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A Comprehensive Review on Surface-Engineered Montmorillonite and Layered Double Hydroxides as Smart Nanocarriers for Active Corrosion Protection in Polymer Coatings

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Advancements in corrosion science increasingly leverage nanoarchitecture materials to develop multifunctional protective coatings. This review highlights the efficacy of engineered nanoclays specifically montmorillonite (MMT) and layered double hydroxides (LDHs) as intelligent host materials for eco-friendly inhibitors in the polymer matrices. Their unique layered structures, when surface-functionalized with organic modifiers, achieve two critical objectives. First, they facilitate excellent dispersion, forming an impermeable nanoscale labyrinth that physically blocks corrosive agents such as water, oxygen, and chloride ions. Second, the modified interfaces act as selective gateways, enabling controlled, on-demand release of encapsulated inhibitors such as plant extracts or cerium cations triggered by local corrosion stimuli like pH changes or chloride ions. This responsive action delivers autonomous self-repair at defect sites, effectively halting corrosion propagation. The resulting nanocomposites exhibit synergistic enhancements in long-term barrier performance, mechanical strength, adhesion, and thermal stability. Furthermore, many systems demonstrate potent antimicrobial activity. This integrated approach offers a robust, durable, and sustainable alternative to traditional toxic inhibitor systems, significantly extending the service life of industrial infrastructure.

Keywords: Smart Nanocontainers, Self-healing Coatings, Montmorillonite Clay, Layered Double Hydroxides, Green Corrosion Inhibitor

Introduction

Corrosion of metallic materials poses a significant challenge to global industry, infrastructure, and economic stability [1]. This relentless electrochemical degradation process naturally reverts refined metals to their more thermodynamically stable oxidized states, often causing catastrophic failure of critical components across sectors such as transportation, energy, and construction [2]. The resulting financial burden is staggering, with annual losses estimated in the trillions of dollars worldwide, accompanied by serious safety risks and environmental consequences [3]. In the present context, the pursuit of effective corrosion protection is not merely a technical task but a vital act of preservation a form of industrial "forgiveness" that mitigates the inherent vulnerability of metals [4]. Protective strategies, therefore, serve to counteract the material's thermodynamic tendency to deteriorate, thereby extending service life, ensuring structural integrity, and conserving resources [5]. This philosophical perspective highlights the critical importance of developing advanced and sustainable anti-corrosion technologies to safeguard our engineered assets.

Among the most prevalent and effective methods for providing this "forgiveness" there is application of organic polymer coatings [6]. These systems primarily function as high-performance physical barriers, isolating the underlying metal substrate from corrosive species present in the environment, such as water, oxygen, and chloride ions [7]. The protective efficacy is intrinsically linked to the coating's adhesion to the substrate and its inherent resistance to the permeation of these aggressive electrolytes [8]. However, a significant limitation of conventional polymer coatings is their eventual

susceptibility to long-term degradation [9]. Mechanisms such as polymer chain scission, swelling, and the formation of micro-pores and micro-cracks compromise the coating's integrity [9]. This gradual breakdown transforms the initially robust barrier into a permeable membrane, allowing corrosive agents to reach the metal-coating interface and initiate under film corrosion, which can propagate undetected, leading to premature failure [9].

In order to enhance the protective performance of polymer coatings and provide active corrosion inhibition, the incorporation of corrosion inhibitors has been extensively studied [10]. Corrosion inhibitors are broadly categorized into inorganic and organic types. Inorganic inhibitors, such as chromates, phosphates, and molybdates, typically function by forming a passive oxide layer on the metal surface or by precipitating insoluble compounds that block active sites. Their advantages often include high efficiency and good thermal stability. However, major disadvantages include high toxicity (e.g., carcinogenic chromates), environmental persistence, and potential negative interactions with the coating matrix. Organic inhibitors, which encompass a wide range of compounds such as amines, azoles, and carboxylates, generally adsorb onto the metal surface through heteroatoms (e.g., N, O, S) or π-electrons, forming a protective film [10]. Their advantages include diverse molecular structures that allow for tailored design and often lower toxicity compared to some inorganic counterparts. Disadvantages may include higher cost, susceptibility to degradation under UV light or high temperatures, and potential volatility [10]. Driven by growing regulatory pressure and environmental concerns, research has increasingly focused on environmentally friendly, "green" inhibitors [11,12]. Green inhibitors are predominantly classified under the organic inhibitor category, as they are typically derived from natural products, plant extracts (rich in organic compounds such as alkaloids, tannins, and flavonoids), or biodegradable organic compounds [13]. They offer a sustainable and less toxic alternative to traditional hazardous inorganic inhibitors like chromates and lead-based substances [13,14]. Their primary advantage lies in their eco-friendly and often renewable nature. Challenges include variable composition in natural extracts, potentially lower intrinsic efficiency compared to some synthetic counterparts, and the need for careful integration into coating systems. However, a fundamental challenge remains: the direct addition of inhibitors whether conventional or green into the polymer matrix often results in suboptimal performance [15]. The inhibitors can adversely interact with the polymer, impairing its cross-linking and mechanical properties [15]. Moreover, they tend to leach out rapidly from the coating in a short, uncontrollable burst, leaving the system without long-term protection and potentially causing osmotic blistering [15].

To address the critical need for controlled and sustained release of inhibitors, the strategy of inhibitor encapsulation has emerged as a superior approach [16]. This method involves housing the inhibitor within nano or micro-carriers, which are then dispersed throughout the coating matrix [17]. Among the various nanocarriers studies, layered and two-dimensional materials such as MMT clay and LDHs have shown exceptional promise and are the primary focus of this review [18,19]. The emphasis on MMT and LDHs over other micro- and nano-carriers (e.g., silica nanoparticles, polymer capsules, halloysite nanotubes) stems from their unique combination of properties that are particularly advantageous for smart corrosion protection. First, their inherent layered structure provides a high capacity for intercalating or adsorbing inhibitor ions (anions for LDHs, cations for MMT), creating a substantial and stable inhibitor reservoir. Second, the ion-exchange capability of these materials serves as a key mechanism for triggered release, as corrosive stimuli (such as pH changes or specific aggressive ions) can be harnessed to release the stored inhibitors on demand. Third, the lamellar structure of these nanofillers enhances the barrier properties of the coating by creating a more tortuous path for diffusing species. Finally, their surfaces are highly amenable to modification with organic surfactants or silanes, which is critical for achieving compatibility with the polymer matrix (preventing agglomeration) and engineering the "gatekeeping" function for controlled release. This synergy of high loading capacity, stimuli responsiveness, barrier enhancement, and tunable surface chemistry makes MMT and LDHs uniquely suited as intelligent nanocarriers for advanced coating applications [18, 19]. A key step in utilizing these nanocarriers is the surface modification of their layers with organic surfactants or silanes [20]. This surface functionalization is crucial because it not only enhances the compatibility and dispersion of the nanocarriers within the hydrophobic polymer matrix—thereby preventing agglomeration—but also serves as a smart gatekeeper. [20]. The modified surfaces can respond to specific corrosion triggers, such as local pH changes or the presence of chloride ions, to regulate the release of the encapsulated inhibitor, providing an intelligent, on-demand healing effect at the corrosion site [20].

The innovation of yhe present study article lies in its focused synthesis and critical analysis of the latest advancements in the use of surface-modified MMT and LDH as smart nanocarriers for green corrosion inhibitors in polymeric coatings. While green inhibitors, nanocontainers, and smart coatings have each been studied individually, a focused review on how surface engineering synergistically enhances their dispersion and controlled release is urgently needed. This article systematically examines modification strategies, the mechanisms of inhibitor loading and release, and the

resulting anti-corrosion performance, thereby charting a course for future research. The significance of this topic is immense, as it lies at the intersection of materials science, electrochemistry, and environmental sustainability, offering a viable pathway toward developing next-generation protective coatings that are not only highly effective but also environmentally benign, ensuring the longevity and safety of metallic structures for years to come.

2. Functional Clay Family Additives: MMT and LDHs in Corrosion-Resistant Nanocomposites

2.1. The Anti-Corrosion Mechanism of MMT Clay in Polymer Nanocomposites

MMT clay functions as a highly effective anti-corrosion component in polymer nanocomposites primarily through a barrier protection mechanism [21]. Its unique layered silicate structure, when properly dispersed and exfoliated within the polymer matrix, creates a tortuous, maze-like pathway that significantly impedes the penetration of corrosive agents such as water, oxygen, and chloride ions [21]. This physical obstruction delays the diffusion of these electrolytes to the underlying metal substrate, thereby slowing down the anodic and cathodic reactions responsible for corrosion [22]. The high aspect ratio of the individual clay nanoplatelets maximizes the lengthened diffusion path, enhancing the coating's shielding properties [22]. Furthermore, the clay layers can improve the overall cross-linking density and reduce the free volume of the polymer, leading to a denser, less permeable coating film [23]. This synergistic combination of a nanoscale barrier and improved matrix integrity establishes MMT as a fundamental contributor to the long-term durability and enhanced corrosion resistance of protective polymer coatings.

Sheydaei and Edraki [24] synthesized an orange peel-impregnated sodium montmorillonite (Na^+-MMT) nanocomposite and incorporated it into an epoxy polymer matrix to develop a sustainable corrosion-inhibiting coating for mild steel. Their investigation revealed that this eco-friendly composite coating significantly improved the corrosion resistance of the substrate. The most exceptional performance was observed in the epoxy matrix containing 3 wt% of the nanocomposite, which achieved a peak inhibition efficiency of 94%. The research establishes that these plant-based inhibitors within an epoxy matrix present a highly effective and cost-efficient alternative to conventional chemical inhibitors, with the additional advantage of being derived from waste resources.

Edraki and Sheydaei [25] developed a green corrosion inhibitor by modifying Na^+-MMT with date seed powder (DS-MMT). When incorporated into an epoxy coating, it dramatically improved corrosion resistance, with the coating resistance (Rc) of the 3% DS-MMT composite being over 300 times higher than pure epoxy after 24 days. Visual salt spray tests confirmed no corrosion on the modified sample after 400 hours, unlike the heavily rusted pure epoxy (Figure 1). Furthermore, the composite demonstrated significant antimicrobial activity against *Bacillus subtilis* and *Staphylococcus aureus*. This dual mechanical and microbiological protection makes it a highly effective solution for comprehensive steel protection.

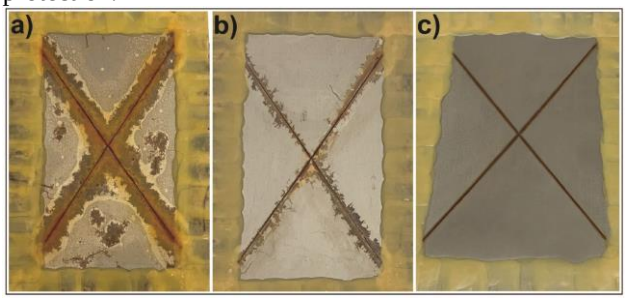

Fig.1. Images of coatings after 400 h of exposure to the salt spray test: (a) neat epoxy, (b) epoxy with 1.5 wt.% date seed-modified montmorillonite (EP/1.5% DS-MMT), and (c) epoxy with 3 wt.% date seed-modified montmorillonite (EP/3% DS-MMT). Reprinted/adapted with permission from Ref. [25]

Based on the study by Sheydaei and Edraki [26], the incorporation of Garcinia cambogia-modified clay (GC-MMT) into an epoxy matrix created a nanocomposite coating with superior properties. The key findings demonstrated a significant enhancement in corrosion protection, with an inhibition efficiency of approximately 88.85% for the coating containing 3 wt.% GC-MMT. Furthermore, the mechanical performance of the epoxy was substantially improved, as confirmed by tensile testing. Thermogravimetric analysis (TGA) revealed a remarkable increase in thermal stability, with the decomposition temperature rising by 50°C due to the presence of the nanofiller. This synergistic improvement in corrosion-mechanical resistance and thermal durability is attributed to the unique and homogeneous distribution of the GC-MMT nanoparticles within the epoxy matrix. Consequently, this research highlights the high efficacy of this bio-modified nanomaterial for developing advanced protective coatings.

Edraki *et al.* [27] successfully developed a novel green corrosion inhibitor by incorporating matcha green tea into Na^+-MMT (M-MMT). The anti-corrosion performance on mild steel in saline solution was then rigorously evaluated. Critical results from electrochemical impedance spectroscopy (EIS) demonstrated a substantial enhancement in corrosion resistance. These findings were further supported by data obtained from Raman spectroscopy. The study ultimately confirms that both M-MMT and matcha are effective green corrosion inhibitors.

Sheydaei *et al.* [28] found that a polyurethane nanocomposite with 3% matcha-modified clay (M-MMT) drastically improved corrosion resistance alongside its mechanical and antimicrobial properties. Electrochemically, after 70 days of immersion, the coating exhibited a remarkable corrosion resistance of 6.68×10^{11} Ω.cm^2, a substantial increase from the 1.62×10^{11} Ω.cm^2 of the pure PU. Mechanically, the tensile strength and elastic modulus rose to 19 MPa and 180 MPa, respectively. Furthermore, the M-MMT coating demonstrated strong biocidal effects against both *Streptococcus pyogenes* and *Klebsiella pneumoniae*.

Sheydaei et al. [29] developed a Ganoderma lucidum-modified clay (GL-MMT)/epoxy nanocomposite coating, demonstrating significant enhancements in key properties. The optimized epoxy/3% GL-MMT formulation exhibited superior anticorrosion performance, confirmed by polarization and salt spray tests, and formed a barrier with enhanced hydrophobicity, evidenced by a water contact angle of 73°.Technically, the coating showed improved adhesion strength and tensile properties, reinforcing its mechanical integrity. Thermally, TGA confirmed increased stability, ensuring performance under thermal stress. Critically, the coating displayed potent antimicrobial activity against Staphylococcus epidermidis and Streptococcus pyogenes, directly combating microbial corrosion. These synergistic improvements are attributed to the uniform dispersion of GL-MMT and polymer intercalation in the clay galleries.

Sheydaei *et al.* [30] demonstrated that incorporating Clitoria ternatea-modified clay (CT-MMT) into a sol-gel coating created a superior multifunctional material. Electrochemical analysis confirmed a dramatic increase in corrosion resistance, where the coating containing 3 wt.% CT-MMT achieved a resistance of 687 Ω·cm^2 after immersion (Figure 2). This performance vastly surpassed the mere 218 Ω·cm^2 of the pure coating. The enhancement is attributed to CT and MMT synergistically blocking anodic and cathodic sites. Concurrently, the coating exhibited potent antimicrobial properties against both Gram-positive *Staphylococcus aureus* and Gram-negative *Salmonella paratyphi-A*. This biocidal effect originates from the phenolic compounds in CT, which disrupt bacterial membranes and neutralize toxins. Consequently, the reasearch successfully developed a dual-action coating that provides excellent corrosion protection and effective microbial inhibition.

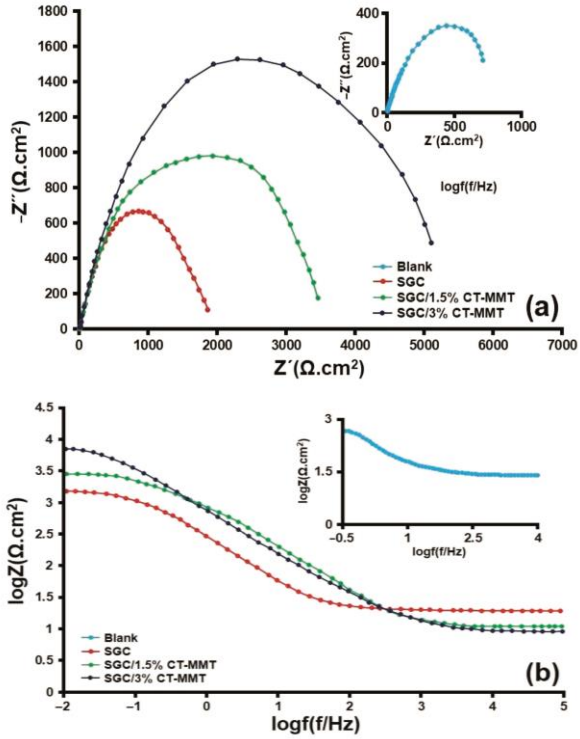

Fig.2. (a) Nyquist and (b) Bode plots of the specimens following 72 h of immersion in a saline solution. Reprinted/adapted with permission from Ref. [30]

Edraki *et al.* [31] successfully synthesized a novel epoxy nanocomposite by incorporating pine pollen-modified Na^+-MMT (PP-MMT) nanofillers into an epoxy resin matrix. Their investigation yielded significant enhancements in the coating's key functional properties. Regarding corrosion performance, electrochemical polarization (Figure 3) and salt spray tests demonstrated a substantial improvement in corrosion resistance, with the optimal coating containing 1.5 wt.% PP-MMT achieving a remarkable inhibition efficiency of approximately 87%. In terms of mechanical properties, the incorporation of PP-MMT effectively reinforced the polymer matrix. The storage modulus (E') was elevated from 2170 MPa for the neat epoxy to 2350 MPa for the nanocomposite. Furthermore, both the tensile strength and Young's modulus witnessed considerable increases, reaching 13 MPa and 126 MPa, respectively. Concerning antimicrobial efficacy, the PP-MMT/epoxy nanocomposite exhibited potent activity against a spectrum of Gram-positive and Gram-negative bacteria, including *Bacillus subtilis*, *Staphylococcus epidermidis*, *Escherichia coli*, and *Shigella dysenteriae*.

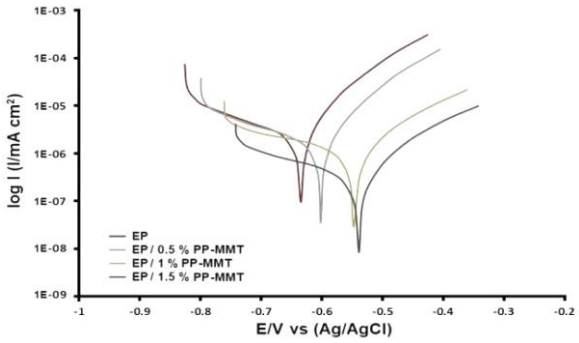

Fig. 3. Polarization curves of the samples immersed in a saline solution. Reprinted/adapted with permission from Ref. [31]

Edraki *et al.* [32] demonstrated that a polyurethane nanocomposite reinforced with ginger-modified clay (G-MMT) exhibits superior protective properties. The optimized coating with 3 wt% G-MMT achieved an exceptional corrosion resistance of 2×10^{10} Ω cm^2 after 120 days of immersion. Furthermore, the incorporation of the hydrophobic G-MMT nanoparticles significantly enhanced water repellency, increasing the contact angle to 72°. The mechanical performance was also substantially improved, with tensile tests confirming that the strength and robustness of the coating increased proportionally with the G-MMT content. This research conclusively establishes the G-MMT nanocomposite as a highly effective coating that synergistically combines enhanced anti-corrosion, hydrophobic, and mechanical properties.

Edraki and Sheydaei [33] successfully developed a vinyl ester (VE) nanocomposite doped with G-MMT nanoparticles. Their results demonstrated a significant enhancement in the coating's multifunctional properties. The nanocomposite exhibited superior dual active/barrier corrosion protection, where the clay structure acted as a physical barrier while the released ginger phenolic compounds actively inhibited anodic reactions. Mechanically, the incorporation of G-MMT notably improved the adhesion strength and tensile properties of the coating. Furthermore, the thermal stability was substantially increased, with TGA showing a rise in the degradation temperature by up to 43°C. Consequently, the G-MMT/VE coating presents a robust protective system with excellent corrosion resistance, mechanical integrity, and thermal performance.

Edraki and Zaarei [34] successfully developed and evaluated novel clay-based nanopigments for epoxy coatings. They modified Na^+-MMT with azole compounds, specifically 2-mercaptobenzothiazole (MBT) and 2-mercaptobenzimidazole (MBI), via an ionic exchange process. The anti-corrosion performance of these hybrid nanopigments was rigorously assessed using EIS and salt spray tests in a 3.5% sodium chloride solution over 60 days. Their critical finding was that the epoxy coating incorporating 3 wt.% of the MMT+MBT nano pigment demonstrated significantly superior long-term corrosion resistance compared to both the MMT+MBI variant and the pure epoxy coating. Furthermore, contact angle measurements revealed that this specific formulation, MMT+MBT, substantially increased the hydrophobicity of the coated steel surface. This enhanced hydrophobicity directly contributed to the improved barrier properties by more effectively repelling the corrosive electrolyte. Consequently, the authors concluded that the MMT+MBT nano pigment was exceptionally effective in simultaneously enhancing both the anti-corrosion performance and surface hydrophobicity of the protective organic coating, offering a robust solution for mitigating corrosion.

Edraki and Zaarei [35] demonstrated that the incorporation of azole derivatives, specifically MBT and MBI, into the interlayer spaces of Na^+-MMT nanocarriers significantly enhanced corrosion protection for mild steel in a 3.5 wt.% NaCl solution. Comprehensive electrochemical analyses, including polarization and EIS, revealed that the solutions containing the MMT + MBT and MMT + MBI nanocarrier systems exhibited superior inhibitory performance compared to solutions containing the inhibitors alone or the bare clay. A critical finding from their investigation into the solution phase, as determined by UV-Vis spectroscopy, was the differential release kinetics of the inhibitors from the clay nanocarriers. It was conclusively shown that the release of MBT species into the neutral aqueous medium was substantially lower and more controlled than that of MBI species. This controlled release profile from the MMT+MBT system is pivotal, as it directly correlates with the formation of a more stable and effective protective film on the steel surface, leading to sustained corrosion inhibition in the aggressive saline environment.

Mehrabian and Sarabi Dariani [36] showed that epoxy coatings modified with 3 wt.% 2-benzylbenzimidazole-modified clay (2-BBI-MMT) exhibited exceptional performance. EIS revealed this formulation provided the highest corrosion resistance after 120 days in a 3.5 wt.% NaCl solution. Furthermore, its barrier properties were outstanding, with a water absorption of only 1.92%, a drastic improvement over the 82% absorption seen in the unmodified clay/epoxy coating. This performance was linked to enhanced mechanical adhesion, as confirmed by pull-off tests. The authors concluded that the 2-BBI modifier improved clay dispersion and compatibility with the epoxy matrix, leading to superior thermal and barrier properties.

Subsequent studies have extensively explored and validated this fundamental barrier mechanism by incorporating various modified forms of MMT into diverse polymer matrices. As comprehensively summarized in Table 1, researchers have enhanced this passive protection by designing advanced nanofillers with active corrosion inhibition and self-healing capabilities.

Table 1: Clay-polymer nanocomposite coatings: formulations, protection mechanisms, and key performance metrics.

Clay Modifier / Type	Polymer Matrix	Metal Substrate & Corrosive Environment	Key Findings	Reference
Zn^{2+} & PO_4^{3-} modified MMT	Epoxy	Low-carbon steel/ Acid rain	Inhibition efficiency >90% in acid rain. protective film formed via Zn^{2+} release and iron phosphate stabilization. optimal at 0.05 M Na_3PO_4. Improves barrier properties of the epoxy coating (6x increase in protection).	37
MMT@Polydopamine@Cerium Phytate (MMT@PDA@PACe)	Epoxy	Mild Steel/ 3.5% NaCl	$\|Z\|_{0\text{-}01\ Hz}$ >10^9 Ω·cm^2 after 60 days. self-healing via Ce^{3+} release. dry adhesion = 5.43 MPa with only 14.1% loss after 30-day immersion.	38
Polyaniline/Phytic Acid modified MMT (PANI/MMT)	Epoxy	Steel / 3.5 wt.% NaCl	High $\|Z\|_{0\text{-}01\ Hz}$ (6.75×10^8 Ω·cm^2 after 150 days). Synergy of barrier (MMT) and passivation (PANI). Coating strength doubled to 3.09 MPa vs. pure epoxy (1.5 MPa).	39
Gallic Acid modified MMT (D-MMT)	Acrylic Latex (AL)	Steel / 3.5 wt.% NaCl	High impedance ($\|Z\| = 10^8$ Ω·cm^2). Post-damage passivation via chelation-reduction forms a dense protective layer. Increased hardness (B to HB), wear resistance, and thermal stability (decomposition temperature from 360°C to 380°C). corrosion current ↓ from 2.31×10^{-7} to 2.34×10^{-9} A/cm²	40
CeO_2 doped MMT (NCM)	Nanoclay Coating	Mild Steel/ 1M HCl	Excellent mixed-type inhibitor. 95-99% inhibition efficiency. Enhanced corrosion resistance in acidic environments.	41
CTAB & AP modified MMT	Epoxy	Cold-rolled Steel/Saline +Alkaline Copper Quaternary (ACQ)	Clay platelets create a tortuous path, blocking Cu^{2+} and O_2. AP-clay provides superior protection. Improved adhesion and wear resistance.	42
BTA-ZIF-8@Ce^{3+}-MMT	Waterborne Epoxy	Q235 Carbon Steel/ 3.5% NaCl	Synergistic anodic (BTA) & cathodic (Ce) inhibition. High $\|Z\|_{0\text{-}01\ Hz}$ (5.12×10^9 Ω·cm^2 after 28 days). Good adhesion retention (49.2% after salt spray test).	43
MMT@Phytic-Glucose@Ce-MOF	Waterborne Epoxy	Q235 Carbon Steel/ 3.5% NaCl	High barrier & active protection ($\|Z\|_{0\text{-}01\ Hz}$ ~10^9 Ω·cm^2). Fluorescent sensor for corrosion detection. Adhesion force 2–3 times higher than bare coating. Improved mechanical properties.	44
Iranian MMT modified with HDTB	Epoxy	Mild Steel/3.5 wt% NaCl	Significantly increased corrosion resistance compared to neat epoxy and unmodified MMT coatings. The optimum concentration was 3 wt.% OMMT. Increased Tg and storage modulus. Significant improvement in mechanical properties at 3 wt.% loading.	45
Acid-modified MMT	Acrylic Electrocoating	Steel / 3.5 wt% NaCl	Optimal at 0.3 wt%; $\|Z\| \approx 10$ GΩ after 21 d uniform dispersion enhances barrier	46
MMT modified with Zn-based Ionic Liquid	Epoxy	Steel / Saline	Outstanding self-healing and anti-corrosion. $\|Z\|$ increased by two orders of magnitude after 360h. Self-healing ability confirmed by SEM/EDX (Zn deposition in scratch).	47
CTAB/DTPMP modified MMT	Epoxy	Steel / NaCl	CTAB-clay significantly improves corrosion resistance.	48

Clay Modifier / Type	Polymer Matrix	Metal Substrate & Corrosive Environment	Key Findings	Reference
with Fe_3O_4/ZnO			Best interfacial adhesion and hardness with CTAB-clay. Denser, less porous layer.	
HDTPP modified MMT (OMMT) + PANI-rGO	Epoxy	Steel / NaCl	Optimal: 5 phr OMMT + 0.5 phr PANI-rGO Optimal sample shows best performance. Synergy between OMMT (barrier) and PANI-rGO. Simultaneous improvement in thermal stability and corrosion protection.	49
Al-pillared MMT modified with Ce^{3+}/Zr^{4+}	Silica Sol-Gel	Mg alloy AZ91D / 3.5 wt.% NaCl	Homogenization method yields coatings with better anticorrosive properties and self-healing ability. Effective self-healing demonstrated by SVET.	50
Urea-modified MMT	Epoxy	Carbon Steel/3.5 wt.% NaCl	Optimal 1-2% clay improves protective properties vs. pure epoxy. Improved protective properties confirmed by EIS and SEM.	51
DCNP based on cationic dyes (Basic Blue 9, Basic Yellow 87) and Cloisite 20A organoclay.	Epoxy	Mild Steel/3.5 wt.% NaCl	Corrosion resistance superior to coatings with Cloisite 20A alone. Hybrid pigments combine color and enhanced barrier properties. Effective loading range of 1–5 wt.%. Improved dispersion (intercalation/exfoliation) of DCNPs compared to pure organoclay. Good dry and wet adhesion.	52
Na^+-MMT impregnated with Basil extract as a green inhibitive carrier (G-MMT).	Sol-Gel Silane Coating	Mild Steel/ 3.5 wt.% NaCl Solution.	R_{ct} value with G-MMT was 5x higher than additive-free sample. Forms a protective film and Fe^{2+}-inhibitor complex on steel surface. Acts as an active nanoreservoir with time-dependent inhibitor release. Enhances the protective behavior of the silane coating. Proven to be an eco-friendly alternative to toxic inhibitors.	53
Na^+-MMT intercalated with Cerium cations (Ce-MMT).	Epoxy	Mild Steel/ 3.5 wt.% NaCl Solution.	93% corrosion inhibition efficiency in electrolyte phase. EIS and salt spray tests confirmed enhanced active protection of the epoxy coating. Ce^{3+} release provides active anti-corrosion performance. Improves the protective behavior of the epoxy coating by providing active corrosion inhibition.	54
MMT Clay (unmodified specified)	Novolac & Resol Phenolic Coatings	Mild Steel/ 3.5 wt.% NaCl Solution.	Adding clay improves corrosion protection for both phenolic coatings. Novolac coating with 3 wt.% clay showed the highest corrosion resistance. Resol coating showed better flexibility and adhesion than novolac coatings.	55
Organically Modified MMT (Organoclay)	Petrolatum	Mild Steel/ 3.5 wt.% NaCl Solution.	Improved barrier and anti-corrosive properties. The coating with 1 wt.% clay showed the best performance. The interlayer spacing of clay increased after incorporation into petrolatum via shear and sonication, indicating good dispersion.	56
MMT Clay (unmodified specified)	Thermoplastic Acrylic Resin	Low Carbon Steel/3.5 wt.% NaCl Solution.	Anti-corrosive properties were obviously increased by nanoclay. The coating with 3 wt.% clay showed the best corrosion resistance. Nanocomposites with 1 and 3 wt.% clay showed the highest adhesion to the substrate (via pull-off and	57

Clay Modifier / Type	Polymer Matrix	Metal Substrate & Corrosive Environment	Key Findings	Reference
			cross-cut tests).	
MMT Clay (unmodified specified)	Bitumen	Steel 37/3.5 wt.% NaCl Solution.	Bitumen/MMT coatings were superior to pristine bitumen. Corrosion protection improved with clay loading up to 4 wt.%. Improved barrier properties. Adhesion was characterized via pull-off test.	58
MMT Clay & Cerium Nitrate (dual filler)	Polyurethane	Carbon Steel/3.5 wt.% NaCl Solution.	Polyurethane/ MMT/Cerium nitrate coatings were superior to neat Polyurethane. Corrosion protection improved with clay (up to 4 wt.%) and cerium nitrate (up to 2 wt.%) loading. Nanocomposites were prepared via mechanical and sonication processes, with dispersion state characterized.	59

Note
CTAB: Cetyl trimethyl amonium bromide
AP: Aniline Pentamer
BTA: 1H-benzotriazole
ZIF-8: imidazolate frameworks
MOF: metal organic framework
Tg: glass transition temperature
HDTB: hexadecyltrimethylammonium bromide
SEM: scanning electron microscope
EDX: Energy Dispersive X-Ray Spectroscopy
DTPMP: diethylenetriamine penta methylene phosphonic acid
HDTPP: hexadecyl triphenyl phosphonium
PANI-r GO: polyaniline-reduced graphene oxide
phr: parts per hundred resin
SVET: scanning vibrating electrode technique
DCNP: Hybrid Dye-Clay Nano-Pigment
R_{ct}: charge transfer resistance

2.2. Dual Active/Passive Corrosion Protection Mechanism of LDHs in Polymer Coatings

LDHs significantly enhance the corrosion protection of polymer coatings through a synergistic combination of passive and active mechanisms. Primarily, the lamellar, platelet-like structure of LDHs dispersed within the polymer matrix creates a highly tortuous pathway, effectively acting as a physical barrier that impedes the penetration of corrosive species such as water, oxygen, and chloride ions [19]. Beyond this barrier effect, LDHs function as intelligent nanocontainers for corrosion inhibitors. These inhibitors are stored within the interlayer galleries of the LDH and are released on-demand via an anion-exchange process, typically triggered by the presence of aggressive chloride anions in the local environment [60]. The released inhibitors then migrate to the coating defect or metal surface, where they form a protective complex or passivation layer, thereby suppressing the anodic and/or cathodic corrosion reactions [60]. This intelligent release system provides a self-healing capability, effectively mitigating corrosion at its inception and offering long-term, active protection that far surpasses the performance of conventional, inert filler-based coatings [61]. More investigations about the functional mechanisms of diverse LDHs systems within polymer coatings are consolidated in Table 2, highlighting key findings from the current literature.

Table 2: Summary of Studies on LDH Composite Coatings for Corrosion Protection

LDH Type & Modifying Factor	Polymer Matrix	Metal Substrate & Corrosive Environment// Tests Methods	Key Findings	Reference
LDH modified with Choline Benzoate (CB) Ionic Liquid	Epoxy	AZ31B Mg alloy /3.5 wt.% NaCl / EIS and salt spray	Significant increase in polarization resistance. Excellent active protection & self-healing. Synergistic mechanism: barrier effect (LDH), anion trapping (Cl^-), and release of benzoate inhibitors.	62
Zn-Al LDH (ZLDH) and its calcined form (CZLDH)	Polyvinylidene Fluoride (PVDF)	Mild Steel/3.5 wt.% NaCl / EIS and Potentiodynamic polarization	ZLDH/PVDF showed superior corrosion resistance vs. CZLDH/PVDF. Intact LDH structure is critical for good dispersion, adhesion, and barrier properties against Cl^-. ZLDH/PVDF showed enhanced interfacial adhesion and reduced porosity.	63
LDH intercalated with Tannic Acid	Polyurethane &	AA2024 Aluminum Alloy /Saline media/ EIS	Polyurethane-LDH-TA provided superior protection vs. epoxy.	64

LDH Type & Modifying Factor	Polymer Matrix	Metal Substrate & Corrosive Environment// Tests Methods	Key Findings	Reference
(TA)	Epoxy		TA release forms a protective Al/ tannate layer at defect sites (active protection). Polyurethane -LDH-TA maintained the structural integrity of the coating.	
Mg-Al LDH intercalated with 8-Hydroxyquinoline (8HQ), Sodium Benzoate (SB), APTS	Not a polymer coating (In-situ LDH coating on alloy)	Mg Alloy/3.5 wt.% NaCl / Potentiodynamic polarization and EIS	All inhibitors improved corrosion resistance. 8HQ performed best due to chelation with Mg^{2+} and self-healing. Compact and uniform LDH coating morphology.	65
Ni-Al LDH modified with Phosphate ions (PO_4^{3-})	Polyurethane	Carbon Steel/3.5 wt.% NaCl /Salt Spray Test & EIS	98% inhibition efficiency. Controlled release of PO_4^{3-} and entrapment of Cl^-. Effective self-healing.	66
Zn-Al LDH loaded with 2-Aminomalonamide (AMA), hybridized with Graphene Oxide (GO)	Waterborne Epoxy	Q235 Steel/ 3.5 wt.% NaCl / EIS and Mechanical	Exceptional long-term barrier ($Z= 3.05\times10^9$ $\Omega\cdot cm^2$ after 100 days). Synergy between LDH/GO labyrinth effect and inhibitor passivation. Enhanced impact resistance, adhesion, and friction resistance.	67
LDH intercalated with Gallic Acid (GA)	Epoxy	Q235 Steel/ 3.5 wt.% NaCl / EIS and a neutral salt spray	GA-LDH/EP showed excellent corrosion protection. LDH blocks diffusion, traps Cl^-, and releases GA anions to chelate with metal ions. Impedance dropped only one order of magnitude in 35 days.	68
Co-Al LDH modified with Sodium Pyrithione (SPT)	LDH coating itself (on Al alloy)	AA7075 Aluminum Alloy / 3.5 wt.% NaCl / EIS and polarization	CoAl-LDHs-SPT coating increased corrosion resistance by two orders of magnitude. SPT provides excellent antibiofouling properties, preventing attachment of bacteria and algae.	69
Mg-Al LDH doped into Micro-Arc Oxidation (MAO) coating, sealed with 8HQ inhibitor film	MAO Coating + 8HQ film	Magnesium Alloy/ 3.5 wt.% NaCl / EIS and polarization	HQ/LDH/MAO composite coating showed synergistic effect. j_{corr} reduced by ~3 orders of magnitude; Z increased by ~4 orders. LDH doping densifies MAO, and 8HQ provides active sealing. Low-porosity and dense composite coating.	70
Ca-Al LDH intercalated with Nitrite ions (NO_2^-)	Epoxy	Carbon Steel/ 0.02 M NaCl/ Salt Spray	LDH-NO_2/Epoxy coating resistance was one order of magnitude higher than LDH/Epoxy. Nitrite is released via ion exchange with Cl^-, providing active protection. Molybdate modification failed, forming $CaMoO_4$.	71
LDH (Hydrotalcite) intercalated with Citric Acid or Tetraborate, modified with Sodium Laurate	LDH coating itself (superhydrophobic)	Aluminum/ 3.5 wt.% NaCl/ polarization and water contact angle	U-LDH-SC/SL film showed the best performance (most positive E_{corr}). Superhydrophobicity (WCA ~154°) slows water infiltration. LDH releases inhibitors for passivation and traps Cl^- Superhydrophobic surface (Sliding Angle < 5°). Uniform film growth.	72
LDH intercalated with Sodium dodecyl sulfate (SDS) & 8-HQ	MAO Coating	AZ31 Mg Alloy/ 3.5 wt.% NaCl/ EIS and Mechanical	CI@LDH-MAO coating's impedance was 3 orders of magnitude higher than MAO after 360h. Formation of LDH sheets and insoluble $Mg(HQ)_2$ compounds provides protection. Excellent tribological properties: low friction coefficient (~0.19) and low wear rate.	73

LDH Type & Modifying Factor	Polymer Matrix	Metal Substrate & Corrosive Environment// Tests Methods	Key Findings	Reference
Zn-Al LDH intercalated with Decavanadate $(V_{10}O_{28})^{6-}$, functionalized with 3-aminopropyl triethoxysilane (APTES)	Epoxy (Epon 828)	Mild Steel/ 3.5 wt.% NaCl/ potentiodynamic polarization	Silane functionalization improves LDH compatibility with epoxy. The modified LDH provides active corrosion protection and improved barrier properties.	74
Mg-Al LDH incorporated with 8-Hydroxyquinoline-5-sulfonic acid (HQS)	LDH-based hierarchical structure on plasma electrolytic oxidation (PEO) coating	Mg Alloy/ 3.5 wt.% NaCl / Potentiodynamic polarization and EIS	Enhanced LDH growth without extreme pressure. Combination of LDH ion-exchange and HQS organic layer augments corrosion resistance. DFT simulations explained interfacial processes.	75
Zn-Al LDH decorated with ZIF-8 metal-organic frameworks (MOF) and doped with Phosphate ions	Epoxy	3.5% NaCl / EIS and Mechanical/Mild Steel	Phosphate-doped LDH@ZIF8/Epoxy showed superior barrier properties ($Z > 10^{10}\ \Omega \cdot cm^2$) and a 57% self-healing index. pH/ion-responsive release of $PO_4^{3-}$and Zn^2. Significantly improved adhesion: ~69% improvement in wet adhesion and 73% reduction in cathodic delamination.	76
Zn-Al LDH intercalated with Diethyldithiocarbamate (DEDTC)	Hybrid Sol-Gel Silane (Bilayer with LDH)	3.5% NaCl / EIS and Potentiodynamic polarization /AA2024-T3 Aluminum Alloy	Bilayer system with inhibitors (DEDTC & Ce^{3+}) dramatically enhanced protection. Corrosion current density reduced from 170 to 6.7 nA/cm^2. Induced active anti-corrosion properties.	77
LDH intercalated with Molybdate (MoO_4^{2-}), core-shell with SiO_2	Sol-Gel Film	3.5% NaCl / EIS and salt spray /Mild Steel	LDH(Mo)@SiO_2 provided superior active inhibition due to controlled release of MoO_4^{2-} and enhanced barrier properties from SiO_2 shell.	78
Mg-Al LDH intercalated with Phosphate ions (PO_4^{3-})	Silane Primer + Epoxy Topcoat	Mild Steel/ 3.5% NaCl/ Salt Spray and Mechanical	Mg-Al-PO_4 LDH provided significant active corrosion protection in a defective coating. Releases Mg^{2+} and PO_4^{3-} (via solubility). Forms an inhibitive film. Improved adhesion strength after salt spray exposure.	79
Zn-Al LDH intercalated with Phosphate (PO_4^{3-}) and Nitrate (NO_3^-)	Hybrid Silane Primer + Epoxy Topcoat	Mild Steel/ 3.5% NaCl/ EIS and potentiodynamic polarization	Zn-Al-PO_4 LDH provided efficient inhibition by releasing PO_4^{3-} anions via ion exchange, outperforming the Zn-Al-NO_3 LDH. Reduced cathodic delamination of the epoxy topcoat.	80
LDH combined with TA	Polyvinyl Alcohol (PVA)	Ti_6Al_4V/ 3.5% NaCl/ potentiodynamic polarization and Mechanical	PVA/TA1/LDH2 composite showed the best overall performance. TA improves corrosion resistance (lowest Icorr: 0.36 $\mu A\ cm^{-2}$). Highest tensile strength and crystallinity. Lowest coefficient of friction and wear rate, indicating superior tribological performance.	81
Mg-Al LDH intercalated with Gallic Acid (GA)	Not a coating; particles dispersed in solution	Mild Steel/ 3.5% NaCl (Simulated Seawater)/ EIS and potentiodynamic polarization	LDH/GA (800 ppm) showed high inhibition efficiency (73-78%). The GA forms a protective layer on the steel surface. The formed GA layer increases the surface	82

LDH Type & Modifying Factor	Polymer Matrix	Metal Substrate & Corrosive Environment// Tests Methods	Key Findings	Reference
			hydrophilicity.	
Mg-Al LDH intercalated with GA	Not a coating; particles dispersed in solution	Mild Steel/ 3.5% NaCl at High Temperature (35°C & 45°C)/ EIS and potentiodynamic polarization	LDH/GA nanocomposite is effective at high temperatures, achieving 72% efficiency at 35°C and 58% at 45°C. Pristine GA performance is severely limited by temperature. Acts as a mixed-type inhibitor.	83
Mg-Al LDH intercalated with GA	Not a coating; particles dispersed in solution	Mild Steel/ Acidic, Neutral, and Alkaline Environments/ EIS and potentiodynamic polarization	LDH/GA acts as a pH-responsive inhibitor, increasing corrosion resistance in all three environments. Confirmed mixed-protection mechanism and the formation of a protective film on the metal surface.	84

3. Key Parameters Influencing the Efficacy of MMT/LDH-Polymer Nanocomposite Coatings: Filler Dispersion, Film Thickness, and Cure Schedule

The performance of polymer nanocomposite coatings incorporating nanofillers like MMT and LDH is critically influenced by filler size, coating thickness, and curing parameters. The particle size and degree of exfoliation of MMT and LDH are fundamental to their reinforcing and barrier efficacy [85]. Smaller, well-exfoliated particles create a more tortuous path for corrosive agents, significantly enhancing the diffusion barrier [85]. However, achieving a homogeneous dispersion is challenging, as nanoparticles are prone to agglomeration, which can create defect sites and undermine coating integrity. Concurrently, the dry film thickness must be optimized; a thicker coating generally provides a better physical barrier but increases the risk of internal stresses, cracking, or incomplete curing [86]. The curing conditions time and temperature directly govern the cross-linking density of the polymer matrix [86]. An optimal cure schedule ensures sufficient reaction of functional groups, leading to a dense, coherent network that improves mechanical strength, adhesion, and long-term barrier properties [85,86]. Inadequate curing leaves residual solvents or unreacted monomers, increasing permeability, while excessive heat can cause thermal degradation of the polymer or nanofillers. Therefore, a synergistic balance among nanofiller dispersion, coating thickness, and a meticulously controlled curing process is paramount to developing high-performance, durable anti-corrosion coatings [85,86]. A summary of these critical parameters and their impacts is provided in Table 3.

Table 3: Key performance parameters for nanocomposite coatings.

Parameter	Description	Typical Value Ranges	Impact on Coating Performance	Reference
Particle Size & Dispersion	Size and exfoliation level of MMT/LDH nanofillers.	Thickness: 1-10 nm (individual layers). Lateral Size: 50-1000 nm. Loading: 0.5-5 wt.% (optimal dispersion).	Smaller, well-exfoliated particles maximize the tortuous path effect, enhancing barrier properties. Agglomeration at high loadings (>5 wt.%) creates defects, reducing performance.	87
Coating Thickness	The final dry film thickness.	Standard Range: 80-150 µm. High-Performance: 150-300 µm.	An optimal thickness provides an effective barrier; films <50 µm may be permeable, while thick films >300 µm can lead to cracking and poor adhesion.	87,88
Curing Temperature	Temperature for cross-linking.	Ambient Cure: 20-30 °C. Thermal Cure (Epoxy): 80-150 °C. High-Temp Cure: 150-200 °C.	Higher temperatures increase cross-linking density, improving hardness and chemical resistance. Excessively high temperatures (>250 °C) can cause polymer/filler degradation.	89
Curing Time	Duration at the curing temperature.	Ambient Cure: 24 hours - 7 days. Thermal Cure: 30 minutes - 2 hours. Post-Cure: 1-4 hours.	Sufficient time is necessary for complete cross-linking. Inadequate time results in a soft film; excessive time can lead to embrittlement without significant added benefit.	87,90

4. conclusion

The pursuit of advanced corrosion protection has evolved from employing passive barriers to designing

intelligent, self-healing systems that actively mitigate degradation. This paradigm shift is exemplified by the strategic use of nanoscale layered materials, specifically MMT and LDHs, as smart carriers for environmentally friendly inhibitors. These engineered nanocarriers provide a dual mechanism of protection: they significantly enhance the coating's barrier properties by creating a tortuous path for corrosive agents, while simultaneously functioning as responsive reservoirs. **The critical innovation lies in the surface modification of these nanocarriers, which ensures their compatibility with the polymer matrix and enables a controlled, trigger-activated release of inhibitors.** This on-demand release provides targeted self-healing at defect sites, effectively prolonging the service life of the coating. In the case of MMT clay, **significant enhancements in barrier, mechanical, and thermal properties have been demonstrated in various polymer matrices**, with particular improvements noted when the clay is modified with natural inhibitors such as orange peel, date seed, and matcha. Similarly, **LDHs have been extensively studied for their dual active/passive protection mechanisms**, where inhibitor release is triggered by corrosive anions, providing effective self-healing. **In both systems, surface functionalization is consistently identified as a key factor for optimizing dispersion and controlled release.** A balance between nanofiller dispersion, coating thickness, and curing parameters is crucial for achieving high performance. **The synergy achieved through this approach—combining robust physical barriers with intelligent chemical release—marks a significant advancement over conventional coatings.** It forges a path toward next-generation protective systems that are not only highly effective and durable but also align with the principles of environmental sustainability. Future developments will likely focus on refining the responsiveness of these nanocontainers and exploring new synergistic combinations to further enhance their multifunctional performance.

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