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Graphene-Based Composite Coatings for Ship Anticorrosion and Antifouling: A Review on Progress and Prospects

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Abstract

The marine environment, on account of its elevated salinity, humidity, and UV radiation, brings about severe corrosion and biofouling to ships and offshore structures. Traditional coatings encounter difficulties in fulfilling the requirements of eco-sustainability, durability, and multifunctional synergy. Graphene, by virtue of its extraordinarily high specific surface area, chemical stability, as well as gas barrier properties, has emerged as a pivotal material for marine protection. The present review carries out a systematic consolidation of the recent advances in graphene-based composite coatings for ship anticorrosion and antifouling. It provides an elaboration on the "labyrinth effect" barrier mechanism of graphene and its integration with low-surface-energy components, for instance, FOTS and LDH. The key bottlenecks-namely functional isolation, weak interfacial adhesion, and elevated scaling costs-are subjected to a critical examination. The potential of a gradient functional design that incorporates an anticorrosion base combined with an antifouling top layer for integrated synergistic protection is explored, and a performance evaluation framework that encompasses impedance, contact angle, and biofouling attachment is put forward. Future directions are outlined, covering interfacial reinforcement, gradient synergy optimization, cost-effective scalable processes, as well as standardized evaluation criteria, thereby offering insights for engineering applications.

Keywords

Graphene-Based Composite Coating; Ship Anticorrosion; Ship Antifouling; Integrated Synergistic Design; Interfacial Reinforcement.

1. Introduction

The harsh marine environment-characterized by elevated salinity, humidity, intense UV radiation, and wave action-serves to accelerate metal corrosion and biofouling on ships and offshore structures, and these phenomena constitute the primary causes of coating failure and the reduction of service life [1,2]. Corrosion on its own accounts for the yearly scrapping of approximately 30% of the world's steel production, and the corrosion rates that are encountered in marine zones are far in excess of those on land [3]. Conventional protective coatings suffer from a number of inherent drawbacks: solvent-based systems release volatile organic compounds, namely VOCs; organic coatings develop microscopic pores that allow the ingress of corrosive agents; and toxic antifoulants, such as organotins, give rise to serious ecological risks [1]. It is therefore of critical importance to develop coatings that are environmentally benign, durable, and multifunctional for the safety of marine infrastructure and for the reduction of lifecycle costs.

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Graphene-a two-dimensional carbon material with a single-atom-layer structure-possesses an ultra-high theoretical specific surface area of 2630 m^2/g, excellent chemical and thermal stability, and outstanding gas barrier properties [3]. The lattice pores of graphene function to effectively block water, oxygen, and chloride ions, while also extending the diffusion pathways of these species by virtue of the "labyrinth effect" [2]; this serves to render graphene highly attractive for anticorrosion applications. Moreover, the combination of graphene with low-surface-energy substances or carriers for corrosion inhibitors holds potential for attaining integrated anticorrosion and antifouling functions [1]. In spite of the notable advances that have been made in single-function coatings, challenges continue to persist in the areas of interfacial adhesion, synergistic design, as well as scalable and cost-effective manufacturing [4]. The present review systematically consolidates the research progress concerning graphene-based composites for ship anticorrosion and antifouling, critically analyzes the current bottlenecks, and outlines the future directions so as to support the engineering translation of these materials.

2. Application Mechanism and Challenges of Graphene in Anticorrosion Coatings for Ship Metals

2.1 The Barrier Anticorrosion Mechanism of Graphene

The superior anticorrosion performance of graphene-based coatings arises from the synergy between its physical barrier effect and its chemical inertness. Aligned graphene sheets that are distributed within the coating matrix form a dense two-dimensional network, and this network significantly prolongs the penetration pathways for corrosive agents-namely H_2O, O_2, and Cl^--via the "labyrinth effect" [1]. At the same time, the sp^2-hybridized honeycomb lattice of graphene confers exceptional chemical stability, and it is capable of maintaining structural integrity even at 200°C in air [3].

Substantial experimental evidence provides support for this efficacy. Li Hao et al. developed a GO-HHU-T heterostructured coating that contained a UV-resistant agent, and this coating retained a high impedance modulus of 6.4×10^9 $\Omega \cdot cm^2$ together with a low water uptake of 2.3 wt.% after 100 days of immersion in a 3.5 wt.% NaCl solution [2]. Xing Yulei et al. employed LPCVD to coat copper with monolayer graphene, and they achieved a 20 mV positive shift in the corrosion potential, an order-of-magnitude decrease in the corrosion current density, and an order-of-magnitude increase-approximately ten times-in the impedance modulus when compared with bare copper [5]. Chen et al. further demonstrated that graphene films grown by CVD were able to protect Cu and Cu/Ni alloys from oxidation after 4 h at 200°C in air, with XPS detecting neither Cu_2O nor CuO [6].

Compositing further enhances the anticorrosion performance. Su et al. carried out the in-situ growth of LDH on graphene and the adsorption of L-aspartate anions; the resulting rGO-LDH-Asp nanofillers proceeded to form a three-dimensional barrier network that obstructed electron transfer [1]. The polyaniline/graphene composite developed by Chang et al. also exhibited excellent protection, and this finding underscores the synergy between graphene and conductive polymers [7].

2.2 The Challenge of Interfacial Bonding

A core bottleneck that hinders the practical application of graphene coatings is the weak interfacial adhesion between the coatings and the metal substrates. Graphene primarily adheres through physical adsorption, that is, via van der Waals forces, and this yields a low binding energy-the adsorption energy between graphene and copper is low, and it mainly originates from van der Waals forces [3]. As a consequence, the coating becomes susceptible to delamination when it is subjected to mechanical stress, abrasion, or prolonged immersion.

Tribological tests conducted by Jin Tao et al. served to illustrate this issue: a graphene coating exhibited stable friction reduction under a 0.3 N load, but it underwent rapid delamination from a stainless steel substrate when the load was raised to 3.0 N, and the friction coefficient reverted to that of the bare metal [4]. Prasai et al. also confirmed that insufficient interfacial bonding can bring about premature coating failure [8].

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Several strategies have been put forward to enhance the adhesion:

(1) Chemical Modification and Coupling Agents: Silane coupling agents, with KH-550 serving as a representative example, have been employed to modify graphene oxide (GO). Through hydrolysis and condensation, Si-O-Si bonds are formed and hydrophobic groups are introduced, and this brings about a significant improvement in the compatibility and interfacial bonding between GO and waterborne epoxy resins [9].

(2) Metal Transition Layer Mediation: The introduction of a transition layer, such as Ni, proceeding from lattice matching principles can promote chemical bonding. Ni possesses a lattice constant that is similar to that of γ-Fe. Depositing a Ni interlayer by means of magnetron sputtering or electrodeposition can facilitate the formation of stronger metal-carbon covalent bonds with graphene, and this strategy has been validated in carbon nanotube/metal systems [4]. Kumar et al. also reported that improved interfacial states were achieved in Ni-graphene composite coatings using this approach [10].

(3) Electrophoretic Deposition (EPD) Optimization: Adjusting EPD parameters, such as the electrode size ratio and the voltage, can improve the dispersion and deposition uniformity of graphene. Jin Tao et al., relying on COMSOL simulations, carried out the optimization of the electrode geometry. A cathode-to-anode size ratio of 2:3 minimized the coating thickness variation to 5.2 μm, yielded more uniform adhesion, and even conferred a temporary self-lubricating repair capability under a 1.0 N load [4].

2.3 Trade-off Between Preparation Process and Cost

The selection of the fabrication method exercises a critical impact upon the coating performance and scalability, and this situation gives rise to a classic trade-off between cost and performance.

(1) Chemical Vapor Deposition (CVD): CVD possesses the capability to produce high-quality graphene films that exhibit excellent anticorrosion properties, yet its complexity, combined with the requirement of high temperatures (650–1000°C) and the elevated cost of the equipment-industrial-scale equipment is expensive-serves to limit the scalability for coating ship hulls [3,5].

(2) Electrophoretic Deposition (EPD): EPD provides the advantages of rapid deposition, good control over the thickness, and low cost. Low-voltage EPD (5–20 V) brings about dense coatings that are suitable for scale-up, whereas high voltages (50–100 V) give rise to agglomeration and porosity, which in turn degrade the performance [4].

(3) Solution Blending and Spraying: These approaches are straightforward, cost-effective, and compatible with waterborne systems. However, the dispersion of graphene continues to constitute a key challenge [11].

In the context of shipyard applications, where scalability and environmental compliance are assigned a higher priority, EPD and spraying methods are the ones that hold greater promise. Their widespread adoption, nonetheless, calls for further optimization of the dispersion techniques and process parameters in order to strike a balance between coating quality and production efficiency. As was emphasized by Mu Songwei et al., carrying out improvements in the dispersibility of graphene oxide-based nanohybrids is of crucial importance for enhancing coatings that are fabricated via solution routes [12].

3. Low Surface Energy Antifouling Technology and its Combination with Graphene

3.1 Paradigm Shift in Antifouling Technology

Traditional ship antifouling was reliant upon the release of toxic biocides, for example, organotins and copper ions, which are effective in inhibiting fouling yet give rise to severe marine pollution. The tightening of environmental regulations has served to drive a paradigm shift, moving from a “toxic release” strategy to “physical barrier” or “fouling-release” strategies. Low surface energy materials,

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such as fluoropolymers and superhydrophobic coatings, function to minimize bioadhesion by way of weak interfacial bonding, and this circumstance makes it possible for the attached organisms to be removed by hydrodynamic forces, thereby offering an alternative that is more eco-friendly [1]. This trend in the direction of eco-friendliness has also been brought to attention by Gong Youning et al. [13].

The combination of graphene with low surface energy components constitutes a logical pathway toward integrated protection: graphene serves to provide the corrosion barrier, while the low-energy surface imparts antifouling properties. Nevertheless, current research continues to be largely compartmentalized; it displays a tendency to focus on either anticorrosion or antifouling in isolation, and the synergistic effects that occur within composite systems have not yet been subjected to adequate exploration [1]. Wang Bing et al. also made the identification that the absence of multifunctional synergy constitutes a gap in the research on graphene-based coatings [14].

3.2 Antifouling Mechanism and Application of Fluorinated Materials like FOTS

Fluorinated silanes, taking FOTS as a prominent example, serve as key low-surface-energy modifiers that are employed for eco-friendly antifouling. The mechanism on which they are based involves hydrophobic isolation and physical repulsion; upon being composited with graphene, synergistic anticorrosion and antifouling effects can be brought about [15,16].

The silanol groups that are present in FOTS undergo hydrolysis so as to form covalent Si–O bonds with the substrate, while the perfluoroalkyl chains proceed to orient outward, a process that raises the water contact angle from approximately 86° to values that exceed 115°. An Si–O–Si cross-linked network functions to lock the fluorinated groups in place, and by doing so it ensures the durability [15]. This low-energy surface brings about a minimization of the adsorption of bioadhesive proteins, and as a result, weakly attached organisms are able to be shed under flow conditions [16].

The optimal application makes use of ultrasonic spraying (with 1 vol.% FOTS, specific solvents, and spray cycles), and the hydrophobicity is preserved after a period of 3 weeks of immersion. Solvent vapor-assisted thermal bonding serves to improve the mechanical robustness-the rupture pressure is in excess of 17 bar-and it also controls the surface roughness such that it stays within the range of 156–178 nm in order to avoid the promotion of fouling [15].Compositing with graphene significantly strengthens the synergistic effect, and both antifouling and anticorrosion functions are achieved. Min et al. reported that a perfluorosilane-modified graphene-MXene 3D network/epoxy composite exhibited a 46.2% reduction in the corrosion current density in a seawater environment and continued to display excellent corrosion resistance after 30 days of immersion [16].

Limitations include poor compatibility with resins-phase separation occurs above 1.5 vol.%, and the optimal level is 1 vol.% [15]-and the susceptibility of the fluorinated groups to UV degradation, which necessitates their combination with UV-resistant components such as graphene [16].

3.3 Other Antifouling Strategies

Within the field of anticorrosion, self-healing technologies that are based on corrosion inhibitor carriers have also been explored, and layered double hydroxides (LDH) serve as a representative example. Yang Heng et al. pointed out that LDH can function as a nanocarrier, adsorbing chloride ions through ion exchange and releasing corrosion inhibitor anions, and in this way self-healing anticorrosion is accomplished [17]. However, the application of LDH in antifouling still presents limitations: its ability to inhibit marine microorganisms is restricted, and the slow ion-exchange rate makes it difficult to satisfy long-term antifouling requirements. Mu Songwei et al. also confirmed that compositing LDH with graphene can bring about an improvement in antifouling performance, yet the problem of insufficient long-term effectiveness persists [12].

The superhydrophobic modification of graphene also demonstrates potential for antifouling. Xing Yulei et al. fabricated CNTs/rGO hybrids with the aid of silane coupling agents, and they achieved a superhydrophobic coating that possessed a contact angle greater than 150° along with self-cleaning

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properties; however, its long-term durability in marine environments still requires further validation [5].

3.4 Challenges in Compositing

Two major challenges emerge when graphene is integrated with antifouling components:

(1) Phase Separation: Low surface energy materials, for instance, fluoropolymers, frequently display poor compatibility with graphene and the polymer matrix, and this leads to phase separation during the formation of the composite. The result is a heterogeneous coating structure that undermines both the anticorrosion and the antifouling performance. Surface modification of graphene is commonly required to improve the compatibility [4], and this challenge is equally pertinent to antifouling composites, as has been noted by Zheng Shufang et al. [11].

(2) Functional Layer Compatibility: In multilayer designs, weak adhesion between the antifouling top layer and the anticorrosion base layer can give rise to delamination under service conditions, such as long-term immersion or wave slap. As Sun Kai et al. cautioned, if the micro- and nano-containers within a "smart" antifouling layer are not well-integrated with the underlying anticorrosion layer, they might create pathways for corrosive media, thereby accelerating the damage to the substrate [1].

4. Integrated Anticorrosion-Antifouling Synergistic Design: Status and Prospects

4.1 Isolation in Current Research

A considerable degree of "functional isolation" serves to characterize a large portion of the current research on graphene-based marine coatings. Approximately 90% of the studies focus their efforts on optimizing either the anticorrosion properties or the antifouling properties in isolation. The anticorrosion work is centered upon the enhancement of the barrier properties and the interfacial strength of graphene [2,4,5], whereas the antifouling research carries out separate explorations of the efficacy of LDH or superhydrophobic materials [1,17]. There exists a pronounced lack of systematic investigation into the synergistic mechanisms that operate between these two critical functions. The coating designs that are currently available are frequently nothing more than "additive" in nature, rather than being truly "synergistic," and the interactions between the anticorrosion and antifouling components continue to remain unclear, with only limited theoretical guidance being available. The review carried out by Wang Bing provides corroboration for the fact that studies on graphene-based self-healing coatings predominantly address single functionalities [14].

Potential conflicts have yet to be brought to a resolution. For instance, the high electrical conductivity of graphene holds the potential to promote galvanic corrosion in the event that it is paired with certain metallic antifouling agents [3]. Conversely, the porous structures that are present in an antifouling layer could serve to compromise the dense barrier formed by graphene, and this circumstance would bring about a reduction in the corrosion protection [1]. Prasai et al. also brought to attention the necessity of addressing the potential role that graphene may play in localized corrosion within composite systems [8].

4.2 Gradient Functional Design: A Promising Pathway

Gradient functional design constitutes a viable strategy for achieving integration. Its core concept revolves around the construction of a layered architecture: a graphene-rich anticorrosion base layer is brought together with resins and/or inhibitor carriers with the aim of forming a dense barrier against corrosive media, and this base is then topped by an antifouling surface layer that incorporates low-surface-energy materials or self-healing micro- and nano-containers, a design that thereby serves to inhibit bioadhesion and to enable damage repair [1]. This approach has come to be recognized as a key direction for the enhancement of performance [13].

The body of existing research supplies foundational support for this concept. The GO-HHU-T heterostructured coating that was developed by Li Hao et al., which brings together UV resistance,

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mechanical reinforcement, and active anticorrosion, is characterized by a layered design wherein the graphene-based layer fulfills the role of a barrier and the functionalized surface responds to environmental triggers [2]. Mu Songwei et al. likewise provided a demonstration that a hierarchical arrangement of GO-based nanohybrids is capable of bringing about an improvement in the synergistic performance [12].

The successful realization of this gradient structure is dependent upon interfacial compatibility. The employment of coupling agents-taking silanes such as KH-550 as an example-or the introduction of transition layers between the base and top coats can serve to strengthen the interfacial bonding by way of chemical linkages, and this works to prevent delamination [2,9].

4.3 Evaluation Criteria for Synergistic Efficacy

There is an urgent requirement for a standardized quantitative framework to carry out the evaluation of the synergistic efficacy. The key performance indicators ought to integrate anticorrosion and antifouling metrics under test conditions that are kept consistent-more precisely, immersion in 3.5 wt.% NaCl along with exposure to simulated marine illumination that employs UVA-340 lamps in accordance with the ASTM G154 standards [18].

The core evaluation criteria are as follows:

• Anticorrosion: the impedance modulus at 0.01 Hz is required to be $\geq 10^8$ $\Omega \cdot cm^2$, and the salt spray corrosion area should be ≤ 5% after 1000 h

• Antifouling: the water contact angle should be ≥ 115° (with an optimal range of 115–130°), and the 72-h biofouling attachment should be ≤ 30%

• Synergistic durability: following 500 abrasion cycles, the decay of the impedance is expected to be ≤ 20%, and the drop in the contact angle should be ≤ 10%

This framework renders it possible to carry out a precise quantification of the anticorrosion-antifouling synergy, and it can be employed to guide the development and qualification of coatings.

5. Results

Graphene-based composite coatings are capable of displaying exceptional anticorrosion performance, primarily by virtue of the physical barrier effect and the chemical inertness of graphene. The “labyrinth effect” that is brought into being by the aligned graphene sheets serves to significantly prolong the penetration pathways of corrosive species, a phenomenon that is evidenced by the high impedance modulus and the low water uptake that are observed in long-term immersion tests [2,3,6]. The process of compositing graphene with other materials, taking layered double hydroxides and conducting polymers as examples, brings about a further strengthening of the barrier properties and the cathodic protection [1,7]. However, the weak interfacial adhesion that exists between graphene and the metal substrates continues to stand as a critical bottleneck. Physical adsorption via van der Waals forces leads to a low binding energy, and this results in delamination under mechanical stress or prolonged immersion [4,8]. The strategies that have been proposed to improve the adhesion include chemical modification with coupling agents [9], the introduction of metal transition layers [10], and the optimization of the electrophoretic deposition parameters [4]. The choice of the fabrication method entails a trade-off between the coating quality and the scalability: chemical vapor deposition yields high-quality films but is costly and complex, whereas electrophoretic deposition and spraying are more amenable to scale-up yet still require further optimization of the dispersion and the process control [5,11,12].

Within the field of antifouling, the paradigm has shifted from the release of toxic biocides to environmentally friendly physical barrier or fouling-release strategies. Low surface energy materials, such as fluorinated silanes like FOTS, create hydrophobic surfaces that minimize bioadhesion, and when these are combined with graphene, they offer the potential for integrated anticorrosion and antifouling functions [15,16]. Systems that are based on layered double hydroxides and that release corrosion inhibitors while trapping chloride ions also display antifouling efficacy, even though their

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long-term performance is limited by the slow kinetics of ion exchange [6,17]. Superhydrophobic modifications of graphene have been explored, but their durability in marine environments still needs to be validated further [5]. The key challenges that arise when compositing graphene with antifouling components include phase separation due to poor compatibility and weak adhesion between the functional layers in multilayer designs [1,4].

In spite of these advances, the current research suffers from functional isolation: roughly 90% of the studies focus on either anticorrosion or antifouling separately, and the investigation of synergistic mechanisms remains limited [1,14]. Potential conflicts, such as the electrical conductivity of graphene promoting galvanic corrosion or porous antifouling layers compromising the barrier integrity, have not yet been resolved [1,3]. A gradient functional design, which features a graphene-rich anticorrosion base layer and a low-surface-energy antifouling top layer, has been proposed as a promising pathway for integrated protection [2,12,13]. The interfacial compatibility between the layers is of crucial importance and can be improved by employing coupling agents or transition layers [9]. There exists an urgent requirement for a standardized evaluation framework that brings together the anticorrosion metrics, for instance, an impedance modulus $\geq 10^8\ \Omega \cdot cm^2$ and a salt spray corrosion area $\leq 5\%$ after 1000 h, with the antifouling metrics, for instance, a water contact angle $\geq 115°$ and a biofouling attachment $\leq 30\%$ after 72 h, so as to carry out the quantification of the synergistic efficacy and to provide guidance for the development of coatings [11,18].

6. Conclusion

Graphene-based composite coatings are possessed of tremendous potential for marine corrosion protection and antifouling, and this is by virtue of their outstanding barrier properties, chemical stability, and versatility. While significant progress has been realized in the understanding of the anticorrosion mechanisms and in the development of eco-friendly antifouling strategies, a number of critical challenges continue to persist, and these challenges take in weak interfacial adhesion, functional isolation between the anticorrosion and the antifouling components, elevated manufacturing costs, as well as the absence of a standardized synergistic evaluation. Future research should accord priority to the reinforcement of the interfaces by means of chemical modification and structural engineering, the optimization of gradient multifunctional architectures, the development of scalable and cost-effective fabrication processes, together with the establishment of comprehensive performance evaluation criteria. The addressing of these issues will be of essential importance for the purpose of bridging the gap between laboratory research and practical application, and it will serve to facilitate the successful deployment of graphene-based coatings for the long-term protection of ships.

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