

# Recent Progress in the Application of Nickel/Nickel Oxide Nanoparticles/Nanocomposites as Corrosion Inhibitors

K. Okon<sup>\*1,2</sup>, C. O. Akalezi<sup>1</sup>, C. A. Maduabuchi<sup>1</sup>, I. B. Onyeachu<sup>1</sup>, T. O. Azeez<sup>1</sup>, S. Nwanonyi<sup>1</sup>, I. C. Ekeke<sup>1</sup>, D. Njoku<sup>1</sup> and I. I. Ayogu<sup>1</sup>

<sup>1</sup>*Africa Centre of Excellence in Future Energies and Electrochemical Systems, Federal University of Technology, Owerri, Imo State, Nigeria*

<sup>2</sup>*Department of Materials and Metallurgical Engineering, Federal University of Technology, Owerri, Imo State, Nigeria*

Corresponding authors: koofsc01@gmail.com; chrisakalezi@yahoo.com

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## Abstract

This review paper examines the potential of using NPNi/NPNiO and NC as CI for MS and other ferrous alloys in a variety of aggressive corrosive media. A survey of previous literature revealed that higher Ct of NPNi has significantly reduced CR and has increased corrosion IE(%) for different ferrous alloys in various corrosive media. These CI effectively protect the substrate, even at low Ct, by forming a dense passive film which adheres to the metal surface and inhibits anodic or cathodic reactions that cause metal corrosion. Their enhanced corrosion protection ability is due to the presence of more available active sites for adsorption of destructive ions, caused by their high surface area to volume ratio. NPNi are very stable, even at higher T and in aggressive salt environments, due to their large surface area, high hardness and excellent thermal stability. These CI have a unique advantage over other traditional organic and green inhibitors, because they are not easily washed away by corrosive fluids during industrial operations, and are chemically stable under harsh conditions and elevated T. Different methods for synthesis and characterization of NPNi, as well as environmental and safety considerations associated with the use of these CI, are also herein discussed.

**Keywords:** corrosion; CI; ferrous alloys; Ni; NiO; NP.

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## Introduction\*

Corrosion is the deterioration or gradual removal of the metallic surface of a material, caused by chemical and/or electrochemical interaction with its environment, which leads to the metal conversion into a chemically stable oxide [1]. Corrosion has become a huge problem of global concern, due to the economic losses and environmental degradation it causes. It has been estimated that each country loses about 2 to 4% of its GDP each year, due to corrosion, and approximately one-third of the generated metal scrap in the world every year is a direct consequence of metal

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\*The abbreviations list is in pages 278-280.

corrosion [2]. The cost incurred in the fight against corrosion in third world countries is 10 times more than the above estimation [3]. In a 2013 survey, it was estimated that an outrageous sum of 2.5 trillion U.S. dollars, roughly about 3.4% of the global GDP, was lost due to corrosion alone, excluding costs associated with human safety and environmental protection [1]. Corrosion causes severe damage to metallic components such as automobile and machine parts, pipelines, and others, when exposed to moisture (rain water) and air [4]. In fact, the failure of several metallic structures or systems which has become rampant in recent years, with attendant consequences such as plane crash, shipwreck, equipment deformation, storage tanks failure, explosions, collapse of civil structures and release of toxic products leading to pollution of food and water ingested by men, plants and animals, can be directly attributed to metal corrosion. Corrosion poses a serious economic and environmental challenge to several industrial applications, such as water treatment, acid pickling, chemical cleaning and other operations in the oil and gas industry, due to exposure of materials and equipment to acidic, neutral and alkaline corrosive media [5]. Combined economic losses attributed to metal corrosion on a global scale are six times higher than total damages caused by natural disasters such as earthquakes, floods and typhoons [6].

Thus, metal corrosion prevention, mitigation and protection strategies are of great significance to ensure reliability and integrity of engineering facilities and infrastructure. Several techniques such as the use of protective coatings and linings, cathodic/anodic protection, alloying and CI have been adopted successfully for the prevention, control and mitigation of corrosion [3, 7, 8]. The use of CI is the most viable alternative to reduce corrosion damage, and also the preferred technique among researchers, owing to its low cost, availability from a variety of sources, easy application and very high degree of effectiveness [9]. A unique advantage of CI is that they can be easily implemented *in situ*, often without disrupting metallic applications, and can be used to control a wide range of corrosion processes in oil, gas, chemical, petrochemical, heavy industrial manufacturing steel, coatings and product additive industries, as well as in water treatment facilities and mining [10].

Some traditional chemical and organic inhibitors usually dissolve in water and are carried away by corrosive fluids during use in industrial applications, while others are not thermally stable at high T. Therefore, the need for chemically and thermally stable CI that can withstand harsh operating conditions, such as elevated T, has exponentially increased the demand for metal oxides and their NP/NC, since they possess the aforementioned characteristics.

NPNiO and NC not only are resistant to corrosive chemicals, but they also possess the advantages of high hardness, excellent thermal stability, good electrical conductivity, low solubility, enhanced catalytic activity, self-healing properties, effectiveness, adjustable particle size and surface composition, longevity, versatility, cost effectiveness and synergistic effects with other existing inhibitors.

In metallic NC coating studies, Ni has acquired a great deal of attention, because of its ability to treat the host matrix for electroless NPNiO coating. Ni coating has

excellent corrosion and wear resistance in a number of commercial applications [11], since it acts as a barrier for special metals, to prevent unfavorable elements from migrating and diffusing among alloys, which may hinder the development of galvanic corrosion. Because of its prominent adhesive power and ductility, Ni layers are also used as a protecting coating and ferrite cloth in airframes [12].

However, challenges of dispersion and agglomeration are associated with the use of NPNiO. To address this challenge, NPNiO are usually embedded within epoxy resin coatings. Epoxy resins are renowned for their excellent adhesion, mechanical properties and chemical resistance, making them ideal matrices for hosting CI [13-16]. Epoxy resins serve as very good CI due to their electro-donors and electro-attractors sites, providing better surface coverage than simple organic CI [14]. In a study by [17], a novel epoxy resin, pentaglycidyl ether pentaphenoxy of phosphorus, has exhibited excellent CI of CS in 0.5 M H<sub>2</sub>SO<sub>4</sub>, with maximum IE(%) of 94.23 and 94.96%, at 10<sup>-3</sup> M, for PDP and PEIS, respectively.

Evaluation of ERH and ERO, as potential CI for CS in 1 M HCl medium, has revealed that both of them were mixed type inhibitors. Their IE(%) in a 1 M HCl solution was due to heteroatoms (O, N), ring compounds and oxiran groups, which donated electrons to the metallic surface. Findings from the study showed that corrosion IE(%) of ERO (94.1%) was higher than that of ERH (89.7%) in a HCl medium at room T.[18].

Two epoxy resins, diglycidyl ether ethylene and diglycidyl ether benzene have proven to be effective CI for C38 steel in 1 M HCl. Findings from the study have showed that these CI act as mixed-type inhibitors, delaying the rate of anodic and cathodic reactions. Their adsorption isotherms are Langmuir's type, and primary mechanisms involve physical adsorption [19].

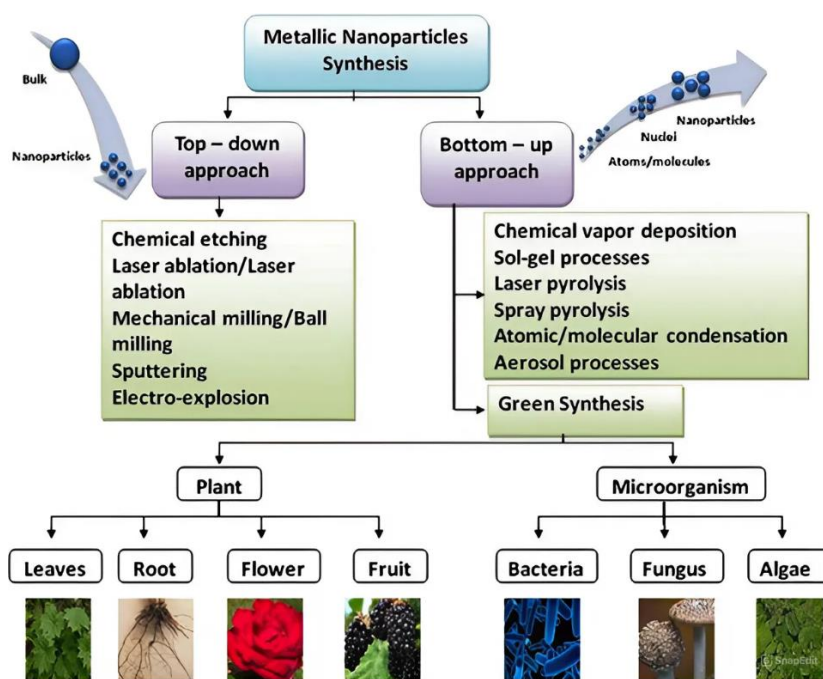
By incorporating metal oxide NP into epoxy resin, synergy may be achieved, resulting in enhanced corrosion resistance, barrier properties and longevity in harsh operating environments, since they block microcracks usually present in epoxy resin [20].

Most published works on NPNiO have focused on the synthesis and characterization of NPNi and NC, studying their photocatalytic, optical and antimicrobial activity, for applications in catalysis, sensors, biomedical remediation of polluted wastewater and biomedicine. This review has focused on exploring the potentials of NPNiO as effective CI for ferrous metals and alloys in different aggressive corrosive media. The methods of synthesis and characterization of NPNi are herein also discussed. Furthermore, the CI mechanism for NPNiO is presented, and recent progress in their use is discussed in detail. Environmental impacts of NPNiO, their disposal and safety concerns associated with their use are also herein highlighted.

## NPNiO synthesis and characterization methods

NP from different metal oxides with particle sizes within the range from 1 to 100 nm [21] are generally synthesized by two major approaches: top-down and bottom-up, as shown in Fig. 1.

Top-down approach involves breaking down a block of a bulk material to get nanosized particles, while bottom-up method involves synthesizing NP by joining up atoms, molecules or clusters [22]. Top-down approach includes synthesis techniques such as mechanical and ball milling, thermal decomposition, laser ablation, chemical etching, sputtering, plasma processing, robust catalytic reactions and lithography. Examples of bottom-up approach methods for synthesis of NPNi include sol-gel, spinning fabrication, chemical vapor deposition, pyrolysis and biosynthesis techniques [23].



**Figure 1:** Top-down and bottom-up approaches for NP synthesis [24].

Table 1 gives a summary of some reported synthesis techniques successfully used for the production of NPNi by various researchers.

**Table 1:** Summary of synthesis and characterization methods for NPNi.

S/N	NP	Technique	Experimental conditions	Characterization techniques	Shape/ size	Ref.
1	NPNiO powder	Green synthesis using <i>Acacia N.</i> bark extract	60-80 °C; drying at 120 °C for 6 h; calcination at 500 °C for 5 h	XRD, SEM, EDAX, IR, UV-vis, PLS, FTIR	Cubic cluster-like structure	[25]
2	Ni	Green synthesis using <i>Calotropis G.</i> leaves	80 °C; pH: 12.0; 90 min	UV-vis, XRD, SEM, FTIR	60 nm	[26]

3	Ni	Biosynthesis using <i>Desmodium G.</i> roots extract	80 °C; 45 min	UV-vis, XRD, FTIR, ZETA potential, magnetometer	Pellet	[27]
4	Cu-NPNi	Mechanical milling	45 °C; 20 h	XRD, TEM, VSM, TGA-SDTA	10 nm	[28]
5	NPNi	Chemical	45 °C; 15 min; drying for 15 days; thermal decomposition at 800 °C for 24 h	XRD, SEM, TGA, IR-spectroscopy	24 nm, monoclinic	[29]
6	NPNi	Chemical reduction	80 °C for 3 h	XRD, SEM, EDS	10-15 nm	[30]
7	NPNi	Hydrazine reduction	Room T for 2 h	XRD, SEM, VSM	Pure FCC structure metallic Ni	[31]
8	NPNiFe <sub>2</sub> O <sub>4</sub>	Sol-gel using PAA as chelating agent	300 °C for 2 h	TGA, DTA, XRD, TEM, magnetometer,	5-30 nm	[32]
9	NPNi	Pulse laser ablation method at low energy Nd:YAG 1064 nm.	20 min; surface plasma resonance: 290 nm	SEM-EDX, UV-vis	Spherical shape of average diameter of 25 nm and standard deviation of 3 nm.	[33]
10	NPNi	Laser-assisted synthesis	40 °C for 10 min; calcination: 450 °C for 2 h; drying: 40 °C for 5 min	UV-vis, DLS, TEM, EDS, XPS	Colloid of spherical, almost purely metallic NPNi	[34]
11	NPNi/NPNiO	Chemical deposition vapor	500 °C	TEM, HR-TEM, SAED, EDS	~10 to ~50 nm	[35]
12	Ni and NPNiO		-----	XRD, TEM, VSM-SQUID magnetometer, BET	Hexagonal/spheroidal to cubic/truncated cubic.	[36]
13	Ni	Non-thermal plasma synthesis	Integration time for spectra measurement: 500 ms/nm <sup>-1</sup> . Total reactor pressure: 20 Torr	SEM-EDS, TEM, SAED	Average size of 3.3-10.9 nm	[37]
14	Hcp particles Ni	Chemical synthesis	120 °C for 30 min	XRD, TEM, FTIR, SQUID magnetometer,	20-50 nm	[38]
15	NPNi dispersed in imidazolium ionic liquids	Simple decomposition of organometallic precursor dissolved in ionic liquids	75-100 °C for 30 min	TEM, XAS, SAXS	Cubic fcc NPNi with 4.9 nm- 5.9 nm in mean diameter and monomodal size-distribution	[39]
16	NPNi	Reduction of Ni chloride with hydrazine in an aqueous solution of cationic surfactants	60 °C for 1 h	XRD, TEM, magnetic measurement	10-36 nm	[40]
17	NPNi	Thermal decomposition	215-240 °C for 45 min	XRD, TEM, magnetic measurement	Near-spherical; 10- 50 nm.	[41]
18	NPNi	Reduction	100-180 °C for 30 min	XRD, TEM, SAED, SQUID magnetometry	Monodisperse; 3 to 11 nm	[42]
19	NPNi	Wet chemical approaches	28 °C; 12-24 h	XRD, FE-SEM, UV-vis	31 nm to 150 nm	[43]
20	NPNi	Laser-driven decomposition of Ni carbonyl	300-350 °C for 1 h	XRD, TEM, BET, XPS	5 to 50 nm	[44]
21	NPNi	Reduction of Ni acetylacetonate in a monosurfactant system	70-500 °C	XRD, TEM, magnetic measurements	10 to 20 nm	[45]
22	NPNi	Reduction of Ni nitrate hexahydrate with vegetable oil	170 °C; calcination at 450 °C for 20 min	XRD, SEM	5 to 58 nm	[46]
23	NPNi	Gamma irradiation	80°C for 2 h	TEM, STEM, XPS, FTIR	9 to 16 nm	[47]

24	NPNiO	Green synthesis	Room T for 2 h; calcination at 400 °C for 2 h	UV-vis, XRD, FTIR, SEM	14 to 18 nm	[48]
25	NPNi	Hydrazine reduction	Reaction T: 120 °C	XRD, FTIR, STEM, TEM	9.2 ± 2.9 to 5.4 ± 0.9 nm at higher T and NaOH Ct	[49]
26	NPNi	Wet chemical solution route	50-70 °C for 24 h	SEM, XRD	80 nm to 400 nm	[50]
27	NPNi	Chemical reduction	80 °C for 24 h	FTIR, PXRD, UV-vis, EDS, SEM, TEM, TG/DTG	30 nm	[51]
28	Biogenic NPNi	Bio-reduction between CRPE and nitrate salt of Ni	25-45 °C for 90 min	UV-vis, DLS, FTIR, XRD, TEM, SEM, EDAX,	48.5 nm	[52]
29	NPNi	Homogeneous precipitation	80 °C for 1 h; calcination: 500°C for 2 h	NMR, TEM, DLS, FTIR, PXRD, ESI-MS, SEM, AFM	Rod-like shape; 35-50 nm	[53]
30	NPNiO thin films	Wet chemical precipitation/spray coating of NP of NiO	Coating chamber: 150 °C	UV-vis, micro Raman, PLS, FE-SEM, EDAX, AFM	-----	[54]
31	Nanocrystalline Ni foil	Electrodeposition	-----	SEM, AFM, XRD	Spherical; 20~30 nm	[55]
32	NPNiO	Microwave irradiation technique	25 °C for 24h; pH: 9.5; drying at 80 °C for 2h ; calcination at °C for 8h	FTIR, TEM, XRD, FE-SEM, EDX, SAED	43 nm	[56]
33	NPNiO	Green synthesis using <i>Eucalyptus G.</i> extract	70 °C; Drying at 100 °C for 24 h	UV-vis, FTIR, XRD, EDX	Pleomorphic; 10-20 nm	[57]
34	NPNi	Green synthesis using <i>Ocimum S.</i>	60 °C for 3h	UV-vis, FTIR, XRD, SEM, TEM,	12-36 nm	[58]
35	NPNi	Plant-mediated synthesis using alfalfa extract	4 h at 60 °C. After reaction, solution vacuum freeze- drying for 24 h	XRD, XPS, TEM, UV-vis, FTIR	sub-10 nm	[59]
36	Nanostructured NiO	Solvothermal synthesis	Thermal treatment: 400 °C for 1 h in air	XRD, FE-SEM, HR-TEM/SAED	7–15 nm	[60]

### CI studies involving NPNi

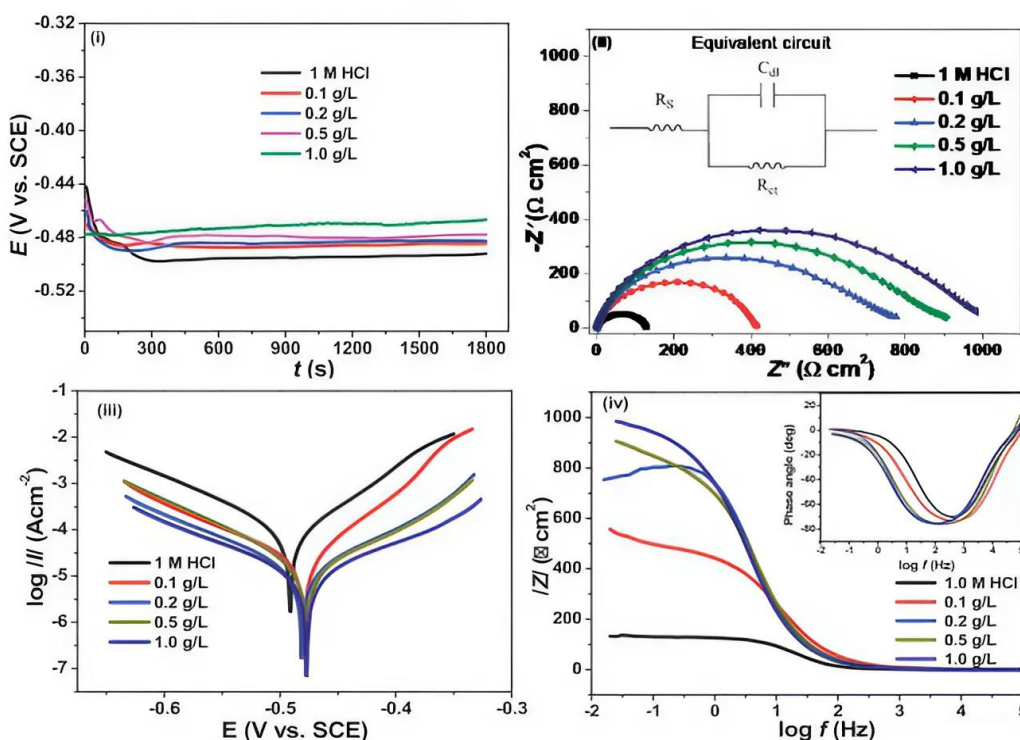
An overview of previous studies on the CI of CS and other ferrous alloys in different corrosive media by Ni /NPNiO/NC is presented in Table 2.

**Table 2:** Summary of previous studies involving the use of Ni /NPNiO/NC CI.

S/N	Inhibitor	Ct	Metal/medium	Technique/method	Max. IE(%)	Findings	Ref.
1	Green synthesized NPNiO	250 ppm	CS/0.5 M HCl	XRD, SEM, EDAX, IR, UV-VIS, WL, PDP, EIS	93.68%	PDP proved that a defensive film was shaped on the cathodic locales of CS surface. EIS affirmed that a protection layer was formed on the surface of the CS to protect it from corrosion	[25]
2	NC from NiO-CNT-SnO <sub>2</sub>	1:1:2	AISI 1020 steel/soil environment	HR-SEM, EDS, XRD, HR-TEM, SAED, XPS, WL, PDP, EIS	93.9%	1:1:2 NiO-CNTs-SO <sub>2</sub> had excellent corrosion resistance behavior. CR decreased from 0.0335 to 0.0022 mpy, showing better IE(%) of steel corrosion.	[61]
3	NiO	250 ppm	CS /1 N HCl.	Gravimetry	19.8%	With higher Ct, WL decreased and IE(%) increased	[62]
4	Onion mesocarp extract-NPNi composite	1000 ppm	X80 steel/ 1 M HCl	EDAX, FTIR, UV-vis, DLS, XRD, SEM, TEM, WL, OCP, PDP, EIS, LPR	88.7%	IE(%) improved with T increase from 303 to 323 K, was constant from 323 to 343 K and decreased from 343 to 363 K. NP physically adsorbed on X80 steel surface and acted as mixed type CI with influence on cathodic reaction involving HER	[63]

5	NiO-PANI composite	500 ppm	MS/ 1 M and 2 M H <sub>2</sub> SO <sub>4</sub>	FTIR, XRD, SEM, WL, OCP, PDP, EIS	88 and 82% for 1 and 2 M H <sub>2</sub> SO <sub>4</sub> , respectively	Effective protection up to eight h with slight changes in IE(%) was seen in WL method.	[64]
6	Epoxy/ZnO-NiO NC	-----	MS/3.5% NaCl solution	XRD, SEM/EDAX, PDP, EIS	-----	Anticorrosion (Epoxy/1.8 ZnO-NiO) coating with outstanding physical barrier qualities can be employed in corrosion protection field	[65]
7	Fe NiO nanopowder	70 ppm	Line pipe steel (API 5L X- 80)/ 3.5 wt.% NaCl.	WL, OCP, EIS	-----	Protection mechanism with adsorption of metallic cations on steel surface forming protective film. EIS spectra revealed that CI was at low Ct, and higher Ct of aqueous solution produced induction behavior.	[66]
8	NPNi	-----	Ni-Cr alloy/ 1 M H <sub>2</sub> SO <sub>4</sub> saturated with CO <sub>2</sub>	OCP, PDP, LPR	88.48%	Ni-Cr alloy was protected from corrosion in acidic media by a low-acidity bath coating layer. Surface morphologies showed that coatings at different acidic scales resisted acid attack due to their excellent adherence to Ni-Cr alloy surface	[67]
9	Nanofiber layer of ZnO-NiO-CuO/ polycaprolactone	-----	MS/1 M HCl	SEM, EDX, FTIR, OCP, PDP, EIS, LPR	94.8%	ZnO-NiO-CuO/PCL electrospinning nanofiber coating is a new metallic oxide NC coating for MS with excellent corrosion resistance	[68]
10	Nano Ni ferrite and magnetite double layer coatings	-----	CS	Laser Raman spectroscopy, XRD, XPS, SEM, TEM, EIS, PDP, Mott Schottky analysis	-----	NiFe <sub>2</sub> O <sub>4</sub> coating improved corrosion resistance compared to CS. CS/Fe <sub>3</sub> O <sub>4</sub> provided optimized thickness with lesser defects and better protection	[69]
11	Biogenic NPNi	3.12mg/L	X80 steel/ 1 M HCl	UV-vis, DLS, FTIR, XRD, TEM, SEM, EDAX, WL, OCP, EIS, PDP	87.3%	CRPE-NPNi efficiently adsorbed onto X80 steel and inhibited its corrosion in 1 M HCl with IE(%) of 87.3%. Had 3-log reduction in <i>Desulfovibrio</i> sp. population	[52]
12	p-MBDM and p- MBDM assembled on NPNiO	3.00g/L	MS/2 M HCl	NMR, TEM, DLS, FTIR, PXRD, ESI- MS, SEM, AFM, WL, EIS, Galvanostatic polarization	98.1%	IE(%) increased with higher inhibitor Ct. With higher T, IE(%) increased for p-MBDM assembled on NPNiO	[53]
13	Nanostructured thin films from NiO	-----	304L SS/ 3.5% NaCl	UV-vis, micro Raman, PLS, FE- SEM, EDAX, AFM OCP, EIS, Tafel analysis	-----	OCP, PDP and EIS revealed enhanced substrate protection by NiO thin film. Salt spray exposure confirmed that NiO films provided better CI of SS 304L than the one from the substrate without NP deposition.	[54]
14	NPNiO	-----	304L SS/5% NaCl	AFM, OCP, EIS, salt spray test, Tafel studies	-----	Polarization curves of coated 304L SS substrates showed positive shift in corrosion potential. Nyquist plot confirmed that coatings improved corrosion resistance of SS. Impedance variation with frequency of SS before and after salt spray test showed that NiO films were quite stable in salt.	[70]
15	NiO	-----	GTD111 superalloy (Ni base, Cr 14 wt%, Co 9.5 wt%)/gas turbine environment	XRD, SEM/EDS, DTA, TGA	-----	NiO addition led to formation of Ni <sub>3</sub> V <sub>2</sub> O <sub>8</sub> , which reduced corrosiveness of the ash materials by trapping vanadium	[71]
16	PAC with NPNiO (NiONP)	250 ppm	C-Steel/ 1.0 M HCl	FTIR, TEM, XRD, FE-SEM, EDX, SAED, WL, PDP, EIS	98.6%	PAC/NiONP served as mixed-type CI. Their adsorption onto C-steel interface followed Langmuir's isotherm model	[56]

CREP mediated NPNi were synthesized, characterized and used for CI of X80 steel, with the composition of C (0.17), Si (0.38), Mn (1.25), P (0.015), S (0.002), Mo (0.20), Nb (0.04), Ti (0.015) and Fe (balance), supplied by Shengxin Technology Co. Ltd, Xinyang, China, in a 1 M HCl solution [52]. X80 is a high-strength, low-alloy steel primarily known for its use in industries that require high-performance materials, such as pipeline and construction industries. CREP-NPNi richer in N and O sites were reported to be more stable and to efficiently inhibit corrosion at high T than CREP. IE(%) of CREP and CREP-NPNi decreased as T increased. Adsorption of CREP-NPNi was found to occur by combined physisorption and chemisorption enabled by C=C, C=O, C-O, O and N-H sites. Electrochemical measurements show that CREP-NPNi had dominant influence on anodic half reaction. Fig 2 depicts charge transfer controlled corrosion mechanism.



**Figure 2:** (i) OCP vs. time (ii) Nyquist plot and (iii) PDP curves for the corrosion of X80 steel in 1 M HCl with different Ct of CREP-NPNi [52].

In contrast, NPNi synthesized from onion mesocarp ethanol extract acted as mixed type CI and physically adsorbed onto X80 steel surface in 1 M HCl, with dominant influence on cathodic reaction involving HER. These NP effectively inhibited HER and X80 steel CR, especially at increased Ct [63].

In another study, changes occurring at the steel surface during interaction with metallic cations from Fe-NiO nanopowder were investigated using two Ct. EIS studies for CS in 3.5 wt.% NaCl linked the corrosion mechanism to the adsorption of



metallic cations onto the steel surface, which formed a protective film. Findings also indicated CI at low Ct and induction behavior at higher Ct [66].

Corrosion protection of CS in 0.5 M HCl using green-synthesized NPNiO by WL and electrochemical techniques was carried out by [25], and the results indicate a decrease in CR and increase in IE(%) with higher inhibitor Ct. The researchers have reported that a protection layer was formed on the CS surface, protecting it from corrosion, with an indication of predominant control over cathodic reaction.

By increasing the Ct of HCl to 1 N, [62] have reported that the CR of MS decreased with higher Ct of NiO inhibitor. IE(%) also decreased with higher Ct of CI, which was in disagreement with results from [56], who has reported that the protection power of CS in 1.0 M HCl increased with a rise in Ct of PAC containing NPNiO, which agrees with findings from similar studies [52, 68].

CI obtained by p-MBDM and p-MBDM assembled on NPNiO on MS in 2 M HCl was investigated by [53]. Their findings showed that IE(%) increased with higher inhibitors Ct. Both CI were of mixed type.

In 1 and 2 M H<sub>2</sub>SO<sub>4</sub>, NiO-Polyaniline (NiO-PANI) composite offered effective protection of MS, with slight variations in IE(%) up to eight h, for Ct above 250 ppm, and acted as a mixed type inhibitor[74]. NPNi (pH of 1) electrodeposited on Ni-Cr alloy had also excellent corrosion resistance in 1 M H<sub>2</sub>SO<sub>4</sub> saturated with CO<sub>2</sub> [77]. Results from the study indicate that corrosion current densities reduced while polarization resistance increased, due to NPNi application on the alloy surface.

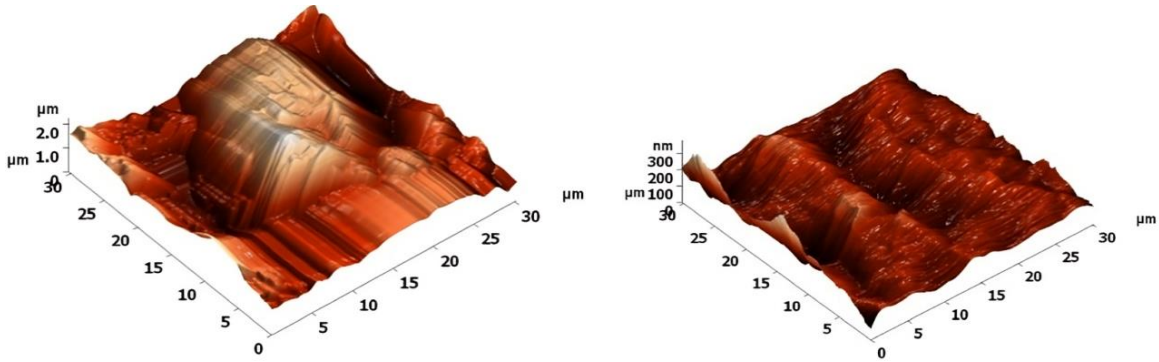
Corrosion resistance of NC from ZnO-NiO coated with epoxy on steel in 3.5% NaCl solution was compared by EIS to that of pure epoxy-coated steel [65], showing excellent CI.

Electrochemical measurements revealed enhanced protection of 304L SS substrate in 3.5% NaCl by nanostructured thin films of NiO [54], which was also confirmed using the 390 h salt spray test.

Figs. 3 and 4 show a thin film of NiO coated on SS 304L steel substrate exposed for 390 h (5% of NaCl at 35 °C) to salt spray test and their AFM images, respectively. Surface roughness values displayed little variation on nanoindentation and salt spray, indicating the anti-corrosive nature of the films.

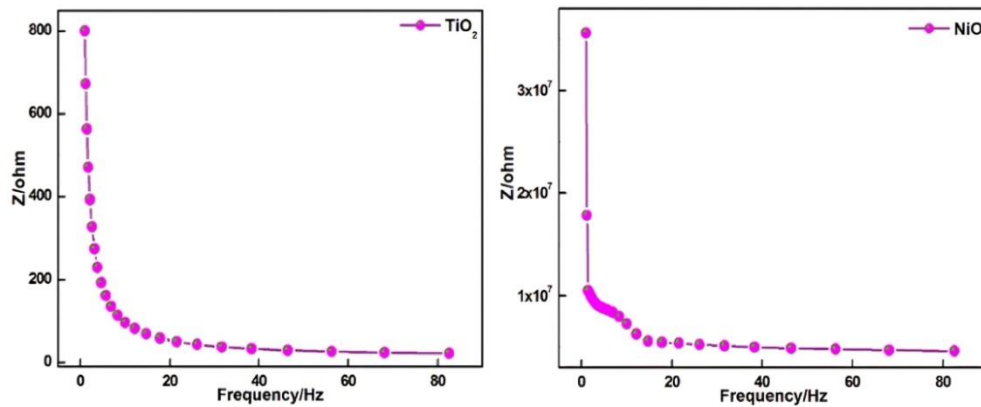


**Figure 3:** NiO thin film after 390 h salt spray test [54].

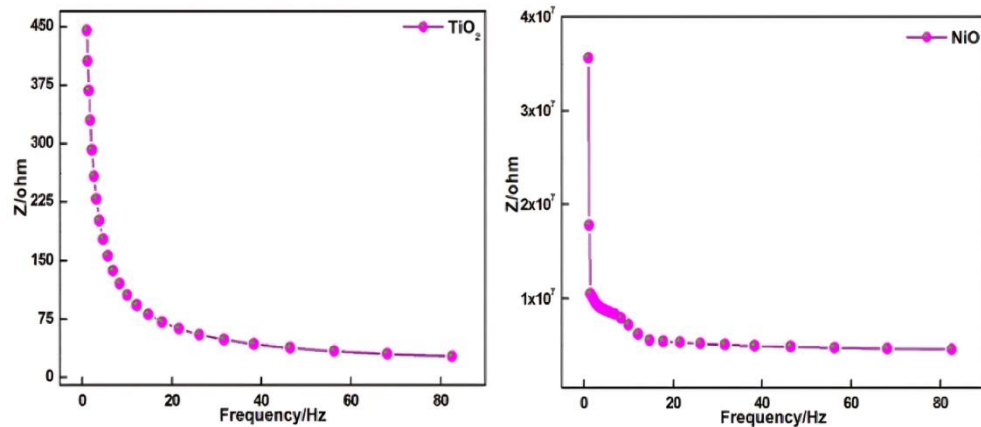


**Figure 4:** AFM images of NiO thin films before and after nanoindentation (after salt spray) [54].

In a related study, the corrosion resistance of NPNiO and TiO<sub>2</sub> spray coated on 304L SS substrates in 3.5% NaCl was compared using Tafel polarization curve and EIS. Both films were quite stable in salt, being recommended for suitable protection of 304 SS in adverse salt environment [70]. Although the impedance value decreased with higher frequencies after corrosion test for TiO<sub>2</sub>, it was static for NiO. This indicates extended durability and protection of NiO thin film over TiO<sub>2</sub> (Figs. 5 and 6).



**Figure 5:** Impedance spectra of TiO<sub>2</sub> and NiO coated SS before corrosion test [70].

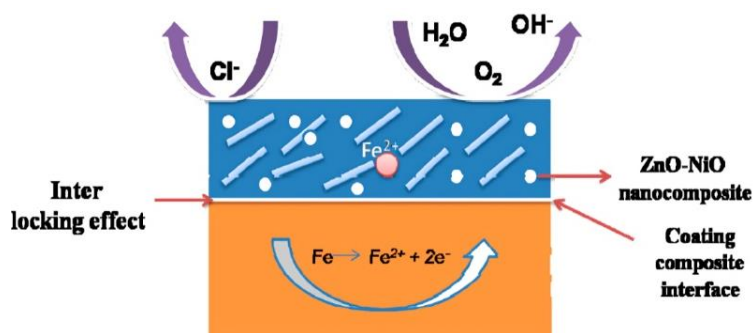


**Figure 6:** EIS of TiO<sub>2</sub> and NiO coated SS after corrosion test [70].

### Mechanism of CI by NPNiO

NPNiO possess remarkable CI properties, due to their unique surface properties, high surface area and good stability under harsh conditions [72]. Its ability to produce a passive and dense oxide layer on the metal surface, which serves as a barrier against corrosive environments, is thought to be the mechanism by which it hinders corrosion. This passive layer inhibits anodic or cathodic reactions that cause metal corrosion [25, 65]. CR is slowed down by NPNiO adsorption onto the metal surface, which lessens the interaction between the metal and corrosive ions. Since NPNiO have a large surface area, more active sites are available for corrosive ions adsorption, leading to excellent corrosion prevention capabilities. Since positively charged NP absorb more readily onto negatively charged metal surfaces, creating a protective layer, NPNiO surface charge is critical to CI mechanism. NP size and location play an important role in their CI mechanism. Smaller particles tend to offer better surface coverage and protection against corrosion [56].

The mechanism for enhanced corrosion protection of epoxy/ZnO-NiO NC coatings was classified by [65]: (i) the high surface energy due to the nanomaterials system leads to the creation of appropriate surface irregularity and hydrophobic surfaces at the epoxy/1.8 ZnO-NiO coating top; and (ii) the formation of an adjacent interconnecting structure with tough adhesion at the coating–composite interface (Fig. 7).



**Figure 7:** Proposed mechanism of the corrosion protection by ZnO-NiO NC [65].

### Environmental and safety considerations

Although NPNi have shown good potential as CI, there is a great concern about their level of toxicity to human health and the environment. A study has examined the response of human lung epithelial A549 cells after treatment with NPNi at the Ct of 0, 1, 2, 5, 10 and 25  $\mu\text{g}/\text{mL}$  for 24 and 48 h. Findings revealed that NPNi induces cytotoxic effects in the cells, which could be mediated through reactive oxygen species generation and oxidative stress. NPNi has reduced mitochondrial function and induced the leakage of lactate dehydrogenase in dose and time-dependent manners [73]. Another study has confirmed that NPNi can induce a variety of toxic effects on animals and humans cells, leading to adverse effects on respiratory, cardiovascular and reproductive systems [74].

NPNi penetration in the skin, especially if there are cuts or abrasions, can cause dermatitis [75, 76]. Prolonged exposure to high Ct of NPNi can also significantly increase the chances of allergies that manifest in various forms such as headaches, gastrointestinal, contact dermatitis and respiratory issues [77]. In addition, prolonged exposure to NPNi without any special respiratory protection or control measures can lead to throat irritation, nasal congestion, post nasal drip, facial flushing, skin reactions and death, in severe cases [75, 78].

NPNi effect on aquatic organisms representing general trophic levels has also been investigated, and the findings reveal that it has a selective toxicity for some and not all of the analyzed test organisms. Higher Ct of NPNi lead to significant toxic damages of tested organisms (except for fish). So, a Ct not exceeding 1.0 mg/L was recommended as the safe level [79].

The application of bio-nanotechnology for NPNi sorting, immobilization, degradation, stabilization and recycling, as well as the use of bacteria and other organisms to reuse these valuable NP from nanowaste, were recommended as measures that can mitigate their adverse effect on aquatic life and the environment [80]. The use of adequate personal protective equipment such as gloves, respirators and lab coats should be emphasized and enforced for workers exposed to NPNi, for ensuring their safety and minimizing occupational health hazards [78].

## **Conclusion**

This review has confirmed that Ni /NPNiO and NC are very effective CI for MS, SS, PS, CS, Ni-Cr alloys and Ni-Cr-Co superalloy protection in a variety of aggressive corrosive media such as acidic, alkaline, soil and gas turbine environments. Most studies have revealed that NPNi act as mixed type CI with dominant influence on cathodic reaction. NPNiO stability has been confirmed by salt spray test. The challenges of NPNiO agglomeration and dispersion have been successfully overcome by embedding them in epoxy resin. NPNi can be synthesized using various physical, chemical and biological methods, but more emphasis on green synthesis is recommended for future researches, due to safety, environmental and health concerns. Future research work includes: studies involving CR and IE(%) mathematical modelling and optimization for various alloys in different aqueous media using NPNi; and molecular modelling of NPNiO adsorption behavior onto various substrates.

## **Abbreviations**

**AFM:** atomic force microscopy

**BET:** Brunauer-Emmett-Teller analysis

**CI:** corrosion inhibitor/ inhibition

**Co:** cobalt

**Cr:** chromium

**CR:** corrosion rate  
**CREP:** *Citrus reticulata* peels extract  
**CS:** carbon steel  
**Ct:** concentration  
**Cu:** copper  
**DLS:** dynamic light scattering  
**DTA:** differential thermal analysis  
**DTG:** derivative thermogravimetry  
**EDS/EDX/EDAX:** energy dispersive x-ray analysis  
**EIS:** electrochemical impedance spectroscopy  
**ERH:** hexaglycidyl N- phenylaminopropoxy-N.N.N-triphenyl propane-1.3-diamine  
**ERO:** octaglycidyl N.N.N.N-tetraphenylamino- N.N.N.N-tetrapropoxy methylene dianiline  
**ESI-MS:** electrospray ionization-mass spectroscopy  
**Fe:** iron  
**FE-SEM:** field emission scanning electron microscopy  
**FTIR:** Fourier transform infrared spectroscopy  
**GC-MS:** gas chromatography-mass spectroscopy  
**GDP:** gross domestic product  
**H<sub>2</sub>SO<sub>4</sub>:** sulphuric acid  
**HCl:** hydrochloric acid  
**Hcp:** hexagonal close packed  
**HER:** hydrogen gas evolution reaction  
**HR-SEM:** high resolution scanning electron microscopy  
**HR-TEM:** high resolution transmission electron microscopy  
**IE(%):** inhibition efficiency  
**IR:** infrared spectroscopy  
**LPR:** linear polarization resistance  
**MS:** mild steel  
**NC:** nanocomposite  
**Ni:** nickel  
**NiFe<sub>2</sub>O<sub>4</sub>:** nickel ferrite  
**NiO:** nickel oxide  
**NMR:** nuclear magnetic resonance  
**NP:** nanoparticles  
**NPNi:** nanoparticles from nickel  
**NPNiO:** nanoparticles from nickel oxide  
**OCP:** open circuit potential  
**p-MBDM:** p-methoxybenzylidene-4,40-dimorpholine  
**PAC:** primary aminated modified cellulose  
**PDP:** potentiodynamic polarization  
**PLS:** photoluminescence spectroscopy

**PS:** pipeline steel  
**SAED:** selected area electron diffraction  
**SAXS:** small angle x-ray scattering  
**SEM:** scanning electron microscopy  
**SQUID:** superconducting quantum interference device  
**SS:** stainless steel  
**STEM:** scanning transmission electron microscopy  
**T:** temperature  
**TEM:** transmission electron microscopy  
**TGA:** thermogravimetric analyzer  
**TiO<sub>2</sub>:** titanium dioxide  
**UV-Vis:** ultraviolet-visible spectroscopy  
**VSM:** vibrating sample magnetometry  
**WL:** weight loss  
**XAS:** X-ray absorption spectroscopy  
**XPS:** X-ray photoelectron spectroscopy  
**XRD:** X-ray diffraction

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### **Conflict of interest**

The authors declare that there are no conflicts of interest.

### **Authors' contributions**

**K. Okon:** conducted literature searches; contributed with data or analysis tools; performed analysis of outcomes; drafted the manuscript as lead author. **C. O. Akalezi:** designed and conceptualized the study. **C. A. Maduabuchi, I. B. Onyeachu, S. Nwanonyi and D. Njoku:** proofread and supervised the manuscript. **T.O. Azeez and I. C. Ekeke:** proofread, edited and supervised the manuscript. **I. I. Ayogu:** proofread the manuscript and provided technical guidance on analyses.

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