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Polyethylene-Silica-Zirconia Based Protective and Hydrophobic Nanocomposite Coatings for Corrosion Control on Aluminium and Mild Steel

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Abstract

In this work, we prepared polyethylene oxide-silica-zirconia based hydrophobic coatings for corrosion controlled on aluminium (Al) and mild steel (MS) substrates (sheets) by the wet chemical process using (3-glycidoxypropyl)-trimethoxysilane (GPTMS), zirconium (IV) propoxide (ZP) and polydimethyl siloxane (PDMS) with trimethylsiloxy terminated as precursors. Depending on the mol percentage of ZP used for preparation, the sols were designated as GZ-0, GZ-5, GZ-10, and GZ-20. Crack free homogeneous coatings of thickness 3-4 µm having refractive index values 1.48 to 1.535 ± 0.005 were prepared using these sols after drying at 60°C/ 1 h followed by heat treatment at 120 °C/ 2 h. Formation of Si-O-Si, ZrO- Zr and inter-linked Si-O-Zr network during sol the stage as well as post thermal treatment condition were confirmed by FTIR and ATR analysis. The hardness of the coatings was ≥6H (ASTM D 3363) with good adhesion to the substrate. After immersion in 3.5 wt % NaCl solution uncoated Al and MS showed significant deterioration, whereas no damage was observed for all coated substrates even after 20 days. Static water contact angle (WCA) of the heat treated (120 °C/2h) coatings were around 108 ± 2°. With increasing the hydrophobicity of coating corrosion resistant significantly enhanced to certain extent (25 ±1 days) but mechanical property (hardness) decreased. Considering above, the said composite materials can be used to coat or strengthen various metal surfaces for enhancing their performance against environmental impact, durability, and functional life for different applications areas.

Keywords: Sol-gel, dip-coating, nanocomposite, water contact angle, corrosion resistance.

Introduction

Among the metals Al and MS are being used in large quantity from domestic to heavy industrial applications as well as in the areas of electronics, power generation and distribution, transportation, equipment and miscellaneous components particularly owing to their highly recyclability (Sheffer et al., 2003). But these metals are susceptible to chemical reactions in atmospheric conditions leading to application of suitable protective coating is very important to alleviate the effects of rust and corrosion (Hammer et al., 2010; Zheng & Li, 2010; Alemdar et al., 2007; Hammer et. al., 2012; Figueira et. al., 2015; Kunst et al., 2015). These protective coatings help to retain the quality and durability of the material and can significantly increase the usable lifetime of materials along with reducing the maintenance and replacement costs. In this regard metal oxide coatings, particularly, more effective chromate compounds are largely used to protect corrosion prone metal surfaces. However, in recent decades, due to the carcinogenic effects environmental regulations have restricted the use of hexavalent chromium- containing species from corrosion inhibiting packages (Figueira et. al., 2015). Hence, a chromium-free technology with superior corrosion performance is highly desirable.

Several methods have been applied for protecting Al and MS and its alloys against corrosion such as deposition of thin films or by coating. Different techniques are known for applying thin coatings on aluminium and other related substrates such as dip-and spin coating, sputtering, pyrolytic deposition, co-extrusion, PE-CVD etc. Using sol-gel technique attempts have been made to protect the Al-substrate by deposition of inorganic-oxides. This process requires higher temperature due to which coatings turn brittle (Zeng & Li, 2010). To over this, inorganic-organic approaches have been carried out. Organosilane coatings with functional organic groups (vinyl, methacryloxy and glycidoxypropyl) exhibit better corrosion resistance and improved flexibility (Liu et. al., 2005). Oxide nanoparticles embedded within such a system can further improve corrosion properties of the coating (Gonzalez et al., 2011).

Numerous inorganic-organic hybrid coatings produced by sol-gel processing have been studied extensively for prevention of corrosion in metals that naturally form a passive layer on their surface (Zheng & Li, 2010; Hammer et al., 2012; Kunst et al., 2015; Amiery et al., 2014; Pandit et al., 2015; Esmeryan, & Chaushev, 2023; Pepe et al., 2005; Chou et al, 2003). But most

of these reported coatings have been cured at relatively high temperature, not less than 130°C. To improve the protection properties, silane-based layers have been fabricated by adding inorganic or organic inhibitors to the silane films (Chou et al, 2003; Mustaq et al., 2020). However, many cases inhibitors negatively influence the stability time of the sol–gel network. So, attempts have been made for introduction of hydrophobic character into the protective coating for better corrosion resistant property of metals (Li, et al., 2015; Figueira et al., 2015; Kunst et al., 2015; Pepe et al., 2005; Agbogo et al., 2025). Apart from the wide range of applications, this process also offers various benefits, which include reduction in maintenance cost, elimination of tedious manual effort and also reduction in the time spent in cleaning work. For this purpose, various materials and different methods have been used to develop coatings on different metal surfaces. But reported protective hard hydrophobic coatings with sufficient surface hardness (usable for practical application) data are very limited. So, preparation of suitable composition is necessary to obtain hard coatings having good surface hardness with hydrophobic character on metal substrates for different applications. Beside the above, some other international published patents have been reported multilayer coatings to improve corrosion resistance property of metals for different applications (Richert et al., 2010; Sue et al., 2015). Moreover, in many reported corrosion resistant coatings on metals, coatings have been cured at relatively high temperature, not less than 130°C (Hack et al., 2012; Huesmann et al., 2015).

In this work, we attempted to prepare 'polyethylene oxide-silica-zirconia' coatings (PEO- SiO_2-ZrO_2) with varying GPTMS/ZP molar ratios, on Al and MS substrates (sheets) by wet chemical route to study corrosion resistance and focus on the effect of zirconia and PDMS incorporation into the silica matrix. The synthesis process is discussed as a function of sol (lacquer) processing and thermal curing steps. The characteristics of the coatings were evaluated using FTIR studies, hardness and adhesion tests, refractive index and contact angle measurements along with SAXS, TEM analysis and corrosion test in 3.5 wt % NaCl solution.

Materials and Methods

Materials

For the preparation of nanocomposite sols (lacquer's) following chemicals were used as received. Zirconium (IV) propoxide (70% in 1-propanol) (ZP), (3- glycidoxypropyl) trimethoxysilane (GPTMS), polydimethyl siloxane (PDMS) with hydroxy terminated (MW550 g/mol) and aluminium acetylacetonate ($Al(acac)_3$) were supplied by Sigma-Aldrich. Acetyl acetone, 1-propanol, HCl (35.4%) were supplied by MERCK Specialties Pvt. Ltd. Mili-Q (Millipore) water (18.2 MΩ) was used throughout the study.

Preparation of 'SiO_2-PEO-ZrO_2' based inorganic-organic coating sols (GZ)

For the preparation of sols, first 0.175 mol of (3-glycidoxypropyl)-trimethoxysilane (GPTMS) was mixed with 0.0406 mol of 1-propanol in a 100 ml beaker. To this, a mixture of Zirconium (IV) propoxide (ZP) (0.0132 mol), acetylacetone (0.0066 mol) and 1-propanol (0.0466 mol) was added after stirring for 1h at RT (25 ±2 °C). After 20 min HCl (5.5×10^{-4} mol)−water (0.394 mol) and 1-propanol (0.133 mol) were mixed and stirred for 2 h at RT. This sol was designated as GZ-5 and aged for a day. In similar ways, sols containing 5, 10 and 20 mol % with respect to the total mol content (ZP and GPTMS) were prepared and designed as GZ-10 and GZ-20, respectively. While, for the preparation of GZ-0 sol, ZP was not used.

Sols were used for coating deposition on aluminium and mild steel sheet, soda-lime glass, two sides and single side polished silicon wafer substrates (Figure 1).

To increase the hydrophobicity of the coating PDMS 5 wt% w. r. to silica has been incorporated into the GS-10 sol and designed as GZH-10 and evaluate the effect of corrosion resistant property.

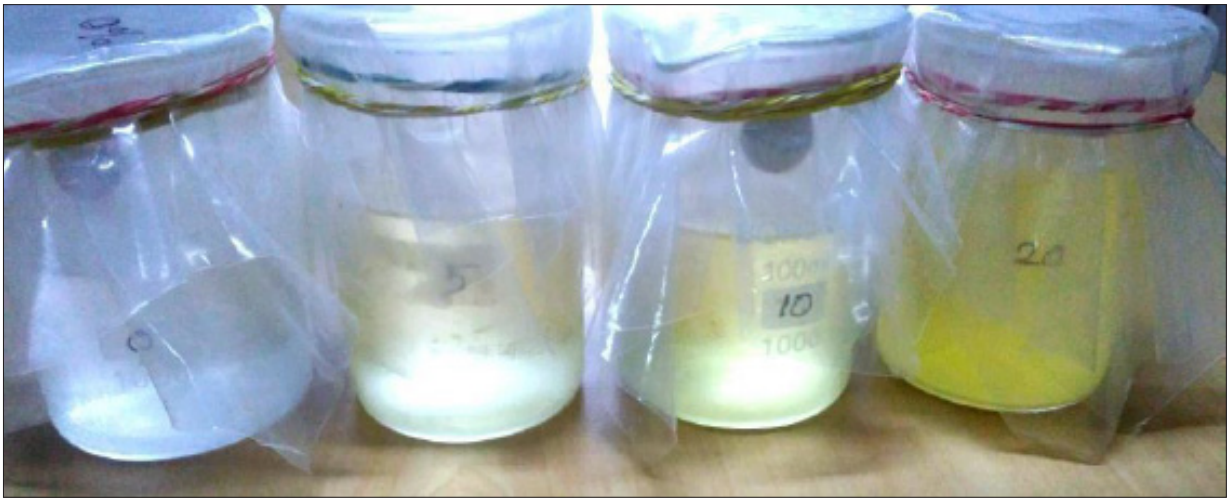


Figure 1: Digital image shows the final nanocomposite sols (GZ-0, GZ-5, GZ-10 and GZ-20 from left) kept in 100 ml beakers.

Preparation of Coatings

Prior to coating deposition, aluminium substrates (sheets) were first treated with N/50 NaOH solution by immersion for 5-6 min followed by washing under tap water. Finally, it was rinsed with distilled water and wiped cleaned with acetone. For MS, substrates were first immersed in 4 wt% H_2SO_4 for 4 mins followed by washing thoroughly with tap water then with distilled water. The cleaned substrates were coated with the sol by using dip-coating technique. The withdrawal velocities ranged from 12-20 cm/min. The prepared samples were first dried at 60°C for 1 h followed by heat treatment (cured) at 120 °C/2 h (ramped from 60 °C at 2 °C min^{-1}) in an oven. The thicknesses of the coatings so obtained were in the range 3-4 µm (thickness varies with withdrawing velocity) having RI value in the range of 1.481.535 ± 0.005. Similar coatings were deposited on silicon wafers (both side polished, intrinsic, IR transparent), single side polished silicon wafers and soda-lime glass substrates for the FTIR studies, RI and thickness measurements, respectively. ATR spectrum was obtained by placing the liquid sample on a ZnSe crystal plate.

Characterization

The refractive index of the coating was measured with the help of an Ellipsiometer (J.A Woollam) by depositing similar coatings on one side polished silicon wafer. Thickness of the coating was measured by Field emission scanning electron microscopy (cross sectional FESEM) analysis using Carl Zeiss, Germany, SUPRA-35VP instrument. Transmission (SAXS) of the trimethylsilyl modified hydrophobic sols were measured with a Rigaku SmartLab X-ray diffractometer operating at 9 kW (200 mA, 45 kV) using Cu Kα (λ = 1.5406 Å) radiation. For this study sols were taken in a sealed silica glass capillary of diameter 1 mm, and the results were analyzed using Rigaku's Nanosolver software. Infrared absorption spectra of the film deposited on both side polished Si-wafers were recorded by FTIR spectrometry (Nicolet, 380) with a resolution of 4 cm^{-1} and 200 scans. Transmission electron microscopic (TEM) measurements were carried out with a JEOL 2010 transmission electron microscope equipped with an EDS (energy dispersive X-ray scattering) facility. TEM samples were prepared by scratching a low thickness (□150 nm) film and placing the scratched off films on a carbon coated copper grid. Surface area of the coating was measured using Quanta chrome instrument, Nova 4000e, USA. Water contact angle (WCA) measurements were done with a Data Physics Instruments GmbH, Germany (OCA 15 Pro model) using 6 µL water droplets at room temperature. (WCA) was taken after averaging 10 measurements in different areas of 100 x 100 mm^2 coating surface. Corrosion tests of the coated and uncoated MS and Al substrates were carried out by immersion in 3.5 wt % NaCl solution.

Results and Discussion

Properties of the Sols

In this work the nanocomposite coating sols were prepared keeping in view longer shelf life at room temperature (RT) (25±2 °C). As prepared sols viscosity were in the rage of 5-6 cPs with sol pH of 2.5-3 which is close to the isoelectric point (IEP) of silica to increase the shelf life or stability of the sol. In this approach the sol remains suitable for more than 1 month for continuous coating deposition at RT. This is to be noted here that sol was stable > 6 month when it was kept in closed condition at refrigerator (4 °C) after preparation. However, the viscosity of the sol tends to increase with time and temperature of storing. It has been observed up to a viscosity value of 35 ±2 cPs of the sol can be useful for the deposition of spot-free hard and hydrophobic coating on metals. PDMS 5 wt% w. r. to silica has been incorporated into the sol to increase hydrophobicity of the coatings for evaluating the effect of corrosion resistant property.

Small-Angle X-ray Scattering (SAXS) studies were done to estimate the particle size distributions of the silica-zirconia NPs in the GZ-10 sol. Figure 2a shows the experimental transmission SAXS profiles obtained from the sol. The simulated curve using a spherical particle model is in good agreement with the measurement (Figure 2a). From the SAXS profile (Figure 2b), the average size of the particles for sol GZ-10 was estimated to be 5.2 nm with a dispersion of 19 % are embedded with inorganic-organic hybrid network.

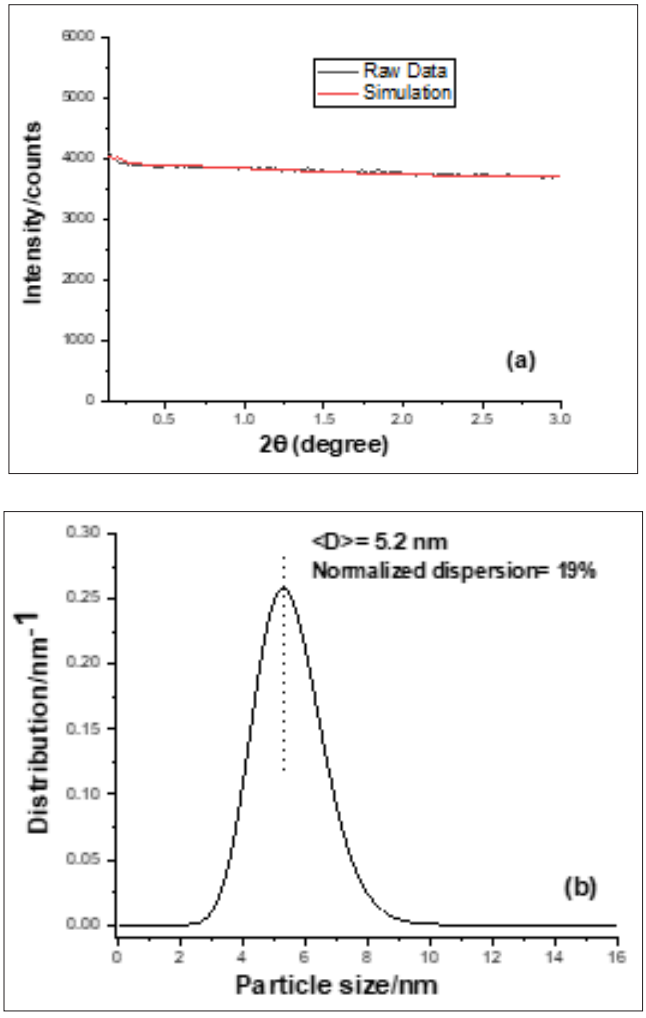


Figure 2: (a) Transmission SAXS profile of the GZ-10 sol along with the simulated curve using a spherical particle model and (b) size distributions of the silica nanoparticles estimated from the SAXS profile using the NANO solver software.

FTIR Studies

IR spectroscopy was performed for structural characterization of coatings. Figure 3 shows the ATR spectra of sols GZ-0, GZ-5, GZ-10 and GZ-20. The all spectra showed Si-O-Si asymmetric stretching at 1085 and 797 cm^{-1}, Si-OH vibrations at 940 cm^{-1}, and peaks at 910 and 855 cm^{-1} are due to the epoxide bands originated from GPTMS. Peak at ~1200 which is due to Si-CH_2 vibrations (Zheludkevich et al., 2005; Goswami et al., 2011; Cartereta & Labrosse, 2010; Colthup et al, 1990; De et al., 2024). Moreover, in ZP containing sols (GZ-5, GZ-10 and GZ-20) showed peaks at 1026 and 1060 cm^{-1} due to Si-O-Zr network. The appearance of peak at 1098 is due to Zr-O-Zr. So, all above peaks support formation of silica, zirconia and silica-zirconia network at the sol stage. In addition, as we have used acac to control the fast hydrolysis rate of Zr-alkoxide, ATR spectra of all ZP containing sols showed a sharp pair of peaks near 1586 and 1533 cm^{-1} due to (C-C and C-O) stretching's arising from Zr-acac chelate.

Figure 4 shows the FTIR spectra of as prepared and thermal treated (120 °C/2h) representative films (GZ0 and GZ10). On thermal treatment at 120°C for 2 h, all epoxy related peaks were almost decreased or disappeared due to polymerization of epoxy groups to polyethylene oxide (PEO) (Zheludkevich et al., 2005; Goswami et al., 2011). Moreover, intensity of the peak at 940 and 650 cm-1 (Si-OH and Zr-OH) decreased (Figure 10. (b)) whereas peaks at 1100-1112 and 472 cm–1 (Si-O-Si); peaks at 1098 and 430 cm^{-1} (Zr-O-Zr) and at 1060 cm-1 (Si-O-Zr) increased indicating formation of SiO_2, ZrO_2 and inter linked SiO-Zr network (Rodic et al, 2014; Fu et al., 2014). It may be concluded from ATR and FTIR analysis that this hybrid approach helps in forming a uniform network.

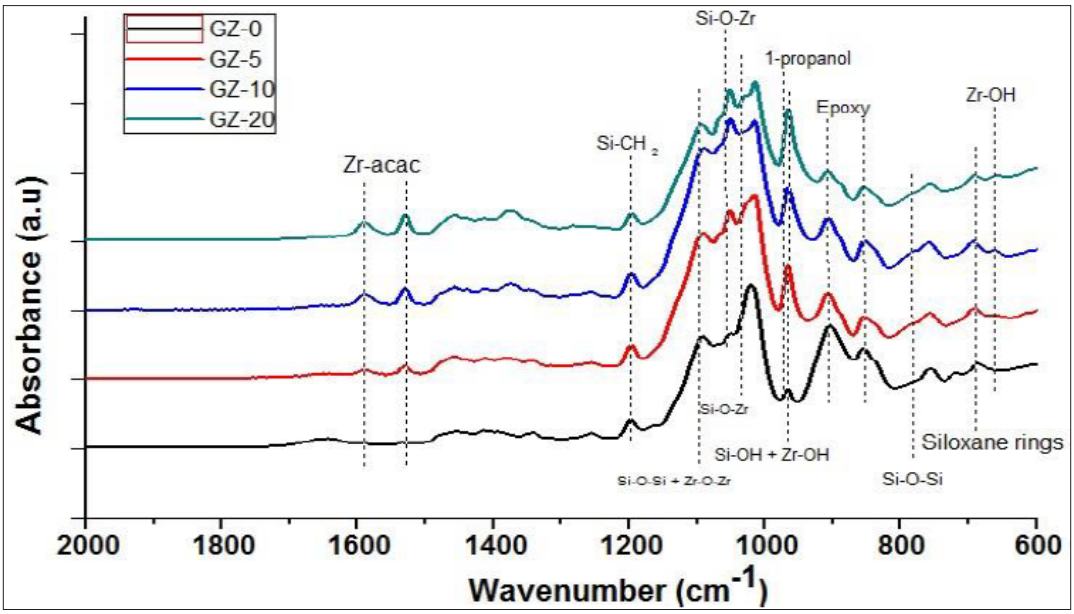


Figure 3: ATR spectra of the GZ-0, GZ-5, GZ-10 and GZ-20 sols. For such measurements the sol was placed on ZnSe crystal plate.

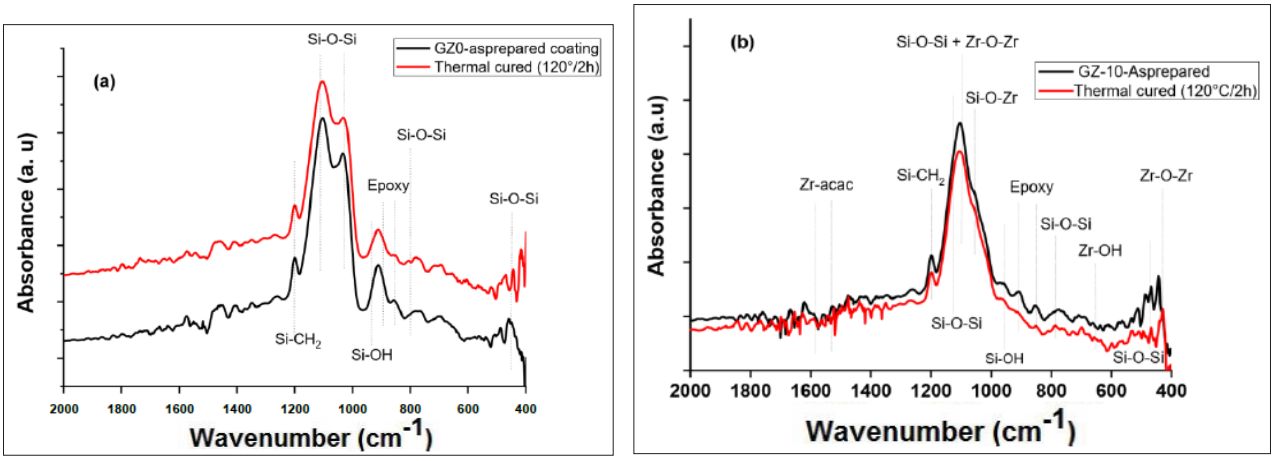


Figure 4: FTIR spectra of the coatings as prepared and heat treated at 120 °C for 2h. The coatings derived from (a) GZ-0 and (b) GZ-10 sol were deposited on both sides polished intrinsic Si wafer.

FESEM Study

For making good adhesion of coatings to the substrates, chemical treatment on MS and Al substrates with 6 wt% H_2SO_4 and N/50 NaOH solution, respectively were done before coating deposition. Figure 5 shows FESEM images of MS and Al surfaces before and after the treatment with H_2SO_4 and NaOH, respectively. After surface treatment, besides the formation of surface roughness higher percentage of oxygen (with respect to un-treated surface) was observed from EDX analysis, making excellent adhesion of coatings to the substrates as compared to un-treated MS and Al (Figure 1S, see Supplementary Information). Based on the FTIR and FESEM analysis of as prepared and cured coatings, a schematic representation of probable bonding mechanism between the nanocomposite coating and metal surfaces is shown in Scheme 1. By controlling sol composition, as for representative, we have achieved coating thickness to about 3.9 µm as shown in cross sectional FESEM image (Figure 6). Surface image also shows silica-zirconia NPs are embedded into the inorganic-organic hybrid matrix and responsible for developing hardness and surface protection. PDMS incorporated hydrophobic modified (GZH-10) coating was deposited for further enhancement of the corrosion resistant property.

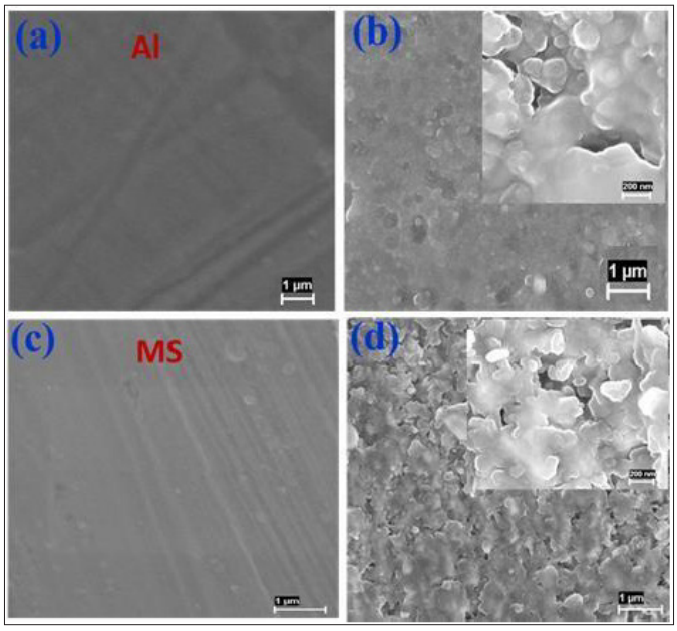
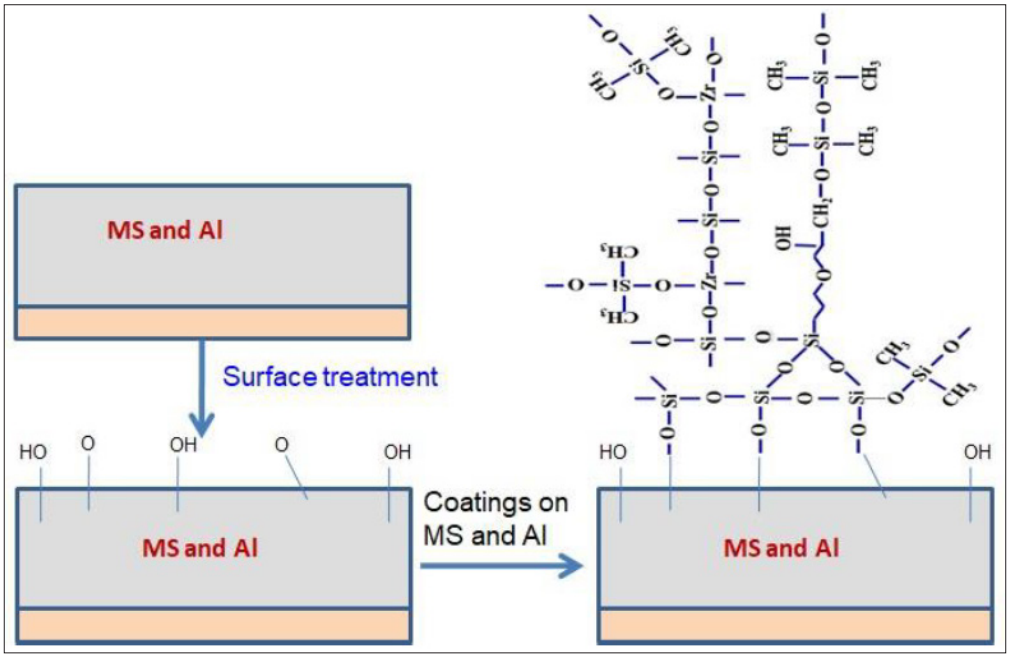


Figure 5: FESEM images of Al and MS surfaces before (a and c) and after (b and d) chemical treatment by N/50 NaOH and 6 wt% H_2SO_4 solution on Al and MS substrate, respectively.



Scheme 1: Schematic representation of probable bonding mechanism between the nanocomposite coating (derived from GZH-10) and metal surfaces.

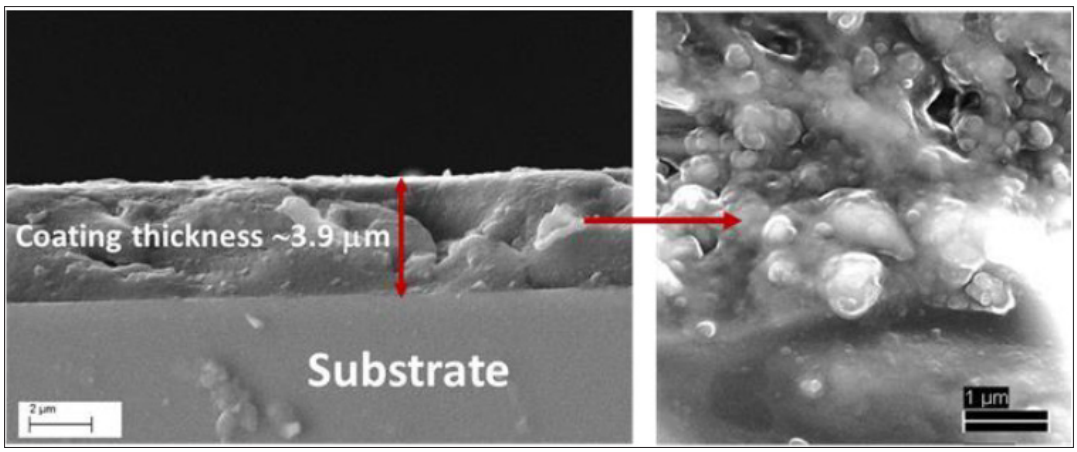


Figure 6: Cross sectional FESEM images of the GZ-10 coating along with coating surface in different magnification.

TEM Study

To characterize the coatings material TEM study was performed. TEM images with different magnifications and the corresponding electron diffraction (ED) patterns of the cured coating are shown in Figure 7. From the ED pattern it is very clear that the coating is amorphous in nature. TEM studies do not show any porous structure. For further confirmation its nonporous nature, the surface area/porosity of the scratched off hard coatings were done. The multi-point BET surface area of the coating was found to be extremely low (typically <0.9 m^2 g^{-1}) indicating nonporous characteristics of the coatings.

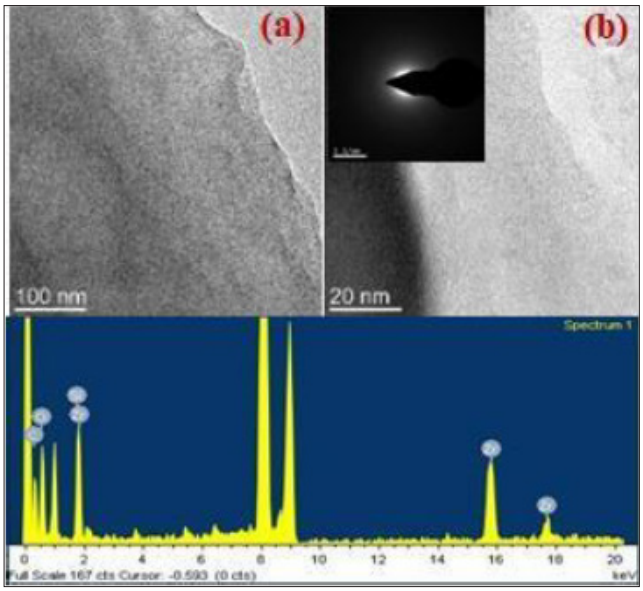


Figure 7: TEM images of the GZ-10 coating in different magnifications (a-b) along with EDS spectrum and the selected area diffraction pattern presented in the inset of Fig. b.

Evaluation of the Coatings

The thicknesses of the heat treated (120 °C/2h) coatings (Figure 12) were in the range of 3-4 µm (by changing withdrawal speed) having refractive index value of 1.48-1.535 ± 0.005 (Table 1). Coatings were done on MS and Al substrates up to a dimension of 130 x 130 mm^2 (Figure 8). The coatings passed the cross-cut adhesion test in accordance with the DIN 53151 or ASTM D 3359 specification which indicates formation of chemical linkage/bonding between coating materials with the substrate. The hardness of the coatings was ≥6H (ASTM D 3363). Static water contact angle (WCA) was measured to understand the hydrophilic characteristics of the coatings. WCA values (using 6 µL water drop) of the cured coatings (GZ- 0 to GZ-20) were in the range of 60 ±3° as shown in the following Figure 9. In case of coating on substrates derived from PDMS modified sol (GZH-10), WCA value of 108 ±2° was obtained. As for representative, water contact angle hysteresis (CAH) of coated MS with different WCA is shown in figure 10. Low CAH value (14 ±2°) of these coatings also indicates non stickiness and water repellence properties of the coatings. It was observed that WCA value of the coated MS and Al was unaltered after heating at 200°C/2 h.

Corrosion test of the coated and uncoated Al and MS substrates was carried by immersion in 3.5 wt% NaCl solution. Uncoated Al and MS showed severe damage starting from day one (Figure 2S, see Supplementary Information), whereas no damage was observed for all coated Al and MS substrates even after 20 days of immersion (Figure 11 & Figure 12). This indicates significant protection performance of these coatings. With increasing hydrophobicity (WCA 108 ±2°) (Figure 13) of the coating by incorporating PDMS (GZH-10) corrosion resistant was also increased to a certain extent (up to 25 days) but mechanical property related to the hardness also decreased (4H).

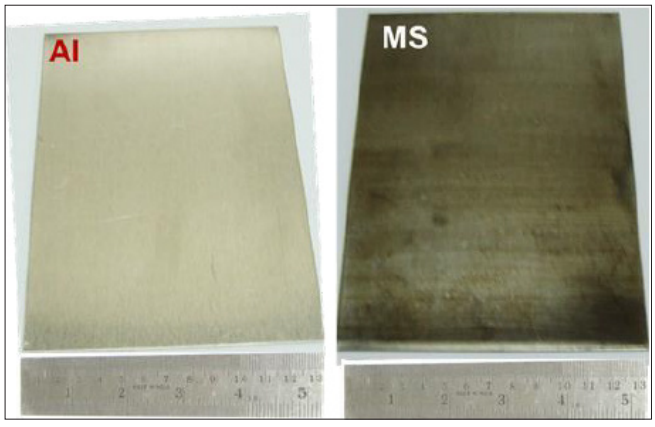


Figure 8: Photographs of GZ-10 coated Al and MS sheet (size: 130 mm x 130 mm).

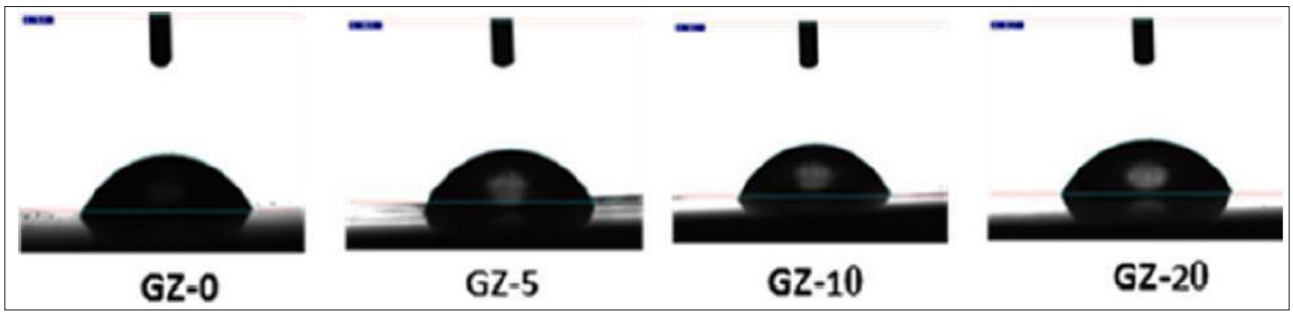


Figure 9: Water contact angles (WCA) of the heat-treated coatings derived from GZ-0 GZ-5, GZ-10 and GZ-20 sols on aluminium substrates.

Table 1: Properties of the thermal treated (120°C/2h) coatings derived from GZ-0, GZ-5, GZ- 10 and GZ-20 sols deposited on Al and MS substrates (sheet).

Name of the Test/ physical properties	Specifications	Result
Visual Appearance (sols)	-	Colourless for GZ-0 and yellowish for the rest.
Refractive index (ellipsometry)	-	1.48-1.535 ± 0.005 RI increases with ZP content in the sol: RI for GZ-0, GZ-5, GZ-10 and GZ-20 based coatings are 1.480, 1.4994, 1.524 and 1.535, respectively with an error bar of ± 0.005
Thickness	-	3-4 µm (thickness increases with withdrawal speed)
Adhesion	ASTM D3359 cross-cut adhesion	ASTM class 5B (highest standard)
Hardness	ASTM D3363 Pencil hardness	≥6H
Water contact angle (WCA)	6 µl water droplets sessile drop method	GZ-0, GZ-5, GZ-10 and GZ-20 coated samples show 60 ±3° and for GZH-10 coating is 108 ±2° with CAH 14 ± 1°
Corrosion test (GZ-10 and GZH- 10)	3.5 wt% NaCl solution	All coated samples can resist for more than 20 days, and it increases to 25 days for GZH10 coated samples

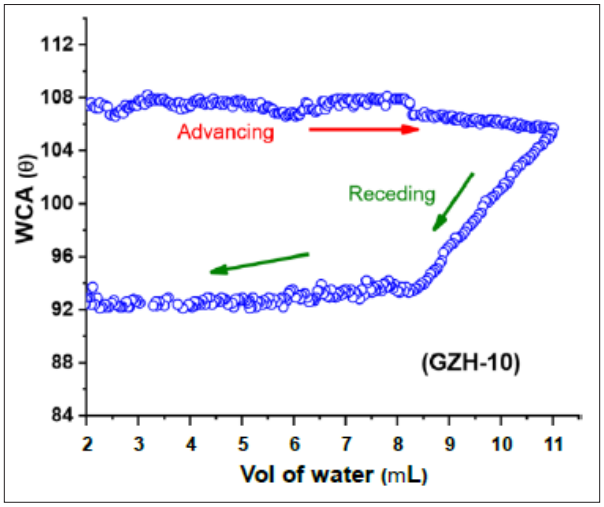


Figure 10: Plots showing water contact angle hysteresis of GZH-10 coated MS having coatings hardness 5H.

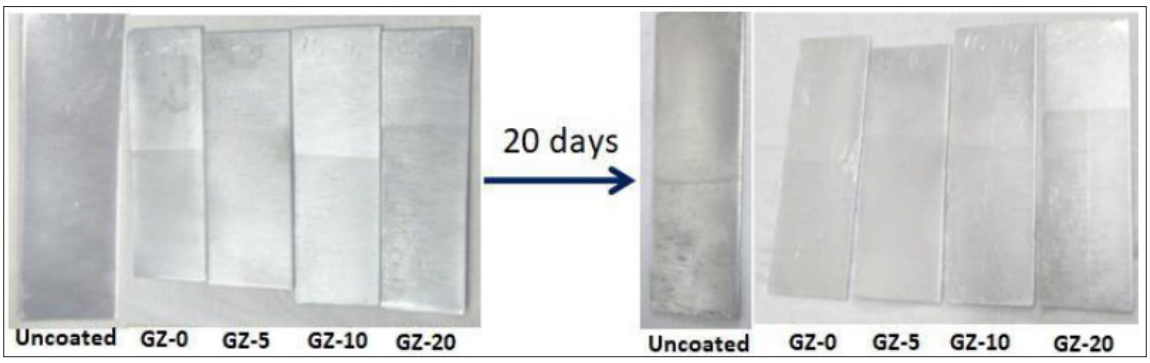


Figure 11: Digital images of uncoated and coated Al sheets (derived from GZ-0, GZ-5, GZ10 and GZ-20 sols) before and after 20 days kept in 3.5 wt% NaCl solution.

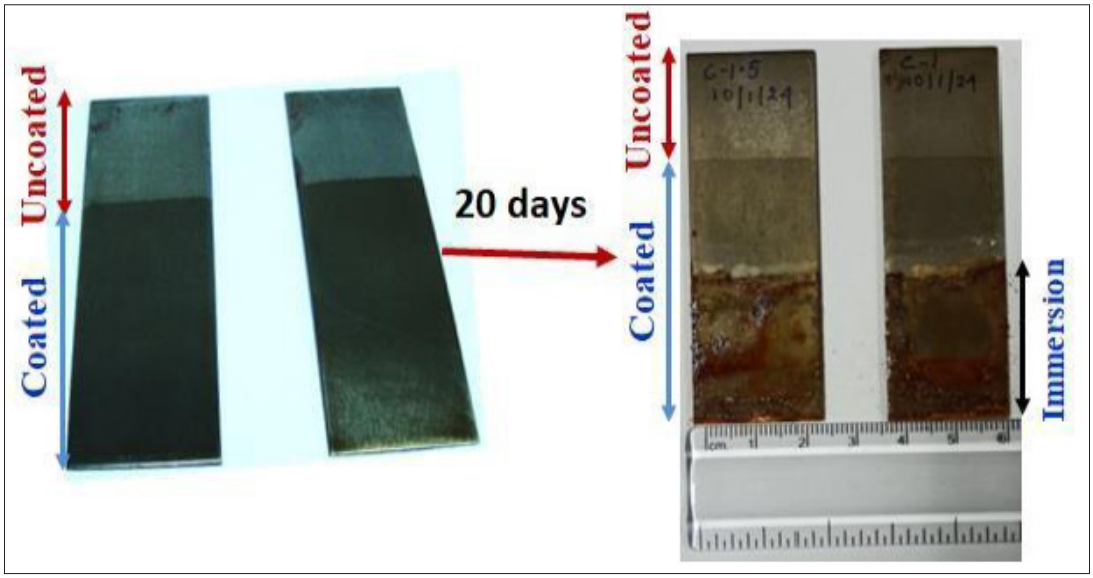


Figure 12: Photographs of GZ-10 coated MS after 20 days kept in 3.5 wt% NaCl solution (half of the coated part immersed).

Figure 13: Photographs of hydrophobic GZH-10 coated MS sheet (size: 100 mm x 100 mm).

Conclusions

'Polyethylene-oxide silica-zirconia' based corrosion resistant inorganic-organic hybrid nanocomposite coatings on Al and MS substrates (sheets) were developed by the sol-gel dipcoating technique. FTIR and ATR studies confirmed the Si-O-Si, Zr-O-Zr and inter-linked Si-O-Zr network formation during sol stage as well as in thermally cured condition. Coatings having RI 1.48-1.535 ± 0.005 and thickness of 3-4 µm were crack free, homogeneous and transparent. The hardness of the coatings was ≥6H (ASTM D 3363) with good adhesion to the substrates (ASTM D 3359; class 5B, highest standard). Uncoated Al and MS showed severe damage after day one following immersion in 3.5 wt% NaCl solution, whereas no damage was observed for all coated (using GZ-5 to GZ-20) Al and MS substrates even after 20 days. This indicates the boosted protection performance of these coatings. Static WCA of the heat treated (120 °C/2h) GZH-10 coatings was 108 ±2°, signifying hydrophobic character of the coatings. With increasing the hydrophobicity of coating corrosion resistance was also increased to a certain extent (up to 25 days) however, mechanical property decreased. Such heat stable corrosion protective hydrophobic metal surface can find different potential for domestic to industrial applications.

Associated Content

Supplementary Information: The Supplementary Information is available free of charge at https://unisciencepub.com/journal-of-materials-and-polymer-science/

Oxygen wt% of uncoated or bare MS and Al substrates before and after chemical treatment and partially coated MS substrates (a) and (b) kept at ambient condition for 1 and 2 months, respectively.

Author Contributions

Dr. Samar Kumar Medda: Investigation, conceptualization, methodology, visualization, writing – original draft and funding acquisition, Dr. Srikrishna Manna: Experimental, investigation, writing – review & editing, visualization. Pushpack Banerjee: Investigation, experiment and writing.

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