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Graphene derivatives reinforced metal matrix nanocomposite coatings: A review

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Abstract

Due to the extraordinary mechanical, thermal, and electrical properties of graphene, graphene oxide (GO), and reduced graphene oxide (rGO), these materials have the potential to become ideal nanofillers in the electrodeposited nanocomposite coatings. This article provides an overview of literature on the improvements of properties associated with graphene, GO, and rGO-reinforced coatings, along with the processing parameters and mechanisms that would lead to these improvements in electrodeposited metal matrix nanocomposite coatings, where those affected the microstructural, mechanical, tribological, and anti-corrosion characteristics of coatings. The challenges associated with the electroplating of nanocomposite coatings are addressed. The results of this survey indicated that adding graphene into the plating bath led to a finer crystalline size in the composite coating due to increasing the potential development of specific crystalline planes and the number of heterogeneous nucleation sites. This consequently caused an improvement in hardness and in tribological properties of the electrodeposited coating. In graphene reinforced metallic composites, the severe adhesive wear mechanism for pure metallic coatings was replaced by abrasive wear and slight adhesive wear, where the formation of a tribolayer at the contact surface increased the wear resistance and decreased friction coefficient. Furthermore, superhydrophobicity and smaller grain size resulted from embedding graphene in the coating. It also provided a smaller cathode/anode surface ratio against localized corrosion, which has been found to be the main anti-corrosion mechanism for graphene/metal coating. Lastly, the study offers a discussion of the areas of research that need further attention to make these high-performance nanocomposite coatings more suitable for industrial applications.

1. Introduction

Nanocomposite/nanostructure coatings have recently attracted attention due to their mechanical, physical, and chemical properties [1-3]. There are several methods to prepare nanocomposite coatings, such as sol-gel, cold/thermal spray method, chemical vapor deposition (CVD), physical vapor deposition (PVD), electroless deposition, laser processing [4-12], and electrodeposition methods. The electrodeposition technique in particular has attracted significant attention since it can be done at ambient temperatures, it leads to a reduction in the chance of interfacial reactions, and it is an economical and less wasteful generative process requiring only a simple set-up [13-18].

A significant amount of research has been done on the electrodeposition of metal and alloy coatings such as nickel (Ni), zinc (Zn), copper (Cu), nickel-cobalt (Ni-Co), nickel-zinc (Ni-Zn) [19-27]. The effects of different nanofillers on mechanical properties, wear resistance, and corrosion resistance of the coatings were investigated. It was found that the properties of the coatings can be significantly improved by adding nanofillers such as silicon carbide (SiC) [28,29], silicon dioxide (SiO_2) [30], aluminum oxide (Al_2O_3) [31-32], titanium nitrate (TiN) [33], carbon nanotubes (CNTs) [34] to the matrix.

The extraordinary mechanical, electrical, and thermal capabilities of graphene-based materials have attracted the attention of researchers, resulting in an exponential increase in the number of papers relevant to graphene-based materials in recent years. Graphene based materials can be effectively incorporated into electrodeposited materials. The increased understanding of graphene-based nanofiller and its homogenous dispersion in metal matrix coatings using the electrodeposition process opened up a wide variety of possible applications in fields ranging from medicine to energy. The regulated synthesis of these coatings, with well-defined nanomaterial size, shape, and crystallinity, provided ideal templates for the creation of hydrophobic surfaces, resulting in improved anti-corrosion capabilities [35-45].

The superior mechanical properties (high Young's modulus and strong fracture strength) of graphene are related to their strong σ bonds (C–C bonds). Graphene has Young's modulus of ~1 TPa, and the breaking strength is almost 200 times higher than steel. It is also worth noting that the mechanical properties decrease as the number of defects increases [46-47].

Perfectly structured graphene is relatively inert and interacts with other materials mainly by physical adsorption. However, foreign atom defects or surface functional groups realize higher reactivity

[48-52]. For example, Mark A. Bissett *et al* showed that mechanical strain could change the structure of graphene and greatly increase its chemical reactivity [53]. Furthermore, since graphene is inherently hydrophobic and graphene oxide is hydrophilic, it has been commonly used in combination with other materials in appropriate aqueous systems for the synthesis of graphene-based composites [54].

The thermal conductivity of suspended graphene is very high (6000 $W \cdot m^{-1} \cdot K^{-1}$), [43],[55-57]. Balandin *et al* reported that by including 5 vol% graphene, thermal conductivities could be improved by ~500% for graphene-metal composites. This mainly resulted from the intrinsically high thermal conductivity of the graphene component combined with its strong synergetic effects with metal and polymer components [58]. The quality of the graphene, such as the lateral size and defect factor, influenced its thermal conductivity [47]. The theoretical specific surface area (SSA) of defect-free monolayer graphene is 2630 $m^2 \cdot g^{-1}$. Graphene is a zero-gap semiconductor with semiconductive and metallic properties due to its unique 2D structure. The carrier mobility of graphene and suspended graphene at room temperature could reach 15000 $cm^2 \cdot V^{-1} \cdot s^{-1}$ to 200000 $cm^2 \cdot V^{-1} \cdot s^{-1}$ with the charge carriers adjustable between electrons and holes [35], [43],[59-61]. Consequently, graphene has a high electrical conductivity, which is advantageous for enhancing the electrical conductivity of graphene-based composites for EES [62-63]. The number of layers and stacking orders, as well as the volume and form of defects, have a significant effect on the electrical conductivity of graphene [47]. Because of its special 2D microstructure with single-layered atomic thickness, graphene has an opacity of ~2.3% in a broad wavelength spectrum from ultraviolet to near-infrared [64-65]. On the other hand, the researchers also found that the transmittance of graphene decreases linearly as the layers increase. Graphene is being considered as a potential material for transparent applications in the future, including solar cells, batteries, smart windows, and other optoelectronic devices due to its superior optical properties [43],[64],[66-69].

Nanocomposite/nanostructure coatings have an excellent passive barrier function which significantly improves the overall performance and service life of the coating. Graphene nanocomposites can be used in a wide range of applications such as membranes, coatings, anti-corrosion, energy storage, etc. [70-77]. More recently, the research has been extended to demonstrate graphene-containing (G) nano-composites coatings [39],[40],[78-86].

For this paper, current developments in the synthesis of graphene-based metal nanocomposite coatings were reviewed in-depth, with a particular emphasis on the electrodeposition technique and characteristics of these coatings. The objective is to provide a perspective on the effective parameters for the synthesis of nanostructure and nanocomposite coatings via electrodeposition methods and provide an overview of metallic coatings reported thus far. Section 2 of the article gives details on the co-deposition mechanism. The structural and morphological properties of nanocomposite coatings are studied in Section 3. Section 4 of this review deals with mechanical properties while Section 5 covers tribological properties. Section 6 gives detail about the anti-corrosion performance improvement and the final section describes challenges associated with electrodeposition of metal-graphene composite coatings and the current research gap.

2. Electrodeposition mechanism of graphene entry into metallic coating

The electrophoresis migration of charged particles under electric field force is the main factor influencing particles into the electroplated coating, based on the theory of electrochemistry (Figure 1). As the cathode surfaces are charged negatively during the electrodeposition process, if the surfaces of particles have sufficient positive charges, conditions are favorable to the co-deposition of metal ions and particles at high speed. Therefore, it is hypothesized that the particle surface has a positive charge and the charge density per unit area is constant. It can be proved that the deposition speed depends on the size [87-89].

$$F = Eq \tag{1}$$

Furthermore, movement in the liquid is resisted by a frictional force that is proportional to the velocity.

$$F_f = Vf \tag{2}$$

Where f is the frictional coefficient.

From equations 1 and 2 the following can be deduced:

$$\mu = \frac{V}{E} = \frac{q}{f} \tag{3}$$

Where μ is the mobility, V is velocity, q is the net charge on molecule, and E is the electric field strength.

According to the Stokes law:

$$f = 6\pi\eta R \tag{4}$$

From equations 3 and 4:

$$Eq = 6\pi\eta RV \tag{5}$$

R is the radius of the capsule and η is viscosity.

Here, E, q, and η are fixed values. Therefore

$$V \propto \frac{1}{R} \tag{6}$$

Table 1. Properties of single-layer graphene [43].

ρ ($g \cdot cm^{-1}$)	T_m (°C)	A_s ($m^2 \cdot g^{-1}$)	UTS (GPa)	η (TPa)	TC ($W \cdot m^{-1} \cdot K^{-1}$)
1.06	5727	2630	130	1	1000-6000

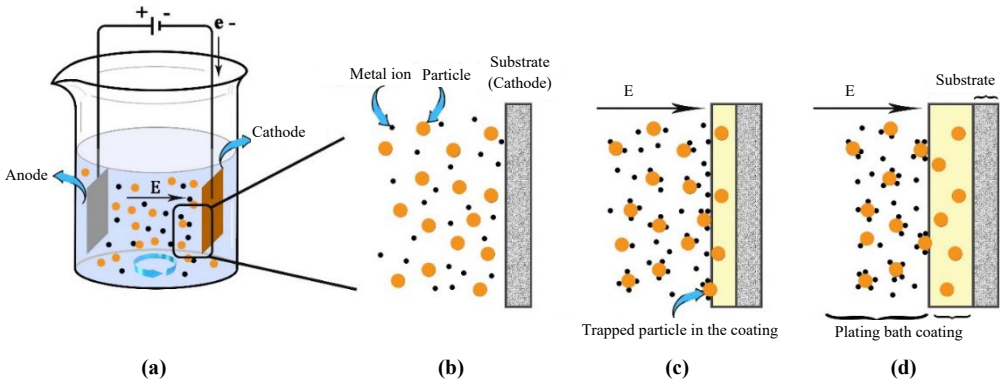

Figure 1. Schematic mechanism of co-deposition of the particle into metal coating by electroplating methods: (a) schematic of electrodeposition set up, (b) initial state of electroplating bath, and (c) attachment of metal ions to the particles by electric field force, d) particle-ion migration toward the cathode.

3. Microstructure of graphene containing composite coatingTable

Graphene-based additives led to a finer crystalline size, changes in phase structure and the morphology to needle shape, pinecone-like or rough surface for metal or metal alloy coatings as shown in Figures 2-4 [90-92]. The mechanism of crystalline size reduction is based on the graphene acting as a nucleation site as well as facilitating the formation of specific crystalline planes, which results in grain refining and heterogeneous growth. The addition of graphene oxide to cobalt (Co) coatings using the electrodeposition technique shows that GO affects morphology, phase structure, and the average grain size of the electrodeposit. The GO nanosheets dispersed in the composite coating change the morphology of the coating from a conical shape to a protruding structure, as well as refining crystalline size from 50 nm for pristine cobalt to 20 nm for a GO/cobalt nanocomposite [93]. The presence of graphene in the coating resulted in the reduction of the cobalt crystallite size, and the pyramidal morphology of the pure Co coating became smaller at low graphene concentrations and changed to a needle shape at the high concentrations [94]. Also, the incorporation of GO led to a refinement of the Zn crystallites in the coating matrix [95].

By using a combination of Ni pre-deposition and an elevated current assistant approach, pinecone-like micro/nanostructures of rGO/Ni composite coating were deposited successfully on a stainless-steel substrate. This structure can improve the hydrophobicity of the coating which has led to enhanced anti-corrosion properties [96]. The addition of a proper amount of rGO provides a large number of nucleation sites, which accelerate the formation of heterogeneous microstructure [97].

The influence of sodium dodecyl sulfate (SDS) usage as a dispersant in the electrochemical co-deposition of nickel shows that when the surfactant concentration in the electrolyte is increased, good graphene dispersion, coarser surface morphology, and reduction in grain sizes are achieved. The microhardness of coatings, adhesive strength, and anti-corrosion performance are also found to increase with the increasing SDS concentration [99]. The bath temperature during the electrodeposition process affects the surface morphology of the G/Ni composite coating (Figure 5). The results show that by increasing the deposition temperature, the average roughness (R_a nm) will increase, as well as the carbon content in the coating [79].

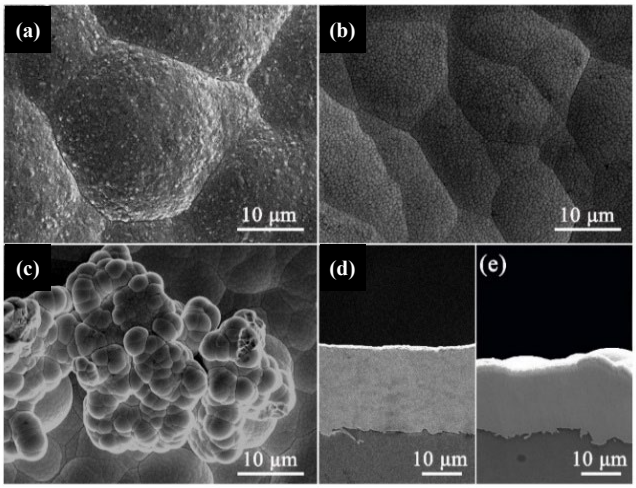

Figure 2. SEM images of the surface morphologies of (a) pure Ni, (b) Ni-B, and (c) Ni-B-GO. The cross sections of (d) Ni-B and (e) Ni-B-GO [90].

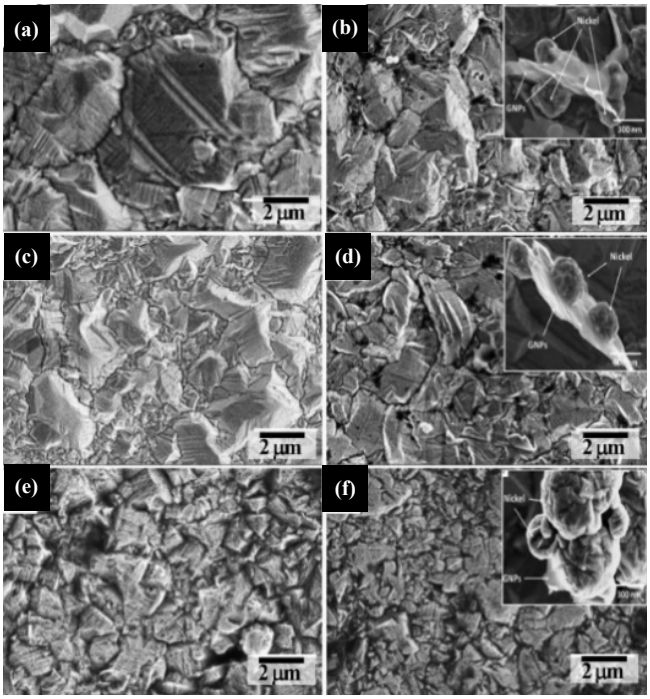

Figure 3. FESEM images of the surface coatings (a) Ni, (b) Ni/GNPs1, (c) Ni/GNPs2, (d) Ni/GNPs3, (e) Ni/GNPs4, and (f) Ni/GNPs5 [98].

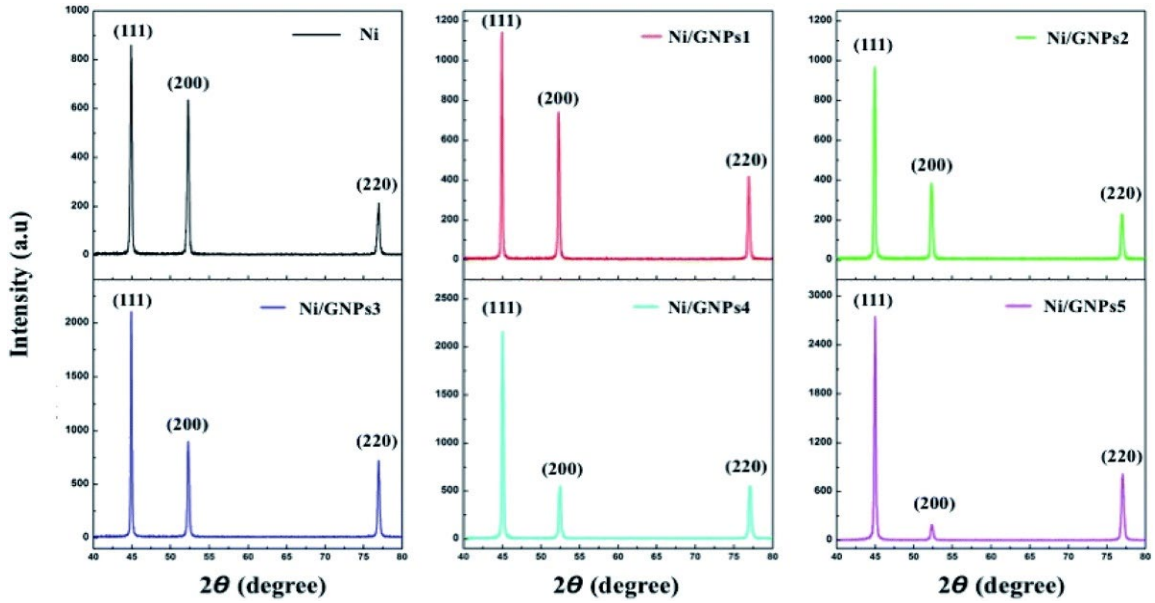

Figure 4. XRD pattern of nickel and graphene/nickel nanocomposite [98].

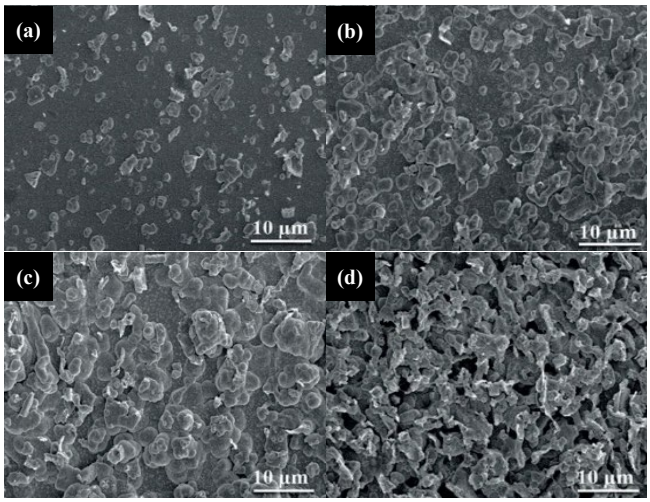

Figure 5. SEM images for the surface morphologies of Ni–graphene composite coatings prepared at deposition temperatures (a) 15°C, b) 30°C, c) 45°C, and d) 60°C [79].

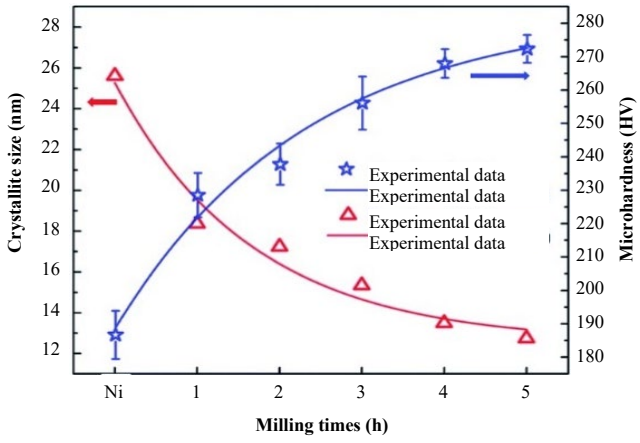

Figure 6. Microhardness and crystallite size of electrodeposited coatings Ni and Ni/GNPs1, Ni/GNPs2, Ni/GNPs3, Ni/GNPs4, Ni/GNPs5 corresponding to the milled graphene at 1 h, 2 h, 3 h, 4 h, and 5 h [98].

4. Mechanical properties

Enhancement of the hardness of graphene-bashed nanocomposite coatings is the result of the excellent mechanical properties of graphene as well as refinement of crystalline size, which is based on increasing the number of heterogenous nucleation sites and possibility of formation of specific crystalline planes itself. According to the Hall-Petch equation, there is a linear relation between hardness and grain size, which is

$$H = H_0 + k.d^{-1/2} \quad (7)$$

where H_0 is the hardness constant, k is a constant (Hall–Petch slope), and d is the average grain diameter [27], [100], [101]. Also, the graphene sheets make a compact interfacial bonding to the matrix and act as a net within the coating. When the indenter penetrates into the composite films, graphene sheets carry the load and hinder the movement of dislocations [102-104]. The nanomechanical analysis of the Ni/Graphene composites shows the improvement of the hardness from 1.81 GPa to 6.85 GPa, and elastic modulus from 166.70 GPa to 252.76 GPa compared to the pure Ni electrodeposits [92].

The size of the graphene nanoplatelets (GNPs) affects the microstructure and hardness of the electrodeposited nickel-graphene nanocomposite coatings (Figure 6). The experimental findings reveal that reducing the size, increasing the surface area, and enhancing the ability of GNPs to disperse led to changes in the microstructure and the hardness of the nanocomposite coatings. For nanocomposite coatings containing graphene with a size of 180 nm, hardness increased by up to 47% compared to pristine Ni coating. The highest microhardness achieved was calculated to be 273 HV. This hardness enhancement is due to the uniform dispersion in the Ni matrix of the small GNP sizes and the reduction in grain size using smaller GNPs [98]. The Vickers micro-hardness ($HV_{0.2}$) of G-Ni composite coating increases with increasing the amount of graphene. This increase in hardness can be due to the finer grain size of Ni/Graphene composite compared to

Ni coating, which causes hindrance to the movement of the dislocations and led to plastic flow resistance and enhanced hardness. The high mechanical strength of graphene may have played a role in the hardness improvement in the composite coating [105]. Furthermore, the mechanical properties of the Ni matrix improved based on the formation of Ni crystalline in its plane (111), facilitated by graphene [106]. Z. Ren *et al* [102] reported an elastic modulus as large as 240 GPa with hardness as large as 4.6 GPa with the addition of G as low as 0.05 g·L^{-1}.

The electrodeposited GO/Copper coating shows a maximum hardness of 3.32 GPa and the ultimate tensile strength of the composite coating was increased by 22.8% [107]. The Ni–W–TiO_2–Graphene Oxide co-deposition with excellent mechanical properties and high wear resistance was produced using ultrasonic-assisted pulse electro-deposition. The nanomechanical test results indicated a maximum improvement in hardness and elastic modulus of 8.1 GPa and ~209 GPa respectively, with the addition of graphene oxide and TiO_2 [108]. As part of this review, previous work done in other studies was analyzed and compared. Table 2 is an overview of electroplating conditions used and microhardness results.

Electrochemical deposition of Ni and Ni/graphene coatings on the textured surface of aluminum alloy, with concentrations of 0, 0.5, 1, and 1.5 mg graphene, show that friction and wear properties of the textured coating with the 1.5 mg graphene content had improved [4]. A Ni-graphene coating synthesized by electrodeposition in the presence of a surfactant demonstrated uniform graphene dispersion in the Ni matrix. The elastic modulus of the coating reached 240 GPa and the hardness reached 4.6 GPa with the addition of 0.05 g·L^{-1} graphene to the plating bath, which are 1.7 times and 1.2 times the pure nickel deposited under the same condition, respectively. The formation of Ni crystalline in the plane (111) (Figure 4) resulted in the enhancement of mechanical properties of the Ni matrix [102]. Nickel–graphene composite coatings fabricated by the pulse electro-deposition technique show enhanced properties (microhardness, tribological) dependent on the graphene concentration in the electrolyte. The increased graphene content in the electrolyte resulted in a significant increase in microhardness and wear resistance, as well as a decrease in the coefficient of friction (COF) [109].

5. Tribological properties

Graphene-based nanofillers enhance the hardness of composite coatings and based on Archard's law, the wear rate is inversely proportional to the hardness. Also, the severe adhesive wear mechanism for pure metallic coatings changes to abrasive wear and slight adhesive wear in graphene reinforced metallic composites. Graphene is a self-lubricating material. The uniformly distributed nanosheets in the composite coating lead to the formation of a tribolayer and a significant decrease of the friction coefficient [101], [115], [116].

The Nickel-GO nanocomposite coatings on the SS440 samples were prepared using pulse electrodeposition (PED). The tribological results revealed that the GO particles improved the friction and wear resistance through the formation of a tribolayer at the contact interface [117]. Studies on Ni-graphene composite coatings showed that COFs of the composite coatings were reduced by increasing the amount of graphene reinforcement which caused improvement in wear resistance properties (Figure 7) [105], [106]. According to the research done by J. Chen *et al* [105], the friction coefficient decreased with the increased amount of graphene. The friction coefficients shown by the coatings containing 0.1 g·L^{-1} and 0.2 g·L^{-1} graphene were higher than the COF of pure Ni coating (0.7), and for the coating having 0.3 g·l^{-1} graphene was nearly the same as pure Ni coating. Thus, the addition of graphene should be more than 0.3 g·L^{-1} to be effective in reducing friction. This can be due to the high Young's modulus of graphene hindering the formation of a lubricating film when the amount of graphene is less [57], [105].

Table 2. A summary on processing parameters and microhardness of graphene derivations reinforced metal matrix nanocomposite coatings.

No.	Coating	Current density	Temp /pH	Deposition time	Surfactant	Reinforcement material	Microhardness		Ref.
							Pristine coating	Composite	
1	Nickel	Direct 50 mA·cm^{-2}	45°C/ 3-4	60 min	SDS 0, 0.2, 0.4 g·L^{-1}	rGO 0.2 g·L^{-1}		500 HV	[98]
2	Nickel	Pulse 50 mA·cm^{-2}	45°C/ 4		SDS 0.2 g·L^{-1}	rGO 0.1, 0.25, 0.5 g·L^{-1}		427, 451, 492 HV	[108]
3	Nickel	Direct 50 mA·cm^{-2}	15, 30, 45, 60°C/ 3-4	60 min	SDS 0.4 g·L^{-1}	GO 0.2 g·L^{-1}		~220, 300, 500, 340 HV	[109]
4	Nickel	Direct 10 mA·cm^{-2}	40°C		SDS 0.2 g·L^{-1}	rGO 0.1 g·L^{-1}	287 HV	385 HV	[110]
5	Nickel	Direct 50 mA·cm^{-2}	55°C/ 4.0		N/A	GO 1 g·L^{-1}	1.81 GPa	6.85 GPa	[92]
6	Nickel	Direct 1.5-40 mA·cm^{-2}	50°C/ NA	1-4 h	SDS 0.5 g·L^{-1}	rGO 0.05 g·L^{-1}	3.83 GPa	4.6 GPa	[101]
7	Nickel	Direct 25 mA·cm^{-2}	45°C/ 4-5	90 min	SDS 0.1 g·L^{-1}	GNPs 0.3 g·L^{-1}	186 HV	273 HV	[91]
8	Nickel	Direct 10 mA·cm^{-2}	50-80°C/ NA	2, 4, 6, 8 h	SDBS 0.05 g·L^{-1}	GO 0.1, 0.2, 0.3, 0.4 g·L^{-1}		~207, 210, 218, 223 HV	[104]
9	Co–Ni-P	Pulse 20 mA·cm^{-2}	45°C/ 3	70 min		GO 0.2 g·L^{-1}	450 (HV)	600 (HV)	[111]
10	Nickel		90°C/ 6	5 min		rGO 0.2 g·L^{-1}	6.45 ± 0.39 GPa	8.09 ± 0.65 GPa	[112]
11	Ni–Zn	DC 10 mA·cm^{-2}	30°C /2.5	20 min		GO 0.1 g·L^{-1}	105.3 (HV)	124.8 (HV)	[113]
12	Ni–Zn	DC 10 mA·cm^{-2}	30°C /2.5	20 min		rGO 0.1 g·L^{-1}	105.3 (HV)	127.5 (HV)	[113]

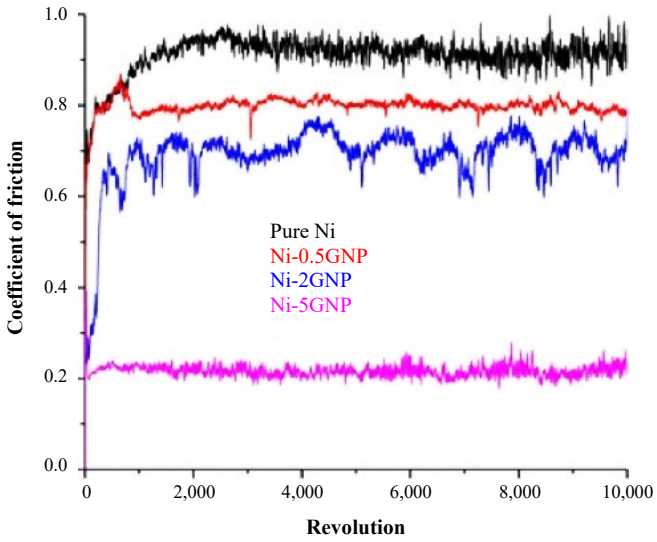

Figure 7. Coefficient of friction plots for (a) pure Ni and Ni-CNT composites, and (b) pure Ni and Ni-GNP composites [118].

Tribological analysis of electrodeposited pure cobalt and cobalt/graphene composite coatings on St37 steel substrates revealed that the co-deposition of graphene particles decreased the volume loss and the friction coefficient of a pure cobalt coating [119]. The GO led to the formation of hcp (1 0 0), (0 0 2), and (1 0 1) textures, which resulted in the functions of grain refining and hardness enhancement. Furthermore, the composite coating showed better corrosion and wear resistance along with a lower coefficient of friction (COF) [93].

Reduced graphene oxide/copper coatings can successfully be electrodeposited from a surfactant-free colloidal solution. Tribological tests showed that a coating with a 0.27 wt% amount of rGO exhibits a low COF and a specific wear rate 10 times to 18 times lower than that of pure copper coatings. This could be due to the formation of a compacted and stable tribolayer consisting of rGO sheets and copper oxides on the worn surface, as a similar phenomenon was seen in the case of nickel/graphene composite coatings. Indeed, the mentioned tribolayer acts as a lubricant and a barrier at the interface of the friction pairs to decrease the direct contact area [109],[120].

The addition of graphene to the Ni matrix can minimize wear while providing low friction, as shown by Figure 8. The low surface energy of the Ni–graphene surface, which decreases the adhesive forces between the wear particles and the surface, is responsible for this behavior. The reduced adhesive forces result in a reduction in wear particle size and agglomeration [121].

6. Anti-corrosion properties

Graphene is an excellent additive candidate to fabricate super-hydrophobic coatings [96,122-127]. Graphene also refines the grain size of the coating and this smaller grain size can provide a smaller cathode/anode surface ratio which is effective against localized corrosion [128,129]. The combined properties of super-hydrophobicity and smaller cathode/anode surface ratio significantly improves overall resistance to corrosion.

Different research has been done on electrodeposited Ni/graphene, Ni/rGO, and Ni/ GO composite coatings to investigate the corrosion performance [130-133]. The composite coating of Ni/graphene on a mild steel substrate showed better anti-corrosion behavior compared to pure Ni coating [111]. Ding *et al* [134] fabricated a superhydrophobic nickel/graphene hybrid film with improved corrosion resistance on mild steel. Superhydrophobicity behavior (water contact angle 162.7°±0.8, sliding angle (SA) of 2.5°±1.0), was shown by the reduced-graphene oxide/Ni composite coating with pinecone-like micro/nano-structures on a stainless steel substrate. As indicated in Figure 9, the coatings exhibited good anti-corrosion performance in 3.5 wt% NaCl solution, with 99.98% inhibition efficiency [96].

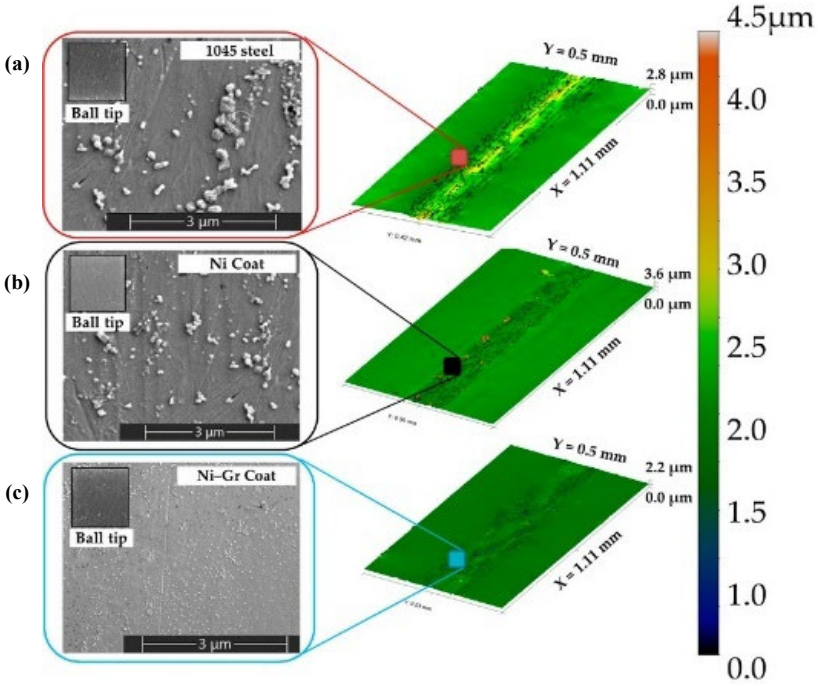

Figure 8. SEM images of the wear particles around the wear track of (a) 1045 steel, (b) Ni coating, and (c) Ni–Gr coating [121].

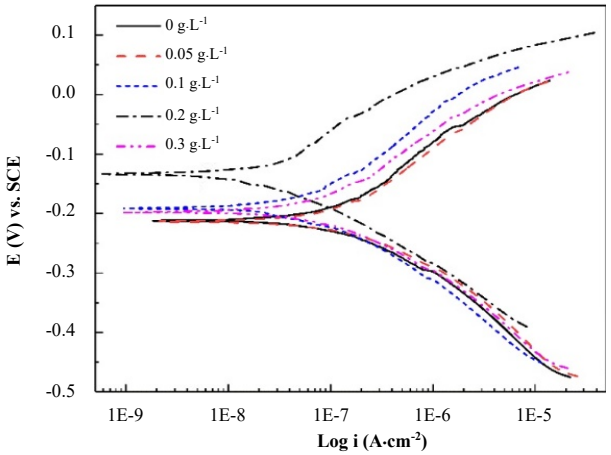

Figure 9. Potentiodynamic polarization curves of coatings obtained at GO concentrations of 0, 0.05, 0.1, 0.2, and 0.3 g·L^{1} at a scan rate of 0.5 mV·s^{-1} in 3.5 wt% NaCl solution [135].

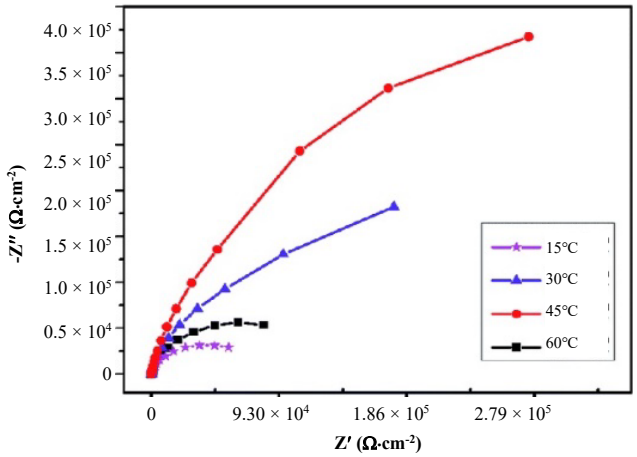

Figure 10. Effect of deposition temperature on the impedance spectra of composite coatings in 3.5% NaCl solution [79].

Table 3. A summary on corrosion behavior of metal matrix nanocomposite coatings reinforced by graphene derivatives.

No.	Matrix	Reinforcement material	Current density	Temp / pH	Deposition time	Surfactant	Microhardness			Ref.
							E_{corr} (mV)	I_{corr} (μA·cm^{-2})	Corrosion rate (mm/year)	
1	Nickel	GO 0.125-1.500 g·L^{-1}	DC 30 mA·cm^{-2}	45°C/ 2.5	60 min	SLS 1 g·L^{-1}	-424 – -419	4.845- 2.883		[139]
2	Nickel	Graphene 0.5 g·L^{-1}	DC 40 mA·cm^{-2}	45°C /4.0	30 min	fluorinated surfactant (16 cm^{3}·dm^{-3})	-225	0.66	8.12	[140]
3	Nickel	Graphene 0.5 g·L^{-1}	DC 40 mA·cm^{-2}	45°C/4.0	30 min	[3-(heptadekafluorineoctyl)-sulphonyl]-aminopropyl-trimethylammonium iodide (21 cm^{3}·dm^{-3})	-220	0.50	6.16	[140]
4	Cobalt	GO 0.2 g·L^{-1}	Pulse 40 mA·cm^{-2}	45°C/ 5.0	2 h		-359.7	3.04	1.56×10^{-2}	[128]
5	Nickel	GO 0.2 g·L^{-1}	Pulse 60 mA·cm^{-2} Frequency 50 (Hz)	50°C/ 3.6	30 min		-131	0.0224		[134]
6	Nickel	GO 0.2 g·L^{-1}	DC 90 mA·cm^{-2}	45°C/ 3-4		SDS 0.4 g·L^{-1}	130	0.2128		[141]
7	Tin	G 0.05 g·L^{-1}	DC 6.5 mA·cm^{-2}	25°C/ 3.5	20 min		-537	0.815	0.896±0.056 μg h^{-1}	[142]
8	Zinc	G 0.05 g·L^{-1}	DC 40 mA·cm^{-2}	25°C/ 3.5	15 min		920	6.82	8.32 μg h^{-1}	[143]
9	Co–Ni-P	GO 0.2 g·L^{-1}	Pulse 20	45°C/ 3	70 min		-352.5	3.05		[111]
10	Nickel	rGO 0.2 g·L^{-1}	mA·cm^{-2}	90°C/ 6	5 min		-442	0.68	0.0078	[112]
11	Ni–Zn	GO 0.2 g·L^{-1}	DC 10 mA·cm^{-2}	30°C/2.5	20 min		42.9	38.2	0.0935	[113]
12	Ni–Zn	rGO 0.2 g·L^{-1}	DC 10 mA·cm^{-2}	30°C/2.5	20 min		-12.3	33.8	0.0629	[113]
13	Zn-Ni-Fe	G 0.075 g·L^{-1}	DC 10 mA·cm^{-2}	43±2°C/ 3.2	45 min		-1100	4.134	5.043 μg h^{-1}	[81]
14	Nickel	G 1 g·L^{-1}	DC 40 mA·cm^{-2}	45°C/4.0	30 min	anionic high fluorinated surfactant (SPA)	-188	0.12	1.48	[144]
15	Nickel	G 1.5 g·L^{-1}	DC 137.5 mA·cm^{-2}	40°C/ 3-4	5 min	SDBS 0.015 g·L^{-1}	-601.69	397.18		[4]
16	Nickel	rGO 0.6 g·L^{-1}	DC 80 mA·cm^{-2}	25°C/	5 min		-184.21	0.20		[145]
17	Ni- Co	G Platelet 0.05 g·L^{-1}	DC 20 mA·cm^{-2}	40°C/ 4			-512	100.1		[146]

The electrodeposition of Ni/graphene composite coatings on carbon steel at various deposition temperatures indicated significant changes in surface morphology, compositions, roughness, phase structures, and the electrochemical properties of the coatings (Figure 10). The results demonstrated deposition at 45°C exhibited refined grain sizes, high microhardness, and better corrosion resistance performance [79].

Other metal or metal alloy matrices are of great interest for electrodeposited composite coatings with graphene, rGO, and GO additives [81,136,137]. Results showed that Zn-GO composite coatings have higher corrosion resistance compared to the pristine Zn coatings. Also, with the increase in the volume fraction of the GO in the composite coatings, the corrosion rate decreased [95]. Moshgi Asl *et al* [138] fabricated a reduced graphene oxide/zinc (rGO-Zn) nanocomposite coating deposited on a steel substrate by using pulse-potential co-electrodeposition. The results showed that the corrosion rate decreased from 0.034 mm/year to 1.62×10^{-4} mm/year. The investigation of electrodeposited Ni-Co and Ni-Co/rGO coatings on Q235 carbon steel substrate showed that incorporation of rGO nanoplatelets has improved corrosion resistance by 17.5 $Rct{\cdot}k^{-1}\Omega^{-1}cm^{-2}$ compared to 4.55 $Rct{\cdot}k^{-1}\Omega^{-1}cm^{-2}$ for Ni-Co coatings. rGO platelets facilitated the formation of a large number of grain boundaries with less crystal growth and provided surface roughness. Lastly, rGO platelets filled the cavities or pores, which subsequently contributed to improved mechanical and corrosion properties [139]. Table 3 provides an overview of electroplating conditions used and anti-corrosion results.

7. Challenges or perspectives

The following lists a few areas of research that should be further explored in order to improve our understanding of the electrodeposited graphene-based nanocomposites:

The effects of reinforcement additives in the plating bath and on the nanostructure of coatings need to be further investigated.

As electric current influences the microstructure of coatings, its impact on morphology, mechanical, and anti-corrosion properties should be explored.

The pH of the plating bath is a vital factor that influences the properties of the produced coating which additional research is needed to investigate.

There is a lack of research on the co-deposition of graphene-based additive into metal matrix coating.

8. Conclusion

Current developments in the synthesis of graphene-based metal nanocomposite coatings, with a special focus on the electrodeposition approach and properties of these coatings, were discussed in detail in this review. Graphene-based reinforcements have been incorporated in the metallic matrices using electrodeposition techniques, based on electrophoresis migration of charged particles. It has been shown that using smaller particles leads to high-speed co-deposition. Furthermore, smaller size and higher amount of graphene-based reinforcement refine the crystalline size and growth of the preferred crystalline plane (111) in the metallic coating. The refinement of crystalline size in addition to the excellent mechanical properties of graphene yields a significant enhancement of the hardness of the coating. Literature showed that the incorporation of graphene impacts the tribological properties of the electrodeposited coating too. Additionally, graphene changes the severe adhesive wear mechanism of metallic coatings to abrasive wear and slight adhesive wear, and also the graphene forms a tribolayer and significantly decreases the friction coefficient. In terms of corrosion characteristics, due to the improvement in superhydrophobicity of the coatings, as well as a smaller cathode/anode surface ratio against localized corrosion, corrosion resistance increases considerably with the addition of graphene particles.

Although the electrodeposition approach can yield defect-free, low cost, and adjustable graphene-based metal nanocomposite coatings, they have not been studied thoroughly and more research is needed to make this process suitable for industrial application. In particular, extensive research is required to understand the co-deposition mechanism and interactions between the metals and the graphene surface, which will have a direct impact on the properties of these coatings such as anti-corrosion, and anti-wear characteristics. Although several methods have been applied to homogeneously electrodeposit the dispersed nanocomposites using various surfactant and functionalization techniques, further efforts must also be directed towards the prevention of restacking graphene and the improvement of the dispersion quality of graphene-based metallic coatings. Furthermore, in the case of biomedical applications, it is critical to understand the biocompatibility and toxicity of these functionalizations and surfactants to make the resulting nanocomposites safe to use.

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