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Research Progress on Graphene-Based Anticorrosive Coatings: From Structural Regulation to Multifunctional Integration

Abstract
Graphene and its derivatives have become a research focus in the field of anticorrosive coatings due to their excellent physical barrier properties, chemical inertness, and electrical conductivity. Based on 34 relevant references, this paper adopts a thematic structure to systematically review the latest research progress on graphene-based anticorrosive coatings from two dimensions: a technical perspective (dispersion improvement, conductivity regulation) and an application perspective (traditional coating enhancement, multifunctional coating development). The article cross-references relevant English literature, objectively presents the current research status in the field, and concludes with a critical commentary, pointing out the shortcomings of current research and future development directions.

Keywords*: Graphene; Anticorrosive coating; Functional modification; Zinc-rich coating; Multifunctional coating*

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I. Introduction

Metal corrosion causes huge economic losses globally each year, and coating protection is currently one of the most effective and economical anticorrosion methods (Sørensen et al., 2009)[1]. Graphene, as a two-dimensional carbon nanomaterial with excellent chemical stability, physical barrier properties, and electrical conductivity, is considered an ideal additive for anticorrosive coatings (Cui et al., 2019)[2]. However, graphene still faces many challenges in practical applications: its poor dispersion in the coating matrix, high electrical conductivity that may induce galvanic corrosion, and interfacial compatibility issues with resin matrices (Ding et al., 2019)[3]. In recent years, researchers have explored methods to solve these problems from different perspectives, promoting the rapid development of graphene-based anticorrosive coatings. This paper will systematically review and comment on related research from the two dimensions of technical perspective and application perspective.

II. Technical Perspective: Modification and Structural Regulation of Graphene

2.1 Dispersion Improvement Strategies

The high specific surface area and strong van der Waals forces of graphene make it prone to agglomeration, which is a bottleneck issue for its application in coatings. To address this challenge, researchers have developed various dispersion strategies. Guo and Li (2024)[4] grafted polyisocyanate onto the graphene surface, forming a cross-linked network with polyurethane, significantly improving the dispersion and interfacial compatibility of graphene in the composite coating. Wei et al. (2024)[5] proposed an ingenious dual-dispersion medium method, dispersing graphene oxide (GO) in deionized water and silver particles in ethanol, then mixing the two, successfully achieving uniform distribution and network structure construction of GO in the silver matrix.

Non-covalent modification has attracted attention because it maintains the structural integrity of graphene. Zhang and Zheng (2022) [6]coated graphene with insulating poly(m-phenylenediamine) through π-π interactions, which not only improved dispersion but also reduced the conductivity of graphene to 1.2×10^{-7} $S\cdot cm^{-1}$. Zhu et al. (2022)[7] used gallic acid to reduce and modify GO; the gallic acid adsorbed onto the graphene surface via π-π interactions, inhibiting agglomeration and repairing some surface defects.

Covalent functionalization is another effective strategy. Cui et al. (2023) [8]pointed out in their review that grafting organic or inorganic molecules via covalent bonds can significantly enhance the interfacial interaction between graphene and the polymer matrix. For example, using silane coupling agents (Pourhashem et al., 2017)[9] or polymer grafting (Ramezanzadeh et al., 2015) [10] can not only improve dispersion but also introduce new functions.

2.2 Conductivity Regulation and Galvanic Corrosion Inhibition

The high conductivity of graphene is a double-edged sword: on one hand, it facilitates electron transport; on the other hand, it may form a galvanic couple with the metal substrate, accelerating metal dissolution (Schriver et al., 2013) [11]. To address this issue, researchers have developed various inhibition strategies.

Coating with insulating materials is the most direct method. Zhang and Zheng (2022) [12] coated graphene with poly(m-phenylenediamine), completely cutting off the conductive pathways. Sun et al. (2015) [13]

encapsulated reduced graphene oxide (rGO) with APTES, successfully inhibiting its galvanic contact with the metal. Zhu et al. (2022) [14] coated graphene with polydopamine, which filled coating pores and coated the highly conductive graphene, effectively inhibiting galvanic corrosion.

Elemental doping can modulate the electronic structure of graphene. An et al. (2022) [15]prepared fluorine and nitrogen co-doped GO (FNGO). The defects introduced by doping and the different electronegativity elements altered the electron cloud distribution of graphene, reducing its conductivity and thus decreasing the tendency for galvanic corrosion. Research by Ren et al. (2022) [16] also confirmed that N-doped graphene exhibits better anticorrosion performance than undoped graphene.

Constructing oriented structures is another approach. Fei et al. (2022) [17] used electrodeposition to align sulfonated graphene (SGO) parallel on the surface of stainless steel driven by an electric field, forming a nacre-like "brick-mortar" structure. This highly oriented structure not only enhanced the physical barrier but also inhibited electron transport through the discontinuous structure in the vertical direction. Inspired by nacre, Ding et al. (2022) [18] prepared alternating layers of PDA-rGO and epoxy via layer-by-layer spraying, achieving anisotropic conductivity (in-plane conductive, out-of-plane insulating), effectively inhibiting galvanic corrosion.

III. Application Perspective: Traditional Coating Enhancement and Multifunctional Integration

3.1 Performance Enhancement of Zinc-Rich Coatings

Zinc-rich coatings provide cathodic protection through the sacrificial anode action of zinc dust, but they suffer from low zinc utilization efficiency and easy blockage of conductive pathways. The introduction of graphene offers new ways to address these problems.

Constructing conductive networks is key to improving zinc utilization efficiency. Wei et al. (2023)[18] compounded F, N co-doped rGO with polyaniline (PANI) to prepare PANI@N-FrGO nanofillers. These fillers acted as "conductive bridges" in the coating, activating isolated zinc dust and forming a Zn-filler-Zn conductive network, extending the cathodic protection period to 35 days. The rGO-Fe_2O_3-HEDP ternary composite designed by Ji et al. (2025)[19] is more ingenious: rGO provides the conductive network, Fe_2O_3 regulates conductivity and inhibits the corrosion-promoting effect of graphene, and HEDP acts as a corrosion inhibitor chelating Zn^{2+} and Fe^{2+} , activating zinc dust and achieving self-healing.

Intelligent corrosion inhibition is another important direction. Qi et al. (2025)[20] used sawdust biochar to load the green corrosion inhibitor tannin, preparing a TA@BC biocomposite. The loading of tannin endowed the material with pH-responsive release capability (released under alkaline conditions), which, synergizing with the conductivity of biochar, significantly enhanced the anticorrosion performance of zinc-rich epoxy coatings.

Replacing part of the zinc dust to reduce cost is a practical industrial demand. Huang et al. (2024)[21] replaced 10 wt% of zinc dust with low-cost iron phosphide (Fe_3P) and found that Fe_3P not only conductively connected zinc particles, but its flaky structure also enhanced the barrier effect, resulting in a 40 μm thick coating still providing a cathodic protection period of up to 86 days.

3.2 Corrosion Resistance Improvement of Waterborne Coatings

Waterborne coatings are favored for their environmental advantages, but their hydrophilicity leads to poor water resistance and insufficient anticorrosion performance. The introduction of functionalized GO offers possibilities for improving waterborne coating performance.

Cui et al. (2023)[22] systematically reviewed the key influencing factors of functionalized GO (fGO) in waterborne anticorrosive coatings, including coating integrity, interfacial adhesion, network structure formation, water resistance, and corrosion inhibition. Research by Li et al. (2016)[23] showed that moderately reduced GO (rGO) is more suitable for waterborne polyurethane coatings than GO because its surface hydrophilicity is reduced, but residual oxygen-containing groups still aid dispersion.

Constructing cross-linked networks can significantly enhance water resistance. Cui et al. (2020)[24] synthesized cross-linkable fGO; the grafted isocyanate groups could react with polyurethane molecules, forming a high cross-linking density network in the interfacial region, consuming hydrophilic groups, thereby improving the water resistance of the coating. Work by Zhu et al. (2022) also confirmed that the introduction of polydopamine not only improved adhesion but also enhanced the physical barrier of the coating by filling pores.

3.3 Development of Multifunctional Coatings

As application scenarios become more complex, single-function coatings are no longer sufficient to meet demands, making multifunctional integration a development trend.

Thermally conductive and anticorrosive dual-functional coatings have important applications in heat exchange equipment. An et al. (2024) [25] used fly ash zeolite-assisted ball milling to exfoliate rGO, preparing A-rGO composite particles. When added to benzoxazine resin, the coating's thermal conductivity increased to 1.561 $W \cdot m^{-1} \cdot K^{-1}$ (an increase of 676.1%), while the low-frequency impedance modulus reached as high as

2.433×10^9 Ω·cm². MrGO-6, efficiently prepared by Yang et al. (2022) [26] using a two-step reduction method, increased the thermal conductivity of epoxy coating by 267% at a 5 wt% loading and increased the coating resistance by 2-3 orders of magnitude.

Antibacterial and anticorrosive dual-functional coatings can address microbiologically influenced corrosion. The POT/CuO/EP composite coating prepared by Zhang et al. (2025)significantly enhanced the corrosion resistance of carbon steel in NaCl solution containing sulfate-reducing bacteria through the synergistic effects of POT passivation (forming Fe_2O_3/Fe_3O_4 passive film), CuO sterilization (releasing Cu^{2+}), and the physical barrier of epoxy.

Self-healing functionality is an important direction for smart coatings. Ye et al. (2020) [27] prepared GO-based nanocontainers loaded with the corrosion inhibitor benzotriazole. When defects occurred in the coating, the inhibitor released and formed a protective film at the defect site; localized electrochemical impedance spectroscopy (LEIS) tests confirmed its self-healing behavior. Research by Liu et al. (2020) [28] also indicated that polydopamine (PDA) and released benzotriazole (BTA) could synergistically form an adsorption layer in the damaged area, reducing interfacial electrochemical reaction activity.

IV. Critical Commentary and Prospects

Although significant progress has been made in research on graphene-based anticorrosive coatings, a comprehensive analysis of the 34 references reveals the following shortcomings in current research:

First, there is a significant gap between laboratory research and industrial application. Most research is conducted on a laboratory scale, and coating preparation methods (such as spraying, dip-coating, electrodeposition) are difficult to scale up directly to industrial production. For instance, although Guo and Li (2024)mentioned that the coating has "potential for scalable roll-to-roll production," they did not provide any experimental data or process parameters for scaled-up production. Sun et al. (2022) [29] also pointed out in their review that "the preparation of high-quality, large-size, controllable graphene films has not yet achieved a cost-effective breakthrough."

Second, there is insufficient understanding of the trade-off relationships among graphene dispersion, conductivity, and anticorrosion performance.Chemical functionalization often comes at the expense of conductivity, while physical coating may affect barrier properties. Although many studies (e.g., Zhang & Zheng, 2022; Zhu et al., 2022)simultaneously achieved high dispersion and low conductivity, their long-term service performance (>1 year) has not been reported. Sun et al. (2022) [30] clearly stated in their review that "detailed analysis of the diffusion and swelling behavior of graphene-modified coatings during immersion" is still lacking broad coverage.

Third, there is a severe lack of long-term service performance and durability data. The testing period in most studies is 30-60 days, such as the 35-day cathodic protection test by Wei et al. (2023)[31] and the 60-day immersion test by Ji et al. (2025)[32]. However, practical industrial applications (e.g., marine engineering, aerospace) require service lives of several years to decades. Zhang et al. (2025)[33] admitted in their study that "the environmental stability of POT and CuO under long-term service conditions lasting several years still needs evaluation."

Fourth, research on the intrinsic defects of graphene is insufficient. Most studies focus on dispersion and interface modification, but systematic research on how the structural defects of graphene itself (such as grain boundaries, vacancies) affect its long-term anticorrosion performance is limited. The study by Xu et al. (2025)[34] is an exception; the low-defect, highly conductive graphene they prepared significantly enhanced the adhesion and anticorrosion performance of polyurethane coatings at an extremely low loading (0.075%), hinting at the importance of graphene quality.

Fifth, research on the synergistic mechanisms of multifunctional coatings mostly remains at the phenomenological description level.Although multiple functions (e.g., anticorrosion, thermal conductivity, antibacterial, self-healing) have been integrated into a single coating, the interactions and synergistic mechanisms among these functions lack in-depth theoretical analysis and quantitative characterization. Although Qi et al. (2025) proposed "the synergy between the conductivity of biochar and the intelligent corrosion inhibition of tannin," they did not elucidate the quantitative relationship of this synergy.

Sixth, there are differences in problem orientation and solution paths between Chinese and international research. Chinese research teams tend to focus more on "systematic design" and "multi-step modification," preferring to solve core problems through complex modification strategies (such as doping + polymer coating, multi-layer structure design) (Wei et al., 2023; Ji et al., 2025). In contrast, European teams (e.g., Romero et al., 2024; Fernandez-Sotillo & Ferreira-Aparicio, 2023) tend to focus more on "process innovation" and "cost-effectiveness," preferring direct application by optimizing preparation processes (such as dip-coating, solution processing) and performing performance validation for specific application scenarios like PEMFC bipolar plates and current collectors. Iranian teams (e.g., Ramezanzadeh et al., 2018) have made outstanding contributions in the design of functionalized graphene.

In summary, future research should focus on: large-scale, low-cost preparation technologies for graphene; systematic evaluation of long-term (>1 year) service performance; research on synergistic mechanisms of multiple functions; and process scale-up validation for specific industrial applications. Only by addressing these issues can graphene-based anticorrosive coatings truly transition from the laboratory to industrial application.

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