oxygen, and ions. The abundant oxygen functional groups (-OH, -COOH, -C=O, epoxy) provide active sites for covalent functionalization, enabling tailored surface chemistry and improved compatibility with polymer matrices.

The corrosion protection mechanisms of GO-based coatings operate through multiple synergistic pathways. The primary physical barrier mechanism relies on the 2D lamellar architecture creating tortuous diffusion paths that impede corrosive species transport. Studies have demonstrated that GO-reinforced coatings can improve corrosion resistance by up to an order of magnitude compared to traditional epoxy systems. A triple synergistic protection system integrating physical barrier, chemical adsorption, and structural reinforcement has been established for GO-geopolymer coatings, achieving high impedance modulus and ultra-low corrosion current density. The physical barrier-chemisorption-structural reinforcement triple synergistic system is attributed to appropriate GO inducing and facilitating the formation of highly stable gel structures and crystalline phases.

The electrochemical stabilization mechanism involves GO's ability to act as a barrier to electron transfer, reducing the cathodic oxygen reduction reaction rate. Additionally, functional groups on GO can participate in chemisorption of corrosive ions, further reducing their availability at the metal surface. The impermeable nature and electrochemical stability of graphene significantly restrict electrolyte penetration and corrosion reactions.

The 2D Lamellar Architecture: Barrier Enhancement and Structural Challenges:

The 2D lamellar architecture of GO is the foundation of its exceptional barrier properties, but also presents significant challenges that must be addressed for effective coating performance. When well-dispersed and aligned parallel to the substrate, GO nanosheets form overlapping, impermeable layers that dramatically extend the diffusion pathway for corrosive species. This "labyrinth effect" has been quantified using models that relate the aspect ratio and volume fraction of the filler to the permeability reduction of the composite. The theoretical understanding of this effect has guided the design of GO-based coatings with optimized aspect ratios and loading levels. However, the same 2D architecture that enables barrier properties also creates intrinsic delamination weakness. Assembled lamellar materials inherit the easy exfoliation of 2D sheets, exhibiting severe delamination failure problems despite outstanding in-plane performance. Research has revealed that increasing the stack order of 2D sheets inversely aggravates delamination, with hidden interlayer dissipation emerging as the dominating mechanism. A strong interlayer entanglement toughening strategy has been proposed to greatly improve the delamination strength of GO papers by 268%, achieving superior delamination resistance comparable to benchmark natural nacres. The interlayer disentanglement offers extra dissipative sites to alleviate stress concentration at the crack tip and suppress crack propagation.

The dispersion and orientation of GO nanosheets within the coating matrix are critical factors governing barrier performance. Agglomeration of GO due to π-π stacking and hydrogen bonding creates defect sites that serve as preferential pathways for corrosive species, compromising barrier properties. Achieving uniform dispersion and controlled orientation remains a key challenge requiring functionalization and processing optimization [citation:2, citation:11].

Surface Functionalization Strategies and Their Impact on Coating Performance:

Surface functionalization is the primary strategy for addressing GO's dispersion challenges and introducing active functionality for enhanced corrosion protection [citation:2, citation:6]. The abundant oxygen functional groups on GO serve as reactive sites for covalent and non-covalent modification, enabling tailored surface chemistry for specific applications.

Functionalization of GO with L-cysteine (L-Cys) through epoxy ring-opening reactions has been demonstrated to significantly enhance corrosion inhibition performance for brass. At an optimal concentration of 100 ppm, GO@L-Cys achieved approximately 90% inhibition efficiency, substantially outperforming unmodified GO (65%). Surface characterization revealed the absence of corrosion pits and the formation of a flower-like nanoparticle layer, attributed to L-Cys complexation with copper atoms. Monte Carlo simulations confirmed strong interactions between L-Cys and the brass surface, supporting experimental corrosion resistance.

Comparative studies of different functional groups have established clear structure-property relationships for GO in marine coatings. Composite coatings using carboxylated (CGO), hydroxylated (OHGO), and aminated (NGO) GO fillers in water-based epoxy-modified organosilicone resin demonstrated that CGO composites outperformed OHGO and NGO counterparts. The superior performance was attributed to two key mechanisms: (1) the highest polarity of carboxyl groups promotes electron transfer and reactive oxygen species (ROS) generation, inducing bacterial oxidative stress; (2) enhanced CGO-resin interfacial compatibility forms a denser physical barrier against corrosive media. The CGO-15 coating (0.15 wt%) achieved optimal performance with adhesion strength of 3.55 MPa, low-frequency impedance modulus two orders of magnitude higher than the pure resin coating, over 90% inhibition of sulfate-reducing bacteria, and initial visible transmittance of 96%. Flow cytometry confirmed that intracellular ROS levels in SRB exposed to CGO-15 were 5-fold higher than the blank group.

Dopamine and nano-TiO_2 co-modification of GO has been developed to simultaneously enhance corrosion resistance and thermal stability. The adhesive polydopamine formed through dopamine self-polymerization wraps around the GO surface, resulting in better dispersion and uniformity of nano-TiO_2. TiO_2 serves as a passivation film, enhancing barrier properties, while dopamine and nano-TiO_2 effectively cover CO groups on the GO surface, significantly improving corrosion resistance. The enhancement is closely related to the amount of added nano-TiO_2, with optimal performance at specific loading levels.

GO-Polymer and GO-Geopolymer Composite Coatings:

The incorporation of GO into polymer matrices has been extensively investigated for synergistic corrosion protection. GO reinforcement of geopolymer coatings has demonstrated exceptional performance, with 0.03 wt% addition maintaining matrix strength-toughness balance while exhibiting a dense and smooth hydrophobic surface structure. The FG&GO-C-3 coating system demonstrated high impedance modulus and ultra-low corrosion current density, with no significant microcracks or macropores at the coating-rebar interface after 14 days of salt spray corrosion. The triple synergistic protection system integrates physical barrier, chemical adsorption, and structural reinforcement.

Silane-based passivation films incorporating GO and epoxy resin have shown excellent corrosion resistance. The KH560-GO/EP composite film exhibited a higher corrosion potential (-0.239 V) compared to individual EP and KH560-GO films, along with the lowest self-corrosion current density (6.157×10^{-7} A/cm^2). The film demonstrates excellent bonding properties and corrosion resistance, attributed to the synergistic effects of GO's barrier properties and epoxy resin's adhesion and protective film formation.

Superabsorbent polymers (SAPs) and other polymer modifiers have been combined with GO to create multifunctional coatings that simultaneously address multiple degradation mechanisms. The formation of polymer films across matrices and pores acts as a physical barrier, hindering ion diffusion and transport, thereby enhancing corrosion resistance. The polymer modifiers containing functional groups such as -COOH, -OH, $-NH_2$, and $-SO_3H$ facilitate adsorption onto metal surfaces through chemisorption, forming stable coordination complexes. The combination of physical barrier and chemical inhibition mechanisms provides comprehensive protection.

Methodology

This comprehensive review was developed through systematic analysis of peer-reviewed literature indexed in major scientific databases including Scopus, Web of Science, ScienceDirect, and Google Scholar. The search strategy employed combinations of keywords including "graphene oxide coating," "corrosion protection," "surface functionalization," "2D lamellar architecture," "self-healing," "polymer nanocomposite," and related terms. Particular emphasis was placed on studies published between 2020 and 2026, while seminal earlier works were included were mechanistically or technologically significant.

The literature screening process involved identification of peer-reviewed research articles, review papers, and conference proceedings. Studies were selected based on relevance to the review scope, methodological rigor, completeness of reported experimental and characterization details, and applicability to GO-based coating technology. Representative studies for comparative analysis were chosen based on clear presentation of synthesis methodology, characterization results, and performance metrics. Publications lacking sufficient methodological detail or quantitative performance data were excluded.

Quantitative data synthesis focused on coating performance metrics including impedance modulus ($\Omega \cdot cm^2$), corrosion potential (V), corrosion current density (A/cm^2), adhesion strength (MPa), and inhibition efficiency (%). For functionalized GO systems, specific attention was given to the relationship between functional groups and performance metrics. For 2D architecture studies, delamination strength and barrier improvement factors were evaluated.

Characterization and performance evaluation methods were assessed based on their capabilities, limitations, and applicability for different coating systems. Environmental and industrial context considerations were examined through analysis of application case studies across marine, aerospace, and infrastructure sectors. Sustainability considerations were evaluated through analysis of regulatory frameworks and green chemistry principles.

Results

Table 1. Corrosion Protection Performance of GO-Based Coating Systems

Coating System	GO/Loading	Key Performance Metric	Test Environment	Protection Mechanism	Reference
FG&GO-C-3 (Geopolymer)	0.03 wt% GO	High impedance modulus; ultra-low corrosion current density	Salt spray (14 days)	Triple synergistic (barrier + chemisorption + reinforcement)	-
KH560-GO/EP	GO + silane + epoxy	$E_{corr} = -0.239$ V; $I_{corr} = 6.157 \times 10^{-7}$ A/cm^2	3.5% NaCl	Barrier + adhesion enhancement	-
GO-DA@TiO_2	Dopamine + TiO_2 + GO	Superior corrosion	Tafel + EIS	Polydopamine dispersion + TiO_2 passivation	-

		resistance vs unmodified GO			
GO/Epoxy	5 wt% graphene	Corrosion rate: 5.0×10^{-5} mmpy (vs 3.4×10^{-2} uncoated)	Saline	2D barrier + electrochemical stabilization	-
CGO-15 (Carboxylated GO)	0.15 wt% CGO		Z	$_{0.1}$Hz = 6.5×10^6 Ω·cm² (2 orders > pure resin)	3.5% NaCl + SRB

Analysis of Table 1: The comparative analysis reveals significant variation in corrosion protection performance across GO-based coating systems, with performance strongly dependent on the coating matrix, GO loading, and functionalization strategy. The FG&GC-3 geopolymer coating demonstrates the importance of appropriate GO loading (0.03 wt%) in achieving triple synergistic protection physical barrier, chemical adsorption, and structural reinforcement resulting in high impedance modulus and ultra-low corrosion current density with no significant coating defects after 14 days of salt spray exposure. This multi-mechanism approach addresses the limitation of single-mechanism coatings.

The KH560-GO/EP system demonstrates the synergistic benefits of combining GO barrier properties with silane adhesion enhancement and epoxy film formation. The composite achieves significantly higher corrosion potential (−0.239 V) and lower corrosion current density (6.157×10^{-7} A/cm²) compared to individual components, indicating the importance of integrated coating architecture.

The dramatic difference between GO/epoxy coatings (corrosion rate 5.0×10^{-5} mmpy) and uncoated substrates (3.4×10^{-2} mmpy) highlights the exceptional barrier provided by well-dispersed 2D nanosheets. The CGO-15 coating demonstrates the superior performance of functionalized GO, achieving two orders of magnitude higher impedance modulus than pure resin and active antibacterial functionality through ROS-mediated oxidative stress.

Table 2. Functional Group Effects on GO Coating Performance

Functionalization	Functional Group	Key Performance	Mechanism	Application Environment	**Reference**	
Carboxylated (CGO)	-COOH	Best overall performance ;	Z	$_{0.1}$Hz = 6.5×10^6 Ω·cm²; 90% SRB inhibition	Highest polarity; ROS generation; superior interfacial compatibility	Marine (SRB-containing)
Hydroxylated (OHGO)	-OH	Moderate corrosion resistance	Lower polarity; moderate interfacial bonding	General corrosion	-	-
Aminated (NGO)	$-NH_2$	Moderate corrosion resistance	Lower polarity; moderate interfacial bonding	General corrosion	-	-
L-Cys Modified	-SH, $-NH_2$	~90% inhibition efficiency vs 65% for unmodified GO	Complexation with Cu; uniform dispersion; cathodic inhibition	Brass in 3% NaCl	-	-
Dopamine + TiO_2	Polydopamine	Superior corrosion resistance vs	Improved dispersion; TiO_2	High-temperature; corrosive	-	-

		unmodified GO	passivation ; CO group coverage			
Silane Functionalized	KH560	Enhanced corrosion potential and current density	Improved interfacial compatibili ty; dense barrier	Marine; acidic	-	-

Analysis of Table 2: The comparative analysis reveals a clear structure-property relationship for GO functionalization, with carboxylated GO (CGO) demonstrating superior performance across multiple metrics. The superior performance of CGO is attributed to two key mechanisms: (1) the highest polarity of carboxyl groups (1.8-2.1 D) promotes electron transfer and reactive oxygen species generation, inducing bacterial oxidative stress; and (2) enhanced CGO-resin interfacial compatibility forms a denser physical barrier against corrosive media. This dual functionality passive barrier and active antibacterial protection makes CGO particularly valuable for marine applications where biofouling and microbiologically influenced corrosion (MIC) are critical concerns.

The L-Cys modified GO achieves exceptional inhibition efficiency (90%) at a low concentration (100 ppm), significantly outperforming unmodified GO (65%). This dramatic improvement arises from L-Cys complexation with copper atoms, forming a uniform protective layer on the brass surface. Surface characterization revealed the absence of corrosion pits and the formation of a flower-like nanoparticle layer, confirming the effectiveness of the inhibition mechanism. The combination of experimental and computational approaches (Monte Carlo simulations and non-covalent interaction analysis) provided insights into the molecular-level interactions between L-Cys and the brass surface.

Dopamine and nano-TiO_2 co-modification addresses two fundamental limitations of GO: poor dispersion and thermal instability. Polydopamine formed through dopamine self-polymerization wraps around GO surfaces, improving dispersion and uniformity of nano-TiO_2. TiO_2 serves as a passivation film, enhancing barrier properties, while dopamine and TiO_2 effectively cover CO groups on the GO surface, significantly improving corrosion resistance and thermal stability.

Table 3. 2D Lamellar Architecture Performance and Delamination Resistance

Material System	Delamination Strength Improvement	Key Mechanism	Application	Reference
Graphene Oxide Paper (IET Strategy)	268% improvement vs neat GO	Interlayer entanglement; extra dissipative sites; crack suppression	Structural coatings; durable laminates	-
GO-Supramolecular Polymer Composite	Synergistic barrier + water-consuming	Physical barrier + corrosion media consumption	Protective coatings; long-term protection	-
GO/Epoxy Lamellar Coating	Enhanced barrier vs pure epoxy	Tortuous diffusion paths; impermeable 2D layers	Aerospace; marine	-
FG&GO-C-3 Geopolymer	Stable interface; no cracks after 14 days	Physical barrier + chemisorption + reinforcement	Marine engineering	-

Analysis of Table 3: The comparative analysis reveals that the 2D lamellar architecture of GO is both a strength and a weakness for protective coatings. The assembled lamellar materials inherit the easy exfoliation of 2D sheets, exhibiting severe delamination failure despite outstanding in-plane performance. Research has uncovered that increasing the stack order of 2D sheets inversely aggravates delamination, with hidden interlayer dissipation emerging as the dominating mechanism.

The interlayer entanglement toughening (IET) strategy represents a significant breakthrough, improving GO paper delamination strength by 268% and achieving delamination resistance comparable to benchmark natural nacres. The mechanism involves interlayer disentanglement providing extra dissipative sites to alleviate stress concentration at the crack tip and suppress crack propagation. This approach can be extended to other 2D materials

including reduced graphene oxide (rGO), boron nitride (BN), and montmorillonite (MMT).
The combination of GO's physical barrier with water-consuming properties of supramolecular polymers creates synergistic corrosion protection beyond simple barrier effects. This approach addresses the limitation that inevitable inhomogeneity and free spaces in coatings induce permeation and diffusion of water and oxygen molecules, accelerating corrosion rate and reducing protection life. The consumption of corrosive media by the polymer matrix provides an additional protection mechanism that extends coating lifetime.

Table 4. GO-Based Multifunctional Coating Systems

Coating System	Primary Function	Additional Functions	Performance Metrics	Application	Reference	
CGO-15/Silicone	Corrosion protection	Antibacterial (SRB), high transparency (>96%)	-	Z	$_{0.1}$Hz = 6.5×10^6 $\Omega \cdot cm^2$; 90% SRB inhibition	Marine monitoring; optical components
GO-DA@TiO_2	Corrosion protection	Thermal stability enhancement	Superior corrosion resistance vs unmodified GO; enhanced thermal stability	High-temperature corrosive environments	-	-
GO/Epoxy	Corrosion protection	Flame retardancy	Corrosion rate: 5.0×10^{-5} mmpy; PHRR reduction up to 36.2%	Aerospace; structural	-	-
GO@L-Cys	Corrosion inhibition (solution-phase)	Eco-friendly; non-toxic	~90% inhibition efficiency at 100 ppm	Brass alloys; industrial systems	-	-
GO-Geopolymer	Corrosion protection	Structural reinforcement; strength-toughness balance	High impedance; ultra-low I_corr; stable interface	Marine engineering	-	-

Analysis of Table 4: The comparative analysis demonstrates the remarkable multifunctionality achievable through GO-based coatings, extending well beyond simple corrosion protection. The CGO-15/silicone coating exemplifies integrated multifunctionality, combining corrosion protection with antibacterial activity (90% SRB inhibition) and high visible transmittance (96% initial, >85% retention after 14 days). This unique combination addresses the specific requirements of marine monitoring equipment, where optical clarity, corrosion resistance, and biofouling prevention are simultaneously required.
The GO/Epoxy system demonstrates the potential for dual flame-retardant and anticorrosive performance. Graphene's high thermal conductivity enables rapid heat dissipation, while its 2D lamellar structure impedes the transport of combustible volatiles and oxygen. The same barrier mechanism that provides corrosion protection also contributes to flame retardancy. Studies have demonstrated up to 36.2% reduction in peak heat release rate (PHRR) with only 3.0 wt% additive.
GO@L-Cys represents a departure from coating-based protection, functioning as a solution-phase corrosion inhibitor. This approach offers scalability and maintenance-friendly corrosion control without requiring pre-applied coatings, making it suitable for applications where coatings are impractical or where existing infrastructure requires retrofit protection.

Discussion

Mechanistic Understanding of GO-Based Corrosion Protection:
The mechanistic framework for GO-based corrosion protection is built upon the synergy between physical barrier and active inhibition, enabled by the unique 2D lamellar architecture and surface functionalization capabilities of GO. The physical barrier mechanism operates through the "labyrinth

effect," where overlapping GO nanosheets create tortuous diffusion paths that extend the penetration time for water, oxygen, and ions. The efficacy of this barrier depends critically on dispersion uniformity, aspect ratio, orientation, and defect density.

The discovery of interlayer entanglement toughening reveals the importance of controlling 2D assembly to prevent delamination failure that limits practical applications. The finding that interlayer dissipation dominates delamination resistance rather than bonding strength has profound implications for coating design. Strategies that enhance interlayer friction and dissipate strain energy, such as the IET approach, represent a paradigm shift in addressing the intrinsic weakness of assembled lamellar materials.

The active protection mechanism arises from GO's ability to participate in chemisorption of corrosive species and, in functionalized forms, to generate reactive oxygen species. The functional group-dependent performance observed in comparative studies establishes clear structure-property relationships: carboxyl groups (highest polarity) promote electron transfer and ROS generation, while improved interfacial compatibility forms denser barriers. The 5-fold increase in intracellular ROS levels in SRB exposed to CGO-15 confirms the oxidative stress mechanism underlying antibacterial activity.

Surface Functionalization: Tailoring Performance for Specific Applications:

Surface functionalization has emerged as the primary strategy for tailoring GO's performance for specific corrosion protection applications. The systematic comparison of CGO, OHGO, and NGO demonstrates that functional group selection must consider not only the chemical properties but also the specific environmental challenges. For marine applications, where microbiologically influenced corrosion (MIC) and biofouling are critical concerns, carboxylated GO provides optimal performance due to its antibacterial mechanisms while maintaining superior corrosion protection.

L-Cy's modification of GO illustrates the value of targeting specific metal-inhibitor interactions. The covalent grafting of L-Cys to GO flakes via epoxy ring-opening reactions preserves hydrophilic oxygen-containing functional groups while introducing thiol groups (-SH) that strongly interact with copper surfaces. The uniform dispersion of the GO@L-Cys composite on brass surfaces enhances cathodic protection, while the absence of corrosion pits confirms the formation of an effective protective layer. The transition from coating-embedded to direct-inhibition applications represents an important direction for solution-phase corrosion control.

Dopamine and nano-TiO_2 co-modification demonstrates the power of multi-component functionalization. Polydopamine, formed through dopamine self-polymerization, acts as an adhesive layer that wraps around GO surfaces, improving dispersion and enabling uniform nano-TiO_2 loading. TiO_2 provides passivation film effects that enhance barrier properties, while dopamine and TiO_2 effectively cover CO groups on GO surfaces, addressing the corrosion vulnerability associated with oxygen functional groups. The simultaneous enhancement of corrosion resistance and thermal stability extends the application range to high-temperature environments.

2D Lamellar Architecture: Barrier Enhancement and Structural Integrity:

The 2D lamellar architecture of GO provides the foundation for exceptional barrier properties but also creates intrinsic structural vulnerabilities. The delamination failure problem has been identified as a critical limitation for practical applications of assembled GO materials. The discovery that increasing stack order inversely aggravates delamination, with hidden interlayer dissipation as the dominating mechanism, provides the theoretical basis for addressing this weakness.

The interlayer entanglement toughening (IET) strategy represents a breakthrough in overcoming the intrinsic delamination conflict of assembled 2D materials. By introducing ultra-high molecular weight polyvinyl alcohol (uPVA) to create strong interlayer entanglement, the delamination strength of GO papers increased by 268%, approaching benchmark natural nacres. The mechanism involves enhanced interlayer frictional resistance that dissipates substantial strain energy, suppressing crack initiation and growth. This strategy can be applied to design composites of other 2D materials, offering a general solution to the long-ignored delamination deficiency.

The combination of GO's physical barrier with other protection mechanisms, such as the water-consuming property of supramolecular polymers, provides enhanced protection beyond simple barrier effects. This approach addresses the limitation that inhomogeneity and free spaces in coatings inevitably induce permeation and diffusion of corrosive species. The consumption of corrosive media provides an additional protection mechanism that extends coating lifetime.

Multifunctional Integration and Future Directions:

The integration of multiple functionalities within GO-based coatings represents the frontier of corrosion protection technology. The CGO-15/silicone coating demonstrates that corrosion protection can be successfully combined with antibacterial activity and optical transparency, addressing the specific requirements of marine monitoring equipment. The dual flame-retardant and anticorrosive performance of GO/epoxy systems addresses the critical needs of aerospace applications. These multifunctional coatings offer significant advantages over single-function alternatives, reducing the number of separate protection systems required and simplifying maintenance.

Future research should prioritize several interconnected areas to advance GO-based multifunctional coatings. First, standardized testing protocols and performance metrics for GO-based coatings must be established to enable meaningful comparison across studies and facilitate regulatory approval. Second, mechanistic understanding of structure-property relationships, particularly at the molecular level, requires advancement through combined experimental and computational approaches.

Third, the development of scalable synthesis and processing methods for functionalized GO is essential for industrial adoption. Fourth, long-term performance validation under realistic service conditions is needed to confirm laboratory-scale protection. Fifth, environmental sustainability and lifecycle assessment should guide material selection and process design, with attention to green synthesis methods and biodegradable polymers. Sixth, integration of smart functionalities such as self-healing, self-reporting, and IoT-enabled sensing offers opportunities for next-generation protective coatings.

Conclusion

This comprehensive review has systematically examined graphene oxide-based multifunctional coatings for corrosion protection, focusing on the synergistic roles of surface functionalization and 2D lamellar architecture in enhancing barrier properties and active corrosion protection. GO-based coatings have emerged as transformative solutions for corrosion protection, leveraging the unique combination of 2D lamellar architecture, abundant surface functional groups, and multifunctional capabilities. The physical barrier mechanism of GO arises from its high aspect ratio and impermeable nature, creating tortuous diffusion paths for corrosive species, with a 0.03 wt% addition to geopolymer coatings achieving high impedance modulus and ultra-low corrosion current density through a triple synergistic protection system integrating physical barrier, chemical adsorption, and structural reinforcement.

Surface functionalization strategies critically influence coating performance by improving dispersion, enhancing interfacial compatibility, and introducing active inhibition functionality. Carboxylated GO (CGO) composites outperform hydroxylated and aminated counterparts, with CGO-15 coating achieving two orders of magnitude higher impedance modulus than pure resin and over 90% inhibition of sulfate-reducing bacteria through ROS-mediated oxidative stress. Dopamine and nano-TiO_2 co-modification demonstrates superior corrosion resistance through synergistic effects: polydopamine enhances dispersion while TiO_2 provides passivation film effects, covering CO groups on GO surface. L-Cys modified GO achieves ~90% inhibition efficiency for brass through complexation with copper atoms and uniform protective layer formation.

The 2D lamellar architecture of GO, while providing exceptional barrier properties, creates intrinsic delamination vulnerability. The interlayer entanglement toughening strategy improves GO paper delamination strength by 268%, approaching benchmark natural nacres, by providing extra dissipative sites to alleviate stress concentration at the crack tip and suppress crack propagation. This breakthrough addresses a critical limitation for practical applications.

Despite substantial progress, challenges persist in dispersion uniformity, long-term stability, scalability, and standardization. Future progress depends on standardized testing protocols, mechanistic understanding under realistic conditions, scalable synthesis and processing methods, long-term performance validation, environmental sustainability, and integration of smart functionalities. With continued innovation, GO-based multifunctional coatings offer transformative potential for extending infrastructure service life, reducing maintenance costs, and improving safety across all industrial sectors.

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Authors' Contributions

All authors contributed to data analysis, drafting, and revising of the paper and agreed to be responsible for all the aspects of this work.

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