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Review Paper

Metal oxides and its blended derivative's coating for anti-corrosion application

Vandana N Mahajan[a*], M J Sable[a], & Suhas R Patil[b]

[a]Department of Mechanical Engineering, Government College of Engineering, Jalgaon, Maharashtra 425 002 , India
[b]Department of Physics, Dr Annasaheb G D Bendale Mahila Mahavidyalaya, Jalgaon, Maharashtra 425 001, India

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Thin film deposition by using different nanomaterials has been an efficient and reliable way for enhancing the anti-corrosive property of the materials as well as strength improvement such as hardness, conductivity and wear resistance. Various materials have been taken as substrates like mild steel, magnesium, Aluminum, Copper, Tin, Carbon steel with thin film coatings of Zn–TiO_2, Ce/Co, Zn-HA/TiO_2, CrN/TiN, Ni- Co have been sampled. These specimens have been studied for numerous properties like surface roughness, wear resistance, adhesion strength, microhardness, hydrophobicity etc. It has been found that the components like muffler, differential, engine chassis, exhaust system, gears do undergo corrosion due to several factors like climate change, oxidation, moisture content etc. The aim of the review has been to highlight the advances in the coatings providing anticorrosive properties to various metallic substrates used specially for mechanical and automobile industries.

Keywords: Substrate, Anti-corrosion, Nanomaterials, Hydrophobicity, Muffler

1 Introduction

There exists a great area which *has been* affected by corrosion ranging from household appliances to industrial installations. Corrosion causes deterioration and degradation of metallic parts[1.] Corrosion can be defined as a natural, spontaneous and thermo-dynamically feasible phenomenon exhibited by all metals except noble ones[2]. It has been found that thin film coatings can ameliorate a material's execution significantly in certain areas of corrosion control and strength improvement. The various parts which are susceptible to corrosion and used in mechanical and automobile industries are muffler, differential, engine chassis, exhaust system, gears. Corrosion is mainly caused due to moisture content of the surrounding and variation in climate , a very prolonged deposition of dust in different parts of automobile in moisture, chlorides content in coastal areas, salts such as NaCl, $CaCl_2$, $MgCl_2$ (used to keep roads free from ice)[3,4]. Various mechanical components facing corrosion have been found as follows: radiators, water pumps, exhaust pipes, body of car, incinerators, boilers, turbines, IC engine parts[5,6]. The nano-coatings on various metallic substrate influence the resulting surface properties like particle size, coalesce particles' distribution, porosity[7.] Stagnation of liquids due to lack of drain hole,over coat thickness and issues related painting process have been responsible for rusting at fire wall area and inner hood of car bodies[8]. The paper has focused on use of lightweight materials such as advanced high strength steels, ultra-high strength steels, aluminum (Al) alloys, magnesium (Mg) alloys, and composites. The problem of arc ignition has been caused due to corrosion into connector blocks and terminals of in electric vehicles[9]. Zinc-magnesium alloy coated steel (ZMG) has been more beneficial than conventional galvanized steel. ZMG suggests mechanism for corrosion protection whereby cathodic activity has been slowed down via the precipitation of insulating, poorly soluble magnesium hydroxide. The cathodic activity has been assigned to the specific oxide layer (possibly magnesium oxyhydroxide doped with zinc) present at the ZMG surface[10].

The root causes in differential of vehicle and body of automotive vehicle are moisture, severe temperature and road debris. Once the paint coatings are removed even slightly, the corrosion takes place if they are subjected to any one of the above causes. The materials which are used for construction of automotive vehicles are generally carbon steel, fiberglass, aluminum, magnesium, copper, cast iron, glass, various polymers and metal alloys. To protect the parts from corrosion, the body of automotive

*Corresponding author (E-mail: er.vandana@rediffmail.com)

vehicles is covered with paints and different metal parts are protected with metallic and inorganic coatings[6]. The most commonly used part like Field muffler (from material 1.4509) after different mileages; it shows corrosion occurred in the interior and exterior cover after 24145 km and 989 km[11].

The research literature is about corrosion resistance improvement, using different nanomaterials coated on various substrates which are used in mechanical and automobile industries. In this paper the previous study which has been done has been surveyed and a brief review of the studies on parameters like coating material, different compositions of coating material, different substrates, and improvement in physical and mechanical properties of materials.

2 Materials and Methods

Many researchers worked on the properties of thin film coatings on various substrates involving different methods to judge its properties in the field of mechanical and automobile engineering. The study was carried out for various coating materials, methods of synthesis and metallic substrate.

In recent years, various alternative methods and coating materials on metallic substrate have been researched. The metallic substrates had been selected in such a way that they have direct applications in automobile and mechanical industries. A lot of coating materials were used by different methods on the substrates like steel, cast steel, ferritic stainless steel, boiler steel, Tin, mild steel, Ti6Al4V, Stainless steel alloy X18H10T (Severstal), aluminum, Copper and Carbon steel. On these materials the synthesis was done by various methods like Electrodeposition process, Vapor deposition method, Sol-Gel, Multiple spin casting, Magnetron sputtering, Situ electro polymerization, Radio frequency magnetron sputtering etc[12-17]. Table 1 explains the brief details of the numerous kinds of coating methods and different combinations of substrates with coating materials.

2.1 Mild steel as substrate

Gao *et al.*[13] reported increase in hardness and corrosion as well as wear resistance of Cu-Sn-Zn coating on mild steel. Habazaki *et al.*[14] reported that Aluminum silicate nanocoating was deposited on stainless steel Type 430 substrate by multiple spin casting. The results showed better corrosion resistance in cyclic corrosion test in Chloride solution. Kapoor *et al.*[18] reported the effect of silicon-zirconium sol-gel coating on hot corrosion of boiler steel (SS304). Cyclic oxidation tests were performed at 900°C in the presence of Na_2SO_4 salt solution on coated and bare samples. Cerium and Zirconium coatings provided excellent corrosion resistance. Arthanareeswari *et al*[19]. reported improvement in anticorrosive properties of nano TiO_2 coated on mild steel substrate due to higher coating weight as well as decrease in crystal size. The various synthesis techniques for coating on SS-304, MS, SS, X-65 pipeline steel were successfully implemented by electroless plating and Sol-gel method. The properties like surface structure, corrosion resistance, Microhardness are studied[18-22,31].

Hannes Falk -Windisch *et al.*[32] reported change in corrosion properties of Co and Ce/Co coated ferritic stainless steel at 850°C and 650°C. Mirak *et al.*[23] reported pronounced hardness of Zn-HA/TiO_2 composite coating than Zn-HA composite coating. They reported better wear resistance of Zn-HA/TiO_2 coatings Zn-HA composite coatings. The electrodeposition method was used to coat NiTi alloy with Zn-HA and Zn-HA/TiO_2 nanocomposite materials.

Sajjadnejad *et al.*[24] deposited Zn and Zn-SiC composite films on steel specimen as well as glass. The glass cell was anode and steel specimen was cathode. The electrodeposition method was used to prepare thin films. Increase in microhardness of Zn coatings was observed by fining deposited structure and inhibiting dislocation propagation. Improvement in anti-corrosion properties of waterborne epoxy coating had been reported. Mesoporous TiO_2 was coated on mild steel substrate[25]. Zhang *et al.*[33] used type 45 steel as a substrate coated by electrochemical machining with Ni-P-Al_2O_3 composite. They found increase in surface roughness of the Ni-P-Al_2O_3. Veedu *et al.*[34] reported good corrosion resistance of *Magnifera indica* coating on steel. Phytochemicals present in the plant acted as corrosion inhibitors. A number of coatings to improve corrosion and wear behaviors were investigated, examined and are still under investigation at academic level as well as industrial level e.g. PVD CrN coatings on 316L stainless steel, CrN/TiN superlattice coatings on AISI 304 L austenite stainless steel[17]. Corrosion and adhesion properties could be upgraded by addition of Mn^{+2} to the phosphate coating composition for mild steel substrate [35]. Zn coated steel substrate showed superior corrosion properties in unnatural sea water as compared to bare steel [36]. Cold sprayed tantalum (T22) coatings provided microstructurally dense

Table 1 — Brief detail of coating on metallic substrates

Substrate	Coating Material	Method	Relative advantages	Ref. No.
Steel	ZnO	Electrodeposition process	Room temperature operation, low cost, single step, good reproducibility, reduction of waste.	2
Type 430 stainless steel	Tetraethoxysilane and aluminium sec-butoxide, aluminosilicate	Multiple spin casting		14
Cast Aluminum	Tetraethoxysilane, Silica	Sol–gel	Easy and cheap.	15
Tin	3-glycydoxypropyltrimethoxysilane (GPTMS) as organic precursor and tetra-n-propoxyzirconium (TPOZ) as inorganic precursor.	Sol-gel		16
Boiler steel ss-304	Silicon -Zirconium	Sol-gel	Easy and cheap.	18
Mild steel	Nano Zinc Phosphate and nano TiO_2, zinc phosphate			19
Mild steel	TiO_2-CuO,CuO-titania nanocomposites			20
Steel	Zinc-hydroxyapatite–titania and Zinc-hydroxyapatite	electroless plating		21
Stainless steel alloy X18H10T (Severstal)	Mixtures of TiO_2 and SiO_2	sol-gel	Sol-gel thin films display excellent anti-wear and friction reduction performance under low loads.	22
Ni-Ti alloy	Zinc, Zinc-hydroxyapatite-titania and Zinc-hydroxyapatite	Electrodeposition		23
Carbon steel	Zn and Zn-SiC	Electrodeposition method		24
Steel substrates with rounded corners and edges	Meso-TiO_2 particles and waterborne epoxy resins			25
304 stainless steel	Zinc acetate, dehydrated TiO_2	sol-gel method		26
Aluminum	TiO_2, Mn_2O_3 and ZnO nanoparticles	Situ electro polymerization		27
Cu	Nano-TiO_2	Dip-coating process		28
Cu	Nano-TiO_2	Dip coating process	A facile and low-cost method.	29
Carbon steel	Zinc,TiO_2,WO_3	Electrodeposition process	Porous free finished products that do not require subsequent consolidation processing, requires low initial capital, provides production rates with few shape and size limitations, precise control, low temperature operation, and high rate of deposition and cost effectiveness.	30

metallic coatings with lower porosity. They show excellent corrosion resistance[37-40]. Ni-20Cr cold spray coating on SA 516 steel produces good oxidation resistance and thicker structure[41]. The crack free dense surface is generated on Ferritic Stainless Steel (FSS) by depositing Mn/Co oxide. The anticorrosion behavior was investigated with post deposition treatments up to 750°C for 750 h. The annealing cycle and the spinal crystalline structure of $(Mn, Co)_3O_4$ showed the slow growth of surface oxide layer[42].

Inorganic, hybrid sol gel films with corrosion inhibitors were maneuvered for steel, Al, Cu, and Mg (as substrate) successfully[43-44]. Ni-P/CeO_2 coating as well as Ni-P with Nano SiO_2 showed improvement in corrosion resistance[45-46]. 316L stainless steel majorly used for implantation purposes in orthopedics due to its anticorrosive properties[47]. Cr and CrSi (with purity of 99.5%), Si/(Cr+Si) mole ratio of CrSi targeted as 2.5 % (PVD method)[48] Polyaniline-montmorillonite nanocomposites (wt%) and glass ceramic was coated on 316L SS by electrodeposition method[49,50]. 316L SS is also coated by glass ceramic using Spin coating method[47] and Multi-arc ion plating methods using PVD CrN and CrSiN on it[48]. Figure 1 shows graph of polarization curves of coated and uncoated 316L.Theanodic current density value of CrSiN is lower than CrN indicating more dense structure with minimum cracks. So CrSiN shows better corrosion resistance than CrN coating in seawater[48].

A bilayer polymer coating by applying polyrrole (PPy) as an interior side on type 304 SS increased the corrosion potential by 400 mV and pitting corrosion by 500 mV[51]. The coating mass increased from 7.56+0.73 g/m^2 to 13.82+0.68 g/m^2 when temperature was increased from 45°C to 85°C also coating thickness was increased from 4.5±0.9 µm to 11.5±1.66 µm . The Phosphate Chemical Conversion (PCC) coating performed excellent in corrosion resistance at 75°C with bath pH of 2.75[52]. Doped/undoped coatings can be applied to 304 SS and mild steel substrates with galvanometric deposition[51,53]. The different methods used for improving corrosion resistance depend on various factors such as coating method, coating material (whether single or composite), the weight or mole percentage of different doped materials, calcination temperatures[54,56]. It was expected that the small sensors used in marine environment had more chances to corrode. So, the aluminum oxide coating was done on 304L SS, Lazar *et al.*[57] fabricated nanostructured surface by using Metal organic chemical vapor deposition (MOCVD). The deposition of Sn or Zn from single target was more impressive than twin target[58].

Performance of nanocomposite coatings could be affected by poor adhesion related to high stresses. TiN, nc-TiN/a-sinX and nc-TiCN/a-SiCN coatings on SS410 showed improved corrosion resistance[59]. Pragmatic painting system was used for encapsulation of corrosion inhibitors. Silica film was fabricated on mild steel substrate of 10 µm thickness by Sol-gel method[60].

Ni-20Cr nanostructured alloy coating on SA 516 Grade 70 boiler steel had been studied for high temperature oxidation resistance at 900°C under cyclic conditions. These coatings found a high-level oxidation resistance[61]. Xu *et al*[26]. fabricated TiO_2/ZnO films by sol-gel method on 304 stainless steel plates (10mm X 10mm X 3mm) which were used as substrate. The formed thin films showed excellent anti-corrosion properties. It was found that corrosion resistance increased 100 times after applying a TiO_2 coating on 316 L stainless steel when compared with the bare steel using the same environment[62]. It is reported that heat treatment of cold sprayed coating at 150°C lowered the corrosion rate when steel substrate is coated by zinc [63]. Figure 2 shows Nyquist test

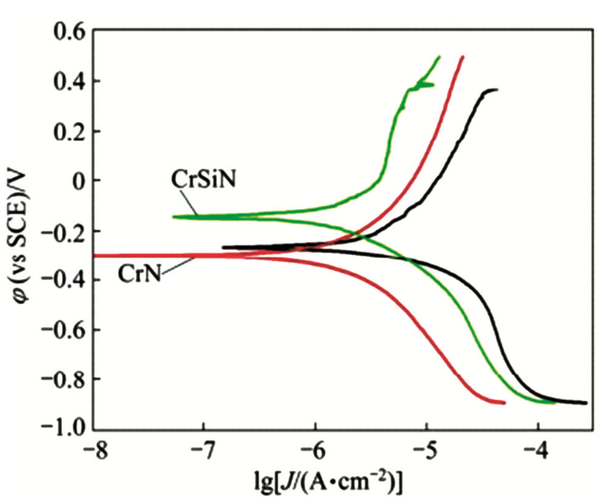

Fig. 1 — Polarization curves of coatings and uncoated 316 L[4]

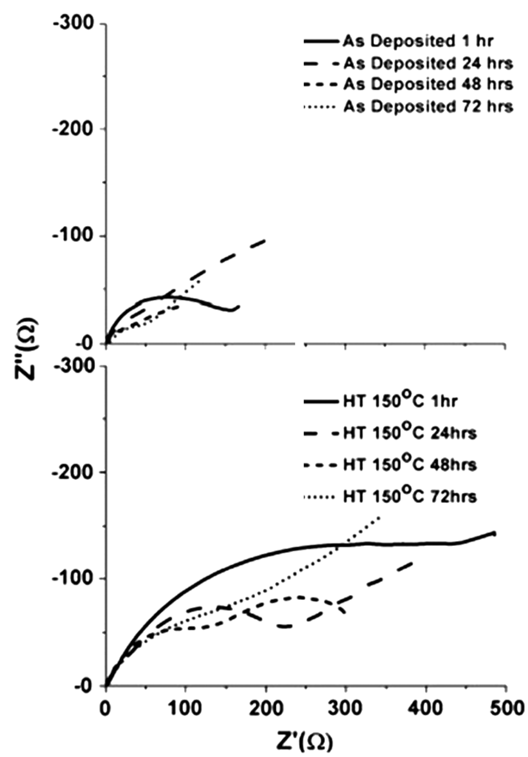

Fig. 2 — Nyquist plots of (a) as-coated, and (b) heat treated Zinc coatings on mild steel substrate[63].

results. Zn coatings in as-coated condition and heat treated condition were exposed to NaCl solution for 1 h, 24 h, 48 h and 72 h. At all high frequencies, all the plots show depressed semicircle which was compared with charge transfer resistance in parallel with double layer capacitance[63].

2.2 Aluminum as substrate

AA 2024-T3 alloy was coated by GLVMO[64], GPTMS and TPOZ[65], TMOS[66], Silane–incorporated epoxy resine[67], SiO_2 and ZrO_2[68,69], MAOS[70] materials. A lot of coating methods like Sol-gel method, layer by layer method[70], painting[67] etc. were implemented mostly[64-69,71,72]. Al (99.999%) and Cu (99.9%) were coated by TAVS (Triacetoxyvinylsilane) with deionized water using sol-gel method. Coated Al showed more resistance at intermediate layer than coated Cu[72]. The review covered chemistry of chromium and the roots of chromium toxicity[73].

Saline solution was modified with CNTs. A detailed review was done covering chromium and its toxicity and various methods to reduce corrosion[74]. Lopez *et al.*[15] used Sol-gel coating method for cast aluminum matrix composite reinforced with 10% SiC. Their study compared the multilayered coatings at different immersion times. Alvarez *et al.*[16] deposited hydrotalcite nanoparticles on tinplate by using Sol-gel method. Their study revealed that the undoped samples had worst behavior. The anticorrosive properties are assessed by electrochemical impedance measurement study. The doped sample shows better results for pitting corrosion. Hosseini *et al.*[27] reported anti-corrosion behavior of polypyrrole films in aggressive environments. TiO_2, Mn_2O_3 and ZnO nanoparticles were synthesized by electro polymerization on Aluminum electrode. They found an enhancement in corrosion resistance.

2.3 Mg as substrate

Researchers focused their study on AZ31 Mg alloy as substrate[75-,83]. Scanning vibrating electrode technique was used to study the behavior of the saline coated coupons. It is observed that the CNTs with rare earth salt slow up the corrosion process of the AZ31 Mg alloy[74]. The emphasis of the research in case of Mg and its alloys was given on various deposition techniques such as electrodeposition method, cold spraying, immersion technique, aerosol deposition, electrochemical painting, anodizing, cladding, organic coatings[74-81,84-87]. Pure Zn coating on AZ31b indicated better corrosion resistance and variation in micro hardness at different temperatures (260°C, 290°C, 320°C, 350°C). Figure 3 indicates that the microhardness values were mostly equal at 2.5 MPa. There was reduction in microhardness values under the different temperatures which indicates limited deformation of Zn powders[76].

Fatigue testing set up (rotating bending machine with the integrated corrosion chamber) is shown in Fig. 4[78]. The maximum stress range was 100 MPa to 165 MPa For corrosion fatigue tests, the stress limit was 40 MPa to 120 MPa .The capacity of the chamber was 18.9L which contained 3.5% of NaCl solution. Early cracking of Aluminum coating was formed due to low tensile strength and hence poor fatigue strength in corrosive environment[78]. The cylindrical work piece of diameter 13 mm and 3.8 mm thick of Mg93Al6Zn1 alloy was coated by PPy film. The corrosion tests were implemented in the Hank's solution pH 6.67 at 37°C by using frequency range from 100 kHz to 0.1 Hz with an ac amplitude of +5mV of electrochemical impedance spectroscopy. The growth in corrosion resistance was observed than the base Mg alloy[84]. The experimentation was put

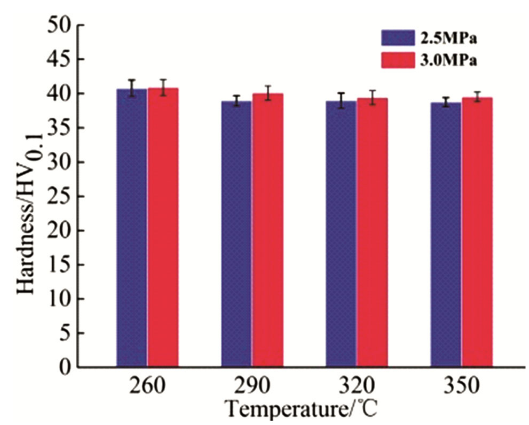

Fig. 3 — Variation in microhardness with increasing temperature, and pressure[76].

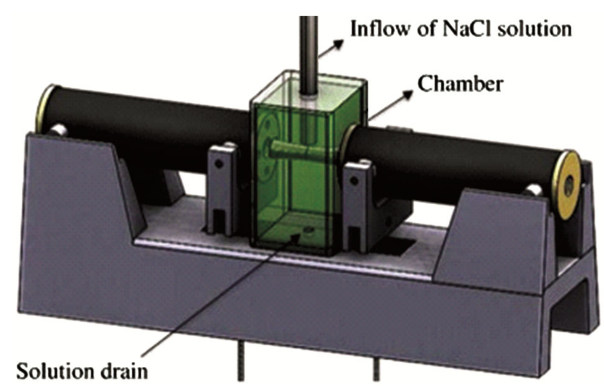

Fig. 4 — Corrosion fatigue testing set up (rotating bending machine with the integrated corrosion chamber)[78].

through on Mg plates and Mesoporous silica nanocontainers with and without Fluoride (inhibitor) dispensed in alkyl solution. It was concluded that MgF_2 was formed and acted as inhibitator[85]. On AZ91D magnesium alloy strip, Ni-Co alloy coating was constructed by electro deposition process. It was observed that hydrophobic surface helped to protect it from corrosion[86].

2.4 Cu and Sn as substrate

Cold sprayed Ta coatings were applied on steel, Al, and Cu substrates. Cold sprayed Ta coatings with bulk material and Cold sprayed Ta coatings showed better corrosion resistance than in 1wt % HF aqueous solution[88].

As Tin plate cans are widely used in food industries, if they corrode, they can contaminate the food stuff. So Tin plates were taken as substrate. They were coated by the organic /inorganic films by Sol-gel method. It was found that tin plate behavior was dependent on the nature of electrolyte. The impedance values reduced with immersion time. Impedance measurements were done at one Hz[82]. Polarization curves and EIS measurements indicated that the superhydrophobic composite surface improved the corrosion resistance when coated by TiO_2 with other materials such as plyvinylidene or PDMS[89,90].

2.5 Carbon steel as substrate

Many researchers had used Carbon Steel as substrate for anticorrosive surface preparation[89-93,28,29]. The different coating methods such as painting, dip coating, spraying[28,29,90,91] were explored. Electrochemical Spectroscopy[89,90,28,29] as well as Salt spray test[91,92] techniques were used to observe corrosion resistance of coated samples.

Yu *et al*.[94] had done the TiO_2/ZnO composite coatings with diverse atomic ratios of Ti to Zn. They were prepared by sol-gel process escorted by thermal treatment at different temperature on carbon steel substrate. The anticorrosive properties were checked for distinct combinations. The 8 layer TiO_2/ZnO with molar ratio 1/1 and 1/3 at 500°C showed good results in anticorrosion.

Punith[2] studied the Zn nanoparticles composite coating process for industrial applications. The Zn nanoparticle coating was generated by Electrodeposition process. The structural, mechanical and anticorrosive properties are studied[2]. The regular high zinc phosphating and recent tricationic phosphate coatings on steel surface are compared and the corrosive behavior of the coated surfaces was analyzed by Salt spray test and EIS[95].

Corrosion could be prevented by using various methods such as nano-alloy, nano-inhibitors, nano-generators, nano-sensors. These methods are not only protecting metals from corrosion but also regularly monitor the structure of the material[96].

Daniyan *et al*.[30] studied the, tribological corrosion and microstructural properties of Zn-TiO_2 and Zn-TiO_2-WO_3 material coatings on carbon steel substrate. They used electrodeposition method to synthesize the nanoparticles. They reported the enhancement of the corrosion resistance due to physical barriers offered by TiO_2 and WO_3. The increase in hardness is reported due to incorporation of TiO_2/WO_3 nanoceramics. Magnesium and its alloys can be coated by different techniques to give pore free and uniform coating[97] Titanium can be used for the exhaust system of a bike due to less weight and fine aesthetics[98].

The often-used material for exhaust system could be stainless steel with chromium called austenitic stainless steel which often contains higher percentage of chromium to maintain good anticorrosive properties at elevated temperatures[99]. But it increases the cost of the system. New combinations of iron and various materials used in automotive industries which are associated with the engine, chassis and other parts to reduce weight together with improved mechanical properties. The surface properties are also the function of number of layers[100]. Iron plate coated by Zn-Ni alloys showed the inverse relationship between anticorrosive property with the grain size and type of structure. Corrosion resistance was evaluated by EIS. The test was conducted in 3.5% NaCl solution at room temperature. The coating by Pulse Reverse Current showed the best corrosion resistance than Direct current and Pulse Current methods[101]. Zinc coated materials like steel exhibited galvanic protection and it tries to trap the corrosion product in the interdentritic region acting as a barrier[102]. The experimentation was carried out on localized corrosion of iron plate. The EIS results and polarization curves indicated Zn-Ni alloy coatings proved to be the best for anticorrosion[101].

Stankiewicz *et al.*[103] explained the various self-healing anticorrosive techniques. Self healing smart coatings can be used for safeguarding of metallic surfaces from corrosion. The smart coatings showed very interactive with a lot of environmental changes.

The smart coatings are environment friendly so can be used in place of toxic chemicals[104].

3 Results and Discussion

As per the literature survey, change in doping concentration affected the mechanical, structural and chemical properties. Thus, it proved the necessity to study these different nano coating materials thoroughly to make better mechanical and structural properties of the thin films.

Tensile strength affected the corrosion behavior of some metals like Aluminum[75]. Coating thickness influences the occurrence of crack. The calcination temperature, coating thickness and surface finish influenced corrosion resistance. Salt spray test, Electrochemical Impedance Spectroscopy used as the effective techniques for assessing anticorrosive property.

4 Conclusion

The impact of various nano materials on metallic substrate has been excellent to improve the mechanical properties of different metals. There is great scope for taking different metals as substrate which are always used in mechanical industries.

Different techniques of coating the metals have been implemented for reducing the unacceptable effect on environment. Nanocoating materials have been successfully synthesized on suitable substrates by different blending combinations and other process parameters.

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